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Maximum limit values for selected hazardous organic contaminants (HOCs) in secondary raw materials used in fertilisers and soil products

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Preface

A revision of the Norwegian legislation regarding the quality of fertilisers and soil amendment products based on organic wastes (secondary raw materials) is in process. As part of this work, The Norwegian Environment Agency engaged COWI to suggest maximum limit values (MLs) for selected hazardous organic contaminants (HOCs) in fertilisers and soil improvers, and they were presented in 2018. As a following up of this work, The Norwegian Environment Agency has, in addition to developing MLs for selected HOCs, asked for more basic knowledge regarding risk evaluation of contaminants, and an updated overview of present concentration of the contaminants which is required in order to suggest limit values to protect environment and health.

The project has been conducted as a close cooperation between NIBIO (project lead), NIVA (Norwegian Institute for Water Research) and NILU (Norwegian Institute for Air Research): Trine Eggen (NIBIO) - project manager and responsible for approach and methodology for environmental and health risk assessments related to establishing MLs for HOCs in fertilisers and soil products, presence of selected HOCs in agricultural soil, as well as fate of HOCs in soil and during composting (Chapters 1, 2, 3, 5.1 and 9); Eldbjørg S. Heimstad and Vladimir Nikiforov (NILU) - responsible for evaluating half-lives (DT_{50}) and adjusting DT_{50} to realistic temperatures (Chapter 4); Christian Vogelsang (NIVA) - responsible for current levels of selected HOCs, potential impacts for the application of sewage sludge in agriculture, fate and transfer of HOCs in wastewater treatment plant, as well as measures to limit the content of selected HOCs (Chapters 5.2, 5.3, 6, 7, 8 and 10).

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Executive Summary

The aims of the project

As part of an ongoing revision of the Norwegian legislation regarding the quality of fertilisers and soil amendment products, the Norwegian Environment Agency wants to identify appropriate maximum limits (MLs) for a limited number of selected hazardous organic contaminants (HOCs) potentially pose a high risk in fertilisers and soil products. In 2018 COWI (Blytt et al., 2018), commissioned by the Norwegian Environment Agency, proposed MLs for selected HOCs. As a follow-up mission from the Agency, the main objectives of the present report have been to:

- i) Evaluate the approach and methodology that was used to develop MLs (Blytt et al., 2018),
- ii) If possible, revise the approach and methodology for developing MLs,
- iii) If revised methodology and necessary knowledge available, propose new MLs for selected HOCs in fertilisers and soil products,
- iv) Assess how the proposed MLs, if enforced, may influence the amount of sewage sludge that can be applied as fertiliser or in soil products,
- v) Describe how commonly applied wastewater and sewage sludge treatment processes at Norwegian wastewater treatment plants (WWTPs) may influence the fate and content of the selected HOCs in the final fertiliser and soil products, and how these treatment processes as well as alternative measures may be optimised to minimise the content of HOCs in the final sludge,
- vi) Collect and summarise knowledge for improving the knowledge background, and identify knowledge gaps.

Conclusions and recommendations

The proposed methodology and approach for developing MLs for organic HOCs in fertilisers and soil improvers (Blytt et al., 2018) is too simplified for the purpose. Reasons for this include selection of half-lives, use of the normative values for contaminated soil (a request by the Norwegian Environment Agency) as a soil quality acceptable for agricultural and growth media, and estimated MLs were for selected HOCs adjusted to higher limit values without any evaluation of possible negative impacts. The adjustments appeared to primarily be based on pragmatic considerations.

Use of half-lives, e.g. 360 days for persistent HOCs as PFOS, PFOA, PCB and deca-BDE, overestimate removal of these chemicals in soil. In this project, we propose to use more conservative half-lives which will prolong the presence of HOCs in the soil and environment significantly.

Enforcement of limit values of HOCs in fertilisers and soil improvers must prevent adverse consequences for the environment and for human health. It is uncertain if the health aspect is adequately assessed by using the established normative values established for contaminated ground and soil. This should be further evaluated. Use of approaches and methodologies that are more similar to those used by the The Norwegian Scientific Committee for Food and Environment (VKM) or The European Food Safety Authority (EFSA) could be possible alternatives. Particularly, the maximum levels of HOCs in growth media should be considered, as the HOC concentrations may be higher than in agricultural soil.

Based on values from Blytt and Stang (2019) and enforcement of the estimated and lowest (i.e. non-adjusted) MLs (Blytt et al., 2018), around 80% of the sewage sludge would have been disqualified from

use as fertilisers and soil improvers, while about 60% of the sludge would have been disqualified if the highest (i.e. adjusted) and the proposed introduced MLs had been enforced.

Under otherwise similar conditions, the soil concentrations of selected HOCs after application of sewage sludge were estimated using the same half-lives as Blytt et al. (2018) and the proposed conservative half-lives from this report. Based on the 95 percentile concentration of galaxolide and deca-BDE in sewage sludge, the soil concentrations 10 years after sludge application with use of the conservative half-lives were 10 times higher, and 6-8 times higher than the suggested accept levels in soil (Blytt et al., 2018). Using the half-lives presented by Blytt et al. (2018), the estimated soil concentrations would be within the accept levels within 2.5-3 years after application.

For some HOCs, there are precursors that might be present in similar or higher concentrations than the HOCs. This might lead to an increase of HOCs due to release from precursors during treatment processes and must be considered.

The presence of precursors to some of the HOCs (PFOA, PFOS, NP) may significantly increase the levels of these HOCs both during treatment and after application and should therefore also be taken into consideration.

The report describes how composting and commonly applied wastewater and sewage sludge treatment processes at Norwegian wastewater treatment plants (WWTPs) may influence the fate and content of the selected HOCs in the final fertiliser and soil products, and how these treatment processes as well as alternative measures may be optimised to minimise the content of HOCs in the final sludge. The HOCs being most rapidly reduced under aerobic conditions, often are more slowly reduced under anaerobic conditions, and vice versa.

Development of MLs for organic HOCs require extensive resources and relevant expertise, including expertise on risk assessment of human health. Within the scope of this project, it has not been possible to propose an alternative methodology for the development of limit values for organic pollutants in fertilizers.

In order to improve knowledge gaps for risk assessments and development of regulations following recommendations are given:

- obtain more experimentally based data for fate parameters (e.g. DT_{50} , BCF, Kd),
- analyse HOC's and important precursors (for e.g. PFOS/PFOA and NP/NPE) in fertilisers and soil products in order to build a solid knowledge of their presence and variation,
- perform a controlled field study where leaching, runoff and transfer to essential edible plants' processes are included and where the HOC applications are controlled and known,
- evaluate existing knowledge of hazard identification and characterisation of the priority HOCs towards humans and vulnerable sub-groups and identify which knowledge is lacking for performing a risk characterisation and develop MLs for growth media.

List of selected HOCs

The Norwegian Environment Agency selected the following compounds and compound groups to be included in the present evaluation:

- Diethylhexyl phthalate (DEHP)
- Perfluorinated octane sulfonate (PFOS)

- Perfluorinated octanoic carboxylic acid (PFOA)
- The fragrances galaxolide (HHCB), tonalide (AHTN) and OTNE
- Short-chain chlorinated paraffins (SCCP)
- Nonylphenol + nonylphenol ethoxylates (NP + NPE)
- Decabromo-diphenyl ether (BDE-209)
- Polychlorinated biphenyls (PCB₇)

Relevant regulations of HOCs in fertilisers and soil products

The sewage sludge directive in European Union (EC 86/278/) regulates application of sewage sludge to agricultural soil and includes limit values for certain potential toxic elements (Cd, Pb, Hg, Ni, Zn, Cu and Cr), but not HOCs. The Waste Framework Directive (2008/98/EC) introduce a new procedure for defining the end-of-waste criteria, where sewage sludge was not on the positive list for compost and digestate. European Parliament has approved new rules that harmonise standards for fertilisers from organic or recycled materials in the EU¹. Other relevant regulations regarding fertilisers and soil products are EC No 1069/2009 (animal by-products regulation) and EC No 1107/2009 COM/2016/0157 final - 2016/084 (COD). Sewage sludge is not included in these harmonised standards.

Some countries have implemented lower MLs than the EU directives, also MLs for selected HOCs, and some countries have restrictions for the use of organic based fertilisers and soil products, for instance, for growing crops for livestock or human consumption. Whether compost and digestate are regulated as a product or as waste, varies. Most EU member countries have similar regulations for digestate as for compost.

Comments to the approach and methodology used by COWI

On request by the Norwegian Environment Agency, COWI used the proposed new normative values for contaminated soil in Norway from 2016 as the basis for developing MLs for fertilisers. The following assumptions were made in order to perform a risk assessment and develop the MLs:

- fertilisers and soil products are evenly distributed in the top 20 cm of soil,
- relatively high O₂ content in soil,
- soil density 1.2 kg/l (mean soil density in Norway) and
- the amount of added fertiliser is the same as the maximum allowed use of sewage sludge category I, in the Norwegian fertiliser regulation (40 tonnes dry matter per hectare per 10 years).

The residual concentration in soil over time was calculated based on the following conditions:

- The initial soil concentration before fertiliser addition and the acceptable soil concentration were:
 - the proposed normative values (from 2016) added an uncertainty in the range of 58 – 104% due to sampling (50, 75 and 100%) and lab analysis (30%) which give the acceptable soil concentrations, or

¹ <https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1565858275007&uri=CELEX:32019R1009>

- soil concentration "zero" and the normative soil values were set as acceptable soil concentrations
- Half-life (DT₅₀) in soil for the selected HOCs were either based on experimental values (HHCB) or predicted using QSAR (Quantitative Structure Activity Relationships) with the software EPISUITE. All DT₅₀ values were based on temperature around or higher than 22°C.
- An assumed average soil loss of 200 kg per decare per year due to runoff.

Comment on half-life (DT₅₀) and suggested temperature corrections

Due to the Norwegian climate, it is important to use realistic half-lives, and whenever possible – based on experiments with application of real sludge – use the highest reported value in further considerations. Most experiments in the laboratory are carried out at +20 °C or +25 °C, or “at ambient temperature”, while typical soil temperature for Norway is much lower. We have selected four representative soil observation stations across Norway, the municipalities Ås, Frosta, Fauske and Tromsø (at approximately 60°, 63°, 66° and 69° North) and calculated that the average annual soil temperature at depth of 10 cm is in the range of +4.3°C in Holt, Tromsø to +7.4 °C in Ås, Oslo area (<http://lmt.nibio.no/>). There is no approved method to account for influence of temperature on degradation in soil, but we suggest using a simplified temperature correction factor, Q₁₀ = 2, which means that dissipation is assumed to be 2 times slower for every 10°C of temperature drop. This translates to 2.5 times (for Ås) or 3 times (for Tromsø) longer dissipation times (DT₅₀) than at + 20°C.

Based on a literature survey, and the temperature correction factor Q₁₀=2, new half-lives (DT₅₀) for the selected HOCs are proposed. They formed three groups: with DT₅₀ = 99 years (PFOS, PFOA, SCCP, BDE-209, PCB-7), 5 years (DEHP, HHCB, AHTN, OTNE) and 75 days (NP, NPE), respectively. The proposed DT₅₀ by COWI, max DT₅₀ reported in literature, and proposed temperature corrected DT₅₀, are shown in the table below.

Contaminant	DT ₅₀ COWI, (d)	Max DT ₅₀ in soil reported in the literature, (d)	Proposed DT ₅₀ , conservative, temperature corrected for Norway
DEHP	365	578	5 years
PFOS	360	Non degradable	99 years (Non degradable) ¹
PFOA	n.a.	Non degradable	99 years (Non degradable) ¹
HHCB	n.a.	1825	5 years
AHTN	239	1825	5 years
OTNE	120	1825	5 years
SCCP	n.a.	6800	99 years (Non degradable)
BDE-209	360	1440	99 years (Non degradable)
PCB 7	n.a.	Non degradable	99 years (Non degradable)
NP	75	25	75 days (same as COWI estimate) ¹
NPE	75	25	75 days

n.a. = not available. ¹ Presence of precursors in fertilizer, at levels much higher than those of regulated substance itself, must be evaluated.

Special attention needs to be paid to precursors of HOCs which possibly release and form the HOCs after application of fertilizer to soil. This can lead to a considerable increase of the HOCs in the soil. Such phenomena are known for NP/NPE and suspected for PFOA/PFOS. Also, BDE-209 calls for special attention due to formation of more toxic and bioaccumulative congeners. Release of HOCs from precursors are known to occur in wastewater treatment processes; for instance, recently reported for PFOS.

Comment on approach and methodology

A mass balance approach is used where added contaminants to soil, DT_{50} of HOCs in soil, and removal via soil loss are used to estimate soil concentration over time. Leaching is not included. Removal of soil via particles does not reduce the concentration in soil, only the amount.

The fate of contaminants, e.g. leaching and uptake by organisms which transfer contaminants to plants, groundwater and surface water, is related to their physiochemical properties, and should be part of an evaluation.

In the report by COWI, “use of fresh water toxicity as an approach to restrict contaminants in fertilisers” is questioned since the pathway (runoff and/or leaching) of contaminants may not be applicable for arable land that is far from surface water. It is difficult without further evaluation to know if this is a correct assumption.

Such a simplified approach should in our opinion only be used if it is known that water transport of contaminants to groundwater (drinking water), and nearby surface waters are insignificant environmental processes. In any case, to operate with different MLs depending on whether the arable soil is close to surface water or not, seems to be very impracticable. Thus, risk evaluation related to HOCs in agricultural soil and growth media should account for all possible exposure pathways.

The normative values are established for evaluation of contaminated ground, and not for risk evaluation of agricultural soil or growth media. It is therefore necessary to ensure that the normative values also protect farm animals and humans including vulnerable sub-populations and children eating soil, which is usually included in risk evaluations related to food and forage. The main transfer of organic contaminants to plants (food and forage) is via roots or as deposits on leaves. Any uptake to edible parts of the plants is usually estimated based on available bioconcentration factors (BCF) or transfer factors (TF). Typically, root vegetables, particularly carrot, show high uptake rates of hydrophobic compounds, while above-ground tissues such as leaves, fruits and seeds show higher uptake rates for hydrophilic compounds. Another point is the amount of sewage sludge added to growth media, which can be up to 30% of total volume, and could therefore theoretically contain higher concentrations of HOCs than in agricultural soil after application of 40 tonnes/ha/10 year. While regulations restrict which crops can be cultivated and how long after application on agricultural land they can be harvested, there are no regulations for what people cultivate in their own containers and kitchen gardens.

Both leaching and plant uptake are thus important transfer processes which in our opinion should be accounted for.

COWI did not consider Norwegian soil temperatures to be relevant in their approach due to generally high uncertainty. In our opinion it is necessary to use as realistic DT_{50} as possible and they should be conservative in order to be precautional and protective for the environment, farm animals and humans.

COWI proposed a set of MLs for selected HOCs (see Table below) based on calculations further described above. These MLs are addressed as non-adjusted MLs. In addition, an adjusted set of MLs were proposed, and the MLs for SCCP, HHCB, AHTN, PCB7 and NP + NPE (in red) were increased. These adjusted MLs are stated as more “acceptable” limit values than can be achieved in organic fertiliser products, and also more in line with what are established in the EU for selected compounds (Blytt et al., 2018). These adjusted MLs were proposed by COWI as MLs if implemented. Both the non-adjusted and adjusted MLs are shown in table below.

Contaminants	Non-adjusted ML values ¹ (mg/kg dw)	Adjusted ML values ² (mg/kg dw)
DEHP	50	50
PFOS	0.1	0.1
PFOA ³	0.1	0.1
SCCP	0.9	2
HHCB	0.5	10
AHTN	0.6	10
OTNE	n.s.	n.s.
BDE-209	0.5	0.5
PCB 7	0.004	0.02
NP + NPE	4	10

n.s.=not suggested. ¹Estimated MLs based of proposed normative values in soil (Blytt et al., 2018); ²Adjusted MLs (Blytt et al., 2018) and in attachment 6 in the tender; ³Proposed limit value for PFAS, not PFOS and PFOA.

It is common that single households, kindergartens, etc. eat vegetables cultivated in growth media/soil mixtures in kitchen gardens and various containers. A higher uptake of hydrophobic contaminants is expected in the edible parts of carrots and other root vegetables and in leaf vegetables such as lettuce than e.g. in cereals (e.g. Eggen et al., 2011, 2012). During the summer months, a portion of the population including children, might have a higher intake of self-produced vegetables containing elevated levels of contaminants. This is an exposure scenario that needs to be considered regarding establishing MLs for fertilisers for different use, and which may justify stricter regulation for HOCs in growth media/soil mixtures than in fertilisers used on agricultural soil. It is recommended that this is addressed and evaluated further.

Experimental or estimated half-lives need to be applied with caution in risk assessments. As a reasonable and conservative (i.e. precautionary) approach, the longest half-life should be used for estimations, until reliable experimental data is available.

Reliable and relevant experimentally-derived transfer factors should always be preferred to estimated factors. However, due to the presence of a large number of organic chemicals, it is important to develop and verify models for predicting the fate of organic contaminants that could represent different classes of contaminants.

Risk assessments need to be scientifically based and follow e.g. guideline documents from ECHA and the European Food Safety Authority (EFSA). It is recommended to perform a scientifically based risk assessment for developing MLs for HOCs in fertilisers and soil products. This has not been possible within the frame of this project.

Fate and transfer in the soil environment

The fate of the selected HOCs in the environment after application to soil are governed by a range of biotic and abiotic processes such as abiotic and biotic degradation, binding to particles and organic matter, humification, water transport via leaching and runoff, uptake of plants or soil organisms, and for volatiles also evaporation. Soil quality and properties, physicochemical properties of the contaminant, climate conditions like temperature, light, precipitation and humidity are all factors with impact on these processes.

Key parameters for evaluating environmental fate are half-lives (DT_{50}), removal rates, and binding and leaching capacity, commonly expressed by distribution and partition coefficients (e.g. K_d , K_{oc} , K_{ow}). Uptake via plant roots or deposits on leaves of organic contaminants are potential transfer and exposure pathways of contaminants towards domestic animals and humans. Uptake in plants, commonly expressed as a transfer factor (TF) or bioconcentration factor (BCF), contributes also as a removal process if plants are harvested and removed from the field. Transfer of contaminants to

nearby water bodies (groundwater, surface water including sediments) is important to include in risk assessments, and is predicted with use of e.g. K_d , annual precipitation, and infiltration rate.

Losses due to degradation can be predicted using compound-specific half-lives in soil (DT_{50}), while the water-mediated transport can be calculated by applying appropriate distribution/partition coefficients (i.e. K_d , P , D). It is important to use compound-specific properties and the properties of the local soil, as well as take local climate conditions into account in such assessments. For instance, the organic matter content of the soil is a major factor influencing the sorption rate and accumulation capacity for HOCs in soil, hence the organic carbon-normalized distribution coefficient (K_{oc}) is often used.

Transfer of the contaminants from soil to surface water must include leaching processes as well as particle transport. Transport of soil particles reduce the content of contaminants in soil but do not change the concentration of the contaminants in soil, PEC_{soil} . Leaching processes are highly dependent on the sorption properties (e.g. K_d , K_{ow}) for a given contaminant. Precipitation, soil properties (infiltration) and landscape topography (slope) will also influence the transfer of HOCs from soil to nearby water bodies.

Transfer of organic contaminants to edible plants (food and forage) is important to include in a risk evaluation related to agricultural soil and growth media. Application of realistic half-lives, distribution/partition coefficients and BCFs to obtain reliable risk assessment results is necessary.

HOCs with need for special attention

BDE-209 requires special attention due to transformation to more toxic and bioaccumulating congeners. Even though the transformation is not predicted to be fast, debromination of BDE-209 over time will occur, and that BDE-99 and BDE-47 will very likely be dominating. It is concluded that BDE-209 is a source for more toxic, lower brominated PBDEs which can produce combined toxicity. An evaluation of BDE-209 in isolation, without accounting for such combination effects, can significantly underestimate the toxicity of BDE-209. Arp et al. (2017) proposed an increase of the normative value for BDE-209 from 0.002 mg/kg dw (human health based) to 0.49 mg/kg dw (ecotoxicologically based) and divided by an extra AF of 100. Whether this substantial increase in normative value is scientifically based or not, is uncertain.

Special attention needs to be paid to precursors of HOCs, which possibly release and transform into HOCs following application of fertilizer and increase levels of HOCs in the soil. Such phenomena are known for NP/NPE and suspected for PFOA/PFOS.

Caution should be exercised when establishing normative limits/threshold-values for crop-producing soils, and to evaluate if these values are relevant for environmental, human and livestock health issues.

Current levels of the selected HOCs in Norwegian sewage sludge

Presence of selected HOCs in agricultural soil

In general, data of the present concentration of HOCs in agricultural soil is scarce, and even when field data are available, it is not possible to evaluate the fate of the HOCs in soil compared to the added concentration and amount of HOCs. Based on the data from a Swedish and a Czech study (published 2015 and 2019, respectively), it is particularly the HOCs with lowest proposed normative soil values (NGI) which have been measured in concentrations close to these values. It is recommended to perform field studies where the concentration of the HOCs in the applied sludge are known, and where leaching and runoff processes of HOCs are studied.

Observed levels in Norwegian sewage sludge

The expected variability in the levels of the selected HOCs in Norwegian sewage sludge are mostly based on the latest of the five-yearly national monitoring campaigns conducted at selected Norwegian WWTPs and sludge treatment facilities (from October 2017 to February 2018). The concentrations in stabilised and sanitised sludge varied considerably between plants, and between samples from the same facility. Some facilities showed relatively high values for some compounds, probably caused by identifiable local sources; e.g. DEHP and PCB7 in landfill leachates, PFOS in runoff from historic usage of PFOS in fire-fighting foams (e.g. at certain airports). The exceedances of the proposed ML and adjusted ML values are summarised in the table below. Although the levels of PFOS and PFOA are generally low, a wide range of potential precursors to PFOS and PFOA may be present in the same sludge. These levels and their potential transformation rates are largely unknown. Moreover, precursors to NP (e.g. NPnE, n=3-20) not included in the analyses may be present.

Compound	Exceedance of proposed ML values (# of samples)	
	Non-Adjusted ML values	Adjusted ML values
DEHP	10 of 95 (10.5%)	
PFOS	1 of 80 (1.3%)	
PFOA	0 of 80 (0%)	
SCCP	16 of 70 (23%)	3 of 70 (4.3%)
HHCB	70 of 70 (100%)	40 of 70 (57%)
AHTN	69 of 70 (99%)	0 of 70 (0%)
OTNE	6 of 6 (100%)	6 of 6 (100%)
BDE-209	19 of 95 (20%)	
PCBs	18 of 95 (19%)	78 of 95 (82%)
NP + NPE	1 of 95 (1.1%)	44 of 95 (46%)

Presence of the selected HOCs in digestate and compost end products

There is limited data on the content of the selected HOCs in compost, and the concentration ranges vary depending on initial concentrations and several process factors and substrate composition. Based on the summary performed in this report, the measured concentrations of the selected HOCs in compost were near or above the suggested adjusted MLs by COWI for NP and PCB7.

Much more data is available for HOCs in sludge digestate than in compost. As summarised in this report, the maximum concentrations measured in the Norwegian sewage sludge digestate are above the proposed adjusted MLs by COWI for all the selected HOCs except PFOA and AHNT.

Sampling and analytical issues

Sampling should generally be conducted in accordance with the guidelines for sampling of sewage sludge, compost and other waste-based fertiliser products (Norwegian Food Safety Authority). Some concerns related to sampling procedures are highlighted and commented in this report.

From an analytical point of view, sewage sludge is challenging because it is not homogenous in its composition. Thus, the concentrations of HOCs may vary widely between WWTPs and over time. Sewage sludge also contains components that may interfere with the analysis of the compounds of interest, making it critical to remove them from the sample extracts by established clean-up procedures. PFOS and PFOA are particularly challenging to analyse and certain precautions should be taken. There are also known and probably also unknown precursors to both PFOS and PFOA in sewage sludge that should be considered. Attempts to develop methods where all potential precursors are

converted to PFOS/PFOA prior to analysis have been used for water samples, but as far as we know, not for sludge.

Currently only the linear 4-NP is analysed at larger Norwegian WWTPs. When analysing NP, branched NPs should also be included.

The accuracy of the analytical method should take both the (proposed) threshold limit and the expected lower concentration range (proposed set at the 10-percentile) of the HOC into account. GC-MS is expected to give adequate LOQs for most of the compounds, possibly not for SCCP and deca-BDE. PFOS and PFOA is analysed by HPLC-MS/MS, but an LOQ of 0.1 µg/kg DS may be challenging due to often high blank contamination level.

Commercial labs are offering analyses of most of the compounds, possibly except for HHCB and AHTN. The costs for the other compounds are relatively high, especially for SCCP and deca-BDE. The costs will probably decrease somewhat if the commercial labs offer special packages for the specific sample matrix. If the market volume increases, the competition between laboratories will probably also bring the costs somewhat down. It is expected that a package cost for all 10 compounds listed will be in the order of 10.000,- NOK.

Potential impacts for the application of Norwegian sewage in agriculture from enforcing the proposed MLs

Almost 95% of the around 50 000 tons dry weight sludge from the 15 facilities participating in the monitoring campaign in 2017 was either used directly as soil conditioner on farmland (88%) or applied in soil products (>6%). This accounted for about 80% of all sludge used for these purposes in Norway that year.

The adjusted ML value for HHCB was exceeded in more than 50% (in dry weight per year) of the produced sewage sludge. Approximately 20% of the sewage sludge exceeded the adjusted ML value for BDE-209. For these two compounds the adjusted MLs were exceeded at many of the treatment facilities that participated in the sampling campaign; 8 of 12 (67%) for HHCB and 8 of 15 (53%) for BDE-209. Though limited data, the adjusted ML value for OTNE was exceeded in all samples (6) that were measured. Hence, there is a need to document the levels of OTNE in Norwegian sewage sludge.

The enforcement of the lower ML values (non-adjusted MLs) (i.e. for SCCP, HHCB, AHTN, PCN7 and NP+NPE) will probably drastically increase the volume of treated sewage sludge that do not pass the limit values. From the 2017 data, all (100%) the sewage sludge exceeded the non-adjusted MLs for HHCB, and almost all (97%) exceeded the non-adjusted MLs for AHTN even if none exceeded the adjusted ML value for AHTN. The ML value for PCB7 was exceeded at almost all the facilities (14 out of 15 plants) and approximately 80% of the total annual amount of sewage sludge. The ML value for NP + NPE was exceeded at 11 of the 15 facilities and amounted to about 30% of the annual sewage sludge.

Implementing the proposed adjusted ML on HHCB would have had the largest impact, denying an estimated 51% of the sludge from being applied either directly on farmland (46%) or in soil products (5%). If all proposed adjusted MLs were enforced, 60% of all the sludge disposed of from these 15 facilities in 2017 would have been disqualified from such use. If the proposed non-adjusted MLs had been enforced, about 80% of the sludge would have been disqualified from such applications that year.

PART II – MEASURES

Measures to limit the content of selected HOCs in final treated sewage sludge through process optimisation

The second part of this report discusses the potential pathway and fate of the selected HOCs from the incoming wastewater through the different treatment steps typically applied at Norwegian WWTPs and conventionally applied sludge treatment processes to the stabilised and dewatered sewage sludge. The main focus is on which treatment steps and conditions that may contribute to minimise the presence of these HOCs in the final products.

The selected HOCs are found at elevated levels in the final treated sewage sludge because;

- there are significant discharges of these HOCs and/or precursors to the wastewater,
- the HOCs sorb to the sludge,
- the loss by volatilisation are limited or not enough to bring the levels sufficiently low,
- the biotransformation occurring during the different wastewater and/or sludge treatment steps are not adequate or efficient enough to bring the levels sufficiently low.

From a strategic point of view, the first and preferred choice of abating HOCs in sewage sludge is to minimise potential sources, i.e. implementing control measures upstream of the WWTP such as regulations on industry, production, import, use and disposal. However, such measures are not a part of the scope of this work.

The second bullet point in the list above – sorption to sludge – is crucial in minimising direct discharges of HOCs to the aquatic environment with the treated effluent. It may be possible, in theory, to (partially) desorb the HOCs from particles and resorb them to an adsorbent with excellent and specific adsorption characteristics and thereby isolate the HOCs before they enter the sludge treatment stage. However, this may have negative effects on the efficiencies of the subsequent wastewater treatment steps, and it may not be feasible from an economic point of view. It will probably be a better option to desorb the HOCs within the sludge line and subsequent resorb the HOC to an adsorbent or apply other types of treatment (see below).

Extensive (bio)transformation to non-toxic and non-bioaccumulating compounds or, more ideally, complete mineralisation is in many ways the preferred mechanism to minimise the content of these HOCs that are present in the wastewater entering the WWTP, since this will minimise production of hazardous waste.

Enhanced evaporation and subsequent sorption/condensation to isolate and supplementary treatment to transform/destroy the more volatile HOCs (i.e. HHCB, AHTN, OTNE and NP) may also be possible.

Removal of selected HOCs through optimised conditions for biotransformation

Optimisation of the potential biotransformations of the HOCs by selecting appropriate treatment processes and conditions is key to reduce the overall amount of HOCs. For many of the compounds (DEHP, SCCP, NP and NPE) anaerobic conditions limit the biotransformation while aerobic conditions promote more extensive and faster biotransformation. Dehalogenation of highly halogenated compounds (e.g. deca-BDE and PCBs) occurs during anaerobic conditions, while further decomposition occurs under aerobic conditions. Similarly, 4-NP tends to accumulate under anaerobic conditions due to partial degradation of NP1EO and NP2EO. However, the strong sorption behaviour

to activated sludge may limit the bioavailability of HOCs that otherwise might be more efficiently biotransformed. This sorption probably also limited the extent of volatilisation of the musk fragrances (HHCB, AHTN and OTNE) during the wastewater treatment. However, if thermal drying of the final sludge is applied this may change considerably, possibly also for NP and NPE. There are many factors apart from applying aerobic or anaerobic conditions that may strongly influence the rate of biotransformation of some of these compounds (e.g. SRT, HRT and temperature), since these factors may impact the composition and structure of the microbial community and the likelihood that some substances will be used as e.g. co-substrates. So far there is very limited documentation of the actual effects that these factors have on the transformation rates of the selected HOCs.

Note that the organic matter content is typically reduced by 50% during anaerobic digestion, hence the concentration of non-degradable compounds (e.g. most of the selected HOCs and heavy metals) will increase correspondingly.

Composting is an aerobic microbial processes which might be relatively effective for the removal of several HOCs such as DEHP, HHCB and OTNE (range of 50-80/90% observed), but also AHNT (25-70%), NP and NPE (60-70%) are removed to some degree. However, the fate of the most persistent HOCs such as PCB7, PFOS, PFOA and BDE-209 during composting, need more documentation.

Measures that harvest the inherent resources while isolating or destroying the selected HOCs

Inherent resources in Norwegian sewage sludge are already today harvested as energy (through biogas production in anaerobic digestion processes), soil conditioner (by applying stabilised and hygienised sewage sludge on e.g. farmland or in soil mixes) and nutrients such as P and N (also by applying stabilised and hygienised sewage sludge on farmland and in soil mixes). The efficiency of the latter is up for discussion, as much of chemically precipitated P appears to have limited availability for plant uptake (Krogstad et al., 2005) and may therefore be leaching to the aquatic recipients instead where it may contribute to eutrophication.

A strict regulation of the content of the selected HOCs in the finally applied sewage sludge may make it necessary and more attractive to find other ways of harvesting these resources (and potentially also others). The last decade or so, a wide range of different processes and technologies have been developed and tested to optimise this type of harvesting.

Precipitation of phosphate salts and derivatives

Phosphate salts can be recovered from different stages along the wastewater and/or sewage sludge treatment lines, but most commonly from the sludge liquor (reject water after dewatering) and from digested sludge. Struvite, the most commonly recovered phosphate salt, forms from equimolar quantities of Mg^{2+} , PO_4^{3-} and NH_4^+ , implying that the efficiency of NH_4^+ removal is relatively low, and the excess N remains in soluble form. In most sewage treatment applications Mg is the limiting element and is therefore added to the process as $MgCl_2$ or MgO . Several processes have been developed and are already operating at full scale in other countries.

HOCs may co-precipitate, typically adsorbed to organic matter. Hence, the organic carbon can be a vector for these HOCs but is thereby also a good proxy for the purity of the precipitated phosphate salt. The STRUBIAS project, organised by the European Sustainable Phosphorous Platform (ESPP), propose a limit value of 3% organic C in the precipitated phosphate salts, which indicate a reduction of one order of magnitude in organic C relative to unprocessed manure and sewage sludge. They argue

that this also “effectively excludes the extensive and expensive testing for a broad range of organic contaminants (e.g. pharmaceutical compounds and personal care products, phthalates, surfactants, etc.), and strengthens market confidence in fertilising materials recovered from biogenic wastes in times of increased concerns about emerging organic contaminants in consumer products and the food chain”.

Thermal oxidation followed by post-extraction of P

These are treatments that combust the organic matter with excess oxygen at high temperatures (800-950°C) for a few seconds in a boiler creating flue-gases containing the majority of the available fuel energy as heat. The phosphorous is extracted from the ashes by wet-chemical processes or thermal processes. Combustion under non-oxygen limiting conditions to low levels of organic C (<3%) in the ashes is a well-demonstrated technique for effective removal and thermal destruction of a broad range of HOCs. The temperatures typically applied in incineration processes are generally sufficient for the destruction of PFOS and PFOA, probably also their precursors. PCB levels are generally low in the ash fraction.

Pyrolysis and gasification

These processes (hydrothermal carbonification or wet pyrolysis, dry pyrolysis and gasification) involve heat treatment under oxygen-limited conditions producing C-rich (>50%) biochar or mineral-rich pyrogenic carbonaceous materials depending on the C-content of the feedstock. The biochar may be applied for soil amendment and as a fertiliser if nutrient rich. The knowledge base regarding the proportional removal of specific organic pollutants is limited and primarily restricted to a few organic pollutants. Importantly, generation of PCDD/F and PCBs may occur throughout the whole operating temperature range (300-750°C) if chlorine is present and their adsorption can be favoured by the presence of elemental carbon and soot particles. Based on the precautionary principle, the STRUBIAS project, concludes that it is justified to exclude highly contaminated feedstocks (e.g. sewage sludge, municipal solid waste and hazardous waste) from the positive input material list to ensure human health and environmental safety. The positive material list of feedstocks to wet and dry pyrolysis processes includes plant-based materials, bio-waste and certain animal by-products (e.g. bone material and manure).

Norsk Sammendrag (kort)

Klima- og miljødepartementet og Landbruks- og matdepartement ga 27. juni 2016

Landbruksdirektoratet, Miljødirektoratet og Mattilsynet i oppdrag å revidere forskrift om gjødselvarer mv. av organisk opphav. I oppdraget ble det lagt vekt på tilrettelegging for økt ressursutnyttelse av restmaterialer i gjødselvarer og at nyttiggjøringen skjer på måter som minimerer forurensning til vann, jord og luft.

I gjødselvarerforskriften er det satt grenseverdier for innhold av tungmetaller i gjødselvarer, men ikke for innhold av organiske miljøgifter. Da gjødselvarer kan inneholde organiske miljøgifter er det behov for å vurdere grenseverdier også for utvalgte organiske miljøgifter. I forbindelse med revidering av gjødselvarerforskriften, har COWI på oppdrag av Miljødirektoratet utarbeidet et forslag til en metode for beregning av grenser for maksimalt tillatt innhold av organiske miljøgifter i gjødselvarer (maximum level, ML), samt foreslått ML for utvalgte miljøgifter (Blytt et al., 2018).

NIBIO (prosjektleder), NIVA og NILU har sammen hatt et oppdrag hvor hovedmålet har vært å:

- i) evaluere tilnærmingen og metodikken som ble brukt for utvikling av forslag til MLs,
- ii) hvis mulig revidere metodikken,
- iii) hvis tilstrekkelig kunnskap og revidert metodikk, også utvikle og foreslå nye MLs for utvalgte miljøgifter,
- iv) vurdere konsekvensene ved innføring av foreslåtte MLs i gjødselvarer i forhold til bruk av avløps slam,
- v) beskrive hvordan ulike behandlingsmetoder og prosesser påvirker skjebnen til de utvalgte miljøgiftene og deres innhold i gjødselvarer, og
- vi) oppdatere bakgrunnskunnskap og mangel på kunnskap.

Oppdraget var begrenset til: DEHP (di-(2-etylheksyl)ftalat), PFOS og PFOA (to stoffer i PFAS-gruppen), SCCP (kortkjedete klorparafiner), galaxolid, tonalid og OTNE (Makromusker), deka-BDE (BDE-209, dekabromodifenyleter), PCB (polyklorerte bifenyler, PCB7) og NP og NPE (nonylfenol og nonylfenoletoksilater).

Prosjektgruppen er av den oppfatning at en bør anvende en mer vitenskapelig tilnærming for utvikling av grenseverdier av miljøgifter i gjødselvarer, enn det som ligger til grunn til de foreslåtte grenseverdiene (Blytt et al., 2018). Følgende momenter er viktig i den vurderingen:

- I tilnærmingen som ble brukt i 2018 (Blytt et al., 2018), var normverdier i jord anvendt som kriterier for god jordkvalitet for dyrking av mat og fôr. Normverdien i jord er etablert som en grenseverdi som forteller om grunnen kan ha en forurensningsrisiko eller ikke. Selv om det i normverdien ligger inne en vurdering av human helse, tar ikke de bakenforliggende beregningene tilfredsstillende hensyn til alle elementene som er nødvendige i risikovurderinger av overføring av miljøgifter fra jord/miljø til mat og fôr, jf vitenskapskomiteen for mat og miljø, VKM, og den europeiske myndighet for næringsmiddeltrygghet (EFSA).
- Halveringstiden (DT_{50}) er en av de viktigste parameterne når det gjelder risikovurdering av organiske miljøgifter i miljøet. DT_{50} har for enkelte stoffer blitt estimert basert på EPISUITE, en QSAR tilnærming hvor lengste DT_{50} er 360 dager. Dette er langt lavere enn hva som er reelt i jord for persistente miljøgifter som for eksempel PCB, PFOS, PFOA og BDE-209. De valgte DT_{50} var heller ikke justert i forhold til at de er etablert for nedbrytning ved 22 – 25 °C, noe som overestimert nedbrytning av miljøgiftene under norske og nordiske forhold. Ved litteraturgjennomgang og ved bruk av en temperatur-koeffisient faktor $Q_{10}=2$, (dvs 2 ganger

langsommere nedbrytning per 10 °C temperatur-reduksjon), har vi foreslått mer konservative og realistiske DT₅₀ for de utvalgte miljøgiftene (se tabell).

Stoff	DT ₅₀ COWI, (d)	Maks DT ₅₀ i jord gitt i litteraturen (d)	Foreslått konservativ og temperaturjustert DT ₅₀ (d eller år) for Norge
DEHP	365	578	5 år
PFOS	360	Ikke nedbrytbar	99 år (Ikke nedbrytbar) ¹
PFOA	–	Ikke nedbrytbar	99 år (Ikke nedbrytbar) ¹
HHCB	–	1825	5 år
AHTN	239	1825	5 år
OTNE	120	1825	5 år
SCCP	–	6800	99 år (Ikke nedbrytbar)
BDE-209	360	1440	99 år (Ikke nedbrytbar)
PCB 7	–	Ikke nedbrytbar	99 år (Ikke nedbrytbar)
NP	75	25	75 år ¹
NPE	75	25	75 år

--: ikke oppgitt, ¹Tilstedeværelse av forløpere til miljøgifter.

- Forløpere til miljøgifter kan foreligge i konsentrasjoner langt over den foreslåtte regulerte miljøgiften, og slike miljøgifter krever spesiell oppmerksomhet. Forløperne kan omdannes til de foreslåtte regulerte miljøgiftene (gjelder for eksempel PFOS, PFOA, NP/NPE). Det er også kjent at BDE-209 kan omdannes til de mer toksiske og bioakkumulerbare kongenerene BDE-99 og BDE-49, og det må også tas hensyn til i vurdering av etablering av grenseverdier i gjødselvarer.
- Grenseverdiene som ble beregnet ved bruk av valgt metodikk, ble for enkelte miljøgifter (HCCB, ATHN, SCCP, PCB7, NP og NPE), oppjustert til høyere grenseverdier (omtalt som justert ML). Denne oppjusteringen anses som pragmatisk fundert og ikke risikobasert.
- Det er indikert i rapporten fra COWI at det for gjødselvarer brukt som vekstmedia kan ha en høyere grenseverdier («Quality class for soil products») for utvalgte miljøgifter enn gjødselvarer brukt i jordbruket. Det anbefales en nærmere vurdering av grenseverdier for vekstmedier.
- Betydningen av en eventuell regulering av bruken av avløpsslam i landbruket ut fra de foreslåtte grenseverdiene for de utvalgte miljøgiftene er vurdert med utgangspunkt i resultatene fra den siste nasjonale overvåkingen av avløpsslam (Blytt og Stang, 2019). Slam fra alle de 15 slambehandlingsanleggene overskred en eller flere av de ikke-justerte grenseverdiene i minst ett tilfelle. Resultatene antydte at innholdet av HHCB, AHTN og PCB7 var de forbindelsene som i størst grad ville gjort at slammet ikke kunne blitt benyttet direkte i landbruket; hhv. 71%, 68% og 69% av slammet ville presumptivt ha blitt nektet anvendelse. Hvis alle de ikke-justerte grenseverdiene ble innført, antydte estimatene at i størrelsesorden 80% av det stabiliserte og hygieniserte avløpsslammet ville kunne blitt nektet direkte anvendelse i landbruket eller i jordprodukter. Hvis de justerte grenseverdiene ble lagt til grunn, var det innholdet av HHCB som alene hadde absolutt størst samlet betydning for overskridelsene av grenseverdiene, estimert til 51% av slammet. Hvis alle de justerte grenseverdiene ble innført, antydte estimatene at i størrelsesorden 60% av det stabiliserte og hygieniserte avløpsslammet ville kunne blitt nektet direkte anvendelse i landbruket eller i jordprodukter.

Konklusjoner og anbefalinger:

Metodikken og tilnærmingen for utvikling av grenseverdier for organiske miljøgifter i gjødselvarer som er foreslått (Blytt et al., 2018), er for forenklet til at det er godt egnet til dette formålet. Årsaken til det er blant annet valget av halveringstider, bruk av normverdien for forurenset grunn og jord (gitt av Miljødirektoratet i oppdraget til COWI) som basis for jordkvaliteten for landbruksjord og vekstmedier, og at grenseverdiene som ble beregnet, ble for enkelte miljøgifter oppjustert til høyere grenseverdier. Denne oppjusteringen anses som pragmatisk fundert og ikke risikobasert.

Halveringstider på 360 dager for miljøgifter som PFOS, PFOA, PCB og deka-BDE, vil overestimerer hvor raskt persistente miljøgifter reduseres i jord. I dette prosjektet foreslår vi bruk av mer konservative halveringstider, og det vil forlenge oppholdstiden for miljøgifter i miljøet betydelig.

Innføring av grenseverdier av miljøgifter i gjødselvarer skal hindre at bruk av resirkulert næringsstoffer og organisk materiale gir negative konsekvenser for miljø og helse. Det er usikkert hvorvidt helseaspektet er tilstrekkelig ivaretatt ved bruk av normverdiene for forurenset grunn, og det må vurderes nærmere. Tilnærminger og metodikk som er mer lik de som benyttes av vitenskapskomiteen for mat og miljø (VKM) og EFSA bør følges. Dette er beskrevet i rapporten. Spesielt, bør grenseverdier for organiske miljøgifter i vekstmedier vurderes nærmere ettersom konsentrasjonen kan teoretisk være høyere enn i jord, og det er ingen restriksjoner på hva som dyrkes i egne kjøkkenhager og dyrkningskasser.

Estimater basert på tall fra Blytt og Stang (2019), vil innføring av de laveste grenseverdiene (Blytt et al., 2018), føre til at omkring 80% av det stabiliserte og hygieniserte avløpsslammet i Norge ikke vil kunne anvendes direkte i landbruket eller i jordprodukter. Innføring av de oppjusterte og foreslått brukte grenseverdiene (Blytt et al., 2018), vil føre til at i omkring 60% av det stabiliserte og hygieniserte avløpsslammet ikke vil kunne anvendes direkte i jord.

Ved ellers like forhold, er halveringstider brukt av COWI og de konservative foreslåtte halveringstidene i dette prosjektet, anvendt til å estimere konsentrasjon i jord 10 år etter tilførsel av avløpsslam. Med bruk av 95% persentil innhold av galaxolid og deka-BDE i slam, er estimert konsentrasjonen i jord ved bruk av de konservative halveringstider mer enn 10 ganger høyere enn ved bruk av de tidligere foreslåtte halveringstidene. Ved å sammenligne disse konsentrasjonen med de foreslåtte akseptverdiene i jord (Blytt et al., 2018), gir de ikke-konservative halveringstidene 10 år etter tilført avløpsslam en estimert jordkonsentrasjon lavere enn akseptverdiene allerede etter 2,5-3 år. Estimert jordkonsentrasjon etter 10 år med bruk av de konservative halveringstidene, var 6-8 ganger høyere enn akseptverdien.

For noen miljøgifter kan konsentrasjonen av forløpere til miljøgiftene være minst like høy som miljøgiftene. Dette vil kunne føre til en økning av konsentrasjonen av miljøgifter ved at forløperne omdannes og frigjør enkelte foreslått regulerte miljøgifter, og dette må tas med i vurderingen.

I rapporten beskriver hvordan kompostering og ofte anvendte renseprosesser for avløpsvann og avløpsslam ved norske avløpsanlegg kan påvirke skjebnen og innholdet til de valgte miljøgiftene i sluttproduktene, og hvordan disse behandlingsprosessene samt alternative tiltak, kan optimaliseres for å minimere konsentrasjonene. De miljøgiftene som raskest omdannes under aerobe forhold omdannes gjerne langt langsommere under anaerobe forhold, og motsatt.

Sammenstilt kunnskap og mangel på kunnskap, vil komme til nytte for utvikling av grenseverdier med en vitenskapelig forankring.

Utvikling av grenseverdier krever omfattende ressurser og en faglig bredde, blant annet ekspertise innen risikovurdering av human helse. Det var ikke mulig innen rammene av dette prosjektet å foreslå alternativ metodikk for utvikling av grenseverdier for organiske miljøgifter i gjødselvarer.

Forslag til videre arbeide for å øke kunnskapsgrunnlaget for gjennomføring av risikovurderinger og regelverk-utvikling:

- Skaffe til veie mer eksperimentelle data for de viktigste parameterne til bruk i risikovurderinger og utvikling av grenseverdier (for eksempel DT_{50} , BCF, Kd),
- Analysere for utvalgte organiske miljøgifter og viktige forløpere (for eksempel PFOS, PFOA, NP, NPE) i relevant gjødselvarer for bedre kunnskap om tilstedeværelse og variasjon,
- Opprett et feltforsøk med kontrollert tilførsel av utvalgte organiske miljøgifter for å frambringe eksperimentelle data om utlekking, avrenning og overføring til viktige fôr- og matplanter, og
- Gjennomgå eksisterende kunnskap om risikoidentifisering og -karakterisering i forhold til human helse, inkludert sårbare grupper, for prioriterte organiske miljøgifter, for å identifisere mangel på nødvendig kunnskap for utvikling av grenseverdier for vekstmedier. Dette kan gjøres som en separat vurdering.

Abbreviations and explanations

ABP	Regulation (EC) No 1069/2009 of the European Parliament and of the Council of 21 October 2009 regulates management of animal by-products and derived products not intended for human consumption
Adjusted MLs vs non-adjusted MLs	COWI (Blytt et al., 2018) proposed a set of MLs for selected HOCs (see Table below) based on calculations which is further described above. These MLs are addressed as non-adjusted MLs. In addition, an adjusted set of MLs were proposed, and where the MLs for SCCP, HHCb, AHTN, PCB7 and NP + NPE (in red) were increased. These adjusted MLs are stated as more “acceptable” limit values than can be achieved in organic fertiliser products, and also more in line with what are established in EU for selected compounds.
AHTN	Tonalide (musk compound)
BDE-209 (or deka-BDE)	Decabromo diphenyl ether
BCF	Bioconcentration Factor. Concentration in organisms, C_o (e.g. plants, given in e.g. mg/kg) over concentration in the environment, Environment (e.g. soil given as e.g mg/kg or porewater given as mg/L). BCF and BSAF are used a bit differently. Regarding ECAH 2017, BCF is related to concentration in pore water and given as L/kg, and BSAF a dimensionless factor where C_s includes both pore water and soil. For expression of uptake of contaminants in plants, BCF is normally used and expressed as concentration in plant, C_o , over concentration in soil given as e.g. mg/kg.
BAF	Bioaccumulation from soil factor – a biota-to-soil accumulation factor. Concentration in organisms, C_o (e.g. plants or earthworm given as mg/kg) over concentration in soil given as mg/kg, C_s .
Biosolids	Sewage sludge after hygienisation
C/D	Commonly used for compost and digestate
CEPT	Chemically Enhanced Primary Treatment
CLP	European Regulation (EC) No 1272/2008 on classification, labelling and packaging of substances and mixtures came into force on 20 January 2009 in all EU Member States, including the UK. It is known by its abbreviated form, ‘the CLP Regulation’ or just plain ‘CLP’
Da	Decare = 1000 m ²
DEHP	Diethylhexyl phthalate
Digestate	The product from anaerobic treatment (anaerobic digestion) of organic waste in a biogas plant
dw	Dry weight
DT ₅₀	Half-lives
ECB	European Chemicals Bureau
EC	European Community

ECHA	European Chemicals Agency
ECHA RAC	European Chemicals Agency Committee for Risk Assessment
EFSA	European Food Safety Authority
EOF	Extractable OrganoFluorine
EUSUS	The European Union System for Evaluation of Substances
EQS	Environmental Quality Standards
FW	Fresh weight
Ha	Hectare = 10 000 m ²
HC5	The fifth percentile, with 50% confidence, of a species sensitivity distribution
HCCB	Galaxolide (musk compound)
HRT	Hydraulic Retention Time
K _d	Soil adsorption coefficient; measure the concentration of the chemical in soil over the concentration in water.
K _{oc}	Organic carbon - water partition coefficient. It is the K _d normalized to total organic carbon content. $K_{oc} = K_d * 100 / \% OC$
P (K _{OW}) and D	Partition coefficient (P) and coefficient (D) refers to distribution of a chemical between water and octanol (most common). P generally refers to the non-ionized species of the chemical, while D generally refers to the concentration ratio of all species (non-ionized plus the ionized).
LOEC	Lowest Observed Effect Concentration
MAC-EQS	Maximum allowable concentrations
MEC	Measured environmental concentration
ML	Maximum limit value
MBBR	Moving bed biological reactor
MBR	Membrane bioreactor
NOEC	No Observed Effect Concentration
NP / NPE	nonyl phenol / nonyl phenol ethoxylates
NP1E	Nonylphenol monoethoxylate
NP2E	Nonylphenol diethoxylate
OC	Organic carbon
OTNE	Octahydrotetramethyl Acetophenone Iso E Super® (musk compound)
PBT	Persistent Bioaccumulative Toxic
PCB	Polychlorinated biphenyls
PFCAs	Perfluoroalkyl carboxylic acids
PFASs	Per- and polyfluoroalkyl substances

PFSAs	Perfluoroalkyl sulfonic acids
PNEC	Predicted No Effect Concentration
PFOA	Perfluorinated octanoic carboxylic acid
PFOS	Perfluorinated octane sulfonate
Priority list	A list established by the Norwegian Environment Agency of the substances that pose a serious threat to environment or health, and the goal is to eliminate or substantially reduce the emissions
PTE	Potential Toxic Elements
QS _{soil}	Quality standard for soil and is a guideline value for limit concentration of contaminants in soil
RAR	Risk Assessment Report
REACH	European regulation of chemicals i.e., Registration, Evaluation, Authorisation and Restriction of Chemicals, enacted in 2007
RMOA	Regulatory management option analysis
SCCP	Short-chain chlorinated paraffins (C10-13)
SOM	Soil Organic Matter, also often expressed only as organic matter (OM)
SRT	Sludge Residence Time
TDI	Tolerable daily intake
TOC	Total Organic Carbon
TF	Transfer factor
VKM	Vitenskapskomiteen for mat og miljø (The Norwegian Scientific Committee for Food and Environment)
WWTP	Waste water treatment plant
WHO	World Health Organization

1 Introduction

There is an obvious need for recirculation of resources in order to create a sustainable society, which also includes recycling nutrients and organic matter back to soil. Historically, recirculation of organic matter and nutrients was primarily linked to manure and compost from garden and green waste (“clean sources”). This has changed, and utilisation of sewage sludge as soil improver and fertiliser has become a common practice, both applied on arable land and as part of growth media. Due to the increasing number of fertiliser and soil-improver products containing materials with potentially hazardous organic contaminants (HOCs), the focus on maximum limits (MLs) for HOCs of such products has increased. In the European Union the use of sewage sludge on agricultural land is governed by European Sewage Sludge Directive 86/278/EC (CEC, 1986) that determines the concentration limits of selected PTEs. Several countries e.g. Denmark, Sweden, Ireland, Germany and France, have introduced a stricter regulation than the Directive. Some countries have also adopted stricter concentration criteria for some persistent organic pollutants, such as polychlorinated biphenyls (PCBs) and other halogenated organic compounds. But there is a long list of other organic contaminants present in e.g. sewage sludge that is not regulated. Compared to some decades ago, when the primary focus in sewage sludge was potential toxic elements (PTEs) such as Cd, Hg, Pb, Cu, Zn and Cr, polychlorinated biphenyls (PCB) and polycyclic aromatic hydrocarbons (PAHs), there is now a wide range of potentially hazardous substances which might have a negative impact on the environment and/or on human health. Many emerging organic compounds in the environment have their origin from household and personal care products or pharmaceuticals, and many might enter agricultural soil via biosolids or other organic-based fertilisers and soil products (Clarke and Smith, 2011; Mohapatra et al., 2016; Semblante et al., 2018). Both national and international commitments and regulations to stimulate recycling of bioresources free for contaminants are in process, which reinforces the need for MLs for HOCs.

Transfer of contaminants from agriculture to water is also an issue. For instance, the EU Water Framework Directive has pointed out the need to strengthen efforts against adverse effect from agriculture in order to reach the goal of improved water quality within 2021. The Norwegian Parliament supports this goal and that runoff from the agriculture is a priority (Parliamentary White Paper No 11 (2016-2017) “Change and development. An agriculture for the future”).

The Norwegian list of contaminants to be prioritized in a national effort to stop emissions contains approximately 35 compounds and groups of compounds (per 2017)². The list is based on a set of criteria that prioritises the contaminants with highest risk. This includes compounds that are persistent and bioaccumulative, have serious long-time effects such as cancer, effects on reproduction and genetics or strongly toxic to the environment. Compounds which affect biological processes in very low concentration, e.g. hormone interfering compounds are also priority contaminants.

A revision of the Norwegian legislation regarding the quality of fertilisers and soil amendment products based on organic wastes (secondary raw materials) is in process. As part of this work, the Norwegian Environment Agency engaged COWI to suggest MLs values for selected HOCs in fertilisers and soil products, and they were presented in 2018 (Blytt et al., 2018). To follow up this work, the Norwegian Environment Agency engaged NIBIO (Norwegian Institute of Bioeconomy) as project leader, NIVA (Norwegian Institute for Water Research) and NILU (Norwegian Institute for Air Research) to conduct an updated overview of present concentration of the contaminants in materials that may be used in agriculture. Updated information was used to consider revised limit values (MLs) to protect environment and health.

²

1.1 The aims of the project

The two main objectives of this project have been i) to evaluate and improve basic knowledge concerning the protection of the environment and health from selected organic contaminants, and ii) to suggest new limit values for the contaminants selected by the Norwegian Environment Agency if necessary and possible.

Secondary objectives in the project have been:

1. to evaluate the method and approach utilized by COWI for establishing limit values in fertilisers and, if possible, propose adjustments to the methodology (search for properties and other relevant issues)
2. to evaluate and make an overview of how different processes and treatment technologies influence the fate and content of organic contaminants (with focus on the HOCs selected by the Norwegian Environment Agency) in sewage sludge,
3. to describe how different environmental processes influence the fate of organic contaminants and environmental and health effects,
4. to search for information and, if enough information available, evaluate the fate of HOCs in other organic fertiliser matrixes than WWT-plant sludge,
5. to update present knowledge and evaluate how limit values influence on the amount of sewage sludge which can be applied as fertiliser,
6. to evaluate analytical cost and how limit values of selected organic contaminants can influence the cost
7. summarise knowledge gaps which are required to close to better predict fate and risk assessment

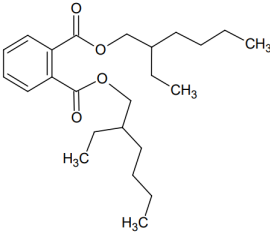
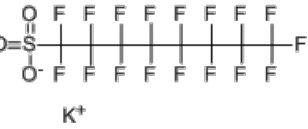

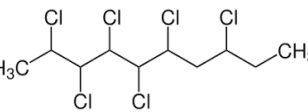
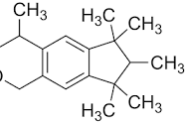
It is urgent that contaminants with high risk are not introduced into the environment, where they can cause irreversible damage on the environment and health.

1.2 The selected hazardous organic contaminants (HOCs)

The contaminants selected by the Norwegian Environment Agency to be included in this evaluation are:

- Diethylhexyl phthalate (DEHP)
 - Perfluorinated octane sulfonate (PFOS)
 - Perfluorinated octanoic carboxylic acid (PFOA)
 - Galaxolide (HHCB) (*not on the priority list*)
 - Tonalide (AHTN) (*not on the priority list*)
 - Short-chain chlorinated paraffins (SCCP)
 - Tetramethyl acetyloctahydronaphthalenes, Octahydrotetramethyl Acetophenone, Iso E Super® (OTNE) (*not on the priority list*)
 - Nonylphenol + nonyl phenoethoxylates (NP + NPE)
 - Decabromo-diphenyl ether (BDE-209)
 - Polychlorinated biphenyls (PCB₇)
- Information and physiochemical properties for the selected organic contaminants are shown in Table 1 and Table 2.

Table 1. Information and structure of the selected HOCs included in this project.

Compound	Name	Abbrev.	CAS No.	Compound type	Chemical formula	Structure
Bis(2-ethylhexyl) 1,2-benzenedicarboxylate	Diethylhexyl phthalate	DEHP	117-81-7	Phthalate	C ₂₄ H ₃₈ O ₄	
1-Octanesulfonic acid, 1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-heptafluoro	Perfluorinated octane sulfonate	PFOS	2795-39-3	Synthetic perfluorinated compound	C ₈ F ₁₇ SO ₃	
	Perfluorinated octanoic carboxylic acid	PFOA	335-67-1	Synthetic perfluorinated compound	C ₈ HF ₁₅ O ₂	
	Short-chain chlorinated paraffins (C10-13)	SCCP	85535-84-8	Chloroparaffins		Mixture of chlorinated alkanes with various degree of chlorination, e.g. 2,3,4,5,6,8-hexachlorodecane 
1,3,4,6,7,8-hexahydro-4,6,6,7,8,8,-hexamethylcyclopenta[g]benzopyran	Galaxolide	HHCB	1222-05-5	Polycyclic musk	C ₁₈ H ₂₆ O	

Compound	Name	Abbrev.	CAS No.	Compound type	Chemical formula	Structure
6-Acetyl-1,1,2,4,4,7-hexamethyltetralin	Tonalide	AHTN	1506-02-1	Polycyclic musk	C ₁₈ H ₂₆ O	
(1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethylnaphthalen-2yl)ethan-1-one	Octahydrotetramethyl Acetophenone Iso E Super®	OTNE	54464-57-2 (68311-19-3)	Synthetic ketone	C ₁₆ H ₂₆ O	
	Decabromo-diphenyl ether	Deka-BDE, BDE-209	1163-19-5	brominated diphenyl ether	C ₁₂ Br ₁₀ O	
	Polychlorinated biphenyls	PCBs	1336-36-3 etc.	poly chlorinated biphenyls	Mixture of chlorinated biphenyls	
	nonylphenol	NP	25154-52-3 104-40-5 4-n-Nonyl phenol	Surfactant	C ₁₅ H ₂₄ O	 Several isomers
Polyethylene glycol nonylphenyl ether	nonyl phenol ethoxylates	NPE	9016-45-9		(C ₂ -H ₄ -O) mult-C ₁₅ -H ₂₄ -O	

Table 2. Physicochemical properties of the selected HOCs. Except for OTNE, values are taken from NGI report on updated background data and proposals for new normative values for contaminated soil.

Compound	Log Kow	K _D (l/kg)	K _{OC} (l/kg)	C _{water sol.} (mg/L)	Henry law constant (H _K) (-)
DEHP	7.5 ⁽¹⁾	1650 ⁽¹⁾	165000 ⁽¹⁾	0.27 ⁽⁶⁾	4.4E-05 ⁽¹⁾
PFOS	0.1 ⁽¹⁾	10 ⁽¹⁾	1000 ⁽¹⁾	1.01e-3 ⁽⁷⁾	3.2E-08 ⁽¹⁾
PFOA	4.3 ⁽¹⁾	1.25 ⁽¹⁾	125 ⁽¹⁾	1.56e-2 ⁽⁷⁾	
SCCP	6 ⁽¹⁾	1995 ⁽¹⁾	199526 ⁽¹⁾		
HHCB (Galaxolide)	5.4 ⁽¹⁾	87 ⁽¹⁾	8710 ⁽¹⁾	1.75 ⁽⁶⁾	1.5E-02 ⁽¹⁾
AHTN (Tonalide)	5.3 ⁽¹⁾	245 ⁽¹⁾	24547 ⁽¹⁾	1.25 ⁽⁶⁾	1.5E-02
OTNE	5.6-5.7 ⁽²⁾ 5.28 ⁽³⁾		5460 ⁽⁴⁾ 3327-9681 ⁽⁵⁾	1.60e-4 ⁽⁸⁾	2.42 E-04 atm·m ³ /mol ⁽⁴⁾ 4.7 E-04 atm·m ³ /mol ⁽⁵⁾
PBDE-209	9.9 ⁽¹⁾	5.25E+9 ⁽¹⁾	5.25E+11 ⁽¹⁾	0.0001 ⁽⁶⁾	4.9E-07 ⁽¹⁾
PCB7	5.7 ⁽¹⁾	3211 ⁽¹⁾	321119 ⁽¹⁾		3.4E-04 ⁽¹⁾
NP+NPE	4.48 ⁽¹⁾	53.6 ⁽¹⁾	5360 ⁽¹⁾	7 ⁽⁶⁾ (NP)	4.0E-03 ⁽¹⁾

¹Arp et al. (2017); ²Klaschka et al., 2013; ³Konieczny et al., 2018; ⁴EPA Chemistry Dashboard, predicted, ⁵EPISUITE, predicted, ⁶EPISUITE experimental data, ⁷EPA Chemistry Dashboard, Experimental, ⁸EPA Chemistry Dashboard, predicted

1.3 Maximum limit (ML) proposed by COWI

On assignment from Norwegian Environment Agency, COWI has proposed MLs for organic pollutants in fertiliser based on organic waste (Blytt et al., 2018). COWI was asked by the Agency to use the proposed soil normative values (also called quality standards or guidelines) (Table 3) in the report from the Norwegian Geological Institute, NGI, Arp et al. (2017), as the basis for the work. Based on the soil normative values, COWI proposed a set of ML values, which for SCCP, HHCB, AHTN, PCB7, NP and NPE were adjusted to “acceptable” values which also would be more in line with established MLs in EU for selected compounds (both shown in Table 4). The adjusted ML values were the limit values proposed as implemented by COWI. Other publications also question the technical basis for the EU limit values and request re-evaluation for some cases (Rigby et al., 2015).

The soil normative values, first introduced by Norwegian Environment Agency in 1999 (TA-1629/1999), are meant to be used as guidelines for soil quality criteria for preliminary assessment of pollution in soils and as an assessment for handling of contaminated areas. The proposed normative values are derived based on the methodology described elsewhere (TA-1629/199; Arp et al., 2017) and illustrated in Figure 1.

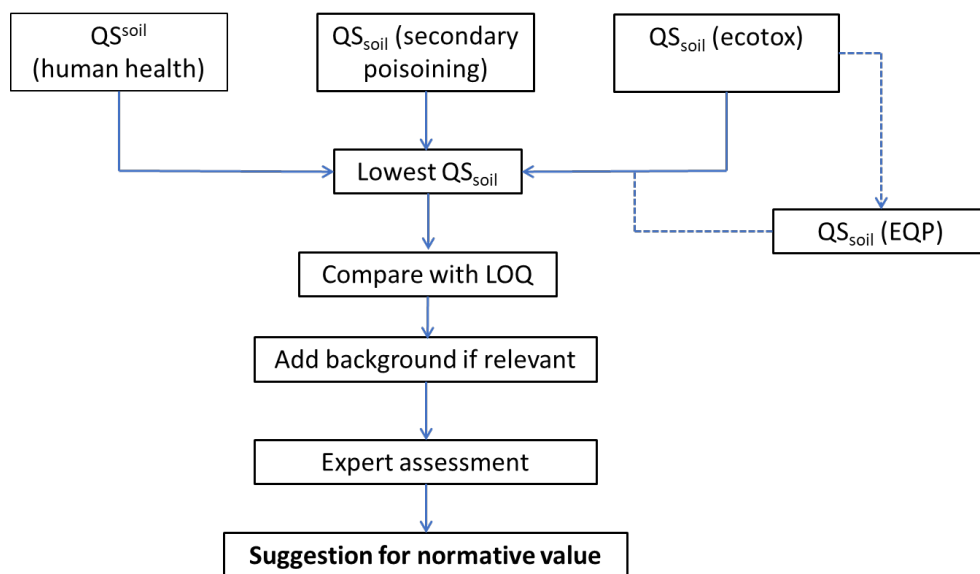


Figure 1. Illustration showing the evaluation scheme for suggestion of normative values (from Arp et al., 2017).

As described by Arp et al., 2017 (2017):

“the first step in deriving a normative value for soil is to derive three types of quality standard soil concentrations (QSsoil). The first is protective of human health QSsoil(human health), the second protective of predators in the terrestrial foodweb QSsoil(secondary poisoning), which is unique to this report compared to previous reports, and the third is protective of soil ecology, and is based on soil ecotox studies QSsoil(ecotox). In addition a QSsoil based on the equilibrium partitioning method QSsoil(EQP) is calculated. The QSsoil(EQP) derivation is based on the assumption that interspecies Predicted No Effect Concentration (PNEC) for chronic toxicity for freshwater organisms, such as the Annual Average-Environmental Quality Standard (AA-EQS) value in the Water Framework Directive (2013/39 EU), can be related to the PNEC for chronic toxicity in soil dwelling organisms via a soil-water partition coefficient, KD. Generally and historically in Norway, QSsoil(ecotox) are favoured over QSsoil(EQP) for deriving soil normative values, as QSsoil(ecotox) more directly apply to soil dwelling organisms. In this report, it was considered important to derive both QSsoil(ecotox) and QSsoil(EQP) to note cases where QSsoil(ecotox) >> QSsoil (EQP). Although no ecological effects are observed in soil, there may be situations where the soil is a contaminant source to nearby freshwater recipients. This most typically occurs for polar substances, such as PFOS and triclosane. Therefore, for some of these polar substances, the QSsoil (EQP) was recommended as the QSsoil value to be considered for the normative value, even if a QSsoil(ecotox) was available. This approach is particularly favoured for environmentally persistent substances, as they will not degrade substantially during transport from the contaminated soil to the water recipient.

After all QSsoil values are derived, the lowest value is compared with the current commercial limit of quantification (LOQ). In this review, this explicitly refers to the limit of quantification by current Norwegian commercial laboratories, and not limits of quantification by cutting edge techniques (e.g. in universities), as most site managers and consultants would not have access to state-of-the-art laboratories.”

The normative values are then assigned based on an expert assessment considering the lowest QSsoil, LOQ, current soil concentration (current/background soil concentration, geogenic concentration, e.g.

PTEs, are added to the lowest QS_{soil} or the LOQ) and compounds specific considerations (e.g. formation of transformation products, unique vulnerabilities to ecosystem or human population in Norway, management consideration). If specific considerations are used, these should be well documented and argued for in order to allow for transparency.

Further description and discussion of the methodology used for suggesting MLs is found in section 3.

In Norway, the proposed normative values for PFOS in soil from 2016 is 0.002 mg/kg dw (Table 3), and Sweden has recently introduced a normative value for PFOS in soil at 0.003 mg/kg dw (Naturvårdsverket, 2019). It is proposed that the Norwegian normative value should be adjusted to 0.01 mg/kg dw³, however, it is not a strong evidence that normative value should be raised from 0.002 to 0.01 mg/kg dw. PFOS is one of the HOCs identified with high concern regarding both environmental and human hazard.

Table 3. Proposed (2016) and current (2007/2011) normative values in soil for selected organic contaminants, given as mg/kg dw (Arp et al., 2017). n.p.=not proposed. Values in red are proposed an increase and values in bold are proposed a decrease.

Contaminants	Normative values (mg/kg dw)		Values are based on
	Current 2007/2011	Proposed 2016	
DEHP	2.8	1	Current: EQP Proposed: LOD (> QS for human health).
PFOS	0.1	0.002 (0.01) ³	Current: Proposed: LOQ and EQP based to protect water recipients ¹
PFOA		0.011	LOQ and EQP based to protect water recipients ²
SCCP	1	0.09	Current: EQP Proposed: Sec. Poisoning based
HHCB		0.01	Sec. Poisoning based
AHTN		0.011	Sec. Poisoning based
OTNE	n.p.	n.p.	
BDE-209	0.002	0.49	Current: Human health based Proposed: Ecotox based (soil(ecotox)-divided by an extra AF of 100 ³)
PCB7	0.01	0.004	Current: EQP based Proposed: 7 x LOQ for individual components ⁴
NP + NPE	0.005	0.035	Current: LOD and ecotox based Proposed: Sec. Poisoning based ⁵

¹PFOS: LOQ and EQP based to protect water recipients as this is a highly persistent substance (use ecotox value of 0.023 mg/kg if emissions to water recipients is not relevant) (table in report) Secondary pois. (in excel file). LOQ >EQP from Blytt et al., 2018,

²PFOA: EQP based to protect water recipients as this is an extremely persistent substance (use ecotox value of 0.13 mg/kg if emissions to water recipients is not relevant); ³To accounts for degradation products which are more toxic than BDE-209; ⁴Few data available, 7 x LOQ for individual components (note that the LOQ is greater than the secondary poisoning based value). LOQ >secondary poisoning (Blytt et al., 2018), ⁵NP+NPE: LOQ >secondary poisoning (Blytt et al., 2018).

³ Suggested by NIBIO in a note from NIBIO "Bistand med rådgiving ved forskriftsfesting av nye normverdier for forurenset jord. 27.01.2019.»

Table 4. Proposed MLs (both non-adjusted¹ and adjusted²) in fertilisers and soil products, given as mg/kg dw (Blytt et al., 2018). n.p.=not proposed. Values in red are higher after adjusted MLs.

Contaminants	Non-adjusted ML values ¹ (mg/kg dw)	Adjusted ML values ² (mg/kg dw)
DEHP	50	50
PFOS ³	0.1	0.1
PFOA ³	0.1	0.1
SCCP	0.9	2
HHCB	0.5	10
AHTN	0.6	10
OTNE	n.p.	n.p.
BDE-209	0.5	0.5
PCB 7	0.004	0.02
NP + NPE	4	10

¹Estimated MLs based of proposed normative values in soil (Blytt et al., 2018); ²Adjusted MLs (Blytt et al., 2018) and in attachment 6 in the tender; ³Proposed limit value for PFAS, not PFOS and PFOA.

1.4 National and international regulations of fertiliser and soil amendment products

1.4.1 Regulation of organic based fertiliser products in Norway

Current Norwegian regulation on organic based fertilisers/soil amendments ([FOR 2003-07-04-951](#)) ([Forskrift om gjødselvarer mv. av organisk opphav](#)), divides these products into four quality classes based on the concentration level of the same seven PTEs that are included in the EU sewage sludge regulative; Cd, Pb, Hg, Ni, Zn, Cu and Cr. Even though there are no MLs for organic contaminants the end user and the waste industry have a duty to follow the precautionary principle and to prevent application of products with potential risk for adverse environmental and health effects.

A revision of the regulation is under process and is planned to also include limits for selected organic contaminants. In the revision it is proposed to have one regulation for production, sale and import of organic based and certain inorganic fertiliser products⁴ and one regulation for storage and use of organic based and inorganic fertilisers product⁵.

As most organic pollutants don't naturally occur in soils, this differs from PTEs which have a normal background range in soils. Some organic pollutants are spread by atmospheric deposition and may be found in soils in natural ecosystems far from the sources of pollution.

4

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1.4.2 Relevant EU regulations

Sewage sludge directive in EU (86/278/EEC)

This EU directive regulates application of sewage sludge to agricultural soil, and include limit values for the elements Cd, Pb, Hg, Ni, Zn, Cu and Cr, and not content of organic contaminants.

The animal by-product regulation (EC No 1069/2009) (abbr. ABP) regulates management of animal by-products and derived products not intended for human consumption. This regulation sets controls on the safe use and disposal of animal-by products to safeguard public and animal health. Such animal by-products may be used as substrate for biogas production but the digestate must comply with the ABP regulation and restrictions related to hygienic and health. Such digestate has restrictions related to use on food and forage production and on grazing areas.

The regulation of organic farming production (Commission regulation EC No 889/2008) has criteria which prohibits sewage sludge, compost or digestate mixed with sewage sludge or other organic waste from mixed municipal solid waste to be used as fertiliser or soil improvements in organic agriculture.

As described by Blytt et al., (2018a), “**The Waste Framework Directive (2008/98/EC)** introduce a new procedure for defining the end-of-waste criteria. The Joint Research Centre's Institute for Prospective Technological Studies (JRC-IPTS) has prepared reports with technical information for supporting their proposal for the end-of-waste criteria for biodegradable waste subject to biological treatment (IPTS, 2013). Regarding restrictions on input material (category 2), municipal waste water treatment sludge, food waste and manure (solid and liquid) were included in the positive list for compost and digestate in the first version of the report regarding the EU end-of-waste framework (IPTS, 2013). However, this list was revised for the final report where it was decided to exclude wastewater treatment sludge and any organic fraction that is separated from mixed waste and not source separated at an EU level, while allowing the existing regulation at a national level for now (IPTS, 2013).”

The European Parliament has approved new harmonised standards for fertilisers from organic or recycled materials in the EU (EU 2019/1009), Regulations EC No 1069/2009 (animal by-products regulation) and EC No 1107/2009 COM/2016/0157 final - 2016/084 (COD). Sewage sludge is not part of these harmonised standards. The diverging national rules have made it difficult to sell and use fertilising products made from organic or secondary raw materials across the EU single market. The new rules will facilitate the market access of fertilisers from recycled or organic materials. The rules focus on PTEs, and particularly limits for cadmium, but include also limits for the organic contaminants PAH and dioxin PCDD/F.

1.4.3 Individual member state regulations

Some countries have implemented lower MLs than the EU directives, also MLs for selected HOCs, and some countries have restrictions on the use of organic based fertilisers and soil products, for instance, for growing crops for livestock or human.

Quality standards and MLs are based on toxicity of the contaminants. There are exceptions, e.g. for PCBs where the aim is to get the substances out of circulation due to their hazardous effects and high risk for serious adverse effect on environment and health. It is reasonable to question if this list should contain more HOCs, or if some HOCs have such high risk that it should not be accepted that they are introduced to the environment via fertilisers and soil products.

Compost and digestate: The regulations for agricultural application of organic fertilisers vary in the EU countries. The most applied and regulated ones are compost and digestate-based organic waste

and sewage sludge. While some countries have strict regulations on input materials (e.g. only source separated organic waste from households) and without further regulation of contaminants, other countries have less strict regulation on input materials and regulate the end products through MLs for some HOCs. Other countries have restrictions on both input and have MLs for the end products (e.g. Denmark and Germany) (Table 5). Whether compost is regulated as a product or waste varies between countries. In Sweden, Denmark, Finland, Germany and United Kingdom, compost is regulated as waste. There are also differences in regulations depending on which input materials it is based on (e.g. sewage sludge, digestate or other sources) (Blytt et al., 2018). Most countries have a positive list over which waste fractions that are permitted to be used as substrates for fertiliser products (Appendix A).

Digestate is extensively used as a fertiliser and soil improver and most EU countries regulate this under their waste or fertiliser laws with similar regulation as for compost.

Sewage sludge: Some countries have more strict regulation of fertilisers and soil products based on sewage sludge than the EU sludge regulative; both related to hygienic aspects and content of contaminants (PTEs and HOCs).

Table 5 summarises limit values, or proposed limit values, of selected HOCs in compost, digestate or sewage sludge for use as fertiliser or soil improvements (Miljøstyrelsen, 2018; Blytt et al., 2018). The United Kingdom and Northern Ireland have developed quality protocols for digestate, which includes criteria for when it ceases to be waste and becomes a product. Sweden has developed a voluntary certification system (CPSR120, owned and operated by Avfall Sverige, the Swedish Waste Management Association), where biogas plants can obtain certification when their digestate meets certain quality criteria (positive list for input materials, disease control and regulation of PTEs and visible impurities such as plastic).

Table 5. MLs, or proposed, MLs for HOCs in sewage sludge, compost and digestate in different countries, given as mg/kg dw (from Miljøstyrelsen, 2018; Blytt et al., 2018 and 2018b).

Contaminants	Sweden	Denmark	Germany	Proposed End-of-Waste ⁴
DEHP		50 ⁴		
PFOS	0.07-0.05-0.02 ^{1*}		0.1 ¹	
PFOA			0.1 ¹	
PFC			0.1 ¹	0.1 ³
SCCP ^{**}	4 ^{1*}			
HHCB			10 ^{5*}	
AHTN			15 ^{5*}	
OTNE				
BDE-209	0.7-0.5-0.5 ^{1*}			
PCB 7	0.4 ⁵ ; 0.06-0.05-0.04 ^{1*}	0.4 ² /0.08 ⁴	0.2 ⁴ ; 0.2 ⁵ /0.1 ^{5*}	0.2 ³
NP + NPE	50 ⁵	10 ^{4,5}		

¹Proposed MLs; ^{**}Given for chlorparaffins; ¹Miljøstyrelsen, 2018. Proposed MLs for 2015-2023-2030; ²indicative, in Blytt et al., 2018; ³Proposed end-of-waste criteria for compost and digestate, Blytt et al., 2018, IPTS, 2013, where PCF is sum of PFOA and PFOS, and PCB7 is sum PCB7; ⁴MLs in compost in different EU member states, Denmark PCB₇ is guideline values, Germany PCB₆, Denmark NPE, in Blytt et al., 2018; ⁵MLs in sewage sludge for agricultural application in different EU member states, Germany 2007 proposed PCB limits for each congener, Sweden PCB₇ 0.4 sum of PCB7 in Blytt et al., 2018.

Important considerations

Regulation of organic-based fertilisers and soil products are necessary in order to obtain a sustainable recirculation of resources without spreading compounds with potential hazard to environment, animals and humans.

Existing quality standards and MLs are mostly based on toxicity of the contaminants. There are exceptions, e.g. for PCBs, where the aim is to take them out of circulation due to hazardous effects and high risk for serious adverse effect on environment and health. MLs for additional HOCs should be considered, or some HOCs with high hazard risk should not be allowed to be introduced to arable land or growth media via fertilisers and soil products.

There are regulations for use of organic based fertilisers in the EU, and several member countries have introduced stricter regulations than the EU. For compost and digestate, some countries have strict regulations on input materials and no further regulations, while other countries have less strict regulations on input materials but regulate the end products with MLs on selected contaminants. Some countries have restrictions both on input and MLs in the end products. Whether compost and digestate are regulated as a product or as waste vary between countries. Most EU member countries regulate digestate and compost alike. Most countries have a positive list over which waste fractions are permitted to be used as substrate for fertiliser products.

PCB7 and PFOS are examples of HOCs that are identified as a great threat and MLs are, or proposed to be, introduced in the EU and different member countries.

Based on the proposed normative values for soil from 2016, COWI proposed MLs for selected HOCs. For SCCP, HHCb, AHTN, PCB7, NP and NPE, COWI suggested to adjust MLs – increase the MLs – to more “acceptable” MLs and also to be more in line with established MLs in the EU for selected HOCs. This adjustment is not based on a risk evaluation, but an assumption that this is acceptable.

Whether the established or proposed MLs by the EU and member countries protects the environment satisfactorily, it is not known. There are e.g. publications that question the technical basis for EU’s MLs for some HOCs and urge for re-evaluation in some cases.

In Norway, the proposed normative value for PFOS in soil is 0.002 mg/kg DW (from 2016), and Sweden recently introduced a normative value for PFOS in soil at 0.003 mg/kg DW. It is recently suggested that the Norwegian normative value should be adjusted to 0.01 mg/kg DW, however, such an increase from 0.002 to 0.01 mg/kg DW need strong evidence. The Norwegian normative values for soil are established for evaluation and remediation of contaminated soil and not agricultural soil or growth media, and thus, explain different focus related to exposure pathways and which evaluations are most in focus.

PART I - RISK ASSESSMENT

2 Approach and methodology for environmental and health risk assessments related to establishing MLs for HOCs in fertilisers

2.1 General principle for environmental and health risk assessments

An environmental and health risk assessment identifies and describes a hazard, estimates the exposure of defined target organisms to the hazard, and finally, compares/evaluates the exposure related to a predicted no effect concentration (PNEC), a tolerable dose or similar hazard describing terms.

Technical guidance documents for performing environmental and health risk assessments have been published (e.g. for chemicals by ECHA 2016). In the guidance documents from European Regulation of Chemicals (ECHA), risk assessment of human exposure indirectly via the environment is based on assessing concentrations in the diet (food, drinking water) and air, and the intake rate of each of them based on a standard consumption pattern (human daily intake of food and water from EUSUS) (Figure 4) (ECHA, 2016). Combining the rate of intake (e.g. vegetables and cereals per day, week etc) and the concentration of the contaminant in food items, an exposure/intake load is predicted (e.g. mg contaminants per day, week etc) and compared with tolerable doses (e.g. Tolerable Daily Intake). For exemplifying this as a risk-evaluation for exposure of contaminants via fertilisers grown on contaminated soil, the concentrations of the contaminants in surface water and ground water are needed in addition to soil. These data are either based on predicted environmental concentration (PEC) or measured environmental concentration (MEC). However, for HOCs, measured data is scarce, so predicted concentrations are most commonly used. In the guidelines for calculating PEC_{soil} , application of sewage sludge (could also be other soil amendments) and dry and wet deposition from the atmosphere are included (ECHA 2016).

The PEC_{soil} is used for risk assessment of terrestrial ecosystems, and indirect human exposure via crops and livestock products (illustrated Figure 2). PEC_{soil} is also often used for evaluating indirect risk for fresh water ecosystems via runoff and leaching. These transfer processes are today predicted with the use of simple equations or algorithms (ECHA, 2016).

There exist more complex models designed for pesticides, which include soil and water transport and predict concentrations of contaminants in crops, but these models are currently not suitable nor ready for use for risk assessment of e.g. HOC or PTE in fertilisers or other soil products. These models might for instance lack input parameters and fate processes which are important for risk assessment of HOC via fertilisers, or they require information and input data which is not available - or relevant. A risk assessment often includes many scenarios such as different regions and climate scenarios, agricultural practices etc. Thus, models necessitate a certain simplicity and information from many tests on compounds and scenarios.

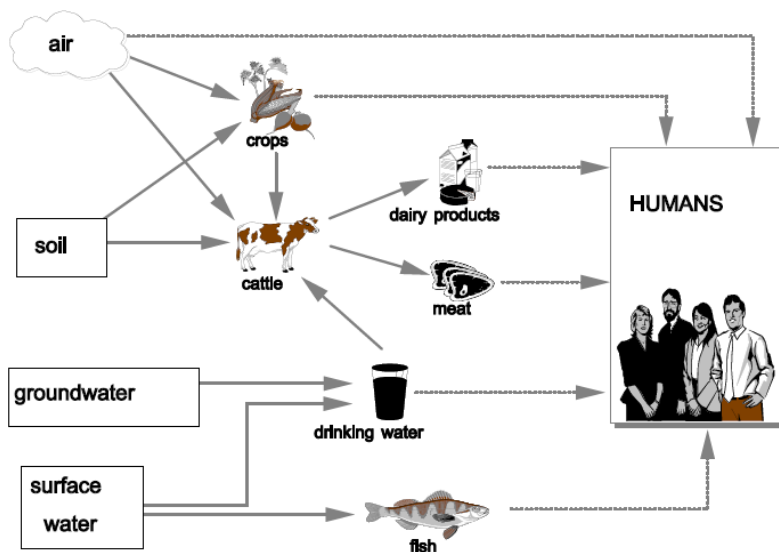


Figure 2. Schematic representation of the exposure routes considered in indirect exposure to humans via the environment (from ECHA, 2016).

The environmental and health risk assessments, “Risk assessment of contaminants in sewage sludge applied on Norwegian soils” (VKM, 2009) and “Risk assessment of copper and zinc from feed to soil and food” (VKM, 2014), both published by The Norwegian Scientific Committee for Food and Environment (Vitenskapskomiteen for mat og miljø, VKM), followed the same approach. The main steps in these risk assessments are:

- Identify the current soil concentration (background/initial soil concentration), and if no data, determine which background concentration level to use in the risk assessment,
- Identify the major sources for a given contaminant which should/need to be included and accounted for in the risk assessment,
- Predict the amounts of contaminants applied to soil (e.g. kg or tonnes per hectare per year),
- Predict via calculations the concentration of contaminants in soil after application (PEC_{soil}),
- Predict transfer to and concentration of in water recipients (predicted environmental concentration in surface water and sediment, $PEC_{surface_water}$, $PEC_{sediment}$),
- Predict transfer to and the concentration of in plants (PEC_{plant}). This includes different forages and major human food plant species,
- In animal and human risk evaluation, the exposure depends on which crops that dominate/are part of the diet for different animal species or groups of human population. This exposure value is compared with tolerable intake per week, month or similar expression for acceptable exposure.
- Identify the hazard to be included in the risk evaluation, e.g. $PNEC_{terrestrial-organisms}$, $PNEC_{aquatic-organisms}$, $TDI_{livestock}$ or TDI_{human} ,
- Health risk assessments often include protection for specific vulnerable groups, for instance children eating soil and pregnant women.
- More commonly, terrestrial or aquatic secondary poisoning of predators is included. This is related to toxic effects of the higher members of the food chain resulting from ingestion of organisms from lower trophic concentrations that contain accumulated substances, also known as bioaccumulation (described in section 2.4).

- Calculate Risk Characterisation Ratio (RCR), defined as the predicted environmental concentrations (PEC) divided by predicted no effect concentration (PNEC) for a given organism exposed to e.g. contaminants in soil, surface water, and sediment. When RCR (PEC/PNEC) < 1, there is an acceptable risk, and if RCR > 1, there is an unacceptable level of risk, or at least a potential risk for hazard effects which need further evaluation.
- For PBT and vPvB substances, a PEC/PNEC approach cannot be carried out with sufficient reliability (ECHA, 2016), and thus, how to handle these substances must be evaluated further.

In addition, the following points need to be considered and decided upon:

- Which organisms to protect?
- Which exposure pathways to include?
- Which fate and transfer processes to include (related to the points above)?
- Which input parameters are sensitive for the evaluation?
- Many parameters and input-data are associated with high uncertainty, resulting in underestimating or overestimating of the risk. The scale of this under/overestimating can vary highly and had to be evaluated,
- When a simplification for fate and transfer processes is needed (for instance developing MLs for HOCs), which processes, and exposure pathways are underestimated or overestimated?
- How to distinguish between application of fertiliser and soil products to agricultural soil, parks, and growth media for use in kitchen gardens or different kinds of containers?
- And finally, a risk assessment should be scientifically-based and based on the best available knowledge. If there is significant lack of knowledge, it can be premature for a scientific-based risk assessment. In the meantime, how can one ensure precautional and conservative utilisation of fertilisers and soil products that potentially contain HOCs?

2.2 Methods and approaches used for estimating fate and transfer processes

The technical guidance documents for environmental and health risk assessments of contaminants (ECHA 2016) contain these tasks/topics:

1. Prediction of input of contaminants to soil

Introduction/input of contaminants via fertilisers or soil products to soil must be estimated based on available information about concentration of contaminants and the regulation for use of fertilisers. Based on this information, the amount (e.g. mg contaminants/ha/yr) is predicted.

2. Prediction of reduction of contaminants in soil

• Degradation

Degradation of organic contaminants depends on several factors: The compounds' inherent degradability, which again depends on its structure and functional groups; the environmental abiotic conditions such as temperature, humidity, light, redox-conditions, organic content, pH, and salinity; and the presence of microbial activity.

With the exception of strongly oxidized contaminants (e.g. brominated, chlorinated, fluorinated), most contaminants are degraded faster (shorter half-lives) under aerobic than anaerobic conditions. The fact that some contaminants have shorter half-lives under anaerobic environmental conditions than aerobic is important to note.

- **Loss of contaminants from soil via water transport**

Loss of HOCs from soil include leaching and erosion/runoff. Contaminants are transported from the soil to nearby water recipients, e.g. groundwater, streams, ponds, surface water, and sediments.

Leaching is predicted based on the binding properties of the contaminants (commonly expressed by K_d or K_{ow} , K_{oc}), annual precipitation, and soil infiltration rate. There exist different simplified equations for predicting leachates. Runoff includes both loss of contaminants via erosion (bound to particles) and dissolved HOCs in water phase.

Leaching predicts removal kinetics and leads to a reduction of the soil concentration of the contaminants. Loss of contaminants bound to particles via erosion does not reduce the concentration in soil. Only HOCs that are removed from soil via e.g. leaching, dissolved in runoff water, degradation, or evaporation lead to lower concentrations of HOCs in the soil. Loss of HOCs bound to particles transfer HOCs from soil to nearby water recipients, and might precipitate in the sediments.

This is important to note since hazard is related to concentrations.

- **Removal of contaminants from soil via harvesting of plants**

Plants have varying potentials for uptake of contaminants. In addition, different tissues in a plant might have highly distinct contaminant contents. In addition to plant species differences, this depends on the physicochemical properties of the contaminant, but also local and regional factors such as soil properties (content of e.g. organic matter, pH, clay) and climate are important. Uptake of contaminants in plants are most commonly expressed as a bioconcentration factor (BCF) or a transfer factor (TF).

In environmental and health risk assessments, removal via harvested plants is predicted based on BCF for plants, concentration in soil and the crop yields (kg biomass).

3. Prediction of change of contaminants in soil over time

The change of contaminants in soil over time, can be predicted by comparing the ratio between kinetic input (k_{input}) and kinetic removal from soil ($k_{removal}$), given as yr^{-1} or d^{-1} . If k_{input} is higher than $k_{removal}$, an accumulation over time is predicted. On the other hand, if $k_{removal}$ is higher than k_{input} , no accumulation is predicted and a reduction over time can be assumed.

Other approaches and equations for predicting the soil concentrations of contaminants over time exist, and they should be evaluated and compared.

4. Prediction of transfer of contaminants to plants and concentration in forage and food plants – exposure to livestock and humans

Concentrations of contaminants in edible plants/plant tissues are predicted based on BCF and soil concentration (PEC_{soil}).

2.3 Fate and transfer of HOCs in soil

2.3.1 Fate processes in soil

The fate of organic contaminants introduced to soil depends on the chemicals' physicochemical properties and several abiotic and biotic environmental factors. For organic substances, their

degradability is important. Rapidly degradable contaminants are mainly a problem if they are frequently loaded/transferred to the environment, or less frequently loaded but in much higher concentrations.

A compound's inherent degradability depends on its structure and functional groups, which determine its availability for abiotic (photooxidation, hydrolysis, reductive) or biotic (enzymatic) transformation, and the environmental conditions: Presence and extent of microbial population and redox condition are two major factors. The microbial activity will vary and is dependent on soil type, organic content and climatic conditions, e.g. temperature, redox condition, soil humidity, pH and salinity.

With the exception of strongly oxidized contaminants (e.g. brominated, chlorinated, fluorinated), most contaminants are degraded more rapidly (shorter half-lives) under aerobic than anaerobic conditions. The fact that some contaminants have shorter half-lives under anaerobic environmental conditions than aerobic is very important to note. This is particularly of importance for the BDE-209, for which it is known that debromination occurs and with a high probability is transformed to the more toxic and bioaccumulating congeners such as BDE-99 and BDE-47 (Kortenkamp et al., 2014) as illustrated in Figure 3. The lack of knowledge related to formation of low brominated PBDEs from BDE-209 in sludge applied to agricultural soil is also addressed by others e.g. Brambilla et al. (2016).

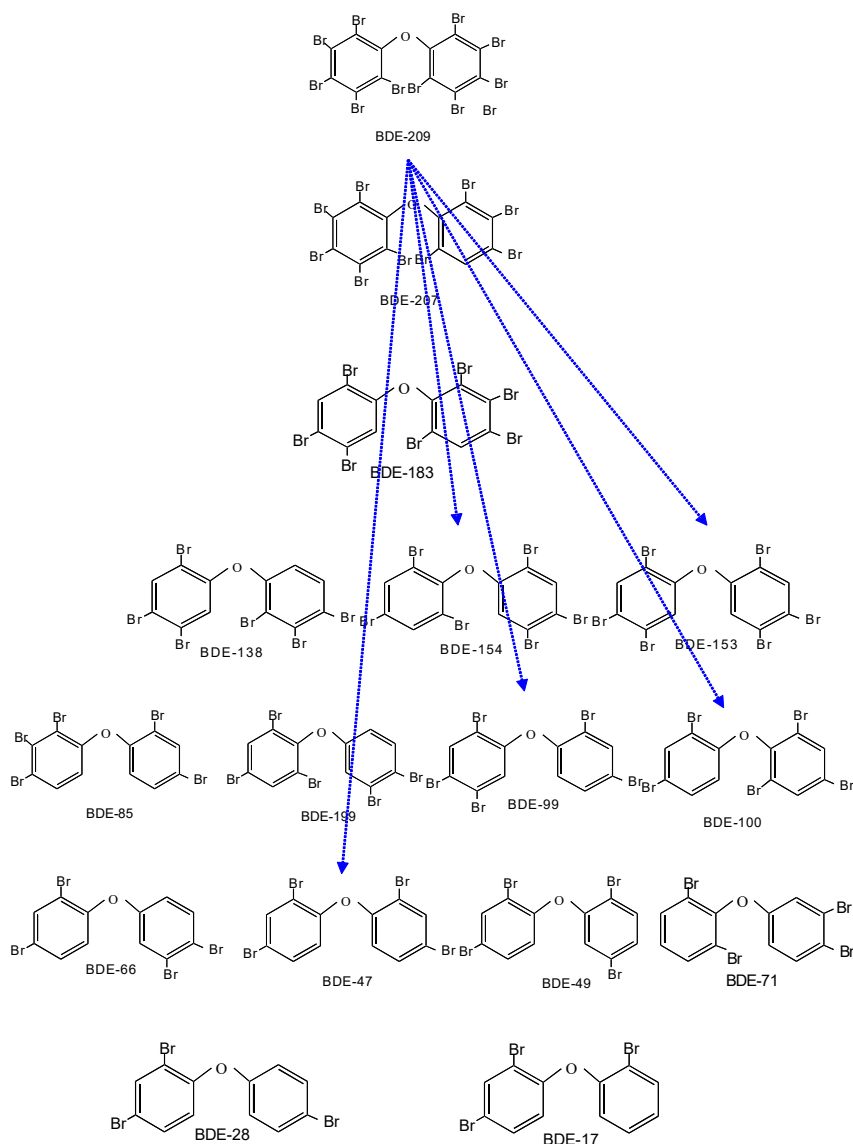


Figure 3. Reductive debromination of BDE-209 (modified from pathways suggested by several e.g. Zhuang et al., 2012; Tokarz et al., 2008; He et al., 2006).

Many contaminants have polar functional groups that form chemical bonds with charged surfaces of particles or organic matter. Non-polar or hydrophobic contaminants associate with organic matter, and may undergo humification via oxidative coupling (Weber et al., 2003). Clay minerals contribute to the sorption capacity, particularly in surface soils with low contents of organic matter and in subsurface material.

The binding capacity of contaminants is usually presented as the distribution coefficient, K_d (ratio for the concentration bound to soil over concentration in water phase). It is often normalised to the organic content of the matrix in order to obtain the organic carbon-water partition coefficient; or as the partition coefficient P (commonly addressed as octanol water coefficient, K_{ow}), which is an experimental value. The distribution of a substance between octanol and water phase indicates the hydrophobicity of a compound. These coefficients might also be predicted via modelling tools. While

the partition coefficient refers to the non-polar species, the distribution coefficient (D) refers to non-polar plus the polar species.

Organic matter differs in its quality and ability for sorption. Over time, K_d values of substances (contaminants) often increase due to formation of covalent bounds to particles, organic matter, or aged residues. Aging is a commonly observed field phenomenon, in which a pollutant becomes with time increasingly more difficult to extract from soil. Both processes, unlike sorption, lead to apparently irreversible associations between the pollutant and soil. A number of factors influence the aging processes such as soil organic matter quality; nanoporosity; soil aggregate structure and presence of clay minerals; pH; humidity; and the properties and spatial structure of the molecules (An et al., 2017).

Organic matter is the most important factor limiting availability and mobility of persistent organic pollutants in soil (Scow et al., 1995) and a substantial percentage of an organic contaminant applied to soil may become associated with the humic fraction of that soil. However, mineral (inorganic) component of the soils also apparently play a significant role in the sequestration of selected organic contaminants (Scow et al., 1995). A compound's binding capacity to soil and organic matter is important for its potential for transfer from the soil. Compounds with low binding capacity, low K_d , and high water solubility, are more leachable and more likely to be transferred to groundwater or via trenches to surface water. Transfer of organic contaminants from soil to water recipients is important to account for in risk assessment: these processes are highly influenced by the binding capacity between soil and contaminant (soil quality and physicochemical properties); infiltration capacity of the soil; and the climate conditions (precipitation, temperature, humidity, light [photooxidation]).

For compounds with high binding capacity, transfer from soil via erosion is an important process. Contaminants with high K_d and low degradation rate (long half-life) will have a high potential for accumulation in soil over time. However, during high or extreme rain events, these may be transferred to nearby ponds and lakes.

For volatile and semi-volatile compounds, evaporation might also be part of the removal process and will depend on the compound's properties as well as the surrounding conditions (e.g. temperature and wind) and is therefore not constant.

2.3.2 Transfer to edible crops

Organic contaminants are taken up via roots or deposited on leaves. Transfer of organic contaminants is primarily related to their hydrophobic/hydrophilic properties and occurs most commonly via root uptake. However, there are also plants, e.g. lettuce, where the contribution of contaminants attached to soil particles also may account for human intake. K_{ow} is the main parameter for predicting a bioconcentration factor (BCF, often used equally as bioaccumulation factor).

For plant uptake it is well known that some of the older BCFs assumed that plants have low uptake of hydrophilic compounds (e.g. Briggs et al., 1982). It is now known that this is not the case, and newer and more advanced plant uptake models exist (e.g. Trapp 2000; Trapp and Legind 2011).

While previous and now banned contaminants were strongly hydrophobic and non-charged, many of today's contaminants are charged (ionic) and highly hydrophilic. Some contaminants are surface active and neither hydrophilic or hydrophobic but have a strong affinity to proteins.

Concentration of organic compounds in root vegetables, particularly carrot, radish and potato, are higher for hydrophobic than hydrophilic compounds, and conversely, usually higher for hydrophilic contaminants in above ground tissues. However, as mentioned about, leafy vegetables can potentially contain hydrophobic HOCs attached to soil particles.

It is demonstrated that there might be high variation in uptake and internal translocation of organic contaminants in plants, even for substances with highly similar structure and properties (Eggen et al.,

2013). It is therefore important to be aware that certain compounds may behave differently than expected. It is also shown an accumulation of a pharmaceutical, metformin, into rape seeds, which is assumed to be due to a mimicking reaction mistaking metformin for a natural plant substance (Eggen and Lillo, 2012). In addition, it has been demonstrated that triclosan forms conjugates of phase II metabolites after uptake in carrots. Analysis of the parent compound (triclosan) alone severely underestimates the extent of uptake in plants (Macherius et al., 2012), as the uptake in carrot was shown to be five times higher than measured for triclosan when the conjugated phase II metabolites was included in the analysis.

Since conjugates of phase II metabolites might become bioavailable under certain conditions, this underestimation of plant uptake can also underestimate potential human exposure.

It is stated in the guidance report by ECHA (ECHA, 2016⁶), that reliable and relevant experimental bioconcentration factors should always be preferred to estimated factors. This is true, however, due to the situation with the presence for a huge number of organic chemicals, it is important to develop and verify models for predicting the fate of organic contaminants which could represent different classes of chemicals/contaminants. Reliable experiments that cover chemicals which represent different relevant physicochemical properties under different relevant environmental conditions should be performed. Such experimental data should be used for verifying or developing environmental fate models.

2.4 Hazard and MLs

In environmental and human health risk assessments, the measured or predicted environmental concentrations (e.g. PEC_{soil} , $PEC_{groundwater}$, $PEC_{surfacewater}$) and the exposure concentration (e.g. mg contaminants exposed for per day or month in average during a life) are compared to the acceptable or non-acceptable exposure levels e.g. Predicted No Effect Concentration (PNEC), No Observed Effect Concentration (NOEC), Lowest Observed Adverse Effect Level (LOAEL), Lowest Observed Effect Level (LOEL), Tolerable Daily Intake (TDI).

Predicted No Effect Concentrations (PNEC)

Within the European regulation of chemicals i.e. REACH, guidance exists on how to quantitatively assess the effects of a substance on the environment by determining the concentration of the substance below which adverse effects are not expected to occur in the environment. This concentration is known as Predicted No-Effect Concentrations (PNECs). A PNEC is obtained through the application of an assessment factor to ecotoxicological endpoints (EC_{50} or NOECs) using organisms with different sensitivities for any type of chemical. The size of the assessment factor depends on duration of the test (acute or chronic), the number of trophic levels tested and the general uncertainties in predicting ecosystem effects from laboratory data. In order to derive risk of contaminants for soil living organisms, such as plants, microorganisms and earthworm, $PNEC_{soil}$ should be determined (Andersen et al 2012).

Cocktail effects

In the natural environment, living organisms are not necessarily exposed to one single pollutant, but to a variety of different contaminants. The exposure to the mixture of chemicals is first and foremost through food (prey), but also from water and the environment they live in, especially for lower trophic organisms. Component-based approaches are suitable methods for evaluating risk of mixtures when

⁶ https://echa.europa.eu/documents/10162/13632/information_requirements_r16_en.pdf

exposure data (i.e. concentrations) in addition to toxicity endpoints or similar toxicity reference values exist for the individual chemical components. (Altenburger et al., 2014).

The method of summing up PEC/PNEC or MEC (Measured environmental concentration)/PNEC ratios, is recommended as a justifiable mixture risk approximation in order to estimate in a first approach whether there is a potential risk for an exposed ecosystem (Backhaus and Karlsson, 2014; Backhaus and Faust 2012). For instance, it is concluded that BDE-209 is a source to more toxic, lower brominated PBDEs which have the capacity to work together with BDE-209 to produce combined toxicity. An evaluation of BDE-209 in isolation, without taking account of such combination effects, can significantly underestimate the toxicity of BDE-209. It is further addressed that other PBDEs pose significant health concerns, especially for young children of age 6 month to 3 years which bear the highest PBDEs exposures of all age groups.

Secondary poisoning

Secondary poisoning (described by $PNEC_{oral}$) is related to toxic effects of the higher members of the food chain in the terrestrial environment, resulting from ingestion of organisms from lower trophic concentrations that contain accumulated substances. Only toxicity studies reporting on dietary and oral exposure are relevant as the pathway for secondary poisoning and is referring exclusively to the uptake through the food chain. Secondary poisoning effects on bird and mammal populations rarely become manifest in short-term studies. Therefore, results from long-term studies are strongly preferred, such as NOECs for mortality, reproduction or growth. If no adequate toxicity data for mammals or birds are available, an assessment of secondary poisoning cannot be made. If a chronic NOEC for both birds and mammals is available, the lower of the resulting PNECs is used in the secondary poisoning assessment to represent all predatory organisms (ECHA, 2008).

Figure 4 illustrates secondary poisoning for aquatic food chain for marine water (from ECHA 2016). Similarly approach for terrestrial food chain are used (Romijn et al., 1994).

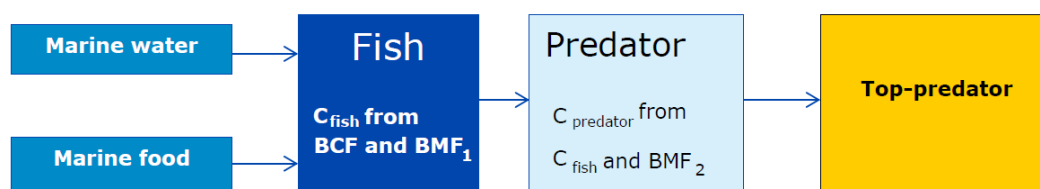


Figure 4. Secondary poisoning illustrated for aquatic food chain for marine water (from ECHA, 2016).

Tolerable doses or intake

Acceptable levels for humans are expressed as different endpoints related to tolerable intake dose, e.g. tolerable daily intake (TDI) which is assessed safe on a long-term basis, normally on a whole lifetime.

In the normative values for soil, human health, QS_{soil} (human health), is the first protection before protection of predators in the terrestrial foodweb QS_{soil} (secondary poisoning), and soil ecology (QS_{soil} (ecotox)). The QS_{soil} (human health) is derived from the methodology established in 1999 (TA-1629/1999) and includes the following exposure pathways; oral uptake of soil and dust, dermal absorption, inhalation of soil dust, inhalation of released vapours from soil, intake of drinking water or fish affected by nearby contaminated soil or intake of vegetables grown on contaminated soil.

Important considerations

Science-based risk assessment must follow certain guidelines and rules to qualify as scientifically based risk assessment; e.g. guidance documents from ECHA and how risk assessments are performed by VKM and EFSA.

Processes such as abiotic and biotic degradation, binding to particles and organic matter, humification, water transport via leaching and runoff, uptake of plants or soil organisms, and for volatiles also evaporation, influence the fate of organic contaminants in the environment. Soil quality and properties, physicochemical properties of the contaminant, climate conditions like temperature, light, precipitation and humidity are additional factors that highly influence these processes.

Key parameters for evaluating environmental fate are half-lives (DT_{50}), removal rates expressed as k (d^{-1} , wk^{-1} , yr^{-1}), and distribution and partition coefficients (e.g. K_d , K_{oc} , K_{ow}) that commonly used to describe binding and leaching processes. Uptake via plant roots or deposit on leaves of organic contaminants are potential transfer and exposure pathways of contaminants towards domestic animals and humans. Uptake in plants, commonly expressed as a transfer factor (TF) or bioconcentration factor (BCF), contributes also as a removal processes if plants are harvested and removed from the field. Transfer of contaminants to nearby water bodies (groundwater and surface water including sediments) is important to account for in risk assessments and is predicted with use of e.g. K_d , annual precipitation, and infiltration rate.

The emerging questions are: which parameters are important to include in risk evaluation? Which are sensitive to changes in environmental conditions and climate? Which equations and models are suitable for description and prediction of these processes?

It is stated in ECHAs technical guidance document (2016) that reliable and relevant experimental bioconcentration factors should always be preferred to estimated bioaccumulation factors. It is important that the quality of the experiment is considered before numbers are used. However, due to the enormous number of potential chemicals present in the environment, it is also important to establish models that can predict fate and risk reliably.

3 Approach and methodology used by COWI for developing MLs

3.1 Approach and assumptions

As presented above, several assumptions need to be made in a risk evaluation. The selections and assumptions made by COWI and which are the basis for the proposed MLs (Table 4) are presented here.

- **Background level (present concentration) of organic contaminants in soil**

COWI applied two different approaches for choosing background concentration in soil:

- 1) no contaminants present in background soil and evaluation of how much contaminants (via fertilisers) can be added before reaching proposed normative values, and
- 2) background soil concentration equivalent to normative guideline values suggested by NGI (Arp et al., 2017) and amount of contaminants that can be added in fertilisers without causing a significant increase in soil. Significant increase in soil was related to safety level.

- **Uncertainty and acceptable soil level**

To adjust for uncertainties, laboratory/analytical uncertainty and sampling uncertainty were taken into account and calculated as:

$$Total\ uncertainty = \sqrt{(lab\ uncertainty)^2 \cdot (sampling\ uncertainty)^2}$$

For laboratory uncertainty 30% was used and for sampling uncertainty 50, 75 and 100% were used. This resulted in a calculated total uncertainty of 58, 81 and 104%, respectively.

Based on the % uncertainty and normative values, an acceptable contamination level was predicted.

- **Assumptions and selection of parameters**

Several assumptions need to be made in order to perform a risk assessment.

In the COWI report the following assumptions and simplifications were made:

- Fertilisers are evenly distributed in 20 cm depth soil
- Relatively high O₂ content in soil
- Soil density 1.2 kg/l (mean soil density in Norway)
- The amount of added fertiliser was the same as regulated for sewage sludge category I; 40 tonnes dry matter per hectare per 10 years (tonnes/ha/10yr), or 4 tonnes dm/ha/yr.
- Half-lives were estimated with the use of QSAR (the software EPISUITE), which does not necessarily give the most reliable results.
- Predicted half-lives were based on a temperature at 22 °C.
- Reduction of contaminants in soil is based on a mass balance approach in which a certain amount of soil is removed each year. For calculating the reduction in soil, measured erosion data from 1990/2000 was used. The measured erosion ranging from 10 to 730 kg daa/yr, and the average at 200 kg soil daa/yr was used in their work (Bechmann et al., 2000).

- The equation used for predicting the concentration of organic contaminant in soil immediately after application of fertilisers to soil, and the change in soil concentration over time in the approach by COWI (Blytt et al., 2018), is not presented in the report. The method used in this report has been provided by the Norwegian Environment Agency:

$$C_{\text{soil}(t)} = C_{\text{soil}(0)} + \left(\frac{\% \text{rate of application of fertiliser}}{100} \right) * C_{\text{sludge}} * 0,5^{\left(\frac{t_n}{DT_{50}} \right)} * \left(1 - \left(\frac{\% \text{soil erosion per year}}{100} \right) * \frac{t_n}{365} \right)$$

Where:

$C_{\text{soil}(0)}$ = initial concentration (normative values from Arp et al. (2017))

Rate of application of fertiliser; 1, 5- or 10-years application cycle

C_{sludge} = concentration of contaminants in sewage sludge

DT_{50} = half life

$C_{\text{soil}(t)}$ = soil concentration of contaminant after time t

Soil erosion per year = assumed amount of soil eroded over a year

3.2 Our comments to the selected approach and methodology

Accept levels and normative values for contaminants in soil

Upon request by the Norwegian Environment Agency, COWI has used proposed normative values for contaminants in soil (see Table 3) as a basis for developing ML values. These normative values for soil and three different levels of % uncertainty (58-104%) was applied as an acceptable soil concentration.

Whether use of normative values for soil and a 58 to 104% uncertainty is a reliable approach for selecting an acceptable soil value is not further discussed in this report. However, it is recommended to evaluate further if the use of normative values and the calculated uncertainties are suitable as acceptable soil concentration for crop production. The normative values for soil are established for evaluating if soil areas are contaminated or not, and exposure pathways to humans include;

- Oral uptake of soil and dust
- Dermal absorption (or uptake of contaminants in the soil through the skin)
- Inhalation of soil dust
- Inhalation of released vapours from soil
- Intake of drinking water affected by nearby contaminated soil
- Intake of fish affected by nearby contaminated soil
- Intake of vegetables grown on contaminated soil

Human exposure via intake of products from farm animals is not included, neither are exposure of children eating soil and other vulnerable groups taken in account.

Particularly for PBT and vPvB compounds, it is very important that an appropriate and conservative (precautionary) approach is applied, and that the normative values are validated for agricultural soil, growth media, and soil mixtures; protect terrestrial and aquatic environments; and ensure animal and human health.

BCF for plant uptake, one of the parameters for establishing $QS_{\text{soil human health}}$, is based on equations from a report from 1999 (TA-1629/1999). Based on new knowledge, improved plant uptake models have been established (see 2.3.2 Transfer to edible crops). In the report from 1999, for instance, the BCF_{stem} and BCF_{root} are both given in fresh weight; it is not clear if this has been accounted for in the normative values used by COWI. For plants with high water content, e.g. leafy vegetables, carrot, and potato, this fact will influence more than for instance cereals with lower water content. There may be other parameters and assumptions which also need to be evaluated more closely.

Parameters used by COWI:

ECHA (2016) has recommended that spread fertilisers are evenly distributed in 20 cm depth in soil, and COWI has used this assumption.

Relatively high O_2 content should normally be assumed. However, for compounds like BDE-209, it is important to take into consideration that they are degraded much faster under anaerobic than aerobic. Under anaerobic conditions, such as in water saturated soil or in clumps of sewage sludge or digestate, or in sediments, there is reason to believe that degradation of BDE-209 will speed up compared to aerobic conditions.

A soil density of 1.2 kg/L is the mean in Norway, and VKM has also used this assumption in their risk assessment of sewage sludge (VKM 2009).

The amount of added fertiliser is the same as that regulated for sewage sludge category I; 40 tonnes dry matter per hectare per 10 years (tonnes/ha/10yr), or 4 tonnes dm/ha/yr, and normally selected for risk assessment of this kind.

Use of 200 kg removal of soil per year is based on an average between 10-730 kg soil/da/year, which are values measured in 1999/2000 from agricultural land with high erosion potential (Bechmann et al., 2000). This value has a high grade of uncertainty, and if erosion is to be included in the evaluation, should be evaluated before use.

Half-lives (DT_{50}):

Half-lives were selected either based on literature or with use of QSAR and EPISUITE as software. This is problematic, since EPISUITE does not predict half-lives higher than 360 days for any compound. EPISUITE is designed to estimate values for neutral compounds and is therefore not suitable to predict properties of charged compounds like of PFOS and PFOA. Although charged compounds can be more labile than hydrophobic ones, many experimental and field studies reveal that both PFOS and PFOA are very persistent in the environment. The precautionary principle should therefore be used, and conservative half-lives are recommended. The use of half-lives from EPISUITE software, as done by COWI, will thus lead to an overestimation of contaminant removal and an underestimation of the residue concentration in soil over time.

Predicted half-lives were based on a temperature of 22 °C, and non-adjusted for Nordic environmental temperatures. This also led to an overestimation of contaminant removal from soil and an underestimation of the residue concentration in soil.

See **Section 4** for our proposed adjustments of the DT_{50} values.

Approach and equations for predicting concentration in soil over time

A mass balance approach was utilized, accounting for the percent of added fertiliser per volume of soil (0.2 m depth x ha (10000 m²)) and removal of soil via erosion (g soil per ha), but not including

leaching and transfer to crops. When predicting the amount of contaminants introduced to soil via fertilisers, the concentration of the contaminants is taken in account; however, the applied equation appears not to take into consideration the concentration of the contaminants in soil or the physicochemical properties of the contaminants when calculating removal via erosion (soil particles). If this is the case, the approach and the equation do not account for the water transport of neither the bound nor the dissolved contaminants. Accounting only for removal of contaminants attached to soil particles is another aspect. This will only remove the total amount of contaminant in a given soil volume but not the concentration in the remaining soil.

Estimating transfer of contaminants via leaching and plant uptake both depend on the concentration in soil and physicochemical properties of the contaminants.

In the report by COWI, use of fresh water toxicity as an approach to restrict contaminants in fertilisers is questioned since the pathway (runoff and/or leaching) of contaminants may not be applicable for arable land that is far from surface water, even if it is stated that for some areas this may be justified because runoff may reach surface waters easily. This might be a reason for considering contaminants' soil concentration and physicochemical properties, but not necessarily for evaluating the water transport from soil. Such a simplified approach should in our option only be used if it is known that water transport of contaminants is an insignificant environmental process.

This approach is problematic, because runoff and leaching of contaminants can cause elevated concentrations of contaminants in groundwater/drinking water, ponds and lakes - and possibly exceed acceptable/maximum levels in drinking water and/or lead to ecotoxicological effects on aquatic or sediment dwelling organisms.

For contaminants with low sorption capacity, e.g. low K_d , and particularly in regions with high precipitation and precipitation excess (infiltration rate), leaching might be an important fate process to include in the risk evaluation.

In the guidance documents for environmental and health risk assessment (e.g. ECHA 2016), the equation for predicting leaching includes annual precipitation, infiltration (precipitation excess) and binding capacity of the contaminants. Thus, depending on the climate (precipitation) in the region and the binding capacity, removal from soil and transfer to nearby water body might vary highly.

In our opinion, transport of contaminants from arable soil to water recipients and prediction of surface water should be included in order to account for protection of drinking water and aquatic organisms.

As commented above, the equation included DT_{50} but needs to be adjusted to more realistic temperature conditions.

Separate ML values for fertilisers and soil products for agricultural use and growth media, soil mixtures

COWI suggested a quality class for soil products used as growing media, which is based on QS_{soil} times 10 (Blytt et al., 2018). For example, for BDE-209, with a normative value of 0.49 mg/kg dw, the suggested ML value for growth media would be 4.9 mg/kg dw.

It is common that families, kindergartens etc. eat self-produced vegetables cultivated in growth media/soil mixtures in kitchen gardens and containers. For many HOCs, higher uptake of contaminants in carrots and other root vegetables and leaf vegetables such as lettuce than e.g. cereals is expected. During the summer period, a portion of the population including children, might thereby have a higher intake of self-produced crops with elevated levels of contaminants. This is an exposure scenario that needs to be considered regarding establishing MLs for fertilisers/growth media for different use. Instead of higher MLs in growth media/soil mixture, which COWI proposed for some of

the HOC (Blytt et al., 2018), there might be reasons for stricter regulation in growth media/soil mixtures than fertilisers used by farmers on agricultural soil. Another point is the amount of sewage sludge added to growth media, which can be up to 30% of total volume, and can theoretically contain higher concentrations of HOCs than in agricultural soil after application of 40 tonnes/ha/10 year. While regulations restrict which crops can be cultivated and how long after application on agricultural land, there are no regulations for what people cultivate in their own containers and kitchen gardens.

It is recommended that this is addressed and evaluated further.

Important considerations

The approach and methodology used for recommending maximum values for organic contaminants from 2018 is meant to be easy and simplified, and several choices and assumptions made are based on today's regulations and previous risk assessments. The major concerns that should be taken into consideration are:

- i. uses of more realistic half-lives, and whenever possible based on experiments with application of contaminated sludge, and use the highest reported reliable values in further considerations,
- ii. if necessary, adjust for Norwegian ambient temperature conditions,
- iii. evaluate how suitable the established normative values are for agricultural soil and growth media, and for protecting all target organisms and humans, including vulnerable subgroups of the population,
- iv. account for transfer of contaminants to water bodies (e.g. ponds, lakes, ground water); this includes evaluation of exposure and toxic effects of aquatic organisms and sediment dwelling organisms, exposure and transfer to fish and further exposure to humans, and exposure of animals and humans via drinking water,
- v. ensure that exposure of humans via crops and forage are taken into account in the risk assessment.

A mass balance approach lacking realistic kinetic processes for a given HOC's physicochemical properties and without predicting environmental concentrations (PECs) in soil, crops and water, is too simplified to be reliable for a risk assessment without any further evaluations.

A portion of the population consumes self-produced vegetables cultivated in growth media/soil mixtures in kitchen gardens and containers. They can have higher intake of crops with elevated contaminant concentrations, especially during the summer. Instead of the same or even higher MLs in growth media/soil mixture and fertilisers, it might be relevant with stricter regulation for HOCs in growth media/soil mixtures than fertilisers used by farmers on agricultural soil. It is recommended that this is addressed and evaluated further.

It was not possible within the frame of the project to suggest how to improve the methodology for developing MLs.

4 Adjusted half-life (DT₅₀) values

4.1 Half-life values proposed by COWI

The half-life values used/proposed used by COWI (Blytt et al., 2018) are shown in Table 6.

In our opinion, these values require re-consideration for the following reasons:

1. none of the values are temperature corrected for Norwegian conditions. Values originated from QSAR are being calculated for at +20° C, values from lab experiments relate to +20°, +25° or even +30° C. Also the values from field experiments and observations relate largely to much warmer locations.
2. In some cases, although experimental data for half-lives were available, predicted QSAR values were applied without a clear explanation for the choice.
3. EPISUITE cannot treat charged substances like PFOS, so the value for PFOS was erroneous by definition
4. assignment of value of “360 d” or “>360 d” or “>365 d” for substances with half-life times of >10 years may lead to dramatic underestimations of accumulation of chemicals in soil.

Table 6. Half-lives used in the work by COWI (Blytt et al., 2018). n.p.=not proposed.

Contaminant	Half-life, DT ₅₀ , COWI, (d)	Exp or QSAR	Temperature corrected
DEHP	365	QSAR	NO
PFOS	360	QSAR	NO
PFOA	n.p.	n.p.	n.p.
SCCP	n.p.	n.p.	n.p.
HHCB	239	Exp	NO
AHTN	120	QSAR	NO
OTNE	n.p.	n.p.	n.p.
BDE-209	360	Chemspider	NO
PCB 7	n.p.	n.p.	n.p.
NP	75	assigned equal to that of NPE	NO
NPE	75	QSAR	NO

Therefore, in this report we present new DT₅₀ values (DT₅₀ = time for dissipation of 50% of the original amount, which includes chemical transformation, volatilization, removal with water flow in dissolved form or as particles, removal with airborne dust etc.), based on experimental data and temperature corrected for Norwegian conditions.

There is a confusion in the literature in reporting either DT₅₀ values or degradation half-life times (T_{1/2}). Half-life time normally is understood as time related to chemical transformation, while “dissipation” includes also a removal of the substance from the object without chemical transformation (for example, by evaporation). In experimental studies it is often difficult to fully

separate chemical and/or biological degradation from other elimination processes. Since it is difficult to prove that reported values correspond solely to degradation, we will call them DT_{50} throughout this report.

4.2 Methods for evaluation of use of half-life values

Soil is a living microcosm. Degradation rate in soil at specific location is difficult to predict. Even in the most careful laboratory studies two parallel degradation experiments carried out at supposedly identical conditions might yield half-lives different by a factor of two.

In real world, even in the same experimental field, degradation (dissipation) would depend on environmental conditions like air and water content in soil, vegetation, temperature etc.

Even in laboratory experiments, for example, in one study half-lives of DEHP in the same soil but with various amendments ranged from 24 to 102 days at otherwise identical conditions.

Therefore, care must be taken when using experimental or estimated half-lives. As a reasonable and conservative approach, the longest half-life is to be used for estimations, until a reliable experimental data is obtained for a given type of sludge on a specific soil type.

In addition, most of the half-lives mentioned in literature was measured or calculated for +20° or +25°C. A rule of thumb is that all chemical and biochemical processes slow down by a factor of 2 with decrease in temperature by 10° C. We have therefore, as a general rule, use temperature correction factor of 2.5 – 3.0, based on average annual soil temperatures in Norway measured at 10 cm depth. The soil temperature data are measured by LandbruksMeterologisk Tjeneste, LMT (<http://lmt.nibio.no/>). The average annual temperatures ranged from +4.3°C in Tromsø to +7.2°C in Ås

Note: in several experimental studies no influence of temperature on degradation rate was found for certain substances and soil samples. This phenomenon was reported for linear alkylsulphonates (LAS) and for Nonylphenol (NP) and Nonylphenolmonoethoxylate (NPE) (González *et al.*, 2012). At least in part this might be due to experimental setup or uncertainty in measurements. For example, for NP the same apparent half-life at higher temperature can be due to formation of additional NP from degradation of higher ethoxylates present in the sample. In scarcity of reliable data, we recommend using temperature correction as part of a precautionous approach.

4.2.1 Proposed half-life values for the selected HOCs from literature

4.2.1.1 DEHP

The half-life of DEHP in the soil environment was estimated to be 50–300 days (Kim *et al.*, 2019). ECHA chemical information database reports biodegradation half-life of 10 months in soil at 12 °C.

In laboratory experiments at 25 °C apparent half-lives varied from 24 to 102 in soil (He *et al.*, 2018) and in the field experiment - from 289 to 578 days in sludge (Chen *et al.*, 2009); temperature correction leads to DT_{50} from ca. 5 months to ca. 5 years in Norwegian conditions.

Degradation depends on the properties of soil. In a laboratory study half-life time at 25 °C was 24 days in soil with high organic content, but 94 days in soil with low organic content (He *et al.*, 2018). This result translates to approximately 100 days and 1 year, respectively for degradation at Norwegian conditions.

The range of reported half-life times was from 24 to 578 days, so, potentially, the half-life time in Norwegian conditions might be as high as 5 years.

The safe approach would be to consider DEHP as very persistent and use $DT_{50} = 5$ years in estimates, until solid experimental data for Norwegian soil/sludge becomes available.

4.2.1.2 PFOS and PFOA

There is no evidence of degradation of these substances in soil.

“Both the REACH Annex XV restriction dossier (ECHA, 2015) and REACH proposal for identification of PFOA as an SVHC (ECHA 2013) state that PFOA was not biodegradable and that, based on high persistence, it was not possible to calculate half-lives in soil or sediment.”

Moreover, there are many precursors that do degrade in soil forming PFOS and PFOA:

“PFOA-related compounds will degrade to PFOA in sludge, soil, water and air (ECHA 2015; ECHA 2013; IPEN 2015), and such degradation has been noted across many products containing PFOA-related substances, including products such as grease proof paper (Dasu et al., 2013; Ellis et al., 2003; Ellis et al., 2004a; Frömel and Knepper, 2010; Gauthier and Mabury, 2005; Jackson and Mabury, 2013; Rayne and Forest, 2010; Renner, 2008; Wang et al., 2005a; Wang et al., 2005b; Washington et al., 2009; Young and Mabury, 2010; Zhang et al., 2013; Butt et al., 2014; Rankin et al., 2014; Washington et al., 2015).”

There is no agreement at present how to account for contribution of such precursors to PFOA/PFOS related risk. The list of such precursors is under discussion and growing. There is no established analytical method for precursors of PFOA/PFOS. This a knowledge gap that needs serious investigation.

Thus, for risk assessment in soil, PFOS and PFOA should be considered non-degradable until solid experimental data for Norwegian soil/sludge becomes available.

Moreover, possible presence of their degradable precursors needs to be considered, as actual concentration of PFOS/PFOA can increase with time as a result of transformation of precursors.

4.2.1.3 SCCP

Experimental data on degradation of SCCPs in soil is scarce. Therefore, we use a recent estimate by the leading international group of fate modelers (Krogseth et al., 2013).

Like components of PCB-7, SCCP components are predicted to have very different degradation rates and half-life times, the average half-life time for SCCPs in soil/canopy at 25 °C was 21833 hours or 910 days, or 2.5 years. For the most persistent SCCP congener group (C₁₃H₁₆Cl₁₂), the predicted half-life time in soil/canopy was 18.8 years. The half-lives were calculated for 25° C, while annual average in Norwegian soil can be below 5 °C. At 20 °C lower temperature one can expect 4 times slower degradation at Norwegian conditions, namely, 10 and 74 years for an average and maximal half-life times, respectively.

The safe approach would be to consider SCCP non-degradable until solid experimental data for Norwegian soil/sludge becomes available.

4.2.1.4 Synthetic musks – HHCB, AHTN, OTNE

Experimental results for these substances show big variation, but as a rule their persistency in soil increases in the following order: OTNE < HHCB < AHTN.

Only one study gave a different order: 336 days for AHTN and 900 days for HHCB (Chen et al., 2014 - Field dissipation and risk assessment of typical personal care products TCC, TCS, AHTN and HHCB in biosolid-amended soils (Feng et al., 2014).

EU Risk Assessment report (EU RAR 2008a) referred to field measurements on sludge amended soil that indicated HHCB disappeared almost completely from soil within one year. The half-life based on unfrozen conditions in sludge amended soil studies was around 140 - 145 days (DiFrancesco et al., 2004). The residues in soil after one year ranged from below 10% to 14% of the initial concentrations.

The highest experiment-based estimates are 2-24 years for AHTN and 2.5 years for HHCB (Chen et al., 2014).

No reliable estimate is available for OTNE, but it is probably less than 1 year.

Attention must be paid to these substances on the grounds of known high concentrations in sludge and soil:

“Tonalide (AHTN) levels measured in biosolids from multiple wastewater treatment plants around the world range from 0.032 to 427 mg/kg, and the average concentration is 9.3 mg/kg. Following the application of biosolids containing tonalide to land, the concentration of tonalide predicted in soil is 0.001–13.1 mg/kg (Langdon et al., 2010). The few reported concentrations of tonalide in soil available internationally are in the range of 0.002 to 0.058 mg/kg dry weight (dw)” (Chen et al., 2014).

Taking a soil temperature into consideration there is a probability that DT_{50} at Norwegian conditions might be >5 years for both AHTN and HHCB, though somewhat less for OTNE. Therefore, AHTN and HHCB should be taken as indicators for pollution with this class of substances.

There are no experimental facts available for more accurate estimate of DT_{50} for these three substances.

It is not possible to provide a good estimate. The safe approach would be to consider HHCB, AHTN and OTNE as very persistent and use $DT_{50} = 5$ years in estimates, until solid experimental data for Norwegian soil/sludge becomes available.

4.2.1.5 BDE-209

Our conclusion is primarily based on the results from a recent and sophisticated study by Andrade et al., 2017.

The authors “...conducted a 3-year study to examine the fate of PBDEs in a small-scale 0.24-ha continuously cropped field after a single biosolids application at 72.3 wet tons/ha and determined dissipation half-lives for BDE-47/BDE-99 and BDE-209. In addition, we conducted a large-scale survey of soils from 26 mostly pasture fields at 10 farms with detailed information on timing and rate of biosolids applications. The estimated residence time from the small-scale experiment was 342 d for BDE-47/BDE-99 and 861 d for BDE-209. In the large-scale study, a subset of fields that received a single biosolids application was used to generate another estimate of residence time: 704 d for BDE-47/BDE-99 and 1440 d for BDE-209. These longer residence time estimates were used in three different first-order decay dissipation scenarios (continuous, limited, and no dissipation) to predict PBDE concentration in fields with single and multiple biosolids applications. Results indicate that dissipation occurs primarily in the first 2 years after application, but residues remaining in the soil after this period are likely to be much more tightly bound and less available for degradation.”

From this study we conclude that:

- half-life time for BDE-209 in Virginia, USA is 1440 days (“residence time” time was used as a proxy for Half-life).
- Assuming the average annual soil temperature is 10°C lower in Norway, this value for Norway would be 2880 days or 8 years.
- Moreover, this time might be longer, as degradation slows down after 2 years, likely due to immobilization of very hydrophobic BDE-209 into non-bioavailable compartments within soil.
- The safe approach will be to consider BDE-209 non-degradable until solid experimental data for Norwegian soil/sludge becomes available.

WARNING: the present conclusion does not consider transformation products of BDE-209. Those lower brominated PBDE congeners pose higher risk than BDE-209 itself (Kortenkamp et al., 2014). Moreover, to take transformation products into account monitoring of all other PBDE congeners would be required, with subsequent assessment of individual and combined risk. Careful consideration of these aspects is beyond the scope of current project. As stated by (Kortenkamp et al., 2014) there is evidence that BDE-209 transformation yields lower brominated PBDE, through both abiotic and biotic processes. BDE-209 can therefore act as a slow release reservoir for the more toxic low-brominated congeners.

4.2.1.6 PCB-7

PCB-7, or sum of concentrations of PCB congeners 28, 52, 101, 118, 138, 153 and 180 is a common proxy for PCB contamination.

However, persistency of individual congeners in soil differs dramatically. One of the earlier 5-year study on application of biosolids on farmland in New-York state (Gan and Berthouex, 1994) yielded T_{1/2} of 8 months for PCB-28, 58 months for PCB-52, 311 months for PCB-101, 69 months for PCB-118 and degradation was too slow to be measured for congeners 138, 153 and 180.

Thus, for 5 out of 7 components of PCB-7 parameter half-lives in NY soil were greater than 5 years, which might correspond to half-lives of more than 10 years in Norway.

This behaviour was nicely illustrated in a more recent paper (Terzaghi et al., 2018). In this study half-life shorter than 10 years was found for PCB-28 only. For the 84% of “PCB-7” it was more than 10 years.

The safe approach would be to consider PCB-7 non-degradable until solid experimental data for Norwegian soil/sludge becomes available.

4.2.1.7 Nonylphenol/Nonylphenoxyethoxylates (NP/NPE)

In a review all T_{1/2} for both NP and NPE in soils were less than 20 days (Staples et al., 2001)

Brown et al., 2009 reported half-life of 16 to 23 days for NP in an agricultural soil amended with biosolids in a greenhouse study depending on treatment.

In more recent publication lifetimes of 4-16 days were reported (Dubroca et al., (2005).

Another study gives 25 days for NP, 7-10 days for NPE (González et al., 2012).

It was shown, however, that the application of sludge may lead to 4-9 times increase in NP and NP-Eon concentration within the first days due to fast hydrolysis of higher ethoxylate oligomers.

NP and NPE are degradable. Experiment-based DT₅₀ is < 25 days. Based on this alone, sludge can be applied every year without taking the previous year's application into account.

However, it would be wise to check if sludge contains significant quantities of higher ethoxylates and use a total concentration of all NP derivatives for calculation of the safe applicable amount.

NOTE. In a study (Venkatesan and Halden, 2013), NP compounds showed observable loss from sewage sludge/soil mixtures (1:2), with mean half-lives ranging from 301 to 495 days. The first-order degradation curves for these three compounds suggest mean half-lives of 301, 495, and 462 days for NP, nonylphenol monoethoxylate (NP1EO), and nonylphenol diethoxylate (NP2EO), respectively. These half-lives represent the net loss of compounds through a combination of abiotic and biotic processes including leaching. About 60 to 70% of the compounds were lost from sludge/soil mixtures within 120 days. The authors further stated that the longer half-life observed in this study compared to other studies for these surfactants could have resulted from the higher application rate of sewage sludge to soil (1:2), temporary oxygen-limited conditions after rainfall events and intrinsic properties of the soil and microbial community. They concluded that though previous studies proposed short half-lives for these endocrine disruptors in soil, the extended half-lives shown in their study may prevail in regions with limited oxygen conditions and higher land application rates of sewage sludge.

Though these results must be kept in mind, they are not related to the case directly, as degradation medium (soil:sludge = 2:1) by its nature was a sludge rather than soil.

4.2.2 Uncertainty in selection of an appropriate half-life values for application of biosoils to soil

It is well known that certain chemicals can be persistent in sludge and degradable in soil and *vice versa*. What will be the properties of the medium when soil and sludge are being combined depends largely on the ratio and the way of mixing.

When soil was sieved at 4 mm, thoroughly mixed with sludge (ca 1 – 3% of the amount of soil) in a pot experiment at regulated humidity and temperature, with addition of indigenous earthworms to the setup – in several replicates both NP and NPE dissipated below 0.1-0.6% of the original amount (Rivier et al., 2019).

In contrast, when sludge and soil were mixed in 1:2 ratio, mean half-life times for NP and NPE ranged from 301 to 495 days (Venkatesan and Halden, 2013).

This example shows the scale of uncertainty and that why dissipation needs to be checked experimentally for relevant natural conditions and application methods.

4.2.3 General conclusion

PFOS/PFOA, SCCP, PCB-7 and BDE-209 shall be considered stable and non-degradable.

PFOS/PFOA precursors need to be considered, though no science-based method for this is available.

DEHP and synthetic musks may have DT₅₀ up to 5 years and should be applied accordingly and monitored carefully.

NP/NPE are likely completely degradable within one year, and new portions of fertilizer can be added. However, presence of higher ethoxylates in the sludge must be considered.

In case the actual levels of these substances in sludge pose a risk for application on agricultural soil, they should be used with care and levels in soil should be monitored to determine reliable dissipation rates.

The proposed DT₅₀ are summarised in Table 7.

Table 7. Proposed DT₅₀ in a precautionous approach to degradation based on literature and corrected for Norwegian temperatures.

Contaminant	Max Half-life in soil reported in the literature, (d)	Proposed DT ₅₀ , conservative, temperature corrected for Norway
DEHP	578	5 years
PFOS	Non degradable	99 years (Non degradable) ¹
PFOA	Non degradable	99 years (Non degradable) ¹
HHCB	1825	5 years
AHTN	1825	5 years
OTNE	1825	5 years
SCCP	6800	99 years (Non degradable)
BDE-209	1440	99 years (Non degradable)
PCB 7	Non degradable	99 years (Non degradable)
NP	25	75 days (same as COWI estimate) ¹
NPE	25	75 days

¹ Presence of precursors in fertilizer, at levels much higher than those of regulated substance itself, must be evaluated.

4.2.4 Temperature adjustment of half-lives

Our approach to accounting for soil temperature is based on the theory presented by Matthies and Beulke (Matthies and Beulke, 2017). The Arrhenius equation assumes that the first-order rate constant of degradation depends on the activation energy E_a of the reaction and the temperature at which the reaction occurs.

$$k = A \exp\left(-\frac{E_a}{RT}\right) \quad \text{Eq. 1}$$

Here in the Arrhenius equation: k = degradation rate constant [d^{-1}]. A = factor equal to the rate coefficient at infinite temperature [d^{-1}]. E_a = activation energy [$kJ\ mol^{-1}$]. R = gas constant = $0.008314\ [kJ\ mol^{-1}\ K^{-1}]$ and T = absolute temperature [K]. The degradation half-life is related to the degradation rate constant as $T_{1/2} = \ln(2)/k$. Unfortunately, parameters of Arrhenius equation for the studied chemicals are not available, therefore we used a simplified temperature correction factor, $Q_{10} = 2^7$, which means that degradation slows down by a factor of 2 with decrease of temperature by $10\ ^\circ C$.

We have selected four representative soil observation stations across Norway, the municipalities Ås, Frosta, Fauske and Tromsø (at approximately 60° , 63° , 66° and 69° North) and calculated the average annual soil temperature at depth of 10 cm is in the range from $+4.3^\circ C$ in Holt, Tromsø to $+7.4\ ^\circ C$ in Ås, Oslo area (<http://lmt.nibio.no/>).

This would correspond to 3 and 2.5 longer DT_{50} for Holt, Tromsø and Ås, near Oslo, respectively. Interestingly, the difference in degradation rate between “the warmest” and “the coldest” soil in Norway is just 20%.

The figure (Figure 5) shows modelled decline in contamination for a hypothetical substance with $DT_{50} = 1$ year at $20\ ^\circ C$ and at location in Ås in three model assumptions: orange line – simple approximation using average annual temperature, grey line – monthly temperatures applied, dd - monthly temperatures applied and it is postulated that no degradation occurs when soil is frozen (average monthly temperature below $0\ ^\circ C$).

⁷ [https://en.wikipedia.org/wiki/Q10_\(temperature_coefficient\)](https://en.wikipedia.org/wiki/Q10_(temperature_coefficient))

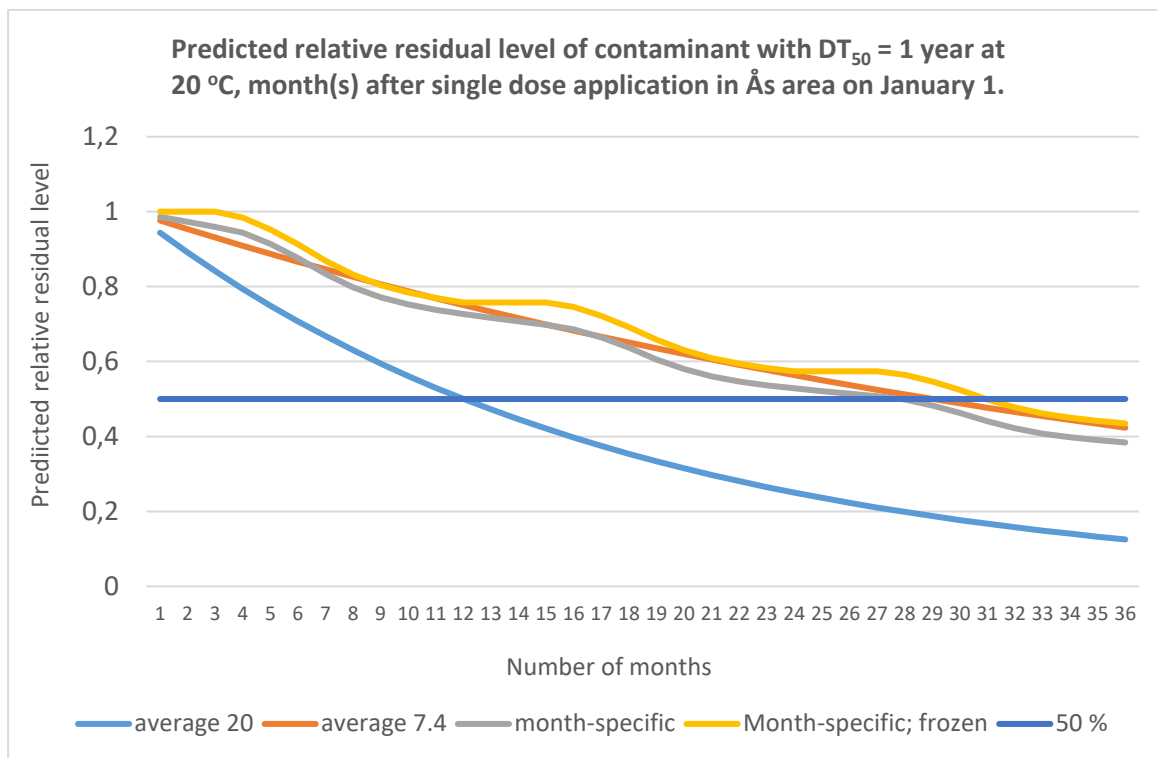


Figure 5. Predicted relative residual level of a contaminant with $DT_{50} = 1$ year at 20°C, month(s) after single dose application in Ås area on January 1st.

Light blue line – at +20 °C

Orange line – for constant average annual temperature +7.4 °C

Grey line – monthly temperatures applied

Yellow line - monthly temperatures applied; no degradation in frozen soil (when average monthly temperature below 0 °C)

Blue line – 50% of initial level

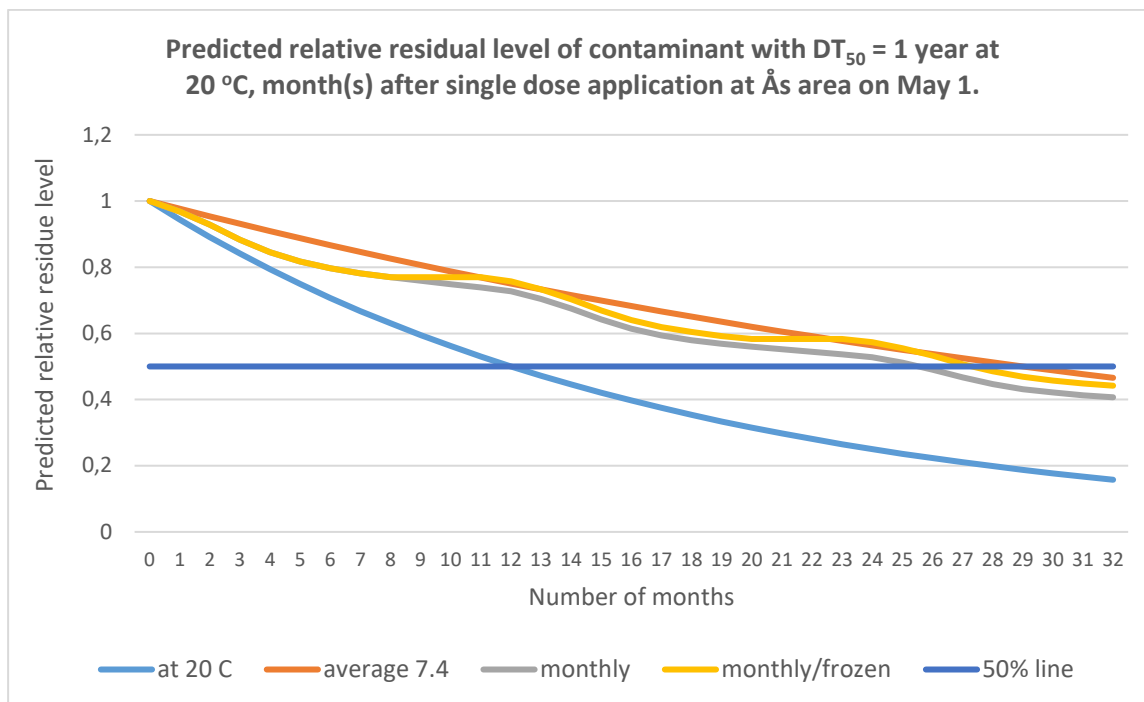


Figure 6. Relative concentration vs time (months) for a substance with $DT_{50} = 1$ year; fertilizer was applied on May 1.

Light blue line – at +20 °C

Orange line – average annual temperature +7.4 °C

Grey line – monthly temperatures applied

Yellow line - monthly temperatures applied; no degradation in frozen soil (when average monthly temperature below 0 °C).

Blue line – 50% of initial level

Taking a seasonal variation (actual monthly temperatures vs year's average) of temperature into account leads to minor variation of DT_{50} for persistent substances (with $DT_{50} = 1$ year at +20 °C) and of their expected levels in soil over time (Figure 6). DT_{50} by these different calculation methods for Ås are from 26 to 31 month instead of 1 year, up to 2.5 time higher. Similar evaluation for year's average of +4.3°C in Tromsø leads to DT_{50} of up to 3 years, or three times higher than at +20 °C.

4.3 Effect of the temperature adjusted DT_{50} values

In order to demonstrate the effect of temperature adjusted DT_{50} values (Table 7) with the DT_{50} values used by COWI (Table 6), similar approaches, assumed same equations, and input parameters which corresponded with COWI's calculations (Blytt et al., 2018), was used to predict the amount of sewage sludge which could be added to soil before reaching predicted acceptable soil levels. The following assumptions and choices have been made in this comparison:

- The normative values proposed in 2016 (Table 3) and not zero concentration were chosen as initial (background) concentration as a worst-case. Since there is a 245-times increase for BDE-209 from current/proposed normative values (0.002 mg/kg dw) to the one proposed in 2016 (0.49 mg/kg dw), it is also chosen to do the same prediction for both normative values in order to demonstrate the effect such a change will cause on predicted DT_{50} ,
- The lowest uncertainty level - 30% laboratory uncertainty and 50% sampling uncertainty - 58 % total uncertainty was used. This uncertainty level was added to the normative value and used as the acceptable soil level,

- The soil concentration after application of fertiliser every 10th year and with use of the 95- and 50-percentile concentration levels of contaminants in sewage sludge (summary of data from 2010 up to 2019, shown in Appendix C) was predicted,
- Both DT₅₀ used by COWI (Table 6) and new adjusted DT₅₀ (Table 7) was applied.

Based on these assumptions, the predicted concentrations in soil at day 0 and after 10 years (3650 days), and the day when soil concentration reached the acceptable soil level, has been predicted (Table 8).

Using the DT_{50s} from COWI (Blytt et al., 2018); DEHP, AHTN and HHCB are the only HOCs evaluated not to meet the acceptable soil level immediately after application. Application of sludge with 95% percentile concentration of the given HOCs, needed 265, 565 and 1425 days, respectively, to meet the MLs. When assuming use of sludge with an average concentration of AHTN and HHCB, the acceptable soil level was estimated meet at 325 and 1158 days, respectively.

For BDE-209 and use of normative value 0.49 mg/kg dw (proposed 2016) and DT₅₀ 360 days (Blytt et al., 2018), the soil concentration was below the acceptable soil level immediately after application, also for application of 95% percentile sludge concentration (1.5 mg/kg dw). Same predictions but with use of normative value 0.002 mg/kg dw (current value), the acceptable soil level was reach after 915 and 1555 days after application of 95% percentile and average sludge concentration (0.41 mg/kg dw), respectively.

If temperature adjusted DT₅₀ is used, the application of sewage sludge with average concentration of the given HOCs, do not meet the acceptable soil level within 10 years for HHCB and AHTN, and for BDE-209 when the normative value 0.002 mg/kg dw was applied. With application of sewage sludge at the 95% percentile concentration (57.1 mg/kg dw), it was estimated that DEHP would meet the acceptable soil concentration 1285 days after application.

Acceptable soil concentrations (58% uncertainty added to normative values proposed 2016) for HHCB and AHTN are 0.016 and 0.017 mg/kg dw soil, and 10 years after application of sludge with 95% a concentration of HHCB and AHTN, the predicted soil concentration was 0.101 and 0.052 mg/kg dw, respectively. The predicted soil concentration for BDE-209 10 years after application of 95% percentile concentration in sludge, was 0.025 mg/kg dw. Compared to acceptable soil level concentration based on 0.002 mg/kg dw normative value (current value), this gives around 8 times higher soil concentration after 10 years application than the acceptable soil level. Comparing the estimated BDE-209 soil concentration after 10 year obtained with use of DT₅₀ 360 days (Blytt et al., 2018), 0.509 mg/kg dw, with the acceptable soil concentration based on the normative value 0.002 mg/kg dw, the difference is 160 times higher. For HHCB and AHTN, soil concentrations after 10 years are 6 and 3 times higher than acceptable soil level, respectively.

These examples support the importance of which DT₅₀ and normative values in soil are used in the approach for selection acceptable concentrations in fertilisers and soil mixtures.

Since the methodology for developing scientifically based MLs for HOCs is not yet established, new MLs are not proposed. However, this report demonstrates how one important parameter, DT₅₀, affect the outcome of a risk assessment.

Table 8. Predicted concentration in soil at day 0 to day 3650 (10 year) of selected HOCs after application of 40 tonnes sewage sludge per hectare per 10 year with use of mean and 95% percentile concentration of HOCs in sewage sludge (Table AC-2 Appendix C) with use of COWIs approach and methodology. Approximately days before reaching acceptable soil concentration, calculated from normative values in soil (normative value + 58% uncertainty) is present. DT₅₀ values proposed by COWI (Table 6) are compared with temperature adjusted DT₅₀ values (Table 7). *For BDE-209 both normative values 0.002 and 0.49 were used.

NewDT ₅₀	Norm. value soil	Accep. conc. soil	95% conc. SS	Mean conc. SS	Conc. Soil after 95% percentile conc. SS added			Conc. soil after mean conc. SS added		
					Day 0	Day 3650	Days to acceptable soil conc.	Day 0	Day 3650	Days to acceptable soil conc.
mg/kg dw										
DEHP	1.000	1.580	57.10	27.60	1.950	1.228	1285	1.459	1.106	below at start
PFOS	0.002	0.0032	0.060	0.014	0.0030	0.0029	below at start	0.0022	0.0022	below at start
PFOA	0.011	0.017	0.0047	0.0013	0.0111	0.0110	below at start	0.0110	0.0109	below at start
SCCP	0.090	0.142	2.43	0.57	0.100	0.098	below at start	0.100	0.098	below at start
HHCB	0.010	0.016	22.00	9.98	0.377	0.101	>>10 yr	0.176	0.051	>10 yr
AHTN	0.011	0.017	9.95	2.50	0.177	0.052	>10 yr	0.053	0.021	>10 yr
*BDE-209	0.490	0.774	1.50	0.41	0.515	0.509	below at start	0.497	0.492	below at start
PCB7	0.004	0.0063	0.056	0.017	0.0049	0.0048	below at start	0.0043	0.0042	below at start
NP+NPE	0.035	0.055	8.46	4.13	0.176	0.035	below at start	0.104	0.035	below at start
*BDE-209	0.002	0.003	1.500	0.406	0.027	0.025	>>10 yr	0.009	0.008	>10 yr

DT ₅₀ COWI	Norm. value soil	Accep. conc. soil	95% conc. SS	Mean conc. SS	Conc. Soil after 95% percentile conc. SS added			Conc. soil after mean conc. SS added		
					Day 0	Day 3650	Days to acceptable soil conc.	Day 0	Day 3650	Days to acceptable soil conc.
Substrate										
					mg/kg dw					
DEHP	1.000	1.580	57.10	27.60	1.942	0.993	265	1.456	0.992	below at start
HHCB	0.010	0.016	22.00	9.98	0.377	0.010	1425	0.176	0.010	1158
AHTN	0.011	0.017	9.95	2.50	0.177	0.011	565	0.053	0.011	325
*BDE-209	0.002	0.003	1.500	0.406	0.027	0.002	1555	0.009	0.002	915

Important considerations

Dissipation of organic contaminants in soil is a complex process which includes aerobic and anaerobic microbial transformation, abiotic chemical transformation, volatilization, dissolution and removal with water, mechanical removal with particles as airborne dust or waterborne suspended solids.

Accordingly, reduction of concentration of a substance depends on these fundamental factors, such as microbiome, composition of sludge and soil, application method, agricultural practices, weather conditions and may vary from sludge to sludge, from soil to soil, from location to location, from season to season and from year to year.

This complexity makes correct experiment and representative sampling difficult, and this is, in our opinion, a reason for large variations in reported dissipation rates (which are often confused with transformation rates).

As a result, it is impossible to select values that can be directly applied to the case of WWTP sludge in Norwegian soil in general, despite many reported experimental and calculated half-life or dissipation times.

A safe and conservative approach is to rely on experimental data, whenever possible – on experiments with application of real sludge – and use the highest reported value in further considerations.

Soil temperature matters. Most experiments in laboratory were normally carried out at +20 °C or +25°C, or “at ambient temperature”. Experiments in the field were carried out in areas with much higher average annual soil temperatures, than those typical for Norway (e.g. +4.2 °C in Tromsø and +7.4 °C in Ås at depth of 10 cm).

There exists no approved method to account for influence of temperature on degradation in soil.

Q10 = 2 – approach was selected under these circumstances, which means that dissipation is assumed to be 2 times slower with every 10°C of temperature drop.

This translates to 2.5 times (for Ås) or 3 times (for Tromsø) longer dissipation times than at +20 °C.

Within this approach, the selected HOCs formed three groups: with DT₅₀ = 99 years, 5 years and 75 days, respectively.

For the substances from the first group (PFOS, PFOA, SCCP, BDE-209, PCB-7; DT₅₀ = 99 years) there will be presumed no dissipation in an observable time, therefore a fertilizer might be applied according to the current level of contamination in sludge until the accumulated amount in soil reaches the ML.

For the substances from the second group (DEHP, HHCB, AHTN, OTNE; DT₅₀ = 5 years), fertilizer can be applied according to the current level of contamination in sludge until the accumulated amount in soil reaches the ML and then the real level and its decline shall be monitored until solid scientific evidence is formed for “personalized” recommendations.

For the substances from the third group (NP, NPE; DT₅₀ = 75 days), it is highly likely that fertilizer can be applied every year, according to the current level of contamination until the accumulated amount reaches the ML. However, actual residual levels in soil need to be checked before every year’s application unless these would be found negligible repeatedly.

A special attention needs to be paid to precursors of selected contaminants and a possibility of release of the latter shortly after application of fertilizer to soil, leading to dramatic increase of their content. Such phenomena are known for NP/NPE and suspected for PFOA/PFOS.

The temperature adjusted DT₅₀ were up to 100 times higher for PFOS and BDE-209 (from 360 days to 36135 days (99 years)). For DEHP, HHCB and AHTN the temperature adjusted DT₅₀s were in the range of 5-15 times higher, and for NP and NPE, the DT₅₀ was similar, 75 days.

With the DT₅₀s from COWI and application of sludge with 95% percentile concentration of the HOCs, DEHP, AHTN and HHCB were the only HOCs that did not meet the acceptable soil level immediately, but 265, 565 and 1425 days, respectively, after application. Application of sludge with average concentration of AHTN and HHCB, the acceptable soil level was met after 325 and 1158 days, respectively.

For BDE-209 the soil concentration was below the acceptable soil level immediately after application – even with application of 95% percentile sludge concentration - when the proposed normative value 0.49 mg/kg dw and DT₅₀ 360 days (COWI) was used. Same predictions (DT₅₀ 360 days) but with use of a normative value 0.002 mg/kg dw (current), the acceptable soil level was reached after 915 days (2.5 years) and 1555 days (4.2 years) after application of 95% percentile and mean sludge concentration, respectively.

Use of the adjusted DT₅₀ and current normative values at 0.002 mg/kg dw, the predicted soil concentration (PEC) for BDE-209 10 years after application of 95% percentile concentration in sludge, was 0.025 mg/kg dw. Which is around 8 times higher than acceptable soil concentration (0.003 mg/kg dw). Comparing the estimated BDE-209 soil concentration after 10 year with DT₅₀ 360 days and normative value 0.49 mg/kg dw (used by COWI), 0.509 mg/kg dw, with the acceptable soil concentration based on the normative value 0.002 mg/kg dw, the difference is 160 times higher.

This example supports the importance of a precautional approach for selection of DT₅₀, but also regarding use of normative values in soil as basis for selecting acceptable concentrations in soil used for crop production.

5 Current levels of the selected HOCs

5.1 Presence of selected HOCs in agricultural soil

Data of the present concentration of HOCs in agricultural soil is scarce, and for some of the selected HOCs in this project not found at all. Based on searches on Web of Science and Google Scholar, a few fields studies were found and are summarised in Table 9.

A long-term microscale field experiment in The Czech Republic where soil analyses of HOCs after application of mineral NPK fertiliser, manure and two different loading rates of sewage sludge has recently been published (Pulkrabova et al., 2019). In the Czech study, the concentration level of AHTN and HHCB were in the range of 2-24 µg/kg dw and 4-50 µg/kg dw, respectively. They concluded that PFCs, brominated flame retardants and synthetic musk compounds tended to increase in the sewage sludge treated plots.

The most abundant PBDEs in the Czech study were BDE-47 and -99 (both identified in 78% of the samples), but also BDE-100 (62% of the samples) and BDE-183 (57% of the samples) were frequently detected. The remaining congeners occurred at detectable levels in less than 50% of the samples. The individual congeners with highest concentration were BDE-209 (from 0.5 to 5.9 µg/kg dw) and BDE-206 (from 0.05 to 2.8 µg/kg dw), but they were only detected in 30% of the samples. Venkatesan and Halden (2014) found that the initial brominated flame retardants amount in sludge/soil mixtures (1:2) persisted over the monitoring duration of 3 years.

Among the analysed musk compounds only HHCB (most abundant, identified in 88% of the soil samples), AHTN (70% of the soil samples), and cashmeran (28% of the soil samples) were identified. The total content of the compounds varied between 6.22 and 80.9 µg/kg dw. The predominant compound in all samples was HHCB, varying between 1.6 and 49.9 µg/kg dw. Although the differences among the individual locations were negligible, the content of musk compounds increased with increasing sewage sludge application rate at all locations.

The branched- and linear-isomers of PFOS and PFOA were detected in 90%, 83%, and 43% of the samples, respectively. The total PFOS content varied between < 0.1–2.5 µg/kg dw and for PFOA between < 0.3–0.9 µg/kg dw. There was no significant difference in the PFAS content among the locations, but fertilisation of the soil with sewage sludge was the highest contributor of PFASs to soil pollution. Another interesting fact is that the ratio between branched- and linear-PFOS was 1:4 in all locations and all types of fertilization, which corresponds to the ratio of PFOS isomers in the technical mixture. This result indicates that PFOS probably does not undergo transformation between its forms in soil.

Higher contents of PFOA and PFOS in soil were observed in a study from the USA (Xiao et al., 2015). PFOA and PFOS were detected in all samples and concentrations ranged between 0.2–28.2 µg/kg dw for PFOA and 5.5–125.7 µg/kg dw for PFOS. Like the study by Pulkrabova et al (2019), concentrations of PFOS were higher than of PFOA.

Enrichment of PFOS, PBDEs (47, 99, 209), HHCB, AHTN and DEHP in soils added sludge compared to control soil is also shown in a report from Naturvårdsverket (Österås et al., 2015). The concentrations of BDE-209 in soils were much higher in this study compared to the reported values from same field in 2000. Qualitatively this was in line with model calculations showing that BDE-209 will persist in soils and thus accumulate over time upon repeated sludge amendments (Sternbeck et al., 2011). In the report from Naturvårdsverket, a Spanish field study is referred to which also demonstrated that BDE-209 accumulates more strongly in soil than BDE47 or BDE99 (Eljarrat et al., 2008). A North American field study is also referred to in which it is stated that accumulation over

time for BDE47 and 99 is reported (Xia et al., 2010). In this field study, the sludge had been applied for 33 years and at much higher application rates than in Skåne.

Österås et al., 2015 referred also to other data where PBDEs, PCB, PFOS and PFOA were observed to accumulate after repeated application of sludge, while no long-term accumulation of NP and DEHP was reported after sludge application. A report from SWECO (2010) and a study by DiFrancesco et al., (2004) report few samples where galaxolide was identified, and assumed field removal.

While the concentrations of the HHCb and AHTN in the control plots were much lower than the plots added sewage sludge, the difference between control and sludge added plots were not less for BDE-209, PFOS, PFOA and PCB7 (Österås et al., 2015). PFOS and BDE-209 were shown to accumulate in soil, but due to contradictory results it was not concluded whether HHCb also accumulate in soil or not.

We recommended field studies in which the concentration of the HOCs in the applied sludge are known, and where leaching and runoff processes of HOCs are studied.

Table 9. Summary of measured soil concentration of the selected HOCs in agricultural soil, given as µg/kg dw. Details about sludge application and sampling is given below¹.

HOC	Substrate	Conc. (µg/kg dw)	Country	Ref.
DEHP	Control	<50	Sweden	Österås et al., 2015
DEHP	SS1	<50-230, B: <50-110	Sweden	Österås et al., 2015
DEHP	SS2	A: <50-100, B: <50-100	Sweden	Österås et al., 2015
PFOS	Control	<2,0	Sweden	Österås et al., 2015
PFOS	SS1	A:<2.0-0.5, B: <2.0	Sweden	Österås et al., 2015
PFOS	SS2	A:<2.0-3.7, B: <2.0-5.1	Sweden	Österås et al., 2015
PFOS	Soil SS1	0.9 - 1.2 (min-max)	Czeck	Pulkrabová et al., 2019
PFOS	Soil SS2	1.7 - 2.5 (min-max)	Czeck	Pulkrabová et al., 2019
PFOS	Control	0.2-0.4 (min-max)	Czeck	Pulkrabová et al., 2019
PFOA	Soil SS1	0.5 - 0.6 (min-max)	Czeck	Pulkrabová et al., 2019
PFOA	Soil SS2	0.8 - 0.9 (min-max)	Czeck	Pulkrabová et al., 2019
PFOA	Control	<0.3 - 0.3 (min-max)	Czeck	Pulkrabová et al., 2019
HHCb	Control	A: <2.0-2.6; B: <2.0	Sweden	Österås et al., 2015
HHCb	SS1	A:<2.0-5.1, B: <2.0-13	Sweden	Österås et al., 2015
HHCb	SS2	A:<2.0-5.9, B: <2.0-70	Sweden	Österås et al., 2015
HHCb	SS-3 yr appl.SS	1.7-2.6	Sweden	Österås et al., 2015
HHCb	Soil SS1	4.1 - 49.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
HHCb	Soil SS2	4.1 - 49.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
HHCb	Control	0.4 - 2.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
AHNT	Control	A: <2.0-28; B: <2.0-16	Sweden	Österås et al., 2015
AHNT	SS1	A:<2.0-5.4, B: <2.0-8.3	Sweden	Österås et al., 2015

HOC	Substrate	Conc. (µg/kg dw)	Country	Ref.
AHNT	SS2	A:<2.0-18, B: <2.0-29	Sweden	Österås et al., 2015
AHNT	SS-3 yr appl.SS	2.8-5.9	Sweden	Österås et al., 2015
AHNT	Soil SS1	2.1 - 12.1 (min-max of median)	Czeck	Pulkrabová et al., 2019
AHNT	Soil SS2	2.7- 23.7 (min-max of median)	Czeck	Pulkrabová et al., 2019
AHNT	Control	0.4 - 2.1 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-209	Control	A: <1.0; B: <1.0	Sweden	Österås et al., 2015
PBDE-209	SS1	A:1-6, B: <1.0-3.6	Sweden	Österås et al., 2015
PBDE-209	SS2	A:1.1-16, B: 1.2-5.9	Sweden	Österås et al., 2015
PBDE-209	Soil SS1	1.5 - 2.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-209	Soil SS2	1.9 - 4.4 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-209	Control	<1-5.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-207	Soil SS1, SS2	0.3 - 0.9 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-207	Control	<0.1-1.3 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-206	Soil SS1, SS2	0.3 - 0.8 (min-max of median)	Czeck	Pulkrabová et al., 2019
PBDE-206	Control	0.3 - 1.8 (min-max of median)	Czeck	Pulkrabová et al., 2019
BDE 47	Soil SS1, SS2	0.16 - 0.38 (min-max of median)	Czeck	Pulkrabová et al., 2019
BDE 47	Control	0.2 - 0.3 (min-max of median)	Czeck	Pulkrabová et al., 2019
BDE 99	Soil SS1, SS2	0.17 - 0.41 (min-max of median)	Czeck	Pulkrabová et al., 2019
BDE 99	Control	0.01 - 0.04 (min-max of median)	Czeck	Pulkrabová et al., 2019
PCB7	Soil SS1	2.2 - 6.4 (min-max of median)	Czeck	Pulkrabová et al., 2019
PCB7	Control	0.6-3.2 (min-max of median)	Czeck	Pulkrabová et al., 2019
NP ²	Control/SS1/SS2	1 of 36 samples>LOQ; 16	Sweden	Österås et al., 2015

¹Study by Österås et al., 2015: Sewage sludge (SS) added every 4th year. SS1 - 4 ton/ha, SS2 12 ton/ha- Soil sampled at 0-0.3 m depth and 0.3-0.6 m depth; in this table numbers are pooled. A= Igelösa, B=Petersborg. Study by Pulkrabová et al., 2019: data from 4 sites, n=3 at each site, min and max referee to min and max median values at these 4 sites. Samples from 0-3 m. ²NP: Only one sample > LOQ, 10µg/kg dw. At field B and SS2, measured on sample at 16 µg/kg dw.

5.2 Observed levels in Norwegian sewage sludge

5.2.1 Norwegian monitoring campaign for contaminants in Norwegian WWTPs

Since 1996-97 a five-yearly monitoring campaign has been conducted by Norsk Vann BA (and supported with funding from the Norwegian Environment Agency) at selected WWTPs and sludge treatment facilities to get an overview of the presence of selected organic and inorganic pollutants in Norwegian sewage sludge. Some of the WWTPs have been participating in every campaign since the start. The last campaign was conducted from October 2017 to February 2018 and reported in March 2019. The 18 sewage treatment plants (see Table 34 in Section 7.3) that participated in this campaign produce in total 65 029 tons dry weight of sludge in 2017, which constitute approximately 54% of the

total 121 508 tons dw produced that year. Figure 7 shows disposition of the sludge, and for example was 54% of all the sludge used as soil conditioner on farmland.

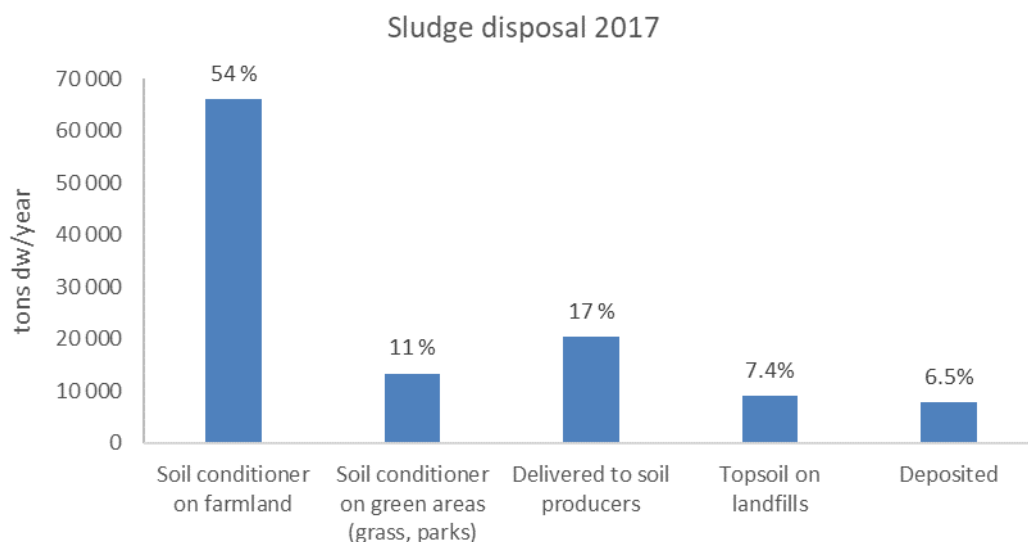


Figure 7. Disposal of sludge from Norwegian WWTPs in 2017. Source: KOSTRA/SSB.

The samples have been collected as monthly composite samples⁸ of daily grab samples during week days over five months during each sampling campaign, hence the contaminant levels should be regarded as average values for each month. The results from the last sampling campaign have been used in the assessment of expected current levels of the selected HOCs in sewage sludge from Norwegian WWTPs for the compounds that were included: DEHP, PFOA, PFOS, SCCP, HHCB, AHTN, BDE-209, PCB7, NP and NPE. Previous sampling campaigns have been used to assess the trend. Hence, the only compound not included is OTNE, for which we have used values from international refereed literature, prioritising levels found in sewage sludge from Scandinavian WWTPs.

Data from the 2012/2013 and 2017/2018 campaign shows some trends for the selected contaminants.

5.2.1.1 DEHP

As indicated in Figure 8, the present levels of DEHP (blue bars) in Norwegian sewage sludge appears to be relatively close to the ML value of 50 mg/kg dw COWI proposed for DEHP in fertiliser products. However, there has been a distinct decreasing trend in DEHP levels since the campaign in 2012-13.

DEHP was detected in all samples (95 of 95). At three of the treatment plants the measured level in at least one sample exceeded the proposed ML value of 50 mg DEHP/kg dw (see Table 10); Lindum Biogas (treated sludge), Solumstrand WWTP (raw sewage sludge) and Årim (raw sewage sludge). The latter is raw septic sludge from two different WWTPs in Ålesund. The two other plants are tightly connected, since the (pre-treated) landfill leachate from Lindum is a significant contribution to the overall influent to Solumstrand WWTP (up to ca. 10% of the volumetric load), and Lindum receives and treats all the sewage sludge that is produced at Solumstrand. Previous reports of the landfill leachate at Lindum have shown high levels of DEHP, suggesting that the sources are linked to deposited material at the landfill.

⁸ Except for siloxanes, which have been collected as grab samples to minimise losses and contamination.

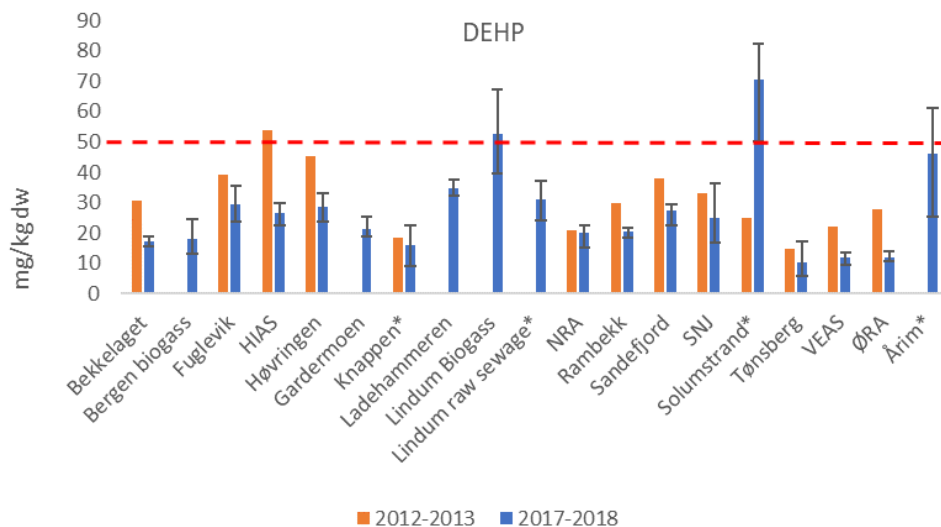


Figure 8. Average levels of DEHP in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018 (blue bars). At the treatment plants marked with * only dewatered raw sewage sludge (i.e. prior to digestion, composting or lime addition) has been sampled. The error bars indicate the maximum and minimum measured values at each plant. Orange bars show the average levels from the campaign in 2012-13. The red dotted line indicates the proposed ML for fertiliser products. Data from Blytt and Stang (2019).

Table 10. Results from the 5-yearly monitoring program: DEHP (Blytt and Stang, 2019). Red numbers indicate when the proposed ML (same for adjusted and non-adjusted) in the fertiliser product is exceeded.

Treatment facility	DEHP (mg/kg dw)				#>ML (50 mg/kg dw)
	Average	Median	Max	Min	
Bekkelaget	17.1	17.3	18.7	15.5	0
Bergen biogas	18.2	16.9	24.7	13.0	0
Fuglevik	29.6	29.4	35.5	23.7	0
HIAS	26.4	26.2	30.0	22.3	0
Høvringen	28.5	27.8	32.9	23.7	0
Gardermoen	21.3	19.1	25.4	18.7	0
Knappen (raw sludge)	16.1	15.8	22.5	8.9	0
Ladehammeren	34.5	33.8	37.7	32.1	0
Lindum Biogas	52.7	52.1	67.4	39.5	3
Lindum (raw sludge)	30.9	30.2	37.2	24.1	0
NRA	20.0	20.7	22.3	15.3	0
Rambekk	20.2	20.8	21.7	18.2	0
Sandefjord	27.4	28.5	29.3	22.4	0
SNJ	24.8	24.3	36.5	16.7	0
Solumstrand (raw sludge)	70.7	73.4	82.6	50.2	5
Tønsberg	10.4	9.1	17.3	5.8	0
VEAS	11.7	11.9	13.6	9.3	0
ØRA	11.9	10.9	13.9	10.5	0
Årim (raw sludge)	46.0	48.3	61.4		2
Average		27.3			
Median		23.7			10 of 95 (10.5%)
Min-max		5.8-82.6			

NRA: Nedre Romerike Avløpselskap; SNJ: Sentralrenseanlegget Nord-Jæren (IVAR), VEAS: Vestfjorden Avløpselskap.

5.2.1.2 PFOS

PFOS was detected in all sludge samples (80 of 80), but the levels were in general much lower than the proposed ML of 100 µg PFOS/kg dw (Figure 9). However, at the WWTPs where airport runoff is a significant part of the contribution to the influent (i.e. Gardermoen and Fuglevik), the levels of PFOS were significantly higher than elsewhere, but only one sample exceeding the proposed ML (Table 11).

Note that, since 2007 PFOS is no longer in use at fire-fighting sites at Norwegian airports, but the soil and groundwaters in the area is polluted from long-term usage (Amundsen et al., 2009), hence, the levels in the runoff may prevail for a long time.

There is a long list of potential precursors to PFOS through abiotic and/or biotic transformation processes (OECD, 2007; Australian Government, 2019⁹). Since PFOS is expected to be more recalcitrant than these precursors, the levels of PFOS is expected to increase due to transformation of these precursors if present. However, so far, their transformation rates in the environment have not been established and appear to be difficult to predict.

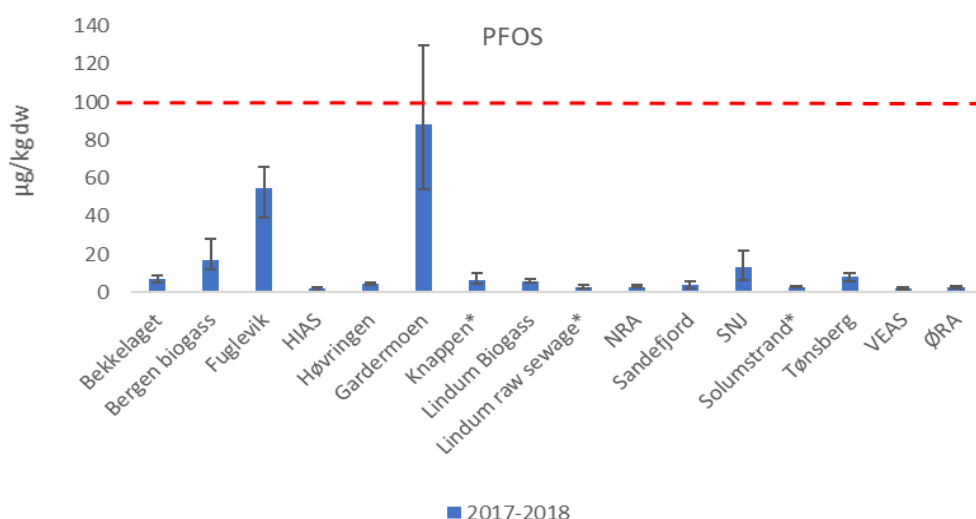


Figure 9. Average levels of PFOS in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018. At the treatment plants marked with * only dewatered raw sewage sludge (i.e. prior to digestion, composting or lime addition) has been sampled. The error bars indicate the maximum and minimum measured values at each plant. The red dotted line indicates the proposed ML for fertiliser products. Data from Blytt and Stang (2019).

⁹ Australian Government (2019) Indirect precursors to perfluorooctanesulfonate (PFOS): Environment tier II assessment. Australian Government, Department of Health, March 8 2019. <https://www.nicnas.gov.au/chemical-information/imap-assessments/imap-assessments/tier-ii-environment-assessments/indirect-precursors-to-perfluorooctanesulfonate-pfos>

Table 11. Results from the 5-yearly monitoring program: PFOS (Blytt and Stang, 2019). Red numbers indicate when the proposed ML (same for adjusted and non-adjusted) in the fertiliser product is exceeded.

Treatment facility	PFOS ($\mu\text{g}/\text{kg dw}$)				#>ML (100 $\mu\text{g}/\text{kg dw}$)
	Average	Median	Max	Min	
Bekkelaget	6.7	6.6	8.7	5.2	0
Bergen biogas	17.2	16.0	28	12.0	0
Fuglevik	55	56	66	39	0
HIAS	1.8	1.9	2.5	1.1	0
Høvringen	4.5	4.4	5.0	4.1	0
Gardermoen	88	100	130	54	1
Knappen (raw sludge)	6.6	6.8	10.0	4.7	0
Ladehammeren	-	-	-	-	-
Lindum Biogas	5.7	5.3	6.9	5.1	0
Lindum (raw sludge)	2.7	2.6	3.6	1.9	0
NRA	2.8	2.5	3.9	2.4	0
Rambekk	-	-	-	-	-
Sandefjord	3.8	3.9	5.5	2.0	0
SNJ	12.9	12.0	22	6.6	0
Solumstrand (raw sludge)	2.9	3.0	3.3	2.5	0
Tønsberg	8.0	8.8	9.9	6.0	0
VEAS	1.9	2.0	2.4	1.3	0
ØRA	2.8	2.9	3.5	2.1	0
Årim (raw sludge)	-	-	-	-	-
Average	14				1 of 80 (1.3%)
Median	4.9				
Min-max	1.1-130				

5.2.1.3 PFOA

The levels of PFOA in the sewage sludge samples were in general low (only 8 out of 80 samples >LOD) and all values were far below the proposed ML of 100 μg PFOA/kg dw (Figure 10 and Table 12). One of the samples from Fuglevik WWTP was higher than all the rest (all other measurements from the same plant were <LOD).

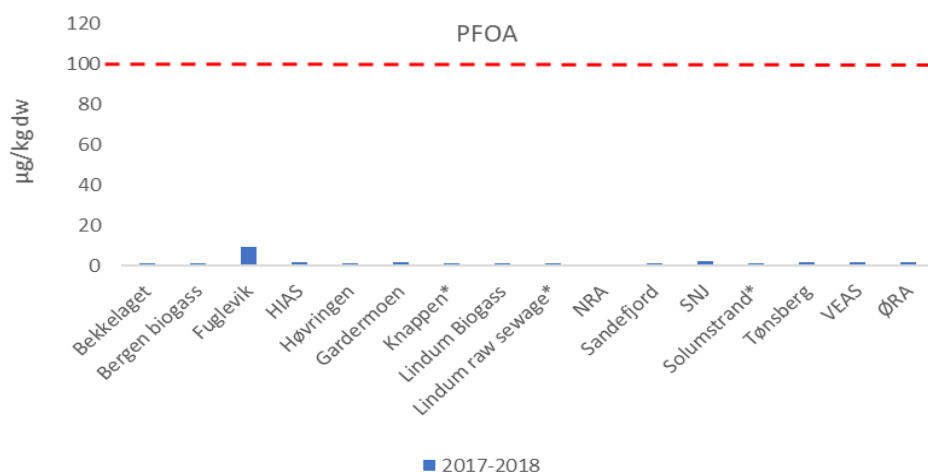


Figure 10. Average levels of PFOA in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018. * only dewatered raw sewage sludge (i.e. prior to digestion, composting or lime addition) has been sampled. Orange bars shows the average levels from the campaign in 2012-13. The red dotted line indicates the proposed ML for fertiliser products. Data from Blytt and Stang (2019).

Table 12. Results from the 5-yearly monitoring program: PFOA (Blytt and Stang, 2019).

Treatment facility	PFOA (µg/kg dw)				#>ML (100 µg/kg dw)
	Average	Median	Max	Min	
Bekkelaget	-	-	1.1	<0.89	0
Bergen biogas	-	-	<0.93	<0.82	0
Fuglevik	-	-	9.2	<1	0
HIAS	-	-	1.5	<1	0
Høvringen	-	-	<1.2	<0.87	0
Gardermoen	-	-	<1.6	<1.1	0
Knappen (raw sludge)	-	-	<1.2	<0.83	0
Ladehammeren	-	-	-	-	-
Lindum Biogas	-	-	<1.3	<1.1	0
Lindum (raw sludge)	-	-	<0.99	<0.67	0
NRA	-	-	<0.76	<0.66	0
Rambekk	-	-	-	-	-
Sandefjord	-	-	<1.2	<1.1	0
SNJ	-	-	2.0	<1.5	0
Solumstrand (raw sludge)	-	-	<0.9	<0.9	0
Tønsberg	-	-	1.5	<0.89	0
VEAS	-	-	<1.5	<1.4	0
ØRA	-	-	<1.6	<1.3	0
Årim (raw sludge)	-	-	-	-	-
Average	-				0 of 80 (0%)
Median	-				
Min-max	<0.66-9.2				

5.2.1.4 SCCP

SCCP was detected in 68 of 70 sludge samples. The levels varied quite a bit between the treatment facilities, but were relatively stable at each location, indicating somewhat stable sources and/or effects from the applied treatment steps (Figure 11). The proposed ML of 2000 µg SCCP/kg dw was exceeded in one sample from Solumstrand WWTP and in two samples from Tønsberg WWTP (Table 13). It is interesting to note that the thermophilic hydrolysis and anaerobic digestion at Lindum Biogas, which also treats the sludge from Solumstrand WWTP, drastically reduced the level of SCCP (Figure 11), whereas the sludge treatments at Tønsberg WWTP and NRA do not include any biological treatment, only the addition of quick lime (the Orsa method). Tønsberg WWTP receives reject water from Greve biogas plant and leachate from two landfills. All the raw sewage sludge samples (Knappen, Lindum raw sewage and Solumstrand) had high values, while all facilities with anaerobic digestion had low values.

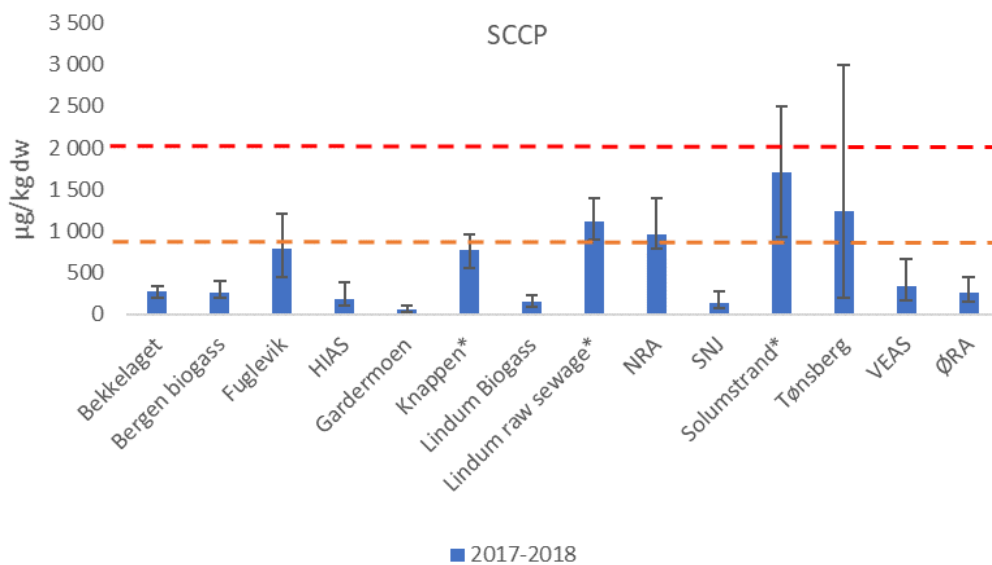


Figure 11. Average levels of SCCP in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018. The red and orange dotted lines indicate the proposed adjusted (2 mg SCCP/kg dw) and a non-adjusted (0.9 mg SCCP/kg dw) MLs for fertiliser products, respectively. Data from Blytt and Stang (2019).

Table 13. Results from the 5-yearly monitoring program: SCCP (Blytt and Stang, 2019). 50% of LOD is used to calculate the average value if <LOD. Yellow background indicates when the proposed adjusted ML in the fertiliser product have been exceeded, while red numbers indicate when the proposed non-adjusted ML have been exceeded.

Treatment facility	SCCP ($\mu\text{g}/\text{kg dw}$)				#>non-adjusted ML (0.9 mg/kg dw)	#>adjusted ML (2 mg/kg dw)
	Average	Median	Max	Min		
Bekkelaget	266	290	330	190	0	0
Bergen biogas	264	210	400	190	0	0
Fuglevik	792	650	1 200	440	2	0
HIAS	182	120	390	110	0	0
Høvringen	-	-	-	-	-	-
Gardermoen	55	61	100	<50	0	0
Knappen (raw sludge)	778	870	950	560	1	0
Ladehammeren	-	-	-	-	-	-
Lindum Biogas	150	160	220	86	0	0
Lindum (raw sludge)	1 118	1 100	1 400	890	4	0
NRA	962	850	1 400	780	2	0
Rambekk	-	-	-	-	-	-
Sandefjord	-	-	-	-	-	-
SNJ	139	110	270	72	0	0
Solumstrand (raw sludge)	1 704	1 600	2 500	920	5	1
Tønsberg	1 236	270	3 000	190	2	2
VEAS	332	240	660	170	0	0
ØRA	258	240	440	150	0	0
Årim (raw sludge)	-	-	-	-	-	-
Average	588 ¹⁰				16 of 70 (23%)	3 of 70 (4.3%)
Median	295					
Min-max	<50-3000					

5.2.1.5 HHCb (galaxolid)

The levels of HHCb have increased considerably (approximately doubled) since the campaign in 2012-13, and at several of the treatment facilities and in 57% of all collected samples the levels exceeded the proposed ML of 10 mg HHCb/kg dw (Figure 12 and Table 14)

In fact, both the average and median values exceeded the ML, and at seven facilities even the lowest measured value exceed this limit (Table 14). If the trend continues the ML may be exceeded in all sludge samples in a few years.

¹⁰ These average, median and maximum values do not coincide with the reported values by Blytt and Stang (2019), however, apparently the same measured values are used to calculate these numbers.

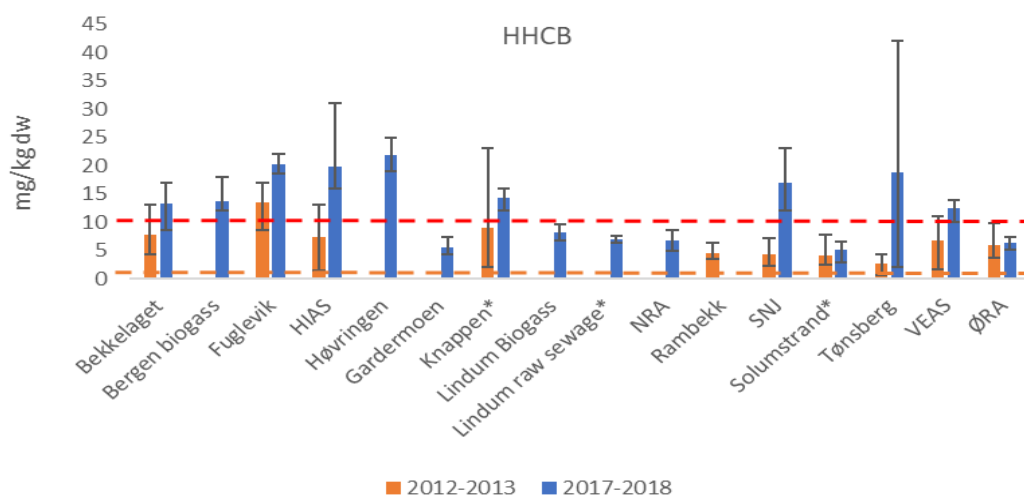


Figure 12. Average levels of HHCb in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018 (blue bars). Orange bars shows the average levels from the campaign in 2012-13. The red and orange dotted lines indicate the proposed adjusted (10 mg HHCb/kg dw) and non-adjusted (0.5 mg HHCb/kg dw) MLs for fertiliser products, respectively. Data from Blytt and Stang (2019).

Table 14. Results from the 5-yearly monitoring program: HHCb (Blytt and Stang, 2019) given as mg/kg dw. Yellow background indicates when the proposed adjusted ML in the fertiliser product have been exceeded, while red numbers indicate when the proposed non-adjusted ML have been exceeded.

Treatment facility	HHCb (mg/kg dw)				#>non-adjusted ML (0.5 mg/kg dw)	#>adjusted ML (10 mg/kg dw)
	Average	Median	Max	Min		
Bekkelaget	13.3	12.0	17.0	8.60	5	4
Bergen biogas	13.6	13.0	18.0	12.0	5	5
Fuglevik	20.2	19.5	22.0	18.5	5	5
HIAS	19.8	17.0	31.0	16.0	5	5
Høvringen	21.8	22.0	25.0	19.0	5	5
Gardermoen	5.62	5.50	7.30	4.20	5	0
Knappen (raw sludge)	14.4	15.0	16.0	12.0	5	5
Ladehammeren	-	-	-	-	-	-
Lindum Biogas ¹	8.10	7.90	9.70	6.70	3	0
Lindum (raw sludge) ¹	6.95	-	7.60	6.30	2	0
NRA	6.70	6.70	8.60	5.00	5	0
Rambekk	-	-	-	-	-	-
Sandefjord	-	-	-	-	-	-
SNJ	17.0	16.0	23.0	12.0	5	5
Solumstrand (raw sludge)	5.14	5.70	6.60	2.80	5	0
Tønsberg	18.8	5.60	42.0	2.00	5	2
VEAS	12.4	12.0	14.0	10.0	5	4
ØRA	6.38	6.50	7.40	5.10	5	0
Årim (raw sludge)	-	-	-	-	-	-
Average	13.1				70 of 70 (100%)	40 of 70 (57%)
Median	12.0					
Min-max	2.00-42					

¹ At Lindum, only three samples of treated sludge (Lindum Biogas) and two samples of raw sludge.

5.2.1.6 AHTN (tonalide)

Also, the levels of AHTN had increased considerably since the campaign in 2012-13 (average value increased by ca. 140% and the median value increased by ca. 150%), but the values were in general much lower than the proposed adjusted ML of 10 mg AHTN/kg dw, but higher than the proposed non-adjusted ML of 0.6 mg/kg dw (Figure 13 Figure 13 and Table 15). However, if the trend continues this limit may be exceeded in a few years at some treatment facilities.

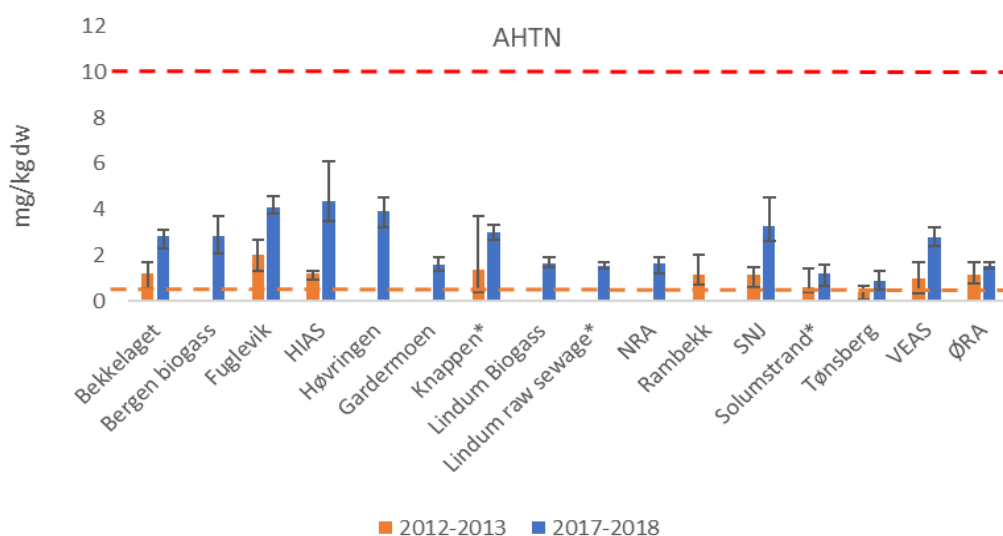


Figure 13. Average of AHTN in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018 (blue bars). Orange bars shows the average levels from the campaign in 2012-13. Orange bars shows the average levels from the campaign in 2012-13. The red and orange dotted lines indicate the proposed adjusted (10 mg AHTN/kg dw) and not adjusted (0.6 mg AHTN/kg dw) MLs for fertiliser products, respectively. Data from Blytt and Stang (2019).

Table 15. Results from the 5-yearly monitoring program: AHTN (Blytt and Stang, 2019). Red background indicates when the proposed adjusted ML in the fertiliser product have been exceeded, while red numbers indicate when the proposed non-adjusted ML have been exceeded.

Treatment facility	AHTN (mg/kg dw)				#>non-adjusted ML (0.6 mg/kg dw)	#>adjusted ML (10 mg/kg dw)
	Average	Median	Max	Min		
Bekkelaget	2.86	3.00	3.10	2.30	5	0
Bergen biogas	2.86	2.90	3.70	2.10	5	0
Fuglevik	4.10	4.00	4.60	3.80	5	0
HIAS	4.38	4.20	6.10	3.50	5	0
Høvringen	3.90	4.10	4.50	3.20	5	0
Gardermoen	1.58	1.60	1.90	1.30	5	0
Knappen (raw sludge)	3.02	3.10	3.30	2.70	5	0
Ladehammeren	-	-	-	-	-	-
Lindum Biogas ¹	1.67	1.60	1.90	1.50	3	0
Lindum (raw sludge) ¹	1.55	-	1.70	1.40	2	0
NRA	1.62	1.80	1.90	1.20	5	0
Rambekk	-	-	-	-	-	-
Sandefjord	-	-	-	-	-	-
SNJ	3.26	3.20	4.50	2.60	5	0

Treatment facility	AHTN (mg/kg dw)				#>non-adjusted ML (0.6 mg/kg dw)	#>adjusted ML (10 mg/kg dw)
	Average	Median	Max	Min		
Solumstrand (raw sludge)	1.23	1.30	1.60	0.66	5	0
Tønsberg	0.88	0.97	1.30	0.51	4	0
VEAS	2.80	2.80	3.20	2.40	5	0
ØRA	1.54	1.60	1.70	1.40	5	0
Årim (raw sludge)	-	-	-	-	-	-
Average	2.55				69 of 70 (99%)	0 of 70 (0%)
Median	2.65					
Min-max	0.51-6.10					

¹ At Lindum, only three samples of treated sludge (Lindum Biogas) and two samples of raw sludge.

5.2.1.7 OTNE

OTNE has not been included in the monitoring campaigns conducted by Norsk Vann BA. Very few data are available for OTNE in Norwegian sewage sludge, the only ones we have found are from the Norwegian EPA screening programme in 2017 (Konieczny et al., 2018), which included three weekly composite samples of the final sludge from both VEAS (11 mg/kg dw) and HIAS (16-18 mg/kg dw). Hence, all these values are above the proposed ML of 10 mg OTNE/kg dw.

Other analysis found of OTNE in sewage sludge in the range of 7300-30700 ug/kg dw (Difrancesco et al., 2004), 3700±690 ug/kg dw (Bester et al., 2008), 656±353 ug/kg dw (Japan) (Ozaki et al., 2017).

5.2.1.8 BDE-209

BDE-209 was detected in 90 out of 95 collected sludge samples during the 2017-18 campaign. The levels varied quite a bit between the treatment facilities and at each location (Figure 14). 20% of the values exceeded the proposed ML of 500 µg BDE-209/kg dw (Table 16). There is no pronounced trend in the levels since the 2012-13-campaign (Figure 14). At Årim, receiving septic sludge from two areas, a very high level in one sample (3400 µg/kg dw) brought the average up to above the threshold value, while the other measured values were below (94-380 µg/kg dw).

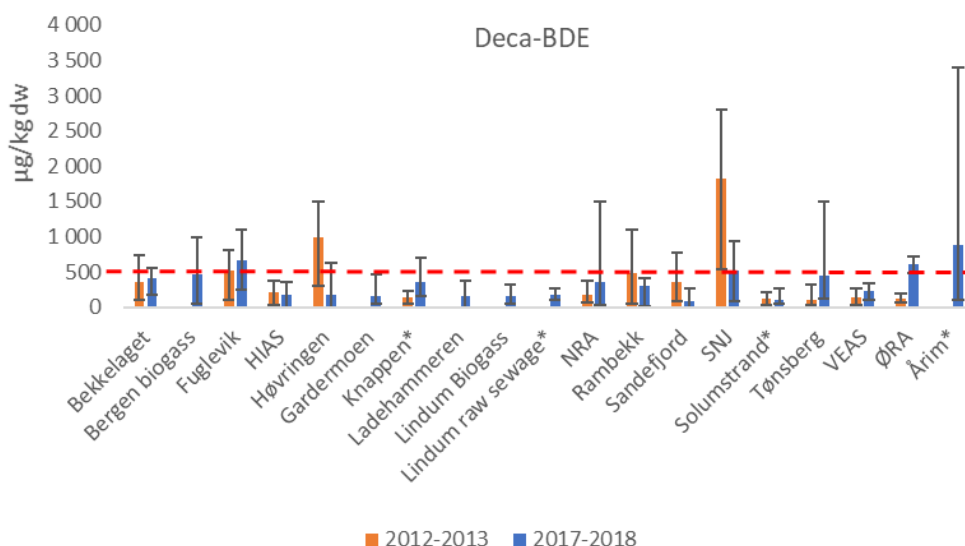


Figure 14. Average of BDE-209 in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018 (blue bars). Orange bars shows the average levels from the campaign in 2012-13. The red dotted line indicates the proposed ML for fertiliser products. Data from Blytt and Stang (2019).

Table 16. Results from the 5-yearly monitoring program: BDE-209 given as $\mu\text{g}/\text{kg dw}$ (Blytt and Stang, 2019). Yellow background indicates when the proposed ML (same for both adjusted and non-adjusted) in the fertiliser product have been exceeded.

Treatment facility	BDE-209 ($\mu\text{g}/\text{kg dw}$)				#>ML (500 $\mu\text{g}/\text{kg dw}$)
	Average	Median	Max	Min	
Bekkelaget	406	500	560	180	2
Bergen biogas	463	330	990	44	2
Fuglevik	670	660	1 100	250	4
HIAS	218	180	360	<2	0
Høvringen	220	93	620	<2	1
Gardermoen	156	100	460	50	0
Knappen (raw sludge)	362	300	700	150	1
Ladehammeren	208	207	370	<2	0
Lindum Biogas	161	130	320	46	0
Lindum (raw sludge)	184	160	270	110	0
NRA	355	75	1 500	30	1
Rambekk	307	370	410	14	0
Sandefjord	135	120	270	<	0
SNJ	526	740	930	92	3
Solumstrand (raw sludge)	105	67	260	40	0
Tønsberg	454	230	1 500	120	1
VEAS	234	230	340	100	0
ØRA	604	620	720	490	4
Årim (raw sludge)	889	320	3 400	94	1
Average	360				19 of 95 (20%)
Median	245				
Min-max	14-3400				

5.2.1.9 PCB7

PCB7 was detected in 91 out of 95 collected sludge samples during the 2017-18 campaign, and 18 of these exceeded the proposed ML of 20 $\mu\text{g PCB7}/\text{kg dw}$ (Figure 15 and Table 16). However, 14 of these samples were collected at Lindum (in both treated and raw sludge) and at Solumstrand WWTP (raw sludge), which as explained in Section 5.2.1.1, are tightly connected. At several of the other treatment facilities the measured levels are rather close to the proposed ML with an average value as high as 17 $\mu\text{g PCB7}/\text{kg dw}$. The values appear to be relatively stable at each location. The measured values at Tønsberg WWTP were surprisingly low (<1-1 mg/kg dw) considering that this WWTP receives leachate from two landfills.

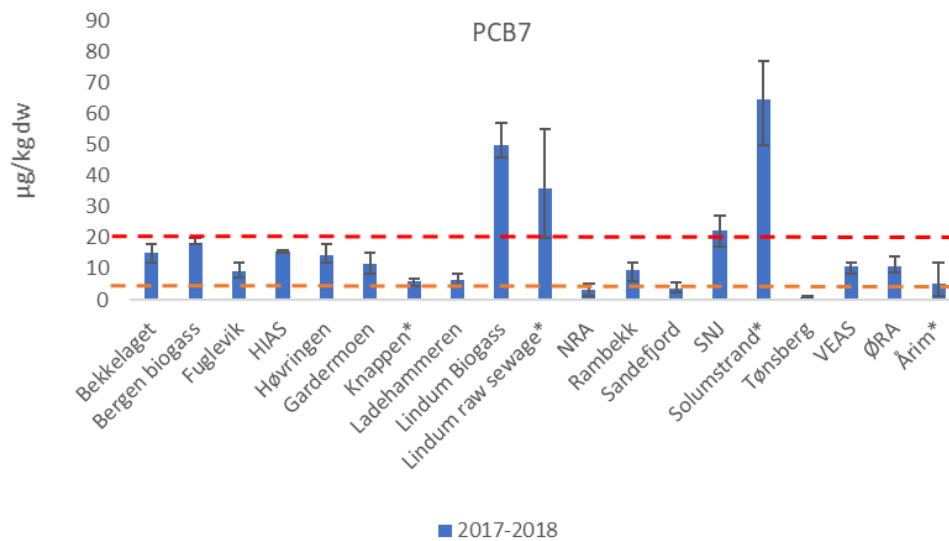


Figure 15. Average levels of PCB7 in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018. The red and orange dotted lines indicate the proposed adjusted (20 µg PCB7/kg dw) and non-adjusted (4 µg PCB7/kg dw) MLs for fertiliser products, respectively. Data from Blytt and Stang (2019).

Table 17. Results from the 5-yearly monitoring program: PCB7 given as µg/kg dw (Blytt and Stang, 2019). Yellow background indicates when the proposed adjusted ML in the fertiliser product have been exceeded, while red numbers indicate when the proposed non-adjusted ML have been exceeded.

Treatment facility	PCB7 (µg/kg dw)				#>non-adjusted ML (4 µg/kg dw)	#>adjusted ML (20 µg/kg dw)
	Average	Median	Max	Min		
Bekkelaget	15	16	18	12	5	0
Bergen biogas	18	18	20	18	5	0
Fuglevik	9.0	9.0	12	7.0	5	0
HIAS	15	15	16	15	5	0
Høvringen	14	14	18	12	5	0
Gardermoen	11.5	11.0	15	8.3	5	0
Knappen (raw sludge)	6.1	6.3	6.8	4.4	5	0
Ladehammeren	6.4	5.9	8.2	4.9	5	0
Lindum Biogas	50	47	57	46	5	5
Lindum (raw sludge)	36	35	55	20	5	4
NRA	3.1	2.4	5.0	1.1	2	0
Rambekk	9.5	9.8	12	6.0	5	0
Sandefjord	3.5	3.0	5.7	2.5	1	0
SNJ	22	22	27	17	5	4
Solumstrand (raw sludge)	64	66	77	50	5	5
Tønsberg	<1	<1	1	<1	0	0
VEAS	10.7	11.0	12	8.3	5	0
ØRA	10.9	11.0	14	8.7	5	0
Årim (raw sludge)	5.4	4.5	12	1.3	3	0
Average	17				78 of 95 (82%)	18 of 95 (19%)
Median	11					
Min-max	<1-77					

5.2.1.10 NP og NPE

Both NP and NPE were detected in all 95 collected sludge samples during the 2017-18 campaign (Table 18). The levels of 4-nonylphenol (4-NP) were considerably lower than NP and below the LOD of 20 µg 4-NP/kg dw in 56 of the samples. In the 2012-13 campaign only 4-NP (not NP) was included in the analyses and these levels were then all below the detection limit (8-50 µg/kg dw).

In the 2017-18 campaign only one sample did exceed the proposed threshold value of 10 000 µg NP+NPE/kg dw (10 mg/kg dw), but at several treatment facilities individual samples were relatively close to the limit (Figure 16). Though the NP values were relatively high, the measured values of the ethoxylates (mono- and di-) dominated the combined NP+NPE values. The analyses did not include longer ethoxylates, which can degrade to the shorter mono- and diethoxylates, nor did they include the nonylphenol carboxylates, which can degrade to NP. Hence, the levels indicated in Figure 16 and Table 18 may underestimate the level of NP + NPE in the sewage sludge.

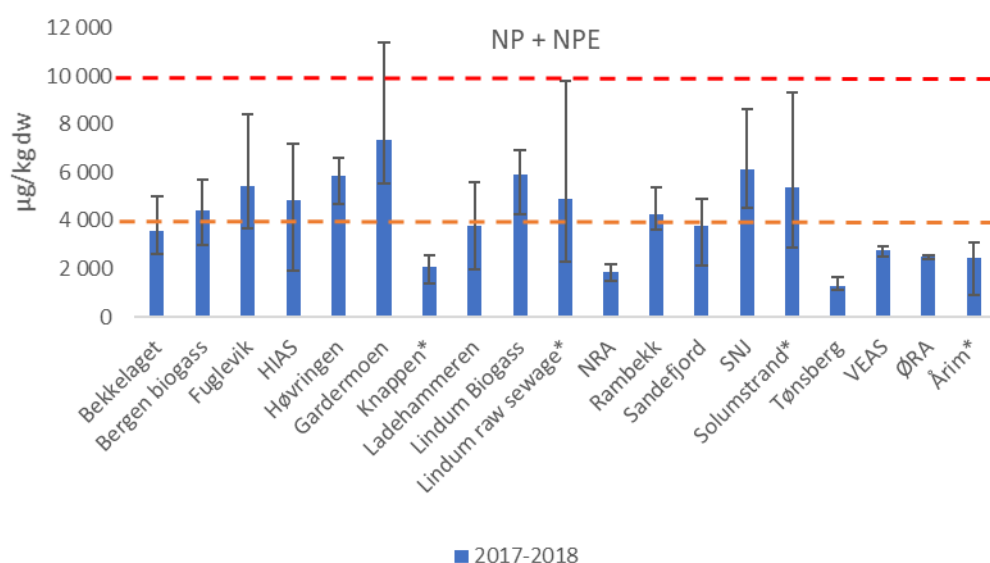


Figure 16. Average levels of NP tech and NPE in sewage sludge at the 18 Norwegian treatment plants that participated in the sampling campaign from October 2017 to February 2018. The red and orange dotted lines indicate the proposed adjusted (10 mg NP+NPE/kg dw) and non-adjusted (4 mg NP+NPE/kg dw) MLs for fertiliser products, respectively. Data from Blytt and Stang (2019).

Table 18. Results from the 5-yearly monitoring program: NP and NPE (Blytt and Stang, 2019). Yellow background indicates when the proposed adjusted ML in the fertiliser product have been exceeded, while red numbers indicate when the proposed non-adjusted ML have been exceeded.

Treatment facility	NP and NPE ($\mu\text{g}/\text{kg dw}$)				#>non-adjusted ML (4 mg/kg dw)	#>adjusted ML (10 mg/kg dw)	
	Average	Median	Max	Min			
Bekkelaget	NP	726	720	770	680	-	
	NP+NPE	3 558	2 850	5 000	2 590	2	
Bergen biogas	NP	936	880	1 200	840	-	
	NP+NPE	4 432	4 690	5 700	2 960	3	
Fuglevik	NP	664	650	800	500	-	
	NP+NPE	5 416	5 030	8 420	3 650	4	
HIAS	NP	894	820	1 200	810	-	
	NP+NPE	4 840	4 750	7 200	1 910	4	
Høvringen	NP	1 260	1 300	1 400	1 000	-	
	NP+NPE	5 840	6 100	6 600	4 700	5	
Gardermoen	NP	1 300	1 300	1 500	1 100	-	
	NP+NPE	7 330	6 600	11 400	5 510	5	1
Knappen (raw sludge)	NP	340	340	400	280	-	
	NP+NPE	2 064	2 330	2 580	1 380	0	
Ladehammeren	NP	842	770	1 200	610	-	
	NP+NPE	3 778	4 000	5 590	2 000	2	
Lindum Biogas	NP	1 840	1 900	2 100	1 600	-	
	NP+NPE	5 900	6 160	6 930	4 270	5	
Lindum (raw sludge)	NP	1 330	1 300	2 200	550	-	
	NP+NPE	4 878	3 930	9 800	2 270	2	
NRA	NP	290	290	420	160	-	
	NP+NPE	1 846	1 880	2 200	1 500	0	
Rambekk	NP	1 400	1 400	1 700	1 200	-	
	NP+NPE	4 250	3 880	5 400	3 610	2	
Sandefjord	NP	894	890	1 100	610	-	
	NP+NPE	3 806	3 850	4 890	2 130	2	
SNJ	NP	2 720	2 600	4 100	1 700	-	
	NP+NPE	6 142	5 680	8 600	4 500	5	
Solumstrand (raw sludge)	NP	1 940	2 100	2 600	1 000	-	
	NP+NPE	5 354	4 340	9 300	2 880	3	
Tønsberg	NP	462	440	570	350	-	
	NP+NPE	1 292	1 240	1 670	1 100	0	
VEAS	NP	1 048	1 100	1 100	970	-	
	NP+NPE	2 766	2 910	2 940	2 480	0	
ØRA	NP	690	630	870	570	-	
	NP+NPE	2 502	2 530	2 580	2 390	0	
Årim (raw sludge)	NP	932	940	1 200	380	-	
	NP+NPE	2 428	2 860	3 090	900	0	
Average	NP	1 079					
	NP+NPE	4 127					
Median	NP	940				44 of 95	1 of 95
	NP+NPE	3 850				(46%)	(1.1%)
Min-max	NP	160-4 100					
	NP+NPE	900-11 400					

5.2.1.11 Summary

Table 21 summarises the number of samples that exceeded the proposed ML for each compound at the treatment plants that were sampled during the 2017-18 campaign. As an overall assessment, a background colour code is used to indicate how close to the proposed ML the sewage sludge at each plant appears to be for the individual compounds. See Table 19. Only two of the treatment plants passed the criteria for the category orange or better; i.e. the WWTPs Ladehammeren and Sandefjord. However, data is missing for some of the compounds.

The treatment facilities are also categorised according to the treatment that had been applied to the sludge prior to sampling.

Table 21 summarises the general considerations related to the exceedances of the adjusted ML values.

Table 19. Colour coding to indicate how close to the proposed adjusted ML by COWI (Blytt et al., 2018) the sewage sludge at a given treatment plant appears to be.

Colour category	Relative to the proposed ML
GREEN	All measured values ¹ were <10% of the adjusted ML.
YELLOW	All measured values were below 75% of the adjusted ML and more than 50% of the values (≥3 if 5 samples) below 25% of the adjusted ML.
ORANGE	All measured values were below the adjusted ML and more than 50% of the values (≥3 if 5 samples) below 75% of the adjusted ML.
RED	At least one measured value above the adjusted ML value or a majority of the measured values (≥3 if 5 samples) above 75% of the adjusted ML.

¹ In all collected sludge samples at the given treatment facility.

Table 20. Expected current levels of selected HOCs in sewage sludge at selected Norwegian treatment facilities relative to the respective proposed adjusted ML values. The number of samples that exceeded the ML during the last sampling campaign in 2017-2018 (Blytt and Stang, 2019) are shown in each square (ref. Tables 10-18). The background colour code indicates how close to the proposed ML the sewage sludge at a given treatment plant appears to be (see Table 19).

Treatment facility	Sludge treatment	Exceedance of MLs										
		DEHP	PFOS	PFOA	SCCP	HCHB	AHTN	OTNE	BDE-209	PCB7	NP+NPE	SUM
Raw dewatered sewage sludge												
Knappen (raw sludge)	Dewatered	0	0	0	0	5	0		1	0	0	6
Lindum (raw sludge)	Dewatered	0	0	0	0	0	0		0	4	0	4
Solumstrand (raw sludge)	Dewatered	5	0	0	1	0	0		0	5	0	11
Årim (raw sludge)	Dewatered	2	-	-	-	-	-		1	0	0	3
Lime stabilisation												
NRA	Lime (Orsa Method)	0	0	0	0	0	0		1	0	0	1
Tønsberg	Lime (Orsa Method)	0	0	0	2	2	0		1	0	0	5
Mesophilic anaerobic digestion												
Høvringen	Pasteurisation + AN	0	0	0	-	5	0		1	0	0	6
Ladehammeren	AN	0	-	-	-	-	-		0	0	0	
Rambekk	AN + thermal drying	0	-	-	-	-	-		0	0	0	
SNJ	AN + thermal drying	0	0	0	0	5	0		3	4	0	12
Sandefjord	AN	0	0	0	-	-	-		0	0	0	
VEAS	AN + lime + filter press drying	0	0	0	0	4	0		0	0	0	4
Thermophilic anaerobic digestion												
Bekkelaget	AN	0	0	0	0	4	0		2	0	0	6
Bergen biogas	Pasteurisation + AN	0	0	0	0	5	0		2	0	0	7
Gardermoen	AN	0	1	0	0	0	0		0	0	1	2
ØRA	AN	0	0	0	0	0	0		4	0	0	4
Aerobic and anaerobic digestion												
Fuglevik	Thermo-A + meso-AN	0	0	0	0	5	0		4	0	0	9
Thermal hydrolysis and mesophilic digestion												
Lindum Biogas	Cambi + AN	3	0	0	0	0	0		0	5	0	5
HIAS	Cambi + AN	0	0	0	0	5	0		0	0	0	5
SUM		7	1	0	3	40	0		20	18	1	

Table 21. Overall considerations of exceedance of adjusted MLs.

Substance	Main considerations
DEHP	Exceedances may be related to landfill leachate as a mediator. Levels are relatively high but are generally decreasing.
PFOS	High levels correlate well with runoff from historic usage of PFOS in fire-fighting foams (e.g. at certain airports). The levels elsewhere are generally low. However, a wide range of potential precursors to PFOS may be present in the same sludge. Their levels and potential transformation rates are largely unknown.
PFOA	The levels are generally low, but as for PFOS, a wide range of potential precursors to PFAS may be present in the same sludge. Also, here the levels and potential transformation rates are largely unknown.
SCCP	The sludges that had been treated by anaerobic digestion had all low levels, while the ML was exceeded in sludges that has not been subjected to biological treatment.
HHCB	The levels have increased considerably the last five years and the proposed ML was exceeded in more than 50% of the samples.
AHTN	The levels have increased considerably the last five years. However, the proposed ML was not exceeded in any samples. But if the trend continues the ML may be exceeded at some plants.
OTNE	Very limited data exist, but all available data exceed the proposed ML value.
BDE-209	The levels were relatively high in most of the sludge samples and exceeded the proposed ML in 20% of the samples. The levels appear to be relatively stable since the 2012-13-campaign.
PCB7	PCB7 were detected in almost all samples, and often close to the proposed ML. However, 14 out of the 18 samples that exceeded this limit were connected to Lindum and Solumstrand WWTP, indicating the local landfill leachate as an important mediator.
NP+NPE	The proposed ML was exceeded in one out of 95 samples, but the general level was relatively high, particularly of NPE (ref. Table 18). However, precursors to NP (e.g. NPnCO and other NPnEO; n=3-20) that were not included in the analyses may be present.

5.3 Sampling and analytical issues

Here we discuss issues related to sampling, sample preparation and analyses related to the selected HOCs in fertilizer products with a focus on final sewage sludge and compost.

5.3.1 Sampling methodology

Norwegian Food Safety Authority (Mattilsynet, 2012) has prepared guidelines for sampling of sewage sludge, compost and other waste-based fertiliser products. The reader is referred to these for guidelines for details. Here we will only comment selected issues of concern to sampling for the analysis of HOCs:

Sampling location in the production process:

- Since some of the HOCs may leave with the reject water during dewatering, samples for the analysis of HOCs should be done on the final dewatered product. However, if lime is used in the stabilising process, the sample need to be taken prior to the addition of lime.
- If the sludge-based product is to be used as raw material in another fertiliser product, sampling should be done prior to mixing with other components, since the quality of the raw material in terms of contaminants (i.e. PTEs and HOCs) need to comply with Norwegian fertiliser regulation criterie for when materials are allowed to be used in fertilisers.

Sampling frequency:

- The sampling frequency must be chosen based on the volume size the sample should represent, either in the form of a given amount of produced product or production over a given period. The frequency may differ from parameter to parameter.
- Important questions stated in the guidelines:
 - Is the process stable or is there a lot of disturbance and constant process changes?
 - Do the raw materials change frequently?
 - Are new types of waste or sludge constantly being added to the plant and how do the raw materials vary throughout the year?
 - What does analytical results from previous years tell?
 - Do the results vary widely?
 - Is there any pattern in the variation?
 - Is there more variation in some parameters than others?
 - Do any of the parameters often lie close to or above the limit values laid down in the regulations?
- As noted in the guidelines: A sample represent the total volume/time period from which it was taken. If you choose to sample rarely, the sample represents a larger volume. When the result of the sample shows too high contents of a HOC (or heavy metal), the analytical result will represent the whole batch. In general, one can say that large variations indicate the need to sample frequently. Good process control can reduce the variations and then one can also decrease the sampling frequency.

Sampling equipment and methodology:

- See the guidelines.

Sampling containers, preservation and storage:

- See Table 22.
- PFOA and PFOS: Fluoropolymeric plastics, including PTFE (polyetrafluoroethene) and rubber materials should be avoided during sampling, sample storage and extraction because they can contaminate the samples with the substances.
- Non-disposable sample containers and lids for semi-volatile analysis should be washed with a phosphate-free detergent solution, followed by thorough rinses with hot tap-water and analyte-free water. The last step should be an acetone rinse. The lids should be in place on the container during

the rinse step (solvent in the container with the lid tightly screwed down) because the solvents can rinse the plastic from the interior screw threads onto the PTFE lining.

- For analysis of volatile organic compounds, sample containers, screw caps and septa (silicone vapour barriers) should be washed with a phosphate-free detergent, rinsed once with tap-water, rinsed at least twice with analyte-free water, then dried at a temperature greater than 105°C. A solvent rinse should generally be avoided because it can interfere with analysis, although a methanol rinse is acceptable.
- Alternatively, single use disposable containers and lids may be used for samples with both semi-volatile and volatile organic compounds.

Table 22. Recommended containers, preservation and storage conditions and maximum storage time for the selected HOCs according to NS-EN ISO 5667-15 (2009).

Compound	Material in container	Preservation and storage conditions	Maximum storage time
PCBs	Glass with PTFE-lined cap	1-5°C, dark and airtight	1 month
Semi- and non-volatile organic compounds: DEHP ¹ , SCCP, BDE-209, NP and NPE	Glass with PTFE-lined cap	Extract and store at 1-5°C, dark and airtight	1 month
		Extract and store at ≤-18°C, dark and airtight	6 months
PFOS, PFOA	Glass with aluminium foil	Extract and store at 1-5°C, dark and airtight	1 month
	HDPE bottle ²	Extract and store at ≤-18°C, dark and airtight	6 months
Volatile organics as-received: HHCB, AHTN, OTNE/Iso E Super	Glass with PTFE-lined cap	1-5°C, dark and airtight	4 days
		Extract with methanol and store at 1-5°C, dark and airtight	1 month
		Extract with methanol and store at ≤-18°C, dark and airtight	6 months

¹ According to one supplier (Arkema, 2014¹¹) of Rilsan plastics, the Rilsan plastic bags do not contain or are not likely to contain any phthalates such as DEHP, ² Fluoropolymeric plastics, including PTFE (polytetrafluoroethylene) and rubber materials, should be avoided during sampling, sample storage and extraction (WRC Group, 2008).

¹¹

<https://www.fosterpolymers.com/upload/medialibrary/1f1/RILSAN%C2%AE%20BESVO%20A%20MED%20Regulatory%20Letter%202-3-2014.pdf>

5.3.2 Recommended extraction methodologies and analytical methods with appropriate LOQs

5.3.2.1 Inherent analytical challenges

From an analytical point of view, sewage sludge as a matrix is challenging because it is not uniform in its composition. The concentrations of HOCs vary widely (see Tables 10-18), and sewage sludge contains a variety of other components that may interfere with the analysis of the compounds of interest, also after extraction; e.g. lipids and other naturally occurring materials, as well as materials that may have been added during the treatment such as surfactants, coagulants, polymers or lime. These components can manifest themselves as interferences at all stages of the analytical process from sample preparation, so it is critical to remove them from the sample extracts using established clean-up procedures (Díaz-Cruz et al., 2009).

Moreover, the analytical procedures and analyses should also include the potential presence of known precursors (in the sludge sample) that may be transformed to one of the selected HOCs after the sewage sludge has been applied as a fertiliser or soil amendment.

In the following comments are given to some of the selected HOCs.

5.3.2.1.1 PFOS, PFOA:

Due to their relatively low volatility, good solubility in water and lack of chromophores the analysis of perfluorinated substances is a challenging task. Several precautions should be taken in order to avoid background contamination in the analytical blanks (van Leeuwen et al., 2006). Contamination sources of PFCs in laboratories have not yet been well characterized, but one of the major sources of contamination known is the contact with laboratory materials made of fluoropolymers, such as polytetrafluoroethylene or perfluoroalkoxy compounds (Díaz-Cruz et al., 2009).

There are many difficult-to-measure and unidentified precursors to both PFOA and PFOS. Studies have been conducted to develop methods where all PFOA-related substances are converted into PFOA by chemical oxidation prior to analysis, hence simulating the actual oxidative transformation potential that may occur in the environment (e.g. Houtz and Sedlak, 2012), but such methods are still in the development at research level and not ready for regulatory application (ECHA RAC, 2015). ECHA RAC (2015) also recommends that the Commission takes advice about the length of time needed to develop suitable analytical methods that can be applied to all matrices, since this might affect the length of the transitional period.

5.3.2.1.2 NP and NPE

It is important to note that NP is not a single chemical structure. It is a complex mixture of highly branched nonylphenols, largely mono-substituted in the para- position, but with small amounts of ortho- and di-substituted nonylphenols. See Table 17. The branched 4-nonylphenol is the most widely produced and marketed nonylphenol, though many manufacturers incorrectly use the linear identity when referring to the branched nonylphenol (US EPA, 2010). Laboratories report NP in different and potentially inconsistent ways, typically either as 4-n-nonylphenol, nonylphenol or nonylphenol tech. Blytt and Stang (2019) noted that 4-n-nonylphenol and nonylphenol tech often is interpreted as equivalents, but when they used both analytical methods on the same 95 sludge samples from Norwegian WWTPs, they found 4-n-nonylphenol above the LOD (20 µg/kg DS) in only 20 samples, while nonylphenol tech was detected in all samples with a mean value of 1079 µg/kg DS and a median value of 940 µg/kg DS.

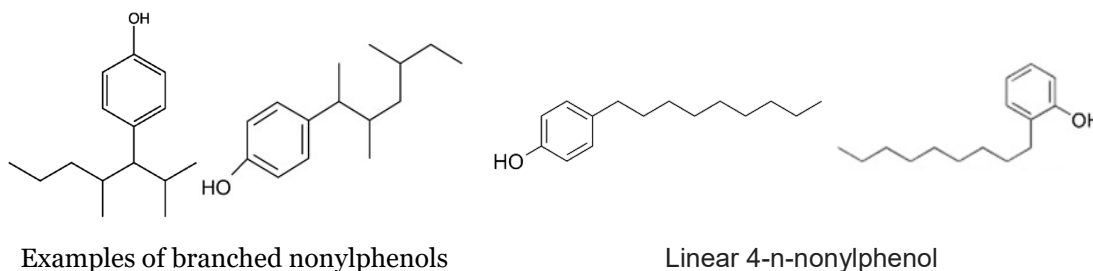


Figure 17. Examples of branched and linear nonylphenols.

5.3.2.2 Required LOQs

The level of quantification (LOQ) for a compound is the minimum concentration of the compound that can be quantitatively determined with suitable accuracy and precision. It is usually defined as 10 times the standard deviation of the blank (same matrix without the compound) or the background signal noise caused by interfering compounds in the sample. Hence, the LOQ is dependent on the matrix and may differ significantly between samples with assumingly similar matrix properties. The level of detection is usually defined as 3 times the standard deviation of the blank.

Table 23 summarises the concentration limit criteria for fertiliser products in quality class 0 as proposed in this report. Due to the uncertainty in the level of interference, we suggest that the LOQ should be at least an order of magnitude lower than the respective concentration limit to make room for inherent statistical errors, as indicated in Table 23. Ideally the analytical precision should be so good that it covers the actual expected concentration range, even for the compounds that are found at very low concentrations. However, since the level in these samples may be below any practical LOD of current analytical methods, we suggest using the observed lower 10-percentile level as a benchmark for the upper LOQ for these compounds (see Table 23).

Table 23. Proposed concentration limit criteria for fertiliser products in quality class 0 and associated maximum LOQ.

HOC	Proposed conc. limit for quality class 0 fertiliser products	Maximum LOQ	Lower 10-percentile in sewage sludge	Proposed upper LOQ
	µg/kg DS	µg/kg DS	µg/kg DS	µg/kg DS
SCCP	2 000	670	31	30
HHCB	10 000	3 300	1 940	1 900
AHTN	10 000	3 300	335	330
DEHP	50 000	16 700	9 500	9 000
PFOS	100	33	3.5	3.0
PFOA	100	33	0.12	0.1
NP+NPE	10 000	3 300	1 780	1 700
PCB7	20	7	3.5	3.5
BDE-209	500	167	84	80

5.3.2.3 Sample preparation and analysis for the selected HOCs

The selected compounds are usually extracted by liquid solvent extraction. The choice of method varies between labs, but it is unclear how well these extraction methods are validated for all applied matrices.

Table 24 summarises typical analytical methods used to separate and detect the selected HOCs and the reported associated LOQs.

Table 24. Typical analytical methods used to separate and detect the selected HOCs, and reported associated LOQs.

HOC	Separation and detection ¹	LOQ (µg/kg DS)	Proposed upper LOQ (µg/kg DS)	Ref.
DEHP	GC/MS	100-500	9 000	ISO 13913:2014
PFOS, PFOA	HPLC-MS/MS	0.1	3.0 (PFOS) 0.1 (PFOA)	Eurofins: DIN 38414-14 mod.
SCCP	GC-MS and ECNI	30-3 000	30	ISO 18635:2016
HHCB	GC-MS	?	1 900	
AHTN	GC-MS	?	330	
BDE-209	GC-EI-MS	0.3-100	80	ISO 22032:2006
PCB7	GC/MS	0.5 (each conger)	3.5	Eurofins: EN 16167
NP, NPE	GC/MS	100	1 700	ISO/TS 13907:2012

¹ GC-MS: gas chromatography-mass spectrometry; ECNI: negative ion chemical ionization mode; EI: electron impact mode

5.3.3 Estimated analytical costs

5.3.3.1 Current costs

Examples of current costs for the selected HOCs commercial labs are listed in Table 25. As far as we are aware of, analyses of HHCB and AHTN in soil or sewage sludge are not provided by any commercial lab in Norway.

Table 25. Analyses offered at commercial labs.

Substance(s)	Matrix	LOQ (µg/kg dw)	Uncertainty	Costs (NOK/sample)	Accredited	Ref.
DEHP	Soil	800	30%	Not available	Yes	ALS
PFOS, PFOA	Soil	0.5	30%	Not available	Yes	ALS
	Soil	3	30%	1 990,-	Yes	ALS
PFOS, PFOA + 13 other PFAs	Soil	10	20%	2 890,-	No	ALS
SCCP	Soil	50	30%	4 000,-	Yes	ALS
HHCB	-	-	-	-	-	-
AHTN	-	-	-	-	-	-
BDE-209 ¹²	Soil	5	Not available	7 300,-	Yes	ALS
PCB7	Sediment	4 (0.5/cong.)	Not available	1 350,-	Yes	ALS
	Soil	2	40%	1 210,-	Yes	ALS
NP, NPE	Soil	10	15-25%	3 250,-	Yes	ALS

5.3.3.2 Potential future costs for the recommended analyses

If the volume of samples to be analysed for the selected HOCs increases drastically, as they probably would if regulated, the incentives for the commercial analytical laboratories to offer affordable analytical packages would increase. However, due to the typical streamlined production at such labs, they have found it most economic to do the whole sample processing and analysis from scratch for each sample and each analysis even if you could, at least in principle, save a lot of work by combining sample preparations that are identical for different types of analyses. A rough guestimate of future package costs for all the selected HOCs (see Table 25) would be around 10 000,- NOK.

Important considerations

The present concentration of HOCs in agricultural soil in general is scarce. Based on the available data it is seen that the most persistent HOCs such as PFOS, PFOA, PBDEs (209 but also 47, 99), HHCB, AHTN and DEHP will accumulate in soil with repeated application of sludge. Though there are some contradicting results for some of the HOCs, as a precautionary principle one should lean on experimental or field data that are considered reliable and that indicate the highest protection level of the environment and health.

There is a need for improved understanding of the role of precursors, particularly to PFOS and PFOA, including standardised methods to simulate the actual oxidative transformation potential that may occur in the environment.

¹² Includes also BDE 28, 47, 99, 100, 138, 153, 154, 183, TBBPA, m-TBBPA, HBCD, TBA and PBB-15, 49 and 52.

6 Potential impacts for the application of Norwegian sewage sludge in agriculture from enforcing the proposed MLs

6.1 Sewage sludge disposal in Norway in 2017

Of the 18 facilities that participated in the 2017-2018 campaign (applied sludge treatments are summarised in Table 20), 15 facilities stabilised and sanitised the sewage sludges so that they in principle could be applied as fertilisers or soil products. The total annual production of final sewage sludge from these 15 facilities was 60 808 tons dw in 2017, making up almost exactly 50% of the total 121 328 tons dw sewage sludge produced in Norway that year. As summarised in Table 26, in 2017 almost 95% of the approximately 50 000 tons of finally disposed of sludge from these 15 facilities was either used directly as soil conditioner on farmland (88%) or applied in soil products (>6%), accounting for 80% of all sludge in Norway that year for those particular uses.

Table 26. Final disposal ways for sewage sludge in 2017 from the 15 sludge treatment facilities included in the 2017-2018 sampling campaign, as well as sum of final disposal ways for sewage sludge for all of Norway. Data provided by Gisle Berge, Statistics Norway.

Treatment facility	Farmland	Soil production	Green areas	Top soil landfills	Incinerated	Other purposes	Total
	ton dw/ year						
Bekkelaget	6181	0	0	0	0	0	6181
Bergen biogass	1350	710	0	0	0	0	2060
Fuglevik	892	0	0	0	0	0	892
HIAS	1387	491	0	0	0	0	1878
Høvringen	1947	0	874	0	0	90	2912
Gardermoen	978	0	0	0	0	0	978
Ladehammeren	1746	0	784	0	0	52	2583
Lindum Biogass	1832	0	181	0	0	222	2235
NRA	7324	0	0	0	0	0	7324
Rambekk	685	696	0	0	0	108	1489
Sandefjord	256	0	0	0	0	0	256
SNJ	0	1283	0	0	0	371	1654
Tønsberg	4918	0	0	0	0	0	4918
VEAS	12641	0	0	0	0	0	12641
ØRA	1624	0	0	0	0	0	1624
Total	43 761	3 180	1 840	0	0	844	49 625
Ratio of total sludge disposal	88,2%	6,4%	3,7%	0%	0%	1,7%	100%
Ratio of all Norway	66,3%	15,5%	13,7%	0%	0%	6,7%	40,8%
All Norway	65 984	20 454	13 413	8 965	0	12 562	121 508
Ratio of total sludge disposal	54,3%	16,8%	11,0%	7,4%	0%	10,3%	100%

6.2 Exceedances of proposed ML values

The numbers and colour coding in Table 20 in **Section 5.2.1.11** summarises the levels of the selected HOCs relative to the respective adjusted ML values; the red colour indicating that at one or more of the five samples collected during the campaign exceeded the ML value or that a majority of the measured values (≥ 3 if 5 samples) were above 75% of the ML. If these data are extrapolated to represent a whole year, hence assuming e.g. no seasonal variations, the annual amount of final sewage sludge from each treatment facility that exceeded the adjusted ML value for a particular HOC ($SS_{annual,HOC>TLV}$; tons dw/year) can be roughly estimated using the following equation:

$$SS_{annual,HOC>TLV} = SS_{annual} \cdot \frac{n_{HOC>TLV}}{N_{HOC}} \quad [\text{Eq. 2}]$$

SS_{annual} is the annual sludge production (tons dw/year), $n_{HOC>TLV}$ is the number of samples that exceeded the adjusted ML for the HOC and N_{HOC} is the total number of samples in which the particular HOC was analysed.

6.2.1 Implementation of the adjusted ML values

Table 27 summarises the calculations for all the selected HOCs (except for OTNE), using the sludge production in 2017 related to the adjusted ML values.

The total annual amounts of finally stabilised and sanitised sewage sludge that exceeds the adjusted ML values in 2017 varies significantly between the different HOCs. The adjusted ML value for HHCB was exceeded in more than 50% of the sewage sludge. Approximately 20% of the sewage sludge exceeded the adjusted ML value for BDE-209. For these two compounds the adjusted MLs were exceeded at a large ratio of the treatment facilities that participated in the sampling campaign; 8 of 12 (0.67) for HHCB and 8 of 15 (0.53) for BDE-209.

There is very limited data for OTNE at Norwegian treatment facilities. However, all the three collected finally stabilised and sanitised sewage sludge samples at to WWTPs included in the survey (HIAS and VEAS; Konieczny et al., 2018) exceeded the proposed adjusted ML value (see **Section 5.2.1.11**). Due to few analyses there is a need to better document the levels of OTNE in sewage sludge and verify if a large fraction of the sewage sludge in Norway exceeds the proposed ML.

For the other compounds much smaller annual amounts of sewage sludge exceeded the adjusted MLs and at a limited number of the facilities (≤ 2).

Table 27. Estimated annual amounts of finally stabilised and sanitised sewage sludge in which the respective adjusted ML values for the selected HOCs were exceeded at the treatment facilities included in the 2017-2018 sampling campaign (Blytt and Stang, 2019). The annual sewage sludge production in 2017 at the different facilities were used in the calculations.

Treatment facility	Sludge prod. ton dw/year	Estimated annual amounts of sewage sludge (ton dw/year) exceeding the respective adjusted ML								
		DEHP	PFOS	PFOA	SCCP	HHCB	AHTN	deca-BDE	PCB7	NP+NPE
Bekkelaget	6 181	0	0	0	0	4 945	0	2 472	0	0
Bergen biogass	2 893	0	0	0	0	2 893	0	1 157	0	0
Fuglevik	779	0	0	0	0	779	0	623	0	0
HIAS	2 174	0	0	0	0	2 174	0	0	0	0
Høvringen	1 799	0	0	0	-	1 799	0	360	0	0
Gardermoen	1 185	0	237	0	0	0	0	0	0	237
Ladehammeren	1 750	0	-	-	-	-	-	0	0	0
Lindum Biogass	3 310	1 986	0	0	0	0	0	0	3 310	0
NRA	7 304	0	0	0	0	0	0	1 461	0	0
Rambekk	1 909	0	-	-	-	-	-	0	0	0
Sandefjord	1 077	0	0	0	-	-	-	0	0	0
SNJ	3 500	0	0	0	0	3 500	0	2 100	2 800	0
Tønsberg	7 281	0	0	0	2 912	2 912	0	1 456	0	0
VEAS	16 966	0	0	0	0	13 573	0	0	0	0
ØRA	2 700	0	0	0	0	0	0	2 160	0	0
Total > ML	-	1 986	237	0	2 912	32 575	0	11 790	6 110	237
Total sludge prod.	60 808	60 808	57 149	57 149	54 273	54 273	54 273	60 808	60 808	60 808
% of total	-	3.3 %	0.4 %	0.0 %	4.8 %	53.6 %	0.0 %	19.4 %	10.0 %	0.4 %
Ratio of facilities >ML	-	0.07	0.08	0.00	0.09	0.67	0.00	0.53	0.13	0.07

6.2.2 If implementing the non-adjusted ML values

Table 28 summarises the calculations for all the selected HOCs (except for OTNE), using the sludge production in 2017 related to the non-adjusted ML values. The non-adjusted ML value is the same as the adjusted ML value for DEHP, PFOS, PFOA and deca-BDE.

The enforcement of the non-adjusted ML values (i.e. for SCCP, HHCB, AHTN, PCN7 and NP+NPE) will probably drastically increase the volume of treated sewage sludge that do not pass these limit values. From the 2017 data, all (100%) sewage sludge exceeds the non-adjusted ML value for HHCB, and almost all (97%) exceeded the non-adjusted ML value for AHTN even if none exceeded the adjusted ML value for AHTN. Also, the ML value for PCB7 was exceeded at almost all the facilities (14 out of 15 plants), counting for approximately 80% of the annual amount of sewage sludge. The ML value for NP + NPE was exceeded at 11 of the 15 facilities and about 30% of the sewage sludge.

Table 28. Estimated annual amounts of finally stabilised and sanitised sewage sludge in which the respective non-adjusted ML values for the selected HOCs were exceeded at the treatment facilities included in the 2017-2018 campaign (Blytt and Stang, 2019). The annual sewage sludge production in 2017 at the different facilities were used in the calculations.

Treatment facility	Sludge prod. ton dw/year	Estimated annual amounts of sewage sludge (ton dw/year) exceeding 75% of the respective non-adjusted ML									
		DEHP	PFOS	PFOA	SCCP	HHCB	AHTN	deca-BDE	PCB7	NP+	NPE
Bekkelaget	6 181	0	0	0	0	6 181	6 181	2 472	6 181	2 472	
Bergen biogass	2 893	0	0	0	0	2 893	2 893	1 157	2 893	1 736	
Fuglevik	779	0	0	0	312	779	779	623	779	623	
HIAS	2 174	0	0	0	0	2 174	2 174	0	2 174	1 739	
Høvringen	1 799	0	0	0	0	1 799	1 799	360	1 799	1 799	
Gardermoen	1 185	0	237	0	0	1 185	1 185	0	1 185	1 185	
Ladehammeren	1 750	0	-	0	0	-	-	0	1 750	700	
Lindum Biogass	3 310	1 986	0	0	0	3 310	3 310	0	3 310	3 310	
NRA	7 304	0	0	0	2 922	7 304	7 304	1 461	2 922	0	
Rambekk	1 909	0	-	0	0	-	-	0	1 909	764	
Sandefjord	1 077	0	0	0	0	-	-	0	2 15,4	431	
SNJ	3 500	0	0	0	0	3 500	3 500	2 100	3 500	3 500	
Tønsberg	7 281	0	0	0	2 912	7 281	5 825	1 456	0	0	
VEAS	16 966	0	0	0	0	16 966	16 966	0	16 966	0	
ØRA	2 700	0	0	0	0	2 700	2 700	2 160	2 700	0	
Total >75% ML	-	1 986	237	0	6 146	56 072	54 616	11 790	48 283	18 259	
Total sludge prod.	60 808	60 808	57 149	57 149	54 273	56 072	56 072	60 808	60 808	60 808	
% of total	-	3,3 %	0,4 %	0,0 %	11,3 %	100,0 %	97,4 %	19,4 %	79,4 %	30,0 %	
Ratio of facilities >ML	-	0,07	0,08	0,00	0,20	1,00	1,00	0,53	0,93	0,73	

6.3 Estimated impacts for the application of Norwegian sewage sludge in agriculture from enforcing the proposed MLs

Table 29 shows the calculated amounts of sewage sludge from 15 treatment facilities that would have been disqualified from being applied on farmland and as ingredients in soil products in 2017 if the adjusted MLs had been enforced at that time. Implementing the proposed adjusted ML on HHCB would have the most negative effect hindering an estimated 51% of the sludge from being applied directly on farmland (46%) or used in soil products (5%). If all adjusted MLs were enforced, 60% of all sludge disposed from these 15 facilities would have been disqualified from use. Table 30 shows the calculated amounts of sewage sludge that would have been disqualified from being applied as soil conditioners on farmland and as ingredients in soil products if the non-adjusted MLs had been enforced, increasing the ratio of sludge disqualified from agricultural application to about 80% that year.

Table 29. Calculated amounts of sewage sludge from the 15 sewage sludge treatment facilities participating in the 2017-2018 campaign that was used on farmland and in soil products that would have been potentially been disqualified from such application if the adjusted MLs had been enforced at that time.

Treatment facility	Amounts of sewage sludge (ton dw/year) > adjusted MLs in 2017									
	DEHP	PFOS	PFOA	SCCP	HHCB	AHTN	deca-BDE	PCB7	NP+NPE	Estimated loss
Disqualified from use on farmland										
Bekkelaget	0	0	0	0	4 945	0	2 472	0	0	4 945
Bergen biogass	0	0	0	0	1 350	0	540	0	0	1 350
Fuglevik	0	0	0	0	892	0	714	0	0	892
HIAS	0	0	0	0	1 387	0	0	0	0	1 387
Høvringen	0	0	0	0	1 947	0	389	0	0	1 947
Gardermoen	0	196	0	0	0	0	0	0	196	196
Ladehammeren	0	0	0	0	0	0	0	0	0	0
Lindum Biogass	1 099	0	0	0	0	0	0	1 832	0	1 832
NRA	0	0	0	0	0	0	1 465	0	0	1 465
Rambekk	0	0	0	0	0	0	0	0	0	0
Sandefjord	0	0	0	0	0	0	0	0	0	0
SNJ	0	0	0	0	0	0	0	0	0	0
Tønsberg	0	0	0	1 967	1 967	0	984	0	0	1 967
VEAS	0	0	0	0	10 113	0	0	0	0	10 113
ØRA	0	0	0	0	0	0	1 299	0	0	1 299
Total	1 099	196	0	1 967	22 601	0	7 863	1 832	196	27 392
% of total	2 %	0 %	0 %	4 %	46 %	0 %	16 %	4 %	0 %	55 %
Disqualified from use in soil products										
Bekkelaget	0	0	0	0	0	0	0	0	0	0
Bergen biogass	0	0	0	0	710	0	284	0	0	710
Fuglevik	0	0	0	0	0	0	0	0	0	0
HIAS	0	0	0	0	491	0	0	0	0	491
Høvringen	0	0	0	0	0	0	0	0	0	0
Gardermoen	0	0	0	0	0	0	0	0	0	0
Ladehammeren	0	0	0	0	0	0	0	0	0	0
Lindum Biogass	0	0	0	0	0	0	0	0	0	0
NRA	0	0	0	0	0	0	0	0	0	0
Rambekk	0	0	0	0	0	0	0	0	0	0
Sandefjord	0	0	0	0	0	0	0	0	0	0
SNJ	0	0	0	0	1 283	0	770	1 026	0	1 283
Tønsberg	0	0	0	0	0	0	0	0	0	0
VEAS	0	0	0	0	0	0	0	0	0	0
ØRA	0	0	0	0	0	0	0	0	0	0
Total	0	0	0	0	2 484	0	1 054	1 026	0	2 484
% of total	0 %	0 %	0 %	0 %	5 %	0 %	2 %	2 %	0 %	5 %
Total loss	1 099	196	0	1 967	25 085	0	8 917	2 858	196	29 876
% of total	2 %	0 %	0 %	4 %	51 %	0 %	18 %	6 %	0 %	60 %

Table 30. Calculated amounts of sewage sludge from the 15 sewage sludge treatment facilities participating in the 2017-2018 campaign that was used on farmland and in soil products that would have been potentially been disqualified from such application if the non-adjusted MLs had been enforced at that time.

Treatment facility	Amounts of sewage sludge (ton dw/year) > proposed non-adjusted MLs in 2017									
	DEHP	PFOS	PFOA	SCCP	HHCB	AHTN	deca-BDE	PCB7	NP+NPE	Estimated loss
Disqualified from use on farmland										
Bekkelaget	0	0	0	0	6 181	6 181	2 472	6 181	2 472	6 181
Bergen biogass	0	0	0	0	1 350	1 350	540	1 350	810	1 350
Fuglevik	0	0	0	357	892	892	714	892	714	892
HIAS	0	0	0	0	1 387	1 387	0	1 387	1 110	1 387
Høvringen	0	0	0	0	1 947	1 947	389	1 947	1 947	1 947
Gardermoen	0	196	0	0	978	978	0	978	978	978
Ladehammeren	0	0	0	279	1 396	1 396	0	1 746	698	1 746
Lindum Biogass	1 099	0	0	0	0	0	0	1 832	1 832	1 832
NRA	0	0	0	0	3 319	3 319	1 465	2 930	0	3 319
Rambekk	0	0	0	0	0	0	0	685	274	685
Sandefjord	0	0	0	695	1 738	1 738	0	51	103	1 738
SNJ	0	0	0	0	0	0	0	0	0	0
Tønsberg	0	0	0	0	0	0	984	0	0	984
VEAS	0	0	0	0	2 608	2 608	0	12 641	0	12 641
ØRA	0	0	0	1 548	1 548	1 548	1 299	1 624	0	1 624
Total	1 099	196	0	3 958	35 377	33 924	7 863	34 244	10 938	37 304
% of total	2 %	0 %	0 %	8 %	71 %	68 %	16 %	69 %	22 %	75 %
Disqualified from use in soil products										
Bekkelaget	0	0	0	0	0	0	0	0	0	0
Bergen biogass	0	0	0	0	710	710	284	710	426	710
Fuglevik	0	0	0	0	0	0	0	0	0	0
HIAS	0	0	0	0	491	491	0	491	393	491
Høvringen	0	0	0	0	0	0	0	0	0	0
Gardermoen	0	0	0	0	0	0	0	0	0	0
Ladehammeren	0	0	0	0	0	0	0	0	0	0
Lindum Biogass	0	0	0	0	0	0	0	0	0	0
NRA	0	0	0	0	0	0	0	0	0	0
Rambekk	0	0	0	0	0	0	0	0	0	0
Sandefjord	0	0	0	0	0	0	0	0	0	0
SNJ	0	0	0	0	0	0	0	700	280	700
Tønsberg	0	0	0	0	0	0	0	0	0	0
VEAS	0	0	0	0	0	0	0	0	0	0
ØRA	0	0	0	0	0	0	0	0	0	0
Total	0	0	0	0	7 387	7 387	1 054	3 180	2 380	1 901
% of total	0 %	0 %	0 %	0 %	15 %	15 %	2 %	6 %	5 %	4 %
Total loss	1 099	196	0	3 958	42 764	41 311	8 917	37 424	13 318	39 205
% of total	2 %	0 %	0 %	8 %	86 %	83 %	18 %	75 %	27 %	79 %

Important considerations

As summarised in Tables 10-18, the levels of the selected HOCs vary considerably and are also influenced by local sources (e.g. landfill leachate for DEHP and PCB7, and historic usage of PFOS in fire-fighting foams at certain airports), and the applied treatment steps (both wastewater and sludge) may impact these levels (see Part II – Measures of this report).

The levels of HHCB and BDE-209 were in general quite high and exceeded the proposed MLs in 57% and 20% of the samples, respectively, from the 2017-18 sampling campaign.

Even if the measured levels of PFOS, PFAS and NP + NPE (mono- and di-ethoxylates) during this sampling campaign were not as alarming as for HHCB and BDE-209, the unknown level and fate of potential precursors to these compounds may underestimate the potential levels of PFOS, PFOA, NP and NPE in the soil environment when the sewage sludge is applied as a fertiliser or a soil amendment product.

The estimated annual amount of sewage that may exceeded the adjusted and non-adjusted ML values should be regarded as an indication only, since better documentation is needed to give a more thorough and science-based advice to set appropriate ML values. Furthermore, a more longterm monitoring of each compound is needed to establish a baseline and associated variability from this baseline for each compound in treated sewage sludge at each facility.

This first rough assessment indicates that the levels of BDE-209, HHCB and PCB7 exceeded the adjusted ML values at 11, 10 and 7 of the 15 treatment facilities, respectively.

PART II - MEASURES

7 Fate and transfer of the selected HOCs from wastewater to untreated sludge

7.1 Commonly applied treatment units and treatment train design in Norway

Figure 18 illustrates a typical treatment train at a conventional large secondary WWTP in Norway. Both primary and secondary treatment units remove particulate matter that is directed to the sludge treatment line. Scrap material and anything that is collected in the sand trap is usually sent to a landfill, while floating material collected in the grease trap is often handled together with the primary sludge due to its richness in fat and hence benefit in biogas production.

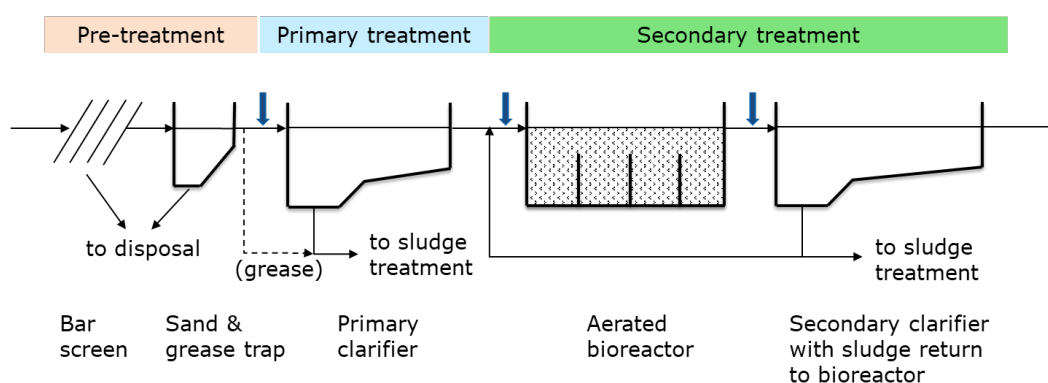


Figure 18. Illustration of a typical conventional WWTP applying pre-treatment, primary treatment and secondary treatment. The blue arrows indicate where chemical treatment is typically applied.

As summarised in Table 30, conventional (Norwegian) WWTPs are not designed to remove micropollutants such as the selected HOCs in focus here, but to remove large debris, floating matter, suspended solids, biodegradable organic matter or nutrient salts that were causing the dominating environmental problems at the time when they were first introduced (i.e. smothering and eutrophication in the aquatic recipient). However, as indicated by the expected removal of suspended solids (SS) of the different treatment trains, if dimensioned and managed properly, compounds that adsorb to the suspended solids may also be efficiently removed at WWTPs applying chemical and/or biological treatment processes.

As shown in Figure 19, approximately 75% of all domestic wastewater in Norway are treated chemically or both biologically and chemically. These plants also remove a large fraction of the biodegradable organic matter (measured as the five-days biological oxygen demand – BOD₅); even the pure chemical treatment plants. However, only at the plants applying biological treatment (about 40% of the wastewater) can one expect any biodegradation of the compounds during the wastewater treatment. Most of the remaining 25% of the wastewater receive only mechanical treatment or no treatment at all with limited removal of sewage sludge and BOD₅. The location of these WWTP are shown in Appendix B.

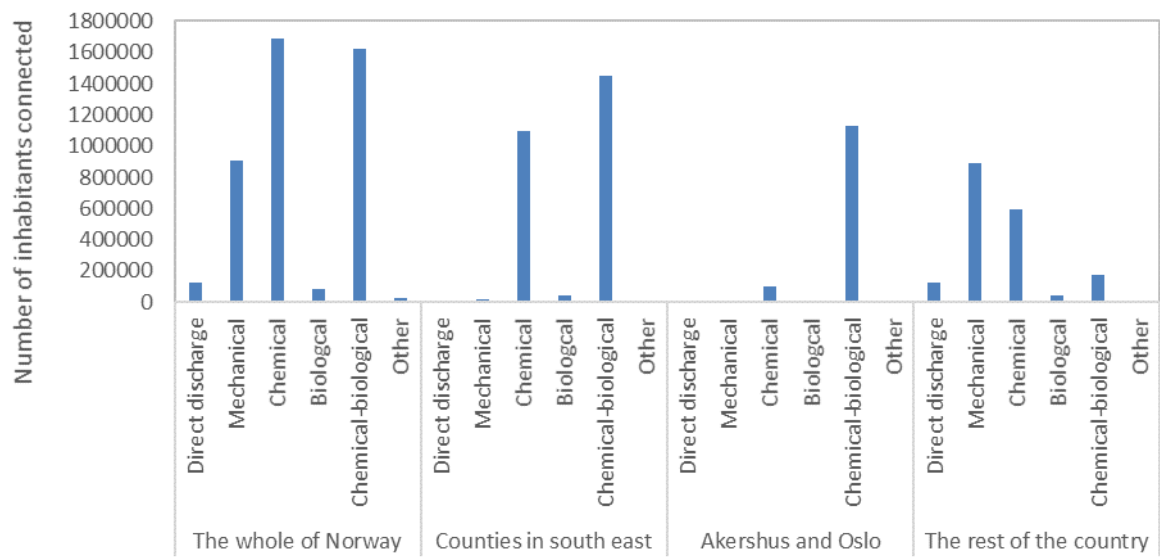


Figure 19. Dominating WWTP types serving >50 PE in Norway in 2017. Source: KOSTRA (SSB).

Table 31. Respective targets compounds and expected removal of suspended solids (SS) and biodegradable organic compounds (measured as the 5-day biological oxygen demand; BOD5) by the most common treatment processes applied at Norwegian WWTPs. Source: adapted from Ødegaard et al., 2009).

Type of WWTP	Most advanced treatment unit	Treatment train ⁽¹⁾	Target compounds of most advanced treatment unit	Expected removal	
				SS (%)	BOD5 (%)
Mechanical	Pre-treatment	PT	Coarse matter, grit/sand, floating matter	10-20	10-20
	Septic tank	PT/S	Settleable and floating matter	35-55	15-25
	Primary treatment	PT – S		40-60	15-25
Chemical	CEPT ⁽²⁾	PT ↓ F – S	Phosphorous, organic matter	80-85	65-75
	Secondary chemical precipitation	PT – S ↓ F – S		85-90	70-80
Biological	Conventional activated sludge process	PT – S – CAS – S	Biodegradable organics	80-90	85-90
	Biofilm process	PT – S – BF – S		80-90	85-90
Biological-chemical	Pre-precipitation	PT ↓ F – S – CAS/BF – S	Phosphorous, biodegradable organics	85-90	90-95
	Co-precipitation	PT – S ↓ CAS – S		85-90	80-90
	Intermittent precipitation	PT – S – BF ↓ F – S		90-95	90-95
	Post-precipitation	PT – S – CAS/BF – S ↓ F – S		90-95	90-95
Biological-chemical with N-removal	Pre-denitrification AS	PT – DN-CAS – N-CAS – S ↓ F – S	Phosphorous, biodegradable organics, nitrogen	90-95	>95
	Post-denitrification BF	PT – S – N-BF – DN-BF ↓ F – S		90-95	>95
	Pre- & post-denitrification BF	PT – DN-BF – N-BF – DN-BF ↓ F – S		90-95	>95

1) **PT** = pre-treatment and typically includes units such as coarse bar screens, grit chambers and grease skimmers. May also include a fine screen (0.1-0.5 mm pore size), which tentatively will improve the removal of SS up to 40% and BOD₅ up to 20%. **S** = separation and may consist of fine screens, sedimentation (most common in Norway), flotation or filtration. **↓** = addition of coagulant. **F** = flocculation chamber to build up settleable flocs. **CAS** = conventional activated sludge process. **BF** = biofilm process. **DN** = denitrification process (step two in N-removal). **N** = nitrification process (step one in N-removal)

2) CEPT = Chemically enhanced primary treatment

7.2 Factors that influence the rate of transfer from wastewater to sludge

The fate of a specific compound in a WWTP is influenced by several factors;

- the wastewater composition including the concentration of the compound itself,
- the types, sequential order and physical arrangements of the different treatment processes that are applied,
- the operational conditions during treatment, and
- the compound's physicochemical properties.

The selected HOCs in focus here are all found in the final sewage sludge, hence they have been transferred from the wastewater that entered the WWTP and/or they may have been transformed from precursors found in the influent, due to the specific treatment steps applied by the WWTP.

7.2.1 The concentration in the influent to the respective treatment unit

The concentration of a specific compound may vary considerably with time in the influent, depending on the local release patterns and processes occurring during transport to the WWTP (e.g. increases/losses due to compound transformations, dilution due to stormwater intrusion and overall losses due to sewer overflows). The concentration may also differ considerably within the WWTP due to the influences of previous treatment steps (see Table 31). In that regard, it is also crucial to consider internal transfer lines that may transfer large quantities of the compound back to earlier treatment steps which may cause accumulation in certain parts of the WWTP. The more advanced WWTPs often have many such transfer lines, both within the wastewater treatment line (e.g.) and from the sludge treatment line back to the wastewater treatment line. There are many local variations in these, but the more common ones are the return of activated sludge from the secondary clarifier back to the bioreactor, treatment of biofilm/sandfilter backwash water using the upstream primary settler, recycling of nitrate-rich water during biological nitrogen removal and transfer of the free water phase after sludge thickening and rejected water after sludge dewatering. In general, if a specific HOC is in fact removed, an increase in its concentration favours its removal efficiency (i.e. per cent removed) unless the concentration passes a certain threshold when it impairs the removal due to toxicity (i.e. in biological processes). However, as is discussed in the following, there are several factors influencing the removal efficiency of a specific HOC.

7.2.2 Sorption to sludge

All WWTPs produce sludge which is separated from the wastewater and treated separately. Hence, any compounds associated with this sludge are also removed from the wastewater. The compound's ability to sorb to the particulate matter present in the wastewater or generated during the different treatment processes, influences the removal rate. The sorption may occur through (i) absorption due to hydrophobic interactions of the aliphatic and aromatic groups of a compound with the lipophilic cell membrane of the microorganisms or the lipid fractions of the suspended solids and (ii) adsorption due to electrostatic interactions of positively charged groups of chemicals (e.g. protonated basic functional groups) with the negatively charged surfaces of the microorganisms (Verlicchi et al., 2012). Factors such as pH, redox potential, stereochemical structures and nature of both the sorbent and the sorbed molecule influence the sorption (Kümmerer, 2009). The quantity of a compound that will be sorbed to suspended solids (SS) in the wastewater at equilibrium (C_{sorbed} [$\mu\text{g/L}$]), is assumed to be proportional to the concentration of the compound in solution ($C_{dissolved}$ [$\mu\text{g/L}$]) (Ternes et al., 2004a):

$$C_{sorbed} = K_d \cdot SS \cdot C_{dissolved} \quad [\text{Eq. 3}]$$

K_d is the solid–liquid partition coefficient of the compound [L/kg SS], defined as the partition of the compound between the sludge (or more correctly; soil) and the water phase. Sewage sludge is the concentration of suspended solids in the raw wastewater. According to Ternes et al., (2004a) the removal by sorption of compounds with K_d values lower than 500 L/kg·SS (i.e. $\log K_d < 2.7$) are negligible in municipal WWTPs. Accordingly, compounds with $\log K_d > 3$ tend to sorb to sludge (Hai et al., 2014). The octanol-water partition coefficient P_{ow} are also frequently used instead of K_d , and it is generally accepted that $\log P_{ow}$ values < 2.5 imply low sorption to sludge, while compounds with $\log P_{ow} > 4$ tend to sorb strongly to sludge.

However, for dissociable compounds such as PFOS, PFOA, NP and NPE the K_d value is highly dependent on the pH; acidic functional groups such as COOH- and OH- are negatively charged above their pK_a and neutral below their pK_a , while basic functional groups such as NH_2^- are positively charged below their pK_a and neutral above their pK_a . As demonstrated by Wang et al., (2000), the isoelectric point (pI, i.e. the pH when the net charge is zero) of both primary, secondary and tertiary sludge particles appears to be within 2.9 ± 0.3 , indicating that they will be negatively charged under most circumstances during wastewater and sludge treatment. Hence, sorption due to electrostatic interactions are probably important for the sorption of positively charged compounds to negatively charged sludge and particles in the wastewater, while negatively charged compounds (e.g. PFOS, PFOA) will tend to be repulsed by the negatively charged sludge. Hence, for such compounds the pH-dependent derivation of P_{ow} is more appropriate (Ternes et al., 2004a):

$$D_{ow} = \frac{P_{ow}}{1 + 10^{pH - pK_a}} \quad [\text{Eq. 4}]$$

Tadkaew et al., (2010) varied the operating pH in a membrane bioreactor (MBR; see Section 7.2.4) from 5 to 9 and found that the removal of ionisable HOCs changed with pH, whereas those of non-ionisable HOCs were independent of pH.

As Equation 3 suggests, the sorption is also highly dependent on the sludge concentration, indicating that the sorption of hydrophobic compounds will increase by increasing the sludge age or sludge residence time (SRT) in the biological treatment step. However, increasing the SRT may drastically influence also its microbial and well as structural composition, which may affect the sorption kinetics. Here, more studies are needed to elucidate the impact of SRT on other sludge properties, such as floc size and density, and their implications on the sorption of HOCs (Semblante et al., 2015).

The hydraulic retention time (HRT; the mean residence time of the liquid phase in the reactor) controls the available time for sorption and may influence the amount of HOCs in the sludge if the sorption kinetics indicate that it would primarily occur within that time frame.

The sorption of HOCs on sludge due to hydrophobic and electrostatic interactions is an enthalpy-driven process and is therefore also temperature-dependent (Hulscher and Corneliusen, 1996).

7.2.2.1 SCCP, HHCB, AHTN, BDE-209 and PCBs

Both SCCP, HHCB, AHTN, BDE-209 and PCBs have very high $\log P_{ow}$ and K_d values and low water solubilities and are therefore expected to sorb strongly to sludge (Arp et al., 2014; Arp et al., 2017):

- SCCP: $\log P_{ow}$ value of 4.39-8.69 depending on the chlorine content and chain length (ECHA, 2008), K_d value of 1995 L/kg TS and water solubility of 0.15-0.47 mg/L.
- HHCB: $\log P_{ow}$ value of 5.4, K_d value of 4920 ± 2080 L/kg TS (Ternes et al., 2004) and water solubility of 1.75 mg/L (EU RAR, 2008a)

- AHTN: log P_{ow} value of 5.3, K_d value of 5300 ± 1900 L/kg TS (Ternes et al., 2004) and water solubility of 1.25 mg/L (EU RAR, 2008b)
- BDE-209: log P_{ow} value of 9.9, K_d value of 5.2×10^9 and water solubility of 0.0001 mg/L (EPISUITE experimental data)
- PCBs: log P_{ow} value of 4.7-6.8, K_d value of 3211 L/kg TS and water solubility of 0.01-0.4 mg/L

7.2.2.2 DEHP

Due to its high log K_{ow} (7.5) DEHP is expected to sorb strongly to sludge. However, DEHP also adsorbs strongly to organic acids such as humic acids, and to proteins, both of which typically have high mobility in water and are ubiquitous in wastewater (EU RAR, 2008d). Hence, chemical precipitation (**Section 7.2.3**) may improve the removal of DEHP and increase its sorption to sludge. Huang et al., (2008) indicated that HRT in the range from 5 to 14 h achieved minor removal of DEHP in their lab-scale anaerobic-ac-aerobic activated sludge system, while higher HRT increased DEHP accumulation in the system and DEHP retention in the waste sludge.

7.2.2.3 PFOS and PFOA

One would expect that the negatively charged and highly water soluble PFOS and PFOA (370 mg/L and 4140 – 9500 mg/L, respectively (Moermond et al., 2010; ECHA, 2013)) would not tend to sorb to sludge. However, experimentally derived K_d values for the partitioning of PFOS and PFOA in activated sludge indicate much stronger sorption behaviour than the anticipated sorption based on the calculated log D_{ow} values using equation 2 (-6.9 and -2.2 for PFOS and PFOA, respectively); 200-4050 L/kg for PFOS and 150-870 L/kg for PFOA (Arvaniti et al., 2012; Zhou et al., 2010; Yu et al., 2009). Zhou et al., (2010) postulated that cations such as Ca^{2+} and Mg^{2+} present in the sludge could act as ion bridges and facilitating sorption. In controlled laboratory tests they showed that the sorption decreased with increasing pH and suggested that amino and amide groups in proteins on the surface of bacteria in the activated sludge may be involved in the sorption. However, the strong hydrophobic properties of the fluorinated carbon chain of PFOS and PFOA also suggest that hydrophobic interactions played an important role in their sorption to sludge. As indicated by the K_d value ranges, PFOS tends to sorb stronger to activated sludge than PFOA, possibly explained by the more hydrophobic property of perfluorinated sulfonates (e.g. PFOS) than perfluorinated carboxylates (e.g. PFOA) with the same number of carbon atoms (Zhou et al., 2010). The large variability in K_d values may be explained by the characteristics of the sludge. For instance, Arvaniti et al., (2012) found that PFOA sorbed stronger to primary sludge (1390 ± 1220 L/kg) than activated sludge (391 ± 320 L/kg) and even somewhat stronger than PFOS sorbed to primary sludge (675 ± 260 L/kg).

7.2.2.4 NP and NPE

With a log P_{ow} value of 4.48 and K_{oc} of 5360 L/kg, both NP and NPE tend to sorb strongly to sludge. As surfactants, NPEs may form micelles at elevated concentrations, which may explain the observed reduced sorption behaviour of shorter NPEs (shorter than NP9EO) (Beigel et al., 1998).

Expected sorption of the selected HOCs to primary and secondary sludge is summarised in Table 32.

Table 32. Summary of expected sorption to primary and secondary sludges. See the text for references to the values given.

Substance	Expected sorption
DEHP	Its high log K_{ow} (7.5) indicates that it will sorb strongly to sludge. However, DEHP also adsorbs strongly to organic acids such as humic acids, and to proteins, both of which typically have high mobility in water.
PFOS, PFOA	The surface-active properties and relatively high water solubilities of these compounds complicates their sorption properties. PFOA appears to sorb stronger to primary sludge than PFOS, but PFOS tends to sorb stronger to activated sludge than PFOA.
SCCP	High log P_{ow} (4.39-8.69) and K_d values (1995 L/kg dw) and low water solubilities (0.15-0.47 mg/L) and are therefore expected to sorb strongly to sludge.
HHCB	High log P_{ow} (5.4) and K_d values (4920±2080 L/kg dw) and low water solubilities (1.75 mg/L) and is therefore expected to sorb strongly to sludge.
AHTN	High log P_{ow} (5.3) and K_d values (5300±1900 L/kg dw) and low water solubilities (1.25 mg/L) and is therefore expected to sorb strongly to sludge.
OTNE	High log P_{ow} (5.3-5.7) and K_d values (assumed in same range as AHTN and HHCB) and low water solubilities (1.6 ⁻⁴ mg/L) and is therefore expected to sorb strongly to sludge.
BDE-209	High log P_{ow} (9.9) and K_d values (5.2 x 10 ⁹ L/kg dw) and low water solubilities (<0.001 mg/L) and is therefore expected to sorb strongly to sludge.
PCB7	High log P_{ow} (4.7-6.8) and K_d values (3211 L/kg dw) and low water solubilities (0.01-0.4 mg/L) and are therefore expected to sorb strongly to sludge.
NP+NPE	High log P_{ow} (4.48) and K_d values (5360 L/kg dw) and are therefore expected to sorb strongly to sludge. However, the surface-active properties of NPE reduce the sorption of particularly shorter NPEs.

7.2.3 Chemical precipitation/coagulation

Positively charged coagulants such as iron, aluminium and calcium salts are frequently used to improve the removal of phosphorous from wastewater by precipitation/coagulation and to improve the settling properties of the suspended solids and colloidal matter by building larger aggregates. Due to the generally low concentrations of HOCs in wastewater, any precipitation of these compounds found in the dissolved state might be limited. Though, compounds associated with the suspended solids will be removed more efficiently. For instance, Suárez et al., (2009) observed extensive removal of HHCB ($79.2 \pm 9.9\%$) and AHTN ($83.4 \pm 14.3\%$) by coagulation at full scale.

The coagulation process is operated within a relatively strict pH interval to control its performance (i.e. P removal), depending upon the type of coagulant(s) used, where in the treatment train it is applied, the water alkalinity and the targeted effluent phosphorous concentration. Generally, precipitation/coagulation using iron and aluminium salts requires acidic conditions (typically 5.5-7), precipitation/coagulation with calcium salts requires alkaline conditions (typically 10-11), while combinations between calcium and iron or aluminium salts have a pH optimum in the range between these depending upon their ratios (Arvin and Henze, 2002). This may affect the sorption of ionisable HOCs (i.e. PFOS, PFOA, NP and NPE) (Semblante et al., 2015) (**Section 7.2.2**).

7.2.4 Biotransformation

The HOCs might be transformed or degraded in the presence of microorganisms that can use them as growth substrates. A prerequisite for transformation is that the compound is available at an appropriate concentration in the water (given by the microorganisms' affinity for the compound), the environmental conditions are generally suitable, and that the compound has a structure that make it available for degradation by the microorganisms' enzymes (Barret et al., 2012). Many xenobiotic compounds lack the latter and are therefore often persistent to biotransformation. The commonly very low influent concentrations of HOCs ($< 1\text{mg/L}$) are also a challenge and two scenarios for the biological transformation or degradation of the trace pollutants are likely (discussed related to pharmaceuticals and personal care products (PPCPs) by Ternes et al., 2004b): If a primary substrate is available for the bacteria to grow on, co-metabolism may occur, in which case bacteria break down or partially convert the trace pollutant but do not use it as a carbon source. In another likely scenario, mixed-substrate growth takes place and the bacteria use the trace pollutant as a carbon and energy source and may mineralize it completely.

Previous studies generally show that the biotransformation of HOCs preferentially occurs under aerobic than anoxic conditions (Suarez et al., 2010; Phan et al., 2014; Stadler et al., 2015). Phan et al., (2014) showed that anoxic conditions promoted the sorption of hydrophobic compounds on sludge, which possibly facilitated their biotransformation under aerobic conditions due to sludge recirculation between aerobic and anoxic conditions.

Molecular features that generally increase or decrease the resistance to aerobic biodegradation are summarised in Table 33.

The sorption behaviour of HOCs has a dual effect on biotransformation. Readily biodegradable compounds that are sorbed to bacterial surfaces facilitates their reaction with extracellular enzymes and uptake into cells, but the sorption of slowly biodegradable compounds on sludge flocs decreases their bioavailability and exacerbates accumulation in sludge (Barret et al., 2012). Hence, the physical (particle/floc size, porosity and flexibility) and chemical (composition (e.g. sorption sites) and spatial hydrophobicity) properties of the sludge matrix are likely to strongly influence the bioavailability of HOCs associated with different entities of the sludge matrix. A three-compartment model that distinguishes between freely dissolved HOCs, HOCs sorbed to particles and HOCs sorbed to dissolved and colloidal matter (DCM) developed by Barret et al., (2010a,b), is shown in Figure 20. Barret et al.,

(2010b) demonstrated that the sorption to both the particulate matter and to DCM was relatively similar PAHs and NP comparing primary settled sludge and activated sludge (with an extremely short SRT of 0.4 days), but the sorption increased by approximately a factor 10 to both the particulate matter and DCM when the sludges had been anaerobically digested. Delgadillo-Mirquez et al., (2011) experimentally tested and validated the model for anaerobic digestion of primary and secondary sewage sludge using PAHs as model compounds and found that it was primarily the free PAHs and PAHs associated with DCM that were bioavailable.

Table 33. Molecular features that generally increase or decrease the resistance to aerobic biodegradation (based on Wennberg, 2017; Schwarzenbach et al., 2003; Tadkaew et al., 2011; Eggen and Vogelsang, 2015)

Expected effect	Molecular feature	Examples from the selected HOCs
Increased resistance	Halogens, particularly chlorine and fluorine and if there are more than three in a small molecule. Highly substituted structures are usually more resistant than simpler compounds.	PFOS, PFOA (both fluorinated), SCCP, PCBs (both chlorinated) and BDE-209 (brominated)
	Extensive chain branching; quaternary carbon is particularly problematic.	HHCB, AHTN, OTNE and to some degree DEHP and NP
	Tertiary amine, nitro, nitroso, azo and arylamino groups	
	Polycyclic residues, especially when there are more than three fused rings.	HHCB (3 rings), AHTN, OTNE, BDE-209 and PCBs (all 2 rings)
	Heterocyclic residues	HHCB
	Aliphatic ether bonds (except in ethoxylates)	
Reduced resistance	Groups susceptible to enzymatic hydrolysis such as esters and amides	DEHP
	Oxygen atom present in the form of hydroxyl, aldehyde, or carboxylic acid groups and probably also ketone	PFOA, AHTN, OTNE, NP, NPE
	Unsubstituted linear alkyl chains (especially with ≥ 4 carbons) and phenyl rings	NPE

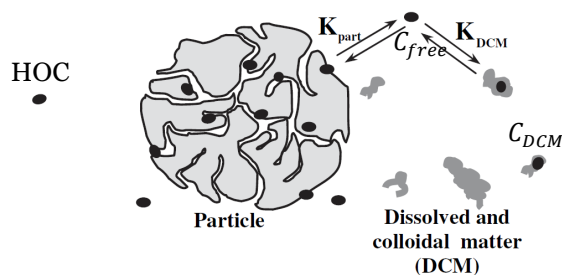


Figure 20. Illustration of the three-compartment model for sorption of HOCs in sewage sludge developed by Barret et al., (2010). HOCs may be sorbed to the particulate matter or to DCM, governed by K_{part} or K_{DCM} , respectively. Adapted from Barret et al., (2010a,b).

Typically, a rather complex transformation pathway involving many different enzymes and microorganisms is needed for complete mineralisation of compounds such as the selected HOCs. Often the biotransformation process will decelerate or completely halt due to unfavourable or limiting conditions (Cirja et al., 2008; Eggen and Vogelsang, 2015):

- The microorganisms present in the sludge lack the necessary enzyme(s) to attack the compound or need time to produce them.
- There are more bioavailable substrates present that require less effort to utilise; hence, no need to initiate the production of an inherent but resting enzyme apparatus.
- Other redox conditions are favourable; aerobic (oxygen as electron acceptor), anaerobic (e.g. sulphate or chlorinated organic compounds as electron acceptor) or anoxic (nitrate as electron acceptor).
- The transformation product is inherently inert or not accessible.
- The transformation product has accumulated to toxic levels in the sludge.

Biotransformation may also contribute to increase the concentration of a specific compound, since the transformation of other compounds (i.e. precursors) can have the specific compound as its transformation product. This is highly relevant for PFOS and PFOA.

Effect of SRT and reactor configuration on the biotransformation

The sludge age or the average solids retention time (SRT) is one of the most important parameters in biological wastewater processes. In conventional activated sludge (CAS) systems that are not required to remove nitrogen, the bioreactors are typically operated with SRTs of just a few days. In CAS systems that require nitrogen removal, the SRTs are usually in the order of 10-20 days to retain slow-growing nitrifying bacteria in sufficient amounts in the sludge. The conventional biological nitrogen removal includes both an aerobic stage, in which ammonia is oxidized to nitrate by nitrifying bacteria, and an anoxic stage, where nitrate is reduced to nitrogen gas by denitrifiers. The nitrifying bacteria may play a key role in the biotransformation of HOCs in WWTPs. It has been shown that their enzyme ammonium monooxygenase (AMO), which is necessary for the oxidation of ammonia, is able to co-oxidize recalcitrant pharmaceuticals such as iopromide and trimethoprim (Batt et al., 2006).

In order to retain the activated sludge in the system, the CAS system includes a (secondary) clarifier to separate the sludge and recirculate it back to the bioreactor. In Norwegian WWTPs the moving bed biological reactor (MBBR) is increasingly common. This is a biofilm reactor, where the biofilm grows on a suspended solid support, thereby making the secondary clarifier unnecessary. The granular

sludge bioreactor is a hybrid system of the CAS and the MBBR in which the granulation of the sludge creates a solid support itself (a few mm in size) that makes it possible to retain it within the bioreactor using appropriate reactor configurations. Another hybrid system is the membrane biological reactor (MBR) in which the biomass is retained by membrane filtration (usually microfiltration or ultrafiltration) either within the bioreactor itself or outside the bioreactor. Both the granular sludge bioreactor and the MBR have so far been in very limited use in Norway.

In the CAS and MBR systems, the SRT is regulated by the amount of the produced biomass that is removed from the systems. However, in contrast to CAS, MBRs also retain microorganisms that are not associated with sludge flocs if their doubling time (i.e. growth rate) is shorter than the SRT. This has been shown to have an impact on the heterogeneity of the microbial community in the MBR system as compared to the CAS system (Göbel et al., 2007). The large fraction of planktonic microorganisms and smaller flocs found in MBR compared with in CAS systems (10-100 µm vs 100-500 µm; Zhang et al., 1997) also increases the viable fraction of the biomass in MBR due to improved mass-transfer conditions and accessibility to the substrates (Cicek et al., 1999; Clara et al., 2005; Pollice and Laera, 2005). Furthermore, the SRT in MBR systems is usually kept even higher than in CAS systems with nitrogen removal (typically 15-80 days), which leads to higher biomass concentrations. The higher biomass concentration decreases the food to microorganism ratio, which makes it more likely that the microorganisms will target less energy-rich substrates (Weiss and Reemtsma, 2008). Also, MBBRs have been shown to be significantly more efficient for the biotransformation of recalcitrant pharmaceuticals than CAS systems, supposedly due to other microorganisms than nitrifiers (Falås et al., 2012), indicating a more heterogenic microbial community caused by the “indefinite” SRT in such systems. However, there is no definite causality between SRT and microbial diversity due to the many factors affecting the structure and composition of the microbial community. There are also studies that clearly contradict the assumption that sludges with high SRT have higher microbial diversity than those with low SRT (Falås et al., 2012).

Effect of HRT on the biotransformation

The hydraulic retention time (HRT) controls the time for sorption to the sludge/biomass and biotransformation in the water. Usually, the HRTs in CAS and MBR systems are in the range of 2-24 h and 7-15 h, respectively, while the HRT in MBBRs are typically in the range of a few hours. In general, compounds having slow-to-intermediate biotransformation kinetics will experience less effective biotransformation at shorter HRTs (Fernandez-Fontaina et al., 2012).

Effect of temperature on the biotransformation

As discussed in detail for the biodegradation of the selected HOCs in soil (Section 2.3.1), the temperature has a huge impact on many processes including the enzymatic reactions taking part in the biotransformations. Importantly, the solubilities of the compounds also increase with increasing temperature, making them more available for enzymatic attack.

Typically, biological processes occur more rapidly at increasing temperatures, however, there is a gradual, but complete, shift in the microbial community when going from a psychrophilic environment (<0-20°C, max. growth rates at ca. 15°C) to a mesophilic environment (ca. 12-45°C, max. growth rates at ca. 40°C) and further to a thermophilic environment (ca. 40-70°C, max. growth rates at ca. 60°C) (Brock and Madigan, 1991). Seasonal changes in influent water temperature is known to affect the growth rate of the nitrifiers, hence, a prolonged SRT is usually applied (and needed) at Norwegian WWTPs with biological nitrogen removal during the winter months. Such temperature fluctuations will affect the microbial sludge community, but very few studies have looked at how this influences the biotransformation of HOCs (Grandclément et al., 2017).

7.2.4.1 DEHP

Based on available biotransformation studies using standard tests DEHP is assumed to be readily biodegradable under aerobic conditions (i.e. >60-70% transformation within 28 days and a typical preliminary lag period <7-10 days) (EU RAR, 2008c). Some studies reported partial mineralisation (4-54%) of the initial DEHP (measured as evolved CO₂). The primary biotransformation products of DEHP are mono(2-ethylhexyl)phthalate (MEHP), which is considered even more toxic than the parent compound, and 2-ethylhexanol. Under aerobic conditions both MEHP and 2-ethylhexanol are converted further and finally completely mineralised (EU RAR, 2008c). Under anaerobic conditions, the transformation process appears to be limited to the first step producing only MEHP (EU RAR, 2008c). Primary transformation of DEHP has also been observed in CAS simulation tests with typical rates of 70-85% within 24 hours (Saeger and Tucker, 1976; O'Grady et al., 1985) and 91% disappearing within 48 hours (Graham 1973). However, the actual observed biotransformation in WWTPs is often slow, probably due to strong adsorption of DEHP to organic matter (EU RAR, 2008c).

7.2.4.2 PFOS and PFAS

Even though both PFOS and PFAS have electron-donating groups at the end of their backbone (sulphate and carboxyl, respectively), they are both very persistent to biodegradation due to the highly fluorinated and strongly electron-attracting backbone.

Moreover, a significant number and wide spectre of precursors exist that can indirectly contribute through both abiotic and biotic degradation routes to the final concentration of these compounds in the final sludge. For an overview of these, see OECD (2007), Nielsen (2014) and Liu and Avendano (2013). Aerobic transformation of these precursors tends to be much more significant than the transformation occurring under anaerobic conditions.

The most important of these precursors for PFOA are (ECHA RAC, 2015):

- 8:2 fluorotelomer alcohol (8:2 FTOH¹³)
- 8:2 fluorotelomer derivatives (e.g. stearate monoester, citrate triester, acrylate, methacrylate, iodide),
- polyfluoroalkyl phosphates (PAPs¹⁴) and
- side-chain fluorinated polymers (e.g. acrylate, urethane).

The transformation rates of these precursors to PFOA are highly uncertain, but ECHA RAC (2015) assumes that at least 10% of the released 8:2 FTOH, at least 5% of the released 8:2 fluorotelomer derivatives, around 1% of the released PAP substances and 1% of the fluorotelomer-based polymers will be transformed into PFOA within 12 months.

Important indirect precursors for PFOS include (NICNAS, 2019):

- Neutral organic derivatives of perfluorooctanesulfonamide (e.g. MeFOSA, EtFOSE, MeFOSE)
- Methacrylate and acrylate polymers with perfluorooctanesulfonamide side chains
- Methacrylate and acrylate telomers with perfluorooctanesulfonamide side chains
- Carbamate esters of perfluorooctanesulfonamides

¹³ 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecan-1-ol

¹⁴ mono-PAPs (including 6:2 and 8:2), polyfluoro alkyl phosphate esters (6:2 di-PAP) and 8:2 fluorotelomer alcohol di-phosphate (8:2 di-PAP)

- Carboxylic acid, ester and amide derivatives of perfluorooctanesulfonamides (e.g. EtFOSAA, FOSAA)
- Urethane polymers incorporating perfluorooctanesulfonamides
- Phosphate Esters of Perfluorooctanesulfonamides
- Polyglycol Derivatives of Perfluorooctanesulfonamides
- Alkyl Ammonium and Amine Derivatives of Perfluorooctanesulfonamides
- Miscellaneous Perfluorooctanesulfonyl Derivatives
- Polyfluorinated sulphonamides (e.g. PFOSA)

There is a wide range of possible transformation routes, both biotic and abiotic to both PFOA and PFOS (Avendaño et al., 2015). For instance, EtFOSE is first oxidized to the corresponding acetic acid (EtFOSAA), which then undergoes stepwise dealkylation to form PFOSA and finally to PFOS. The biotransformation of EtFOSAA seems to be the rate limiting step and thus this metabolite becomes quantitatively important (Avendaño et al., 2015).

7.2.4.3 SCCP

SCCP are regarded as persistent in the environment, but they have been shown to go through slow biotransformation and completely mineralise under aerobic conditions, but not under anaerobic conditions (EU RAR, 2008d).

7.2.4.4 HHCB and AHTN

There is evidence that HHCB is partially biodegradable. Suarez et al., (2010) studied the transformation of HHCB in lab-scale CAS systems (SRT>20 d) and found >80% transformation under aerobic conditions and >60% transformation under anoxic conditions. Wang et al., (2014) found 83% biotransformation of HHCB and 95% biotransformation of AHTN using a laboratory-scale MBR under anaerobic conditions. However, Smyth et al., (2007) studied the removal of HHCB and AHTN during aerobic and anaerobic sludge digestion and observed only removal during aerobic treatment and not during anaerobic treatment.

The oxidation product galaxolidone (HHCB-lactone) has been observed already in the influent to WWTPs, and increased during treatment (Brändli, 2002; Berset et al., 2004). To our knowledge, the toxicity of HHCB-lactone has not been studied. The increase was largest in the plant with extensive aeration (Brändli, 2002). Biodegradation of 30-53% at WWTPs in Germany have been reported (Fahlenkamp et al., 2005).

7.2.4.5 OTNE

OTNE is regarded as persistent according to the registration in REACH (Klaschka et al. 2013).

7.2.4.6 BDE-209

Anaerobic debromination to less brominated congeners occur under anaerobic conditions (Gerecke et al., 2006; He et al., 2006; Robrock et al., 2008). There is a high probability that BDE-209 is transformed in the environment to form substances which themselves have PBT/vPvB properties, or act as precursors to such substances (ECHA RAC, 2015).

7.2.4.7 PCBs

Heavily chlorinated PCBs can be biotransformed to lightly chlorinated ortho-enriched congeners under anaerobic conditions (by anaerobic reductive processes), which can be biotransformed further

under aerobic conditions (by aerobic oxidative processes) (Abramowicz, 1995). However, the biotransformation rates are very slow and limited, if any, transformation can be expected in WWTPs.

7.2.4.8 NP and NPE

Long chain NPnEOs ($n > 4$) have been shown to be biotransformed to short chain NPE, particularly to the monoethoxylate (NP1EO) and diethoxylate (NP2EO), and the rate of transformation increases with increasing chain length (ECHA RAC SEAC, 2014). Using standard biodegradation tests NP1EO and NP2EO have been found to only be inherently biodegradable (ECHA, 2018). Under aerobic conditions these short-chain NPEs can be further oxidised to the corresponding carboxylic acid (e.g. nonylphenoxyacetic acid (NP1EC) or nonylphenoxyethoxyacetic acid (NP2EC)) as well as carboxylated alkylphenol ether carboxylates (CAMPEnC with $m=5-9$ and $n=0$ or 1). Under anaerobic conditions, NP1EO, NP2EO and NP1EC can be transformed to 4-nonylphenol (4-NP) (ECHA, 2018). 4-NP is only a minor transformation product under aerobic conditions.

The degree of mineralisation is assumed to be low (ECHA, 2018).

7.2.5 Abiotic transformation and volatilisation

Abiotic processes such as photooxidation, hydrolysis and reductive transformation may also contribute to changes to the concentration in the wastewater and to the content of the selected HOCs in sludge but have so far been found to be limited in wastewater compared to biotransformation of HOCs (Semblante et al, 2015; Cirja et al., 2008). Photolysis occurs most efficiently during exposure to UV light, which is expected to have limited relevance during conventional wastewater treatment (but is an important mechanism in AOP; see **Section 10.1.3**). However, many PFOA precursors are surfactants and will conceivably undergo photooxidation in aqueous surface layers.

The volatilization of a HOC depends both on its vapour pressure (Henry's law constant, H_k) and its sorption properties, but only becomes significant when H_k is $>0.001-0.01$ (Stenstrom et al., 1989). Only the volatile fragrance musk compounds HHCB and AHTN among the selected HOCs have H_k in this range (0.015; see Table 2), but volatilisation during wastewater treatment has been reported negligible (Suárez et al., 2010).

7.3 Observed removal by treatment units typically applied at Norwegian WWTPs

Since 2009 all Norwegian WWTPs equal to or larger than 50,000 person-equivalents (PE) have been required to analyse for organic micropollutants in weekly composite samples from the influent and treated effluent three times per year. These WWTPs include both plants with highly advanced chemical-biological treatment, chemical treatment, chemically enhanced primary treatment (CEPT) and mechanical treatment. Among the selected HOCs the following are included; BDE-209, PCB7 (congeners 28, 52, 101, 118, 138, 153 og 180), DEHP and 4-NP. Hence, for these compounds the available data is increasing, as summarised in the following.

7.3.1 DEHP

The observed influent and effluent concentrations and estimated removals of DEHP at selected Norwegian WWTPs are summarised in Table 34. The observed average removal ratios at the advanced chemical-biological treatment plants were in the range of 84.0-97.8% with a variation width between 73.6% and 98.9%. The average removals ratios correlated very well with the average annual removal of SS, possibly with an exception at VEAS (Table 34). For the secondary chemical treatment plants the

average removal ratios were in the same range as the chemical-biological treatment plants for two of the four plants (85.7% and 87.5%), while the removal at Knappen WWTP was somewhat lower (79.7%). The observed average removal ratios at the two chemically enhanced primary treatment (CEPT) plants were in the same range as the more advanced treatment plants (71.5% and 90.9%), but the reported average removal ratios at the simple mechanical treatment plants were significantly lower (17-27%) and the observed removals were much more variable (-13-66%). Also, for the WWTPs applying chemical treatment the observed annual removal of sewage sludge appeared to correlate well with the DEHP removal, but at the mechanical WWTPs the average sewage sludge removal was significantly higher than the DEHP removal.

Table 34. Reported influent and effluent concentrations and estimated removals of DEHP at selected Norwegian WWTPs in the period 2009-2017 (omitting periods when the plant is exposed to large modifications that alter the expected performance). Values are given as averages with min-max values in brackets. "n" is number of samples (each influent and effluent), with number >LOD for influent/effluent in brackets. The actual LOD is not reported. Average annual (min-max) removal of sewage sludge at each WWTP covering the same years as the other data. Data provided by Gisle Berge, SSB.

WWTP (last modified)	Type of WWTP	DEHP					SS	
		Period	n	Infl.	Effl.	Rem.	Removal	
		-	-	µg/L	µg/L	%	% (year)	
Chemical-biological (CAS/biofilm)	Bekkelaget (2009)	PT – DN-CAS – N-CAS – S – F – S	2010-17	24 (24/22)	9.0 (0.35-29)	0.53 (0.05-2.5)	94.1 (87.6-98.8)	95.2 (92.4-98.0)
	SNJ (2017)	PT-S-CAS-BioP-S	2011-16	15 (15/10)	10.0 (2.0-26)	1.52 (0.5-7.0)	84.0 (73.6-93.2)	84.1 (81.4-86.5)
	HIAS (2005)	PT – S – CAS/BF – S – F – S	2012-16	12 (12/11)	10.7 (4.4-18)	0.23 (0.05-0.56)	97.8 (96.0-98.9)	96.7 ¹⁵
	VEAS (2008)	PT – BF – F – S	2009-17	27 (27/25)	5.5 (1.3-15)	0.59 (0.01-2.0)	88.2 (81.8-93.8)	95.1 (92.0-97.1)
	NRA (2003)	PT – S – N-BF – DN – BF – F – S	2011-17	21 (21/13)	7.7 (2.0-19)	0.48 (0.11-1.3)	93.7 (88.9-97.6)	93.5 ¹⁶
	Solumstrand (2011)	PT – S – BF – S – F – S	2012-17	12 (12/11)	9.1 (3.0-29)	0.45 (0.1-1.4)	94.5 (90.2-97.0)	93.1 ¹⁷
	Tønsberg (2016)	PT – S – CAS – S – F – S	2009-15	21 (18/15)	4.2 (0.42-11)	0.58 (0.05)	87.5 (79.0-96.1)	N.A.
Sec. chem.	Knappen (1984)	PT – S – F – S	2009-17	27 (27/22)	5.1 (1.2-19)	1.02 (0.01-1.9)	79.7 (66.7-92.7)	88.1 (80.8-93.2)
	ØRA	PT – S – F – S	2009-17	27 (27/11)	5.6 (0.38-20.6)	0.66 (0.05-2.0)	85.7 (74.6-94.6)	82.5 (76.1-91.3)

¹⁵ Value 2012

¹⁶ Value 2011

¹⁷ Value 2012

WWTP (last modified)	Type of WWTP	DEHP					SS	
		Period	n	Infl.	Effl.	Rem.	Removal	
		-	-	µg/L	µg/L	%	% (year)	
CEPT	Ladehammeren (1995)	PT-F-S	2009- 17	27 (23/27)	19.7 (0.5-64)	4.6 (0.21- 14.8)	71.5 (57.6- 90.8)	76.9 (68.5- 81.4)
	Sandefjord (2001)	PT-F-S	2011- 17	21 (21/20)	4.8 (0.24- 11)	0.42 (0.1-1.0)	90.9 (88.0- 94.9)	85.5 ¹⁸
Mechanical	Høvringen (2004)	PT-S	2009- 17	27 (27/26)	9.9 (1.7-36)	6.9 (0.1-29)	25 (-13-66)	68.5 (52.0- 79.7)
	Bore (2016)	PT-S	2009- 13	12 (8/8)	12.4 (1.0-29)	9.0 (0.92-21)	17 (-17-37)	39.1 (24.2- 63.3)
	Grødaland (2013)	PT-S	2015- 17	24 (24/21)	10.9 (0.5-79)	4.8 (0.32-20)	57.7 (-5-94.5)	62.5 (49.5- 73.7)

PT = pre-treatment and typically includes units such as coarse bar screens, grit chambers and grease skimmers. May also include a fine screen (0.1-0.5 mm pore size), which tentatively will improve the removal of sewage sludge up to 40% and BOD₅ up to 20%. **S** = separation and may consist of fine screens, sedimentation (most common in Norway), flotation or filtration. = addition of coagulant. **F** = flocculation chamber to build up settleable flocs. **CAS** = conventional activated sludge process. **BF** = biofilm process. **DN** = denitrification process (step two in N-removal). **N** = nitrification process (step one in N-removal)

These data say nothing about the actual fate of DEHP at the treatment plants; how much will be biodegraded (during wastewater treatment and/or sludge digestion) and how much will end up in the final sludge. As discussed in Section 7.2.4.1, DEHP is regarded as readily biodegradable based on standardised OECD degradation tests and rapid primary transformations were observed in CAS simulation tests. Nevertheless, as was evident from the results from the five-yearly monitoring campaign of Norwegian sewage sludge summarised in Figure 8 and Table 10, DEHP is ubiquitous in sewage sludge (detected in all 95 samples) and was found at relatively high levels (5.8-82.6 mg/kg dw). Predictions made using the European Union System for the Evaluation of Substances (EUSES) to predict the fate of DEHP in a CAS WWTP (see Table 35) support the high removal rate (93%) and indicate 15% biotransformation and an accumulation of approximately 80% in the sludge.

¹⁸ Average 2009-2010; other years not available.

Table 35. Predicted fate of DEHP at conventional activated sludge (CAS) WWTPs using version 1.0 of the European Union System for the Evaluation of Substances (EUSES) (EU RAR, 2008c).

Fate	DEHP
Adsorbed to sludge	78%
Released to water recipient	6.8%
Degraded in WWTP	15%
Evaporated to air	0.016%
Removal rate	93.2%

7.3.2 PFOS and PFOA

Several studies have reported an increase in both PFOS and PFOA concentrations from the influent to the effluent at WWTPs, indicating significant formation from precursors present in the wastewater (Liu and Avendano, 2013; Eriksson et al., 2017). Bossi et al., (2008) measured the concentration of PFOS and PFOA in the influent, effluent and sewage sludge at six Danish WWTPs. For five of the six plants, the concentration of PFOS increased from the influent to the effluent. At the last plant the concentrations of both PFOS and PFOA in the effluent was below the detection limits. PFOS was detected in sludge samples from all six WWTPs at concentrations ranging from 4.8 to 74.1 µg/kg dry weight.

A recent Nordic Council report, demonstrated how precursors for primary PFCAs but also PFSA, dominated the content of PFCs in sewage sludge in 12 samples from the Nordic countries (TemaNord, 2019). The Norwegian two samples, both from HIAS, were included. Both PFOA and PFOS were in the concentration range as 1 and 3 ug/kg dw, respectively. Total PFAS was 64-86 ug/kg dw. The PFCA (perfluoroalkyl carboxylic acids) and PFSA (perfluoroalkyl sulfonic acids) precursors contributed to approximately 75 and 10%, respectively, of the target PFAS (Per- and polyfluoroalkyl substances) substances analysed for. Different precursors for PFCAs were measured in the range of < LOD and 17 ug/kg dw, where isomers of diPAPs (polyfluoroalkyl phosphoric acid diesters) – and particularly 6:2 diPAPs - showed highest concentration. In this study, the extractable organofluorine (EOF) was in the range of <556-695 µg F/kg. The unidentified organofluorine part accounted for approximately 90% in the sludge sample from HIAS.

7.3.3 SCCP

We have not found any studies that look at SCCP at Norwegian WWTPs, and there appears to be surprisingly few studies elsewhere too. Zeng et al., (2012) studied the fate of SCCP in an advanced chemical-biological WWTP in China and found that approximately 73% of the influent SCCP ended up in the final sludge (caused by sorption to the sludge) and <1% ended up in the treated effluent. About 27% were lost, suggested by the authors to be due to biotransformations during the applied anoxic-aerobic wastewater treatment.

7.3.4 HHCB and AHTN

Very few data are available for HHCB and AHTN in Norwegian WWTPs. They were included in the Norwegian EPA screening programme in 2017 (Konieczny et al., 2018), in which 24-hour flow-proportional composite samples from the influents and effluents at VEAS and HIAS were collected. Only one influent sample from each plant was collected, while two effluent samples from each plant were collected (one of which was collected at the same day as the influent sample).

HHCB

- At VEAS the concentration of HHCB in the influent was 8400 ng/L and 1 000 ng/L in both effluent samples, indicating 88% removal of HHCB. The average concentration (and standard deviation of the mean) in the three collected sludge samples from VEAS was 7.4 ± 0.7 mg HHCB/kg dw.
- At HIAS the concentration of HHCB in the influent was 13 000 ng/L in the influent and 2 500 ng/L and 2600 ng/L in the effluent, hence, an approximate 80% removal of HHCB. The average concentration in the three sludge samples from HIAS was 13.0 ± 1.0 mg HHCB/kg dw.
- If extrapolating these numbers to the whole year of 2017, an estimated total of 714 kg HHCB was removed from the incoming wastewater at VEAS (96.43 mill. m³ treated) and an estimated total of 76 kg HHCB was removed from the incoming wastewater at HIAS (7.29 mill. m³ treated). Taking the total annual sludge production at VEAS (10 966 tons dw) and HIAS (2 174 tonns dw) in 2017 into account, an estimated 126 kg HHCB ended up in the sludge at VEAS and an estimated 28 kg HHCB ended up in the sludge at HIAS. Hence, the apparent annual loss of HHCB during treatment was 587 kg at VEAS and 48 kg at HIAS, or approximately 80% and 60% of the HHCB that was removed at VEAS and HIAS, respectively. This loss may be due to biotransformations, though volatilisation should not be excluded as a potential pathway.

AHTN

- The data for AHTN at VEAS were 2 100 ng/L in the influent and 250 ng/L and 270 ng/L in the effluent, hence, also an approximate 88% removal of AHTN (as for HHCB). The average concentration in the three sludge samples from VEAS was 6.9 ± 1.0 mg AHTN/kg dw.
- At HIAS the concentration of AHTN in the influent was 3 200 ng/L, the effluents 600 ng/L and 610 ng/L, and an almost identical estimated removal of AHTN (81%) as HHCB. The average concentration in the three sludge samples from HIAS was 10.3 ± 0.6 mg HHCB/kg dw.
- If doing the same extrapolation for AHTN as was described above for HHCB, an estimated 117 kg AHTN ended up in the sludge at VEAS and an estimated 22 kg AHTN ended up in the sludge at HIAS. Hence, the apparent annual loss of AHTN during treatment was 59 kg at VEAS and -4 kg at HIAS, or approximately 30% and -20% of the AHTN that was removed at VEAS and HIAS, respectively.

Both VEAS and HIAS are advanced chemical-biological WWTPs, which also include anaerobic digestion of the generated sludge (at HIAS also thermal hydrolysis prior to the anaerobic digestion). However, these estimations should be interpreted with caution as the data are very limited, and a real mass balance is not possible to make.

There are several European studies (Germany, Switzerland, Austria, UK and the Netherlands) that generally report removal percentages of 75-90% (EU RAR 2008a; EU RAR 2008b), hence confirming the observed removal ratios at VEAS and HIAS. Simonich et al., (2002) reported good correlation between removal of HHCB and suspended solids. Carballa et al., (2004) observed a reasonably good correlation between the removal of HHCB ($46 \pm 24\%$) and AHTN ($39 \pm 21\%$) and the removal of total suspended solids ($56 \pm 23\%$) over the primary sedimentation tank treatment. The correlation was even better over the secondary aerobic CAS treatment step with $66 \pm 8\%$ removal of HHCB, $78 \pm 10\%$ removal of AHTN and $78 \pm 12\%$ removal of TSS. However, the potential influence of biotransformation was not evaluated.

Lee et al., (2001) simulated a CAS system in the lab feeding the system with ¹⁴C-labelled AHTN to try to make a mass balance for AHTN. Of the parent AHTN, 87.5% was removed. 42.5% had transformed into metabolites, 44.3% of the AHTN had been removed by sorption and 3.3% had been removed by volatilisation. Ca. 10% of the metabolites were sorbed. In a German study at two large WWTPs in

Düsseldorf and Cologne, HHCB was removed by 80-85% with 53-30% found to be due to biotransformation to HHCB-lactone (Fahlenkamp et al., 2005). Contrary to this finding, Besser (2003) studied the transformations of HHCB occurring at WWTPs along the river Ruhr and found that a maximum of 5-10% of the influent HHCB was transformed by enzymatic processes (i.e. biotransformed) based on the enantiomeric ratios of HHCB-lactone in the effluent. No further biotransformation was evident. This was further supported by the findings in a follow-up study of HHCB and AHTN in a large German CAS WWTP, showing that ca. 50% of HHCB and ca. 80% of AHTN ended up in the sludge, and only up to 7% of HHCB was transformed to HHCB-lactone, while the transformation of AHTN was negligible (Bester, 2004).

7.3.5 OTNE

Very few data are available for OTNE in Norwegian WWTPs. OTNE was though included in the Norwegian EPA screening programme in 2017 (Konieczny et al., 2018) (see HHCB and AHTN above):

- At VEAS the concentration in the influent was 4900 ng/L and 670 ng/L and 720 ng/L in the effluent (the latter value from the same 24-hours as the influent), indicating an approximately 85% removal of OTNE. The average concentration of OTNE in the three collected sludge samples from VEAS was 11 mg/kg dw (all three samples showing the same value).
- At HIAS the concentration in the influent was 9300 ng/L, while 1800 ng/L and 1900 ng/L were found in the effluent (the former value from the same 24-hours as the influent), indicating an approximately 80% removal of OTNE. The average concentration of OTNE in the three collected sludge samples was 17±1.0 mg/kg dw at HIAS.
- An estimated total of 403 kg OTNE was removed from the incoming wastewater at VEAS and an estimated total of 55 kg OTNE was removed from the incoming wastewater at HIAS, and an estimated 187 kg OTNE ended up in the sludge at VEAS and an estimated 37 kg OTNE ended up in the sludge at HIAS. Hence, the apparent annual loss of OTNE during treatment was 216 kg at VEAS and 18 kg at HIAS, or approximately 50% and 30% of the OTNE that was removed at VEAS and HIAS, respectively.

Both VEAS and HIAS are advanced chemical-biological WWTPs, which also include anaerobic digestion of the generated sludge (at HIAS also thermal hydrolysis prior to the anaerobic digestion). However, these estimations should be interpreted with caution as the data are very limited, and a real mass balance is not possible to make.

OTNE has similar chemical-physical properties as HHCB and AHTN (see Table 2 and Table 32) and is therefore expected to show more or less the same partitioning to sludge. OTNE is registered as persistent in the environment according to the registration in REACH (Klaschka et al. 2013), but it may, due to its structural similarities to HHCB and AHTN (see Table 33, also be partially biotransformed during biological treatment in WWTPs.

7.3.6 BDE-209

The observed influent and effluent concentrations and estimated removals of BDE-209 at selected Norwegian WWTPs are summarised in Table 36. The observed average removal ratios at the advanced chemical-biological treatment plants were varied, from 45% (with very limited data) to 93% with a variation width between 22% and 99.6%. The average removals at these plants was 81%, if leaving the very low removal at HIAS out of the calculation. The average removals ratios were generally somewhat lower than the average annual removal of SS, except for at SNJ, where the BDE-209 removal was higher than the sewage sludge removal (Table 36). The BDE-209 removals at the advanced chemical

treatment plants (secondary treatment) and at the chemically enhanced primary treatment (CEPT) plants the average removal ratios were in the same range as at the chemical-biological treatment plants and ranged from 51% to 82%. The average removals at the CEPT plants appeared to be slightly better than at the advanced chemical treatment plants. The average removal ratios at the simple mechanical treatment plants were very variable, ranging from literally no removal (-3%) to 69%, with a variation width from -64% to 87% removal. However, the measured values were often quite low resulting in high uncertainties.

Table 36. Reported influent and effluent concentrations and estimated removals of BDE-209 at selected Norwegian WWTPs in the period 2009-2017 (omitting periods when the plant is exposed to large modifications that alter the expected performance). Values are given as averages with min-max values in brackets. "n" is number of samples (each influent and effluent), with number >LOD for influent/effluent in brackets. The actual LOD is not reported. Average annual (min-max) removal of sewage sludge at each WWTP covering the same years as the other data. Data provided by Gisle Berge, SSB.

WWTP (last modified)	Type of WWTP	BDE-209					SS	
		Period	n	Infl.	Effl.	Rem.	Removal	
		-	-	µg/L	µg/L	%	% (year)	
Chemical-biological (CAS/biofilm)	Bekkelaget (2009)	PT – DN-CAS – N-CAS – S – F – S	2010-17	24 (11/0)	0.042 (<0.01-0.150)	<0.01	90 (79-95)	95.2 (92.4-98.0)
	SNJ (2017)	PT-S-CAS-BioP-S	2011-16	15 (13/4)	0.81 (<0.01-3.4)	0.022 (<0.01-0.08)	93 (73-99.6)	84.1 (81.4-86.5)
	HIAS (2005)	PT – S – CAS/BF – S – F – S	2012-16	6 (6/5)	0.24	0.13	45	96.7 (value 2012)
	VEAS (2008)	PT – BF – F – S	2009-17	27 (19/7)	0.035 (<0.01-0.12)	0.007 (<0.01-0.038)	78 (46-92)	95.1 (92.0-97.1)
	NRA (2003)	PT – S – N-BF – DN – BF – F – S	2011-17	12 (11/3)	0.111 (0.02-0.29)	0.036 (<0.01-0.18)	68 (47-94)	93.5 (value 2013)
	Solumstrand (2011)	PT – S – BF – S – F – S	2012-17	12 (8/2)	0.046 (<0.01-0.15)	0.011 (<0.01-0.078)	74 (22-99)	93.1
Sec. chem.	Tønsberg (2016)	PT – S – CAS – S – F – S	2009-15	21 (18/15)	0.023 (<0.01-0.15)	0.007 (<0.01-0.04)	68 (47-82)	N.A.
	Knappen (1984)	PT – S – F – S	2009-17	27 (18/15)	0.015 (<0.01-0.068)	0.006 (<0.01-0.03)	56 (-16-95)	88.1 (80.8-93.2)
	ØRA	PT – S – F – S	2009-17	21 (9/3)	0.026 (<0.01-0.083)	0.014 (<0.01-0.097)	51 ¹⁹ (14-89)	82.5 (76.1-91.3)
CEPT	Ladehammeren (1995)	PT – F – S	2009-17	27 (16/6)	0.142 (<0.01-1.0)	0.028 (<0.01-0.12)	82 (60-98)	76.9 (68.5-81.4)
	Sandefjord (2001)	PT – F – S	2011-17	21 (13/2)	0.171 (<0.01-0.23)	0.008 (<0.01-0.037)	74 (22-99)	85.5 ²⁰

¹⁹ Omitted results from 2013 as the removal was much lower (-68%)

²⁰ Average 2009-2010; other years not available.

WWTP (last modified)	Type of WWTP	BDE-209				SS		
		Period	n	Infl.	Effl.	Rem.	Removal	
		-	-	µg/L	µg/L	%	% (year)	
Mechanical	Høvringen (2004)	PT – S	2011- 17	21 (?/?)	0.099 (<0.01- 0.63)	0.025 (<0.01- 0.11)	69 (33- 87)	68.5 (52.0- 79.7)
	Bore (2016)	PT – S	2009- 13	12 (3/2)	0.012 (<0.01- 0.10)	0.011 (<0.01- 0.10)	28 (0- 56)	39.1 (24.2- 63.3)
	Grødaland (2013)	PT – S	2015-17	9 (?/?)	0.022 (<0.01- 0.28)	0.021 (<0.01- 0.10)	-3 (-64- 50)	62.5 (49.5- 73.7)

PT = pre-treatment and typically includes units such as coarse bar screens, grit chambers and grease skimmers. May also include a fine screen (0.1-0.5 mm pore size), which tentatively will improve the removal of sewage sludge up to 40% and BOD₅ up to 20%. **S** = separation and may consist of fine screens, sedimentation (most common in Norway), flotation or filtration. = addition of coagulant. **F** = flocculation chamber to build up settleable flocs. **CAS** = conventional activated sludge process. **BF** = biofilm process. **DN** = denitrification process (step two in N-removal). **N** = nitrification process (step one in N-removal)

7.3.7 PCBs

The samples collected at the Norwegian WWTPs were analysed for PCB₇, but the levels were generally very low and either close to or below LOQ, hence, the estimated removals are very uncertain. In the few cases where the levels were somewhat high in the influent (0.024-0.220 µg/L), the calculated removals were highly variable (0->95%) and appeared not to be related to the applied treatment. Other studies typically show high removal that are relatively closely correlated with the removal of sewage sludge (US EPA, 1977). Yoshida et al., (2015), studying the Danish Avedøre WWTPs, found that >90% of the PCBs were associated with the primary sludge (i.e. removed by primary treatment) and only a small fraction was found in the secondary sludge (after aerobic treatment).

7.3.8 NP and NPE

The samples collected at the Norwegian WWTPs were only analysed for 4-NP and not NPE, and most of the samples were below the LOQ (0.01 µg/L), both in the influents and in the effluents (Blytt and Stang, 2019). Vogelsang et al., (2006) studied the removal of NP at five Norwegian WWTPs and observed high removal (>90%) at the chemical-biological treatment plant, relatively good removal (60->90%) at two of the three chemical treatment plants, but negative or rather low removal (-20-50%) at the third, and poor removal (0-20%) at the mechanical treatment plant.

Ahel et al., (1994) reported results for nonylphenol, NPnEO (n=1 to 20), NP1EC and NP2EC at different stages at 11 biological wastewater treatment plants in Switzerland. In untreated sewage and effluents from primary treatment the main components found were generally NPnEO (n=3-20), which accounted for 82% of the total nonylphenol derivatives present, followed by NP1EO + NP2EO (11.5% of the total), nonylphenol (3% of the total) and NP1EC + NP2EC (3.1% of the total). In the effluents after the biological treatment the composition of the nonylphenol based compounds had changed markedly, with NPnEO (n=3-20) only present in trace amounts. NP1EC and NP2EC were now the most abundant substances found (46% of the total), followed by NP1EO + NP2EO (22% of the total) and nonylphenol (3.9% of the total).

Important considerations

Conventional WWTPs are not designed to remove micropollutants, but if dimensioned and managed properly, compounds that adsorb strongly to the suspended solids may be efficiently removed from the water phase at WWTPs applying chemical and/or biological treatment processes. This transfers the compounds over to the sludge, but only biotransformation (or evaporation) will entail real removal. Biotransformation can only be expected to occur during biological treatment (about 40% of the wastewater in Norway).

- It is crucial to consider internal transfer lines that may transfer large quantities of the compound back to earlier treatment steps which may cause accumulation in certain parts of the WWTP.
- The impact of the pH during coagulation on the sorption of ionisable HOCs (i.e. PFOS, PFOA, NP and NPE) should be assessed.
- The impacts of dissolved and colloidal matter (DCM) such as humic acids on the mobility and bioavailability of HOCs need to be assessed in more detail (DEHP of particular interest).
- More studies are needed to elucidate the impact of SRT on other sludge properties, such as floc size and density, and their implications on the sorption of HOCs.
- The biotransformation of HOCs appears to preferentially occur under aerobic conditions, but anoxic conditions may promote the sorption of hydrophobic compounds on sludge, which possibly facilitated their biotransformation under aerobic conditions due to sludge recirculation between aerobic and anoxic conditions. The inherent dynamics need to be elucidated.
- The potential key role of nitrifying bacteria in the biotransformation of the selected HOCs should be evaluated.
- The combined impacts of the MBR system on the biotransformation of the selected HOCs due to the increased heterogeneity of the microbial community, the ratio of planktonic bacteria and reduced floc sizes, as well as the decreased food-to-microorganism ratio as compared to CAS need to be assessed.

Biotransformation may also contribute to increase the concentration of a specific compound, since the transformation of other compounds (i.e. precursors) can have the specific compound as its transformation product. This is highly relevant for PFOS and PFOA, and NP and NPE.

There is a high probability that BDE-209 and PFAS are transformed to substances that themselves have PBT/vPvB properties, or act as precursors to such substances.

To our knowledge, the toxicity of HHCB-lactone has not been studied.

8 Fate during sewage sludge treatment

8.1 Commonly applied processes to stabilise and sanitise sewage sludge in Norway

Processes that have been used to stabilise and sanitise municipal sewage sludge at the 137 registered facilities for treating sewage sludge in Norway are shown in Figure 21. The most common process combinations used at the larger facilities are:

- Thermophilic aerobic pre-treatment and mesophilic anaerobic digestion (e.g. Fuglevik)
- Pasteurisation and mesophilic anaerobic digestion (e.g. ØRA/FREVAR)
- Thermophilic anaerobic stabilisation (e.g. Bekkelaget WWTP)
- Anaerobic stabilisation and thermal drying (e.g. IVAR), but is taken out of service several places due to economy and operational problems
- Thermal hydrolysis (the Cambi process) and mesophilic anaerobic digestion (e.g. HIAS)

At the smaller facilities the following process combinations are more common:

- Lime stabilisation of dewatered sludge
- Windrow composting
- Reactor (rotating) composting
- Long-term storage and simple windrow composting

Wet composting is only used at one municipal WWTP in Norway.

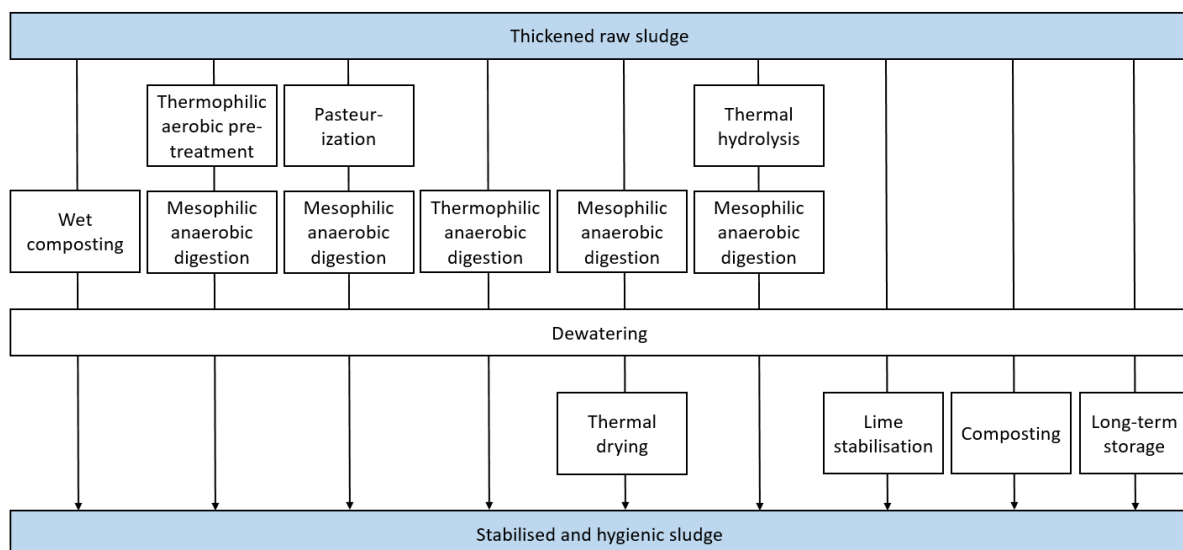


Figure 21. Overview of sludge treatment processes applied in Norway. Adapted from Norsk Vann (2010)

8.2 Expected and observed effects during sludge treatment

8.2.1 Expected biodegradation during aerobic and anaerobic digestion

Anaerobic digestion has become a quite common way of treating Norwegian sewage sludge (see e.g. Table 10), since the amount of sludge is drastically reduced through the production of valuable biogas (contributing to wastewater management cost reductions and promoting the sustainability of the WWTP) at the same time as the sludge is stabilised (including odour minimisation). During aerobic digestion, however, carbon is lost from the sludge as CO₂. Both types of carbon losses from the sludge lead to an apparent accumulation or increase in concentration of compounds that are not reduced at the same rate.

As discussed in Section 7.2.4, aerobic conditions are more likely to promote biotransformation of the selected substances that may be biodegraded (i.e. DEHP, SCCP, HHCB, AHTN and NPE), while anaerobic conditions tend to strongly limit the biotransformation. This was confirmed for HHCB and AHTN by Smyth et al., (2007) who studied their fate during aerobic and anaerobic sludge digestion. One important exception is the potential dichlorination of the highly chlorinated PCBs, which only occurs under anaerobic conditions (but very slowly). Notably, full-scale anaerobic digesters generally have negligible or poor biodegradation of HOCs, and reports of high HOCs removal are limited to laboratory-scale anaerobic digesters (Semblante et al., 2015).

As also mentioned in Section 7.2.2, hydrophobic HOCs such as PAHs, NP and PCBs have been shown to sorb stronger to anaerobically digested sludge than to primary and secondary raw sludges due to the physical and chemical changes that occur with the sludge matrix during anaerobic digestion (Barret et al., 2010b). The particulate matter in the anaerobically digested sludge was smaller (median diameter of 19 µm) than in primary and secondary raw sludges (d₅₀ ca. 100 µm), and while approximately 90% of the entities in the dissolved and colloidal fractions were <1 kDa in molecular sizes, only 5% of the entities in the anaerobically digested sludges were in that size category (Barret et al., 2010b). These observations fit well with the assumption that the smaller-sized fractions may be more readily bioaccessible to microorganisms, and they also suggest that anaerobic treatment enhances the proportion of hydrophobic material responsible for the sorption of hydrophobic micropollutants.

During biological wastewater treatment the substances have already been subjected to aerobic conditions (and possibly also anoxic conditions), and since they sorb strongly to the sludge solids, their retention time in the aerobic bioreactor is primarily determined by the SRT and not the HRT. However, since a large fraction of the influent sewage sludge is removed during primary treatment, also a large fraction of each substance is probably removed before the wastewater enters the biological step.

Moreover, the conditions during sludge digestion is otherwise much different than what the substances are exposed to during biological wastewater treatment;

- The temperature is significantly higher; either mesophilic conditions (typically 35-40°C) or thermophilic conditions (typically 53-58°C, or up to 70°C).
- During anaerobic digestion, the residence time is typically 15-40 days, while much shorter exposure times to the elevated temperature (1.5-2 hours at 55°C) is needed under thermophilic conditions, though the total residence time is typically about 14 days. During aerobic digestion the processes occur faster, and 5-10 days are usually enough.
- The food-to-microorganism ratio changes drastically during the digestion process (though, depending upon the applied feeding strategy), which may promote the proliferation of microorganisms that are able to co-metabolise some of the selected HOCs.

Several studies have investigated removal of HOCs during anaerobic digestion in laboratory scale, however, since rather contractive results have been observed (e.g. Carballa et al., 2007a,b; Gonzales-Gil et al., 2016) and studies are performed under different conditions, with and without spiked HOCs etc, this is not further discussed.

Table 37 summarises observed removal ratios during anaerobic digestion, which are surprisingly high to be under anaerobic conditions considering that limited biotransformations are expected to occur under such conditions. Despite the higher temperature under thermophilic digestion, there is no clear evidence supporting increased biotransformation under thermophilic compared to mesophilic conditions. No data have been found for digestions done under aerobic conditions.

Table 37 Summary of observed removal during anaerobic digestion.

Substance	Removal (%)	Days	Temp °C	Ref
DEHP	61	27	35	Parker et al. 1994
	55-70	16	37	Mailler et al., 2014
SCCP				
HHCB	60-70	10	55	Carballa et al 2006 & 2007
	60-70	20	37	Carballa et al 2006 & 2007
AHTN	60-70	10	55	Carballa et al 2006 & 2007
	60-70	20	37	Carballa et al 2006 & 2007
OTNE				
PBDE-209	40-90	16	37	Mailler et al., 2014
NP	100	60-90	37	Paterakis et al. 2012
	50	60-90	55	Paterakis et al. 2012
	30-65	16	37	Mailler et al., 2014
	0.4	20	37	Hernandez-Raquet et al., 2007
NP1E	3.8	20	37	Hernandez-Raquet et al., 2007
	65-90	16	37	Mailler et al., 2014
NP2E	50-95	16	37	Mailler et al., 2014
	2.6	20	37	Hernandez-Raquet et al., 2007
NPE	58	15-40	37	Paterakis et al. 2012
	92	15-40	55	Paterakis et al. 2012
PFOA				
PFOS				
PCB7	58 ²¹ (mean)		37	Macherzynski et al 2014
	71 ²² (mean)		37	Macherzynski et al 2014

²¹ Solid phase

²² Supernatant

Table 38. Summary of measured concentrations of selected HOCs in digestate from anaerobic digestion (include mesophilic and thermophilic, and in different combinations of treatment of sewage sludge).

Substance	Unit	Conc.	Country	Ref.
DEHP	mg/kg dw	9.3 - 67.4 (min-max)	Norway	Blytt et al., 2019
DEHP	mg/kg dw	1.0-107 (min-max) 22.9 (mean) 9.5 (median)	Finland n=20	Suominen et al., 2014
DEHP	mg/kg dw	18.14 (untreated <0.46) (pilot scale 0.78 m ³)	Italy	Scaglia et al 2018
PFOS	µg/kg dw	1.1- 130 (min-max)	Norway	Blytt et al., 2019
PFC	µg/kg dw	0.97-168 (min-max) 42.9 (mean) 18.5 (median)	Finland n=19	Suominen et al., 2014
PFOA	µg/kg dw	<1-9.2 (min-max)	Norway	Blytt et al., 2019
SCCP	µg/kg dw	<50- 1400 (min-max)	Norway	Blytt et al., 2019
HHCB	mg/kg dw	4.2- 31 (min-max)	Norway	Blytt et al., 2019
AHNT	mg/kg dw	1.3 - 6.10 (min-max)	Norway	Blytt et al., 2019
OTNE	mg/kg dw	11; 16; 18 (n=3)	Norway	Konieczny et al., 2018
BDE-209	µg/kg dw	6.4-11300 (min-max) 1780 (mean) 1040 (median)	Finland n=16	Suominen et al., 2014
BDE-209	µg/kg dw	2 - 1100 (min-max)	Norway	Blytt et al., 2019
PCB7	µg/kg dw	all <50	Finland	Suominen et al., 2014
PCB7	µg/kg dw	5 - 57 (min-max)	Norway	Blytt et al., 2019
PCB7	mg/kg dw	0.46 (untreated 0.17) (pilot scale 0.78 m ³)	Italy	Scaglia et al 2018
NP+NPE	µg/kg dw	< LOQ - 54 (min-max) 11.2 (mean) 7.5 (median)	Finland n=20	Suominen et al., 2014
NP+NPE	mg/kg dw	< 0.67 (<0.46 untreated) (pilot scale 0.78 m ³)	Italy	Scaglia et al 2018
NP+NPE	µg/kg dw	2580 - 11400 (min-max)	Norway	Blytt et al., 2019

8.2.2 Expected effect of thermal hydrolysis as pre-treatment to anaerobic digestion

The so-called Cambi process, a Norwegian invention, is thermal hydrolysis process during which the sludge is kept at 160-170°C under a pressure of 7-7.5 bar for 30 min. The main purpose of introducing this thermal hydrolysis process is to improve the bioavailability of the organic matter in the sludge and thereby increasing the biogas production and reducing the final sludge production. The combined high heat and pressure may also contribute to the hydrolysis of the selected HOCs.

The Cambi process is applied at both HIAS and Lindum biogas, which were both included in the last monitoring campaign conducted by Norsk Vann BA (Blytt and Stang, 2019). Interestingly, at Lindum biogas also untreated sewage sludge samples were collected and analysed, hence it is possible to very roughly estimate the removal by the combined thermal hydrolysis (Cambi process) and mesophilic anaerobic digestion. However, the sludge reduction (i.e. biogas production) is not available. Based on the average values only SCCP appeared to be biotransformed to a significant degree (ca. 90%).

8.2.3 Expected evaporation during thermal drying

Mailler et al., (2014) studied the loss of DEHP, NP, NPE and BDE-209 during thermal drying at three WWTPs in the Paris area. BDE-209 and DEHP remained at the original levels in the sludge during the

drying process, while the volatilisation of NP, NP1EO and NP2EO varied considerably; NP between 20% and 90%, NP1EO between 50% and 95% and NP2EO between 0% and 40%. The high volatilisation may have been caused by the elevated temperature (260°C) as compared to conventional dryers (105°C).

8.2.4 Mobilisation after alkaline stabilisation

Alkaline treatment using e.g. slaked lime or fly ash to increase the pH to ca. 12, or addition of unslaked lime to also develop heat (60°C), is applied to stabilise the sludge. The drastic pH change is expected to alter the sorption behaviour of ionisable compounds such as PFOS, PFOA, NP and NPE. However, the tests that have been conducted on sewage sludge so far has not resulted in any significant immediate mobilisation of these compounds from the sludge, but the mobilisation when mixed with soil appears to increase (Semblante et al., 2015).

8.3 Other substrates than sewage sludge in anaerobic digestion processes

In addition to sewage sludge, common substrates in anaerobic digestion processes are manure, food waste, fish waste and organic sludge from other industries. However, the available data for these substrate types are very scarce.

As described under section 7.2.1, anaerobic digestion is a treatment method commonly used for sewage sludge from WWTPs. However, in addition to sewage sludge, common substrates in anaerobic digestion processes are manure, food waste, fish waste, or organic sludge for other industries. Anaerobic digestion processes produce bioenergy and reduce climate gas emissions but is also a way for recycling nutrients by using digestate as fertilisers.

The removal rates of HOCs during anaerobic digestion are influenced by many parameters and it is not possible to establish degradation rates or DT_{50} for the selected HOCs based on full scale plant data. Several studies have investigated the fate of HOCs at laboratory scale (10-15 L) under different conditions, however, rather contradictory results have been observed, and thus not further discussed (e.g. Carballa et al., 2007a,b; Gonzales-Gil et al., 2016).

A summary of analyses of selected HOCs in digestate is shown in Table 38. Most of the data are from Norwegian (Blytt and Stang, 2019) and Finish (Suominen et al., 2014) digestate where sewage sludge is the only or dominating substrate fraction.

Important considerations

Despite that limited biotransformation is expected during anaerobic conditions, significant removals rates have been observed for several of the selected HOCs (DEHP, HHCb, AHTN, BDE-209, NP, NPE and PCB7). These results need to be verified in well controlled tests as well as at full scale under both mesophilic and thermophilic conditions.

The biotransformation of the selected HOCs during aerobic digestion is largely unknown and should be assessed in well controlled tests as well as at full scale under both mesophilic and thermophilic conditions.

The effects of thermal hydrolysis on the bioavailability of the selected HOCs should be assessed.

The effect of high temperature drying as compared to normal temperature drying on the evaporation of the selected HOCs should be assessed.

9 Fate during composting

9.1 The composting process

Composting of organic waste requires processes that ensure fast degradation and safe sanitation of the organic material. The organic wastes most used as substrate for composting today are garden waste, sewage sludge and separated organic household waste. The aerobic composting process can be done mesophilic at 37 °C and thermophilic over 55 °C. The composting process is dependent on effective gas transmission, O₂ in and CO₂ out of the organic waste surface mix. The quantity of air required is related to the amount of bulking agent and the organic waste. The pH and dry matter of the waste are also important factors that influence the composting. Thermophilic processes are most effective in degradation of organic material from 12 - 30 days.

The high-rate composting phase is characterized by high thermophilic microbial respiration at temperatures above 45 °C (Haug, 1993; Epstein, 1997). Composting plants for source separated organic household waste have reported problems in the establishment of the high-rate composting phase. The problems are often related to slow degradation of organic matter and partly also problems to achieve thermophilic conditions in the composting materials. Several authors have suggested that the problems are related to low pH due to formation of short-chained organic acids (mainly lactic and acetic acid), during pre-storage of waste and during initial phases of composting (Beck-Friis et al., 2001; Beck-Friis et al., 2003; Reinhardt, 2002).

Measures to reduce problems or improve failing compost processes are several. The following suggestions are reported: maintaining mesophilic microorganisms in the composting materials until the pH rises above 5.5 (Smårs et al., 2001); yeast inoculation as an activator in cases of composting failure (Choi and Park, 1998); using a starter culture of active compost with fed-batch composting of food waste to prevent low pH conditions (Sundberg and Jönsson, 2005); and addition of 5 % lime (Ca(OH)₂) or extra bulking agent to establish thermophilic microorganisms and high-rate respiration in composting of acidic source separated organic household (Bøen et al., 2006; Aasen et al., 2006; Bergersen et al., 2009).

These factors will also affect HOCs reduced during composting.

9.2 Removal of selected HOCs during composting and measured concentrations in compost

The dry matter content decreases during composting and persistent contaminants might increase when the concentration is related to dry matter (dry weight). Therefore, analyses of persistent contaminants are sometimes related to ash or loss of ignition instead of dry matter.

There are some studies of HOCs and compost from field-scale composting plants in Denmark and Finland, but also data from other countries is included in the summary in Table 39. Most analyses of HOCs in compost are of composted sewage sludge (some sludges have been anaerobic digested, others not). There are anaerobic digestion treatment plants using other substrates than sewage sludge and which also might contain emerging contaminants, e.g. fish waste.

The temperature during composting, type of sludge, and the contaminant concentration influence the degradation rate (Sadeh et al., 2014; Amir et al., 2005). The composition of the microbial communities (bacteria, actinomycetes and fungi) changes during different temperature phases. Actinomycetes and fungi are likely to be involved in removal of contaminants with optimal degradation at lower temperatures than bacteria, which are more likely to cause degradation of contaminants with optimal

removal at higher temperature. While biodegradation is the major removal process for most HOCs during composting, abiotic processes such as humification might be rather important for the most persistent substances. For volatile compounds evaporation might also be part of the removal.

While some contaminants show removal over a wide range of temperatures thereby indicating degradation by several types of microorganisms, others had a narrower optimal temperature range, suggesting that fewer types of microorganisms are involved in removal. The removal observed during composting is also likely to involve reduction in concentration due to humification. Sadeef et al., (2014) demonstrated also that the metabolites from parent compounds (e.g. metabolites from HBBC) might have different optimal degradation temperatures and suggested that microorganisms involved are not the same for parent compounds and their metabolites.

There are data from composting processes related to field scale composting plants from Denmark (Poulsen et al., 2010; Sadeef et al., 2014) and Finland (Kapanen et al., 2013). For example, analysis was done on selected HOCs from samples of compost taken during the first 24 days of treatment at a full-scale composting facility in Denmark (Poulsen et al., 2010). In Finland, Kapanen and coworkers composted anaerobically digested municipal sewage sludge samples from WWTPs in pilot scale with aeration (turned weekly), in addition to analysing compost samples from two field scale composting plants for municipal treatment (composting aged 3 months, 1 year, and 1-2 years). A pilot scale composting experiment with dewatered sewage sludge in China is also discussed (Cai et al., 2012). Cai and co-workers compared compost turned manually every 5th day (MTC), intermittently aerated compost (IAC) (static pile with forced aeration system) and naturally aerated compost (NAC) over a composting period of 56 days, at which point samples were collected and analysed.

Sadeef and co-workers (2014) took compost samples from a field scale composting plant immediately after the first turn following construction (same treatment plant as the study by Poulsen et al., 2010). The compost samples were further incubated under different temperature regimes, from 18 up to 70°C, and removal rate of selected HOCs were studied. For example, the average removal rate for DEHP at the studied temperatures was 69%, and optimal degradation rates were observed in a relatively narrow temperature range (for DEHP about 37 °C), indicating that few microbial strains that have an optimal growth/activity around this temperature range are efficient degraders of this compound. The differences in optimal degradation temperatures show that some contaminants will have higher removal rate during thermophilic phase while for other contaminants removal will be higher at the mesophilic phase, and others again, may have their highest removal during the mature phase.

Based on this, it would be possible to combine composting processes by different mixtures of substrates to optimize for the development of the compost processes and temperature phases.

The composition and amount of the substrates included in the composting, and at which time during the composting process the samples were taken, differed. Based on the rather scarce data and high variation in composting conditions (e.g. type of substrate/co-substrate, at which time during the composting process the samples were taken, temperature and compost period), only general trends for degradation or removal of the selected HOCs is possible.

Generally, the highest removal rates are reported for DEHP, HHCB, and OTNE, somewhat lower removal rates for AHTN and NP+NPE, and the lowest reduction rates for PCB7 under a standard composting period. The removal rates might be increased by extending the composting period. A summary of available concentrations of the selected HOCs in compost and percent removal during composting is presented in Table 39.

9.2.1 DEHP

Reported DEHP concentrations in compost after 26 and 124 days in a pilot scale composting study were 20 and 11 mg/kg dw, respectively (Kapanen et al., 2013). This corresponds to a calculated 20 and 56% removal related to an estimated initial concentration in the compost after mixing with other substrates. Sewage sludge concentration of DEHP before mixing with other co-substrates but after dewatering was 57 mg/kg dw. A sewage sludge sample from another WWTPs contained 110 mg/kg dw DEHP (Kapanen et al., 2013).

DEHP concentration in field scale compost after 3 months and 1 year were 38 and 16 mg/kg dw (same pile), and in another field scale compost pile after 1-2 year composting, 2 mg/kg dw (Kapanen et al., 2013).

In the pilot scale study by Cai et al., (2012), an initial DEHP concentration at 10.9 mg/kg dw in sewage sludge, was reduced to 4.60 mg/kg dw after mixing with co-substrate and around 2 months natural aerated composting (turned approx. weekly). Initial concentration in compost after mixing with substrate is not present, and % removal is given related to sewage sludge, 57.8%.

Analysis of selected HOCs of samples from compost from full scale composting in treatment plant in Denmark during first 24 days showed that DEHP decreased from 31000 mg/kg dw to around 2000 mg/kg dw (Poulsen et al., 2010) and corresponded in approximately 84% removal. Incubation studies with compost samples from the same Danish composting treatment plant showed a 69% average DEHP removal, and that the optimal degradation rate was only observed in a relatively narrow temperature range, thus indicating that few strains of organisms present in the compost degraded this compound. Estimated optimal temperature was around 42 °C.

It is reported that generally, the removal rates of phthalates with short alkyl side-chains were higher than those with long side-chains (Cia et al., 2012; Staples et al., 1997; Amir et al., 2005b). The removal rates of dimethyl phthalates in naturally aerated compost (NAC) has shown to be lower than during manually turned compost (MTC), 21% versus 77% reduction, respectively. The lower removal rates might be related to the formation of new dimethyl phthalate during composting through demethylation of both side-chains of diethyl phthalates (Cartwright et al., 2000).

Paulsen and co-workers (2010) measured an increase of DEHP on day 2 while concentrations of the remaining micropollutants decrease. It is suggested that the main source of DEHP is not the same as the rest of the micropollutants, and that the yard waste which was observed to contain plastic fragments (a material commonly containing DEHP) could be the DEHP source. However, extensive documentation exists that DEHP also degrades during the composting mesophilic and curing phases, and concentrations will be significantly lower in full length composting.

9.2.2 PFOS and PFOA

A study from the US measuring PFCs in nine commercial composts and one backyard compost, found perfluoroalkyl acids in the range of 28.7 to 75.9 µg/kg, and PFOA and PFOS were detected in all composts (Choi et al., 2019). Concentration of PFOS and PFOA in compost samples based on different types of organic waste (sewage sludge, separated biowaste/green waste) in the range of 0.035-145.66 µg/kg dw and 0.3-25.86 µg/kg dw, respectively, were reported in a European study (JRC 2013). Science-based fate studies of PFOA, PFOS and precursor for PFOS- and PFOA, during composting processes has not been identified.

9.2.3 SSCP

Concentration of NPE+NP in compost after 26 and 124 days in pilot scale composting are reported at 2.0 and 2.6 mg/kg dw, respectively (Kapanen et al., 2013). This correspond to a calculated 69 and 61%

removal based on estimated initial concentration at start based in mixing ration with other substrates. Sewage sludge concentration before being mixed with co-substrate was 15.2 mg/kg dw.

9.2.4 The musk compounds HHCB, AHTN and OTNE

Analyses of HHCB, AHTN and OTNE in samples taken during the first 24 days from compost at a full scale composting facility in showed an initial concentration in of 1, 0.11, and 0.82 mg/kg dw, respectively (Poulsen et al., 2010), corresponding approximately to 89, 68 and 88 % removal, respectively. An incubation study conducted with compost samples from the same composting facility, resulted in an average of 74%, 45% and 95% removal, respectively (Sadeq et al., 2014). Degradation efficiency for the different musk compounds corresponded with temperatures during incubation, which were 52, 70 and 57°C, respectively.

Sadeq et al. (2014) reported that the metabolites from parent compounds might have different optimal degradation temperatures, for instance the metabolite HHCB-LA and it's parent compound HHCB. It was suggested that this is a result of different microorganisms being involved in the degradation processes. In the same study, OTNE was among the contaminants showing highest removal rates over a wide temperature range. This could indicate involvement of different strains of microorganisms, with an average removal rate of 88% over the entire temperature range and an optimal temperature of 57°C. The results for AHTN in the study by Sadeq et al (2014) were more difficult to interpret, but its removal rate seemed to be relatively low (45%), with an optimal temperature of 70°C and optimal degradation rate around 60%.

In the study by Poulsen et al. (2010), AHTN has a reported estimated DT_{50} at 40 days.

In a field scale composting study from Japan, reduction of HHCB and OTNE were both 63% and AHTN 25%, respectively (Ozaki et al., 2017).

In a European study, HHCB and AHTN are reported in compost samples based on different types of organic waste (sewage sludge, separated biowaste/green waste) in concentrations up to 6.8 mg/kg dw and 0.95 mg/kg dw, respectively (JRC 2013). The average concentrations were 2.28 and 0.40 mg/kg dw, respectively.

9.2.5 BDE-209

In a study presented by the JRC in 2013, BDE-209 was not reported as a separate value, but rather as total Deca formulated (Σ BDE-206, BDE-207, BDE-208 and BDE-209). The pooled samples from different types of composts (based on biowaste, green waste, sewage sludge) ranged from 4.1 to 488 μ g/kg dw (JRC 2013). In compost from Switzerland, BDE-209 was detected in the range of 4.2-22 μ g/kg dw and accounted for 72% of the total polyBDE content (Brändli et al., 2007). Higher PBDE concentration during anaerobic digestion of sewage sludge than under aerobic decomposition has been reported (Kim et al. 2013). Debromination from BDE-209 to BDE202 has been detected in sludge, however, the debromination is characterised as minimal. A bioremediation study with use of composting and different amendments (cabbage, sawdust, cabbage and sawdust, cabbage-activated sludge-sawdust) where tested with BDE-209 contaminated sediment (Zhao et al., 2016).

9.2.6 PCB7

From Finland, concentrations of PCB7 after 26 and 124 days in a pilot scale composting study at 0.7 and 0.2 mg/kg dw have been reported (Kapanen et al., 2013). The concentration after 124 days corresponds to a calculated 67% removal based on estimated initial concentration in the compost after mixed with co-substrates. The first analysis showed an increase compared to estimated initial compost

concentration, showing general variation in this type of inhomogeneous matrixes. The concentration in sewage sludge mixing was 1.4 mg/kg dw.

The concentration in sewage sludge from another Finish WWTP was 0.004 mg/kg dw. Concentrations of PCB7 in Norwegian sewage sludge ranged from <0.001-0.077 mg/kg dw, with an average concentration 0.017 mg/kg dw (Blytt and Stang, 2019). The PCB7 concentration in compost from a field scale composting treatment plant after 3 months and 1 year, and in compost after 1-2 year composting from a second treatment plant were 0.06, 0.06 and 0.02 mg/kg dw (Kapanen et al., 2013). PCB7 concentrations up to 46 µg/kg dw in compost samples in a European study are reported (JRC 2013).

9.2.7 NP and NPE

Concentrations of NPE+NP in compost after 26 and 124 days in pilot scale composting are reported at 2.0 and 2.6 mg/kg dw, respectively (Kapanen et al., 2013). This corresponds to a calculated 69 and 61% removal based on estimated initial concentration at start based in mixing ration with other substrates. The concentration in sewage sludge before being mixed with co-substrate was 15.2 mg/kg dw. In addition the challenge of obtaining representative samples in inhomogeneous matrixes, the variation in concentrations might reflect difficulties in interpreting the fate of NP in degradation processes where NPEs are present and cause the formation of NP.

The concentrations of NPE+NP in compost from a field scale facility after 3 months and 1 year (same pile) and 1-2 years composting from a second plant, were 26, 47 and <0.2 mg/kg dw, respectively (Kapanen et al., 2013). In sludge from a second WWTP, 8.9 mg/kg dw NP was measured.

In an incubation study with compost samples from a Danish composting treatment plant (Sadeh et al., 2014), NP showed a low average removal rate – only 36%. However, at the most efficient temperature range (estimated optimal temperature 58°C), a 65% reduction was measured.

Table 39. Summary of measured concentrations of HOCs in compost and percent removal during composting.

Substance	Substrate	Unit conc.	% reduction	Concentration	Study (Country)	Ref.
DEHP	Compost SS	mg/kg dw	84	4.96 (24 d)	Field scale (Denmark. appr. 40% SS)	Poulsen et al., 2010 ¹
DEHP	Compost SS	mg/kg dw	57.8	4.6 (56 d)	Pilot scale (China)	Cai et al., 2012
DEHP	Compost SS	mg/kg dw	20; 56	20 (26 d); 11 (124 d)	Pilot scale (Finland)	Kapanen et al., 2013
DEHP	Compost SS	mg/kg dw		2 (1-2 yr); 38 (3 mo)-16 (1 yr)	Field scale (Finland)	Kapanen et al 2013 ²
PFOS	Compost D	µg/kg dw		2.7 (mean)	Italia	Brambilla et al., 2016
PFOS	Compost D	µg/kg dw		0.87-7.94 (min-max)	Italia	Brambilla et al., 2016
HHCb	Compost SS	µg/kg dw	89	104 (24 d)	Field scale (Denmark. appr. 40% SS)	Poulsen et al., 2010 ¹
HHCb-LA	Compost SS	µg/kg dw	59	49 (24 d)	Field scale (Denmark. appr. 40% SS)	Poulsen et al., 2010 ¹
HHCb	Compost SS	µg/kg dw	63	2689 ± 1163 (1 yr)	Pilot/field scale (Japan)	Ozaki et al., 2017 ³
AHNT	Compost SS	µg/kg dw	68	35 (24 d)	Field scale (Denmark. appr. 40% SS)	Poulsen et al., 2010 ¹⁰
AHNT	Compost SS	µg/kg dw	25	1731± 617 (1 yr)	Pilot/field scale (Japan)	Ozaki et al., 2017 ³
OTNE	Compost SS	µg/kg dw	88	98	Field scale (Denmark. appr. 40% SS)	Poulsen et al., 2010 ¹
OTNE	Compost SS	µg/kg dw	63	451± 179 (1 yr)	Pilot/field scale (Japan)	Ozaki et al., 2017 ³
PBDE-209						
NPE	Compost SS	mg/kg dw	69-61	nd (26 d), 0.4 (124 d)	Finland Pilot scale	Kapanen et al 2013
NPE	Compost SS	mg/kg dw		nd; 3 - 1	Finland Field scale	Kapanen et al 2013 ²
NP	Compost SS	mg/kg dw	66-62	2 (26 d), 2.2 (124 d)	Finland Pilot scale	Kapanen et al 2013
NP	Compost SS	mg/kg dw		0.2; 23-46	Finland Field scale	Kapanen et al 2013 ²
PCB7	Compost SS	mg/kg dw	-17%-67%	0.7 (26 d), 0.2(124 d)	Finland Pilot scale	Kapanen et al 2013
PCB7	Compost SS	mg/kg dw		0.02; 0.006-0.06	Finland Field scale	Kapanen et al 2013 ²
6-NDL-PCBs	Compost D	µg/kg dw		6.6-15.9 (min-max)	Italia	Brambilla et al., 2016 ⁴
6-NDL-PCBs	Compost D	µg/kg dw		9.5 (mean)	Italia	Brambilla et al., 2016

¹Komtek, Denmark; ²Full scale plants, Finland; ³Compost from open windrow thermophilic digestion composting system, from a compost manufacturing company in Hiroshima Prefecture, Japan, ⁴Collected samples, mixed compost/digestate/sludge from WWTPs. SS: sewage sludge not given if sludge have been through anaerobic digestion, D: digestate.

Important considerations

Composting might be an efficient process for reduction of several HOCs, primarily due to biotransformation; but for some compounds, evaporation and humification might contribute. The composting process influences removal efficiency.

Thermophilic processes (temperature > 50 °C) are most efficient for degrading organic material. However, from the literature it is observed that different HOCs have different optimum temperatures for maximal removal.

Reduction rates for DEHP, HHCB (galaxolide), AHNT (tonalide), NP and NPE in the range of 60-90% have been reported in pilot or field scale composting. For strongly oxidized HOCs like PCBs and BDE-209, oxidation processes are not the most efficient processes for removal.

For several HOCs, their fates during composting are not yet studied.

10 Measures to limit the content of selected HOCs in organic waste-based end products

The selected HOCs are found at elevated levels in the final treated sewage sludge because;

- there are significant discharges of these HOCs and/or precursors to the wastewater,
- the HOCs sorb to the sludge,
- the loss by volatilisation are limited or not enough to bring the levels sufficiently low,
- the biotransformation occurring during the different wastewater and/or sludge treatment steps are not adequate or efficient enough to bring the levels sufficiently low.

From a strategic point of view, the first and preferred choice of abating HOCs in sewage sludge is to minimise potential sources, i.e. implementing control measures upstream of the WWTP such as regulations on production, import, use and disposal. Work is ongoing through several national and EU-regulations, and there is a high level of awareness of the environmental exposure through wastewater and sewage sludge. However, such measures are not a part of the scope of this work.

The second bullet point in the list above – sorption to sludge – is crucial in minimising direct discharges of HOCs to the aquatic environment with the treated effluent. It may be possible, in theory, to (partially) desorb the HOCs from the particles and resorb them to an adsorbent with excellent and specific adsorption characteristics and thereby isolate the HOCs before they enter the sludge treatment stage. However, this may have negative impacts on the efficiencies of the subsequent wastewater treatment steps, and it may also be far from feasible from an economic point of view. It will probably be a better option to desorb the HOCs within the sludge line and subsequent resorb the HOC to an adsorbent or apply other types of treatment (see below).

Extensive (bio)transformation to non-toxic and non-bioaccumulating compounds or, more ideally, complete mineralisation is in many ways the preferred mechanism to minimise the content of these HOCs if already present in the wastewater received by the WWTP, since this will minimise the hazardous waste production.

Enhanced evaporation and subsequent sorption/condensation to isolate and additional treatment to transform/destroy the more volatile HOCs (i.e. HHCB, AHTN, OTNE and NP) may also be possible.

10.1 Removal of selected HOCs through optimised conditions for biotransformation

As discussed in **Section 7.2.4**, a prerequisite for the biotransformation of a compound is that (Barret et al., 2012);

- i) the environmental conditions are generally suitable for the microorganisms that can biotransform the compound,
- ii) that the compound is accessible and available at an appropriate concentration to these microorganisms' enzymes, and
- iii) the compound has a structure that makes it available for biotransformation by the microorganisms' enzymes.

Prerequisite i) indicates that there will be very limited, if any, biotransformation occurring during mechanic and chemical treatment. Hence, in the following we briefly discuss potential measures that may be undertaken to optimise the conditions for these prerequisites during biological wastewater and sludge treatment.

10.1.1 Optimising the environmental conditions

Optimum redox condition: As discussed in Section 7.2.4, the redox (i.e. aerobic, anoxic and anaerobic) conditions are very important for the biotransformation of these HOCs. In general, the selected HOCs are more prone to biotransformation under aerobic conditions, but BDE-209 may be debrominated. Highly chlorinated PCBs may also be dechlorinated to less chlorinated PCBs under anaerobic conditions, however, this is less likely. The factors affecting the specific biotransformation of the selected HOCs are summarised in Table 40. Both PFOS, PFOA and NP appears to be very persistent during currently applied treatment processes at WWTPs. Due to the biotransformation of precursors that are present in the wastewater, the concentrations of PFOS and PFOA increase under aerobic conditions, while the concentration of 4-NP increase under anaerobic conditions. For biotransformation and complete mineralisation combinations of successive anaerobic and aerobic treatment steps may be beneficial for brominated and chlorinated HOCs.

Thermophilic vs mesophilic digestion: Temperature impacts all biological processes, but only during sludge treatment is it feasible to control the operating temperature due to the much lower volumes of sludge compared to wastewater. This suggests that thermophilic conditions (typically 53-58°C, or up to 70°C) would result in more rapid biotransformation than mesophilic conditions (typically 35-40°C). However, there is no clear evidence that this is the case for the selected HOCs, at least for anaerobic digestion.

Table 40. Factors affecting the biotransformation rate of the selected HOCs. Composting is not included.

Substance	Factors affecting the biotransformation
DEHP	<ul style="list-style-type: none"> Under aerobic conditions: first transformed to MEHP and 2-ethylhexanol, then stepwise conversions to complete mineralisation Under anaerobic conditions: transformation stops at MEHP (more toxic than DEHP) Biotransformation in WWTPs probably limited by strong sorption of DEHP to organic matter.
PFOS, PFOA	<ul style="list-style-type: none"> Both PFOS and PFOA highly recalcitrant in WWTPs There are many precursors to both PFOS and PFOA in the wastewater, often resulting in higher concentrations of both substances in the effluent than in the influent. The biotransformation of the precursors appears to be more significant under aerobic conditions than under anaerobic conditions.
SCCP	<ul style="list-style-type: none"> Goes through slow biotransformation and complete mineralisation under aerobic conditions. No biotransformation under anaerobic conditions.
HHCB	<ul style="list-style-type: none"> Oxidises to HHCB-lactone Biotransformations have been reported under both aerobic, anoxic (presence of NO₃) and anaerobic conditions.

Substance	Factors affecting the biotransformation
AHTN	<ul style="list-style-type: none"> Reported to be biotransform under both aerobic and anaerobic conditions.
OTNE	<ul style="list-style-type: none"> Presumed persistent, but good removal has been shown during composting
BDE-209	<ul style="list-style-type: none"> Anaerobic debromination to less brominated congeners occur under anaerobic conditions
PCB7	<ul style="list-style-type: none"> Highly chlorinated PCBs may be transformed to lightly chlorinated congeners under anaerobic conditions, and these may be further transformed under aerobic conditions. Biotransformations very slow and limited in WWTPs.
NP+NPE	<ul style="list-style-type: none"> Long chain NPnEOs (n>4) are biotransformed to short chain NPE (NP1EO and NP2EO); transformation rate increases with increasing chain length. Under <u>aerobic conditions</u>: NP1EO and NP2EO are further oxidised to the corresponding carboxylic acid (NP1EC and NP2EC). Under <u>anaerobic conditions</u>: NP1EO, NP2EO and NP1EC transformed to 4-NP.

10.1.2 Optimising the accessibility of the compounds

Use of MBR in wastewater treatment: As demonstrated by Barret et al., (2010b) and Delgadillo-Mirquez et al., (2011), it appears that it is primarily the HOCs associated with the dissolved and colloidal mater (DCM) that is accessible to the enzymes and that may be biotransformed. The need for efficient separation of the activated sludge from the treated water during CAS treatment, counteracts measures that could increase the DCM fraction. However, the high fraction of small-sized flocs and colloidal matter in membrane bioreactors (MBR) has been shown to increase the viable fraction of the biomass in MBR due to improved mass-transfer conditions and accessibility to the substrates (Cicek et al., 1999; Clara et al., 2005; Pollice and Laera, 2005), which suggests the possibility of improved biotransformation also of HOCs in MBRs, as has been documented for HHCB and AHTN under anaerobic conditions (Wang et al., 2014).

Reject water treatment: The dewatering process may return a significant amount of some of the selected HOCs together with the reject water back to the inlet of the WWTP (Yoshida et al., 2015). It is possible to treat this relatively small reject water stream separately to reduce the level of the selected HOCs before returning it the inlet stream. The concentrations here might be quite substantial; for instance, Yoshida et al., (2015) showed that approximately 50% of the DEHP that left the anaerobic digester was found in the reject water.

Alkaline stabilisation: The drastic change in pH that is caused by alkaline stabilisation is expected to alter the sorption behaviour of ionisable compounds such as PFOA, PFOS, NP and NPE, however, there is *so far no evidence that this is happening* (Semblante et al., 2015).

Anionic surfactants: Use of anionic surfactants such as sodium dodecylbenzene sulfonate (SDBS) to extract PFOS directly from the sludge (Pan et al., 2009); has not, as far as we know, yet been tested on sewage sludge. Another non-ionic surfactant and emulsifier that has been extensively used in lab scale test systems to keep strongly hydrophobic HOCs in solution, is Tween 80.

Super-critical fluid extraction: Extraction of PFOS and PFOA directly from the sludge using super-critical CO₂, methanol and HNO₃ (i.e. super-critical fluid extraction) (Chen et al., 2012); has

not, as far as we know, yet been tested on sewage sludge

10.1.3 Optimising the bioavailability of the compounds

Co-metabolism: Since the HOCs typically are present at very low concentrations and their biotransformation are usually not thermodynamically favored, co-metabolism with other substrates is suggested to be the most important biotransformation mechanism (Barret et al., 2012). Such co-metabolism during sludge digestion may be facilitated by supplying a readily biodegradable substrate such as acetate, yeast extract or cellulose (Chang et al., 2004; Dionisi et al., 2006). Co-metabolic pathways may lead to the formation of metabolites that may eventually participate in metabolic reactions resulting in the complete mineralisation of the compounds (Tran et al., 2013).

Decreased food-to-microorganism ratio: The higher biomass concentration due to the usually long SRT in MBRs also decreases the food-to-microorganism ratio, which makes it more likely that the microorganisms will target less energy-rich substrates such as HOCs (Weiss and Reemtsma, 2008). This is likely also the case at the end of sludge digestion processes, though at this stage, the fraction of DCM is minimised (as discussed above).

Thermal hydrolysis: See Section 8.2.2.

Advanced oxidation processes (AOP): AOPs such as ozonation (O_3), UV/ H_2O_2 , Fenton/ H_2O_2 and O_3/H_2O_2 may be used to partially oxidise the selected HOCs to substances that are more easily available for biotransformation in a subsequent treatment step. This O_3 /AOP step may follow a preceding desorption process in the sludge treatment line (see above). A challenge with AOPs is that they are relatively non-discriminative in their action, attacking a broad spectre of compounds and produce also a broad spectre of oxidation products. Hence, AOPs have typically been applied to “clean” water systems to minimise the the necessary oxidant dose (i.e. energy consumption) and limit the potential formation of by-products. However, ozonation has been found to be economically feasible for the removal of residual pharmaceuticals and personal care products in secondary effluents and is now implemented in full scale at some WWTPs in Switzerland and Germany (Rizzo et al, 2018). For ozonation to be feasible the level of organic carbon (and nitrite) should be as low as possible in the water to be treated, and the HOCs should be associated with the DCM fraction. High removals of HHCB, AHTN and NP has been found treating grey water with ozonation (Hernández-Leal et al., 2011). Qiang et al., (2013) used ozonation (0.1 g O_3 /g TS) as pre-treatment to an anaerobic digester and obtained significant desorption and removal of hydrophilic compounds, but the effects on the more hydrophobic compounds such as NP was limited due to their strong sorption to the sludge. Pham et al., (2011) observed a somewhat increased removal of DEHP during aerobic digestion (from 72% to 85%) when pre-treated by Fenton/ H_2O_2 .

10.1.4 Optimising the biotransformation ability of the microbial community

Enzyme activity: As discussed in **Section 7.2.4**, the ammonium monooxygenase (AMO) enzyme of nitrifiers has been found to be involved in the biotransformation of many HOCs, particularly some recalcitrant pharmaceuticals (Batt et al., 2006). Hence, conditions that promote the proliferation of these slow-growing autotrophic bacteria may improve the biotransformation of recalcitrant compounds in general; low concentrations of readily available organic compounds, surplus of both ammonium and dissolved oxygen and a suitable pH range (ca. 7-7.5). Another intracellular enzyme system that has been reported to play important roles in the removal of some HOCs, particularly pharmaceuticals, is cytochrome P450 (Grandclément et al., 2017).

White rot fungi: A biological alternative to activated sludge is the use of white rot fungi (WRF) cultures. These have been reported to biotransform a wide range of persistent contaminants due to the action of fungal oxidative enzymes such as manganese peroxidase (MnP), lignin peroxidase (LiP), versatile peroxidase (VP) or laccase (Grandclément et al., 2017). LiP has been shown to eliminate e.g. phenolic compounds (Christian et al., 2005), DEHP was completely biotransformed by the filamentous fungi *Fusarium oxysporum* and *F. solani* within 10 days incubation (Bouchiat et al., 2016)

and *Trametes versicolor* has been shown to biotransform NP (Cajthaml et al., 2009) and PCBs (Ruiz-Aguilar, 2002). In the latter study, the non-ionic surfactant Tween 80 was used to keep high concentrations (600-3000 mg/L) of PCBs emulsified, and *T. versicolor* biotransformed both low and high-chlorinated congeners of PCBs ranging from 29% to 70% (Ruiz-Aguilar, 2002). Cruz-Morató et al., (2013) found that *T. versicolor* was able to remain active (and biotransforming selected pharmaceuticals at wastewater-relevant concentrations) in non-sterile urban wastewater when kept in a batch fluidized bed bioreactor when fed with real wastewater if nutrient sources such as glucose (as carbon source) and ammonium tartrate (as nitrogen source) were added and the pH was kept at 4.5 to maintain a significant biological fungus activity. The above-mentioned enzymes are extracellular enzymes and may also be produced externally. This has been done with laccase, adding it as a solution to the wastewater (Grandclément et al., 2017). Though, it is also possible to immobilise such enzymes and thereby retaining them in the reactor system as a biofilm system and minimise enzyme consumption (Datta et al., 2013).

However, so far there is very limited documentation of the actual effects these factors have on the biotransformation rates of the selected HOCs, and the feasibility of the measures mentioned above.

10.2 Measures that harvest the inherent resources while isolating or destroying the selected HOCs

Some of the inherent resources in Norwegian sewage sludge are already today harvested as energy (through biogas production in anaerobic digestion processes), soil conditioner (by applying stabilised and hygienised sewage sludge on e.g. farmland or in soil mixes) and nutrients such as P and N (also by applying stabilised and hygienised sewage sludge on farmland and in soil mixes). The efficiency of the latter is subject to debate, as much of chemically precipitated P appears to have limited availability for plant uptake (Krogstad et al., 2005) and may therefore be leaching to the aquatic recipients instead.

A strict regulation of the content of the selected HOCs in the finally applied sewage sludge may make it necessary and more attractive to find other ways of harvesting these resources (and potentially also others). The last decade or so, a wide range of different processes and technologies have been developed and tested to optimise this type of harvesting. It is not the purpose of this report to cover the whole spectre, but we will at least shortly discuss the expected outcome in regard to the potential destruction and isolation of the selected HOCs when applying some of the main types of resource harvesting methods, using the outcome of the STRUBIAS project organised by the European Sustainable Phosphorous Platform (ESPP) as outlined in the [pre-final report](#) with Appendix and Annexes (Huygens et al., 2018):

10.2.1 Precipitation of phosphate salts and derivatives

Phosphate salts can be recovered from different position along the wastewater treatment line and/or sewage sludge treatment line (see green and blue spheres in Figure 22), though, most commonly from sludge liquor (reject water after dewatering) and from digested sludge. PO_4^{3-} precipitates together with Mg^{2+} or Ca^{2+} , possibly also trapping NH_4^+ and/or K^+ in the molecular structure. Struvite, the most commonly recovered phosphate salt, forms from equimolar quantities of Mg^{2+} , PO_4^{3-} and NH_4^+ , implying that the efficiency of NH_4^+ removal is relatively low, and the excess N remains in soluble form. In most sewage treatment applications Mg is the limiting element and is therefore added to the process as MgCl_2 or MgO . Several processes have been developed and are already operating at full scale in other countries, while others are being developed and tested in smaller scale (Egle et al., 2016). The treatment types may be grouped in the following sub-categories with associated products; nutrient salt precipitation/crystallisation (struvite, calcium phosphate), wet chemical leaching (struvite, calcium phosphate), wet oxidation and nanofiltration (phosphoric acid) and supercritical water oxidation (phosphoric acid) (Egle et al., 2016, Blytt et al., 2017).

HOCs may co-precipitate, typically adsorbed to organic matter. Hence, the organic carbon can be seen as a vector for these HOCs but thereby also a good proxy for the purity of the precipitated phosphate salt (Huygens et al., 2018). There is a positive correlation between organic C content in the precipitated phosphate salts and the accumulation of impurities such as antibiotics (Ye et al., 2018), while other impurities have mainly been found in precipitated phosphate salts with an organic C content of >3% (Huygens et al., 2018; STOWA, 2015). The STRUBIAS report proposes a limit value of 3% organic C in the precipitated phosphate salts, which indicate a reduction of one order of magnitude in organic C relative to unprocessed manure and sewage sludge (Huygens et al., 2018). They also argue that this also “effectively excludes the extensive and expensive testing for a broad range of organic contaminants (e.g. pharmaceutical compounds and personal care products, phthalates, surfactants, etc.), and strengthens market confidence in fertilising materials recovered from biogenic wastes in times of increased concerns about emerging organic contaminants in consumer products and the food chain”.

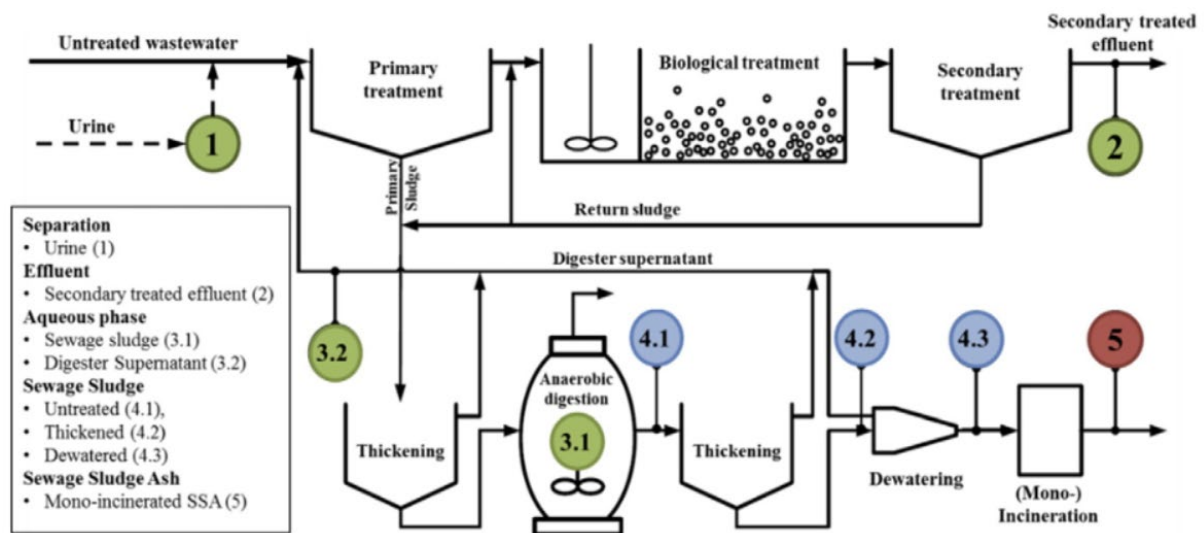


Figure 22. Various access points for the extracting of phosphorous from the wastewater and the sewage sludge lines. Source: Egle et al., (2016).

10.2.2 Thermal oxidation followed by post-extraction of P

These are treatments that combust the organic matter with excess oxygen at high temperatures (800-950°C) for a few seconds. This is done in a boiler creating flue-gases containing most of the available fuel energy as heat. The phosphorous is extracted from the ashes by either of two types of processes (Huygens et al., 2018):

- Wet-chemical processes* in which chemicals (e.g. H_2SO_4 or HCl) are added to acidity the solution to $pH < 2$ to bring the phosphorous to a bioavailable form and separating metals (e.g. Al, Fe and PTEs) by e.g. sequential precipitation, liquid-liquid extraction or ion exchange.
- Thermal processes* in which phosphorous is either transferred into a metallurgic slag with low C content (typically <3%) by reductive smelting at very high temperatures (ca. 1250-2000°C) in a shaft furnace (Scheidig, 2009) or is reduced to elemental P that is separated via the gas phase in an inductively heated shaft furnace (Schönberg et al., 2014). Volatile PTEs such as Zn, Pb, Cd and Hg are separated from the product via the gas phase and further collected in the flue dust, and PTEs with high boiling points such as Fe, Cu, Ni and Cr are separated in the form of a liquid alloy.

Combustion under non-oxygen limiting conditions to low levels of organic C (<3%) in the ashes is a well-demonstrated technique for the effective removal and thermal destruction of a broad range of HOCs (EC, 2017, Huygens et al., 2018). The temperatures typically applied in incineration processes are generally enough for the destruction of PFOS and PFOA, probably also their precursors (Yamada et al., 2005). PCB levels are typically low in the ash fraction (Huygens et al., 2018).

10.2.3 Pyrolysis and gasification

These processes involve heat treatment under oxygen-limited conditions producing C-rich (>50%) biochar or mineral-rich pyrogenic carbonaceous materials depending on the C-content of the feedstock. The biochar may be applied for soil amendment and as a fertiliser if nutrient rich. The less oxygen present in the reactor, the more solid pyrolysis material is produced. The processes can be further categorised by the temperature applied during treatment (Huygens et al., 2018):

- i. In *hydrothermal carbonification* (or *wet pyrolysis*) an aqueous biomass solution is treated at moderate temperatures (ca. 180-300°C) and 10-30 bar pressure for several hours (2-10 h).
- ii. In *pyrolysis* the organic material is thermochemical decomposed at moderate to high temperatures (ca. 300-700°C) producing solid biochar, condensable liquid biooil and non-condensable syngas. The syngas is captured for combustion.
- iii. In *gasification* the organic feedstock is treated at high temperatures (>700°C) while introducing a controlled and limited level of oxygen into the system converting the organics to CO, H₂, CO₂ and a limited amount of solid material.

The knowledge base regarding the proportional removal of specific organic pollutants is limited and primarily restricted to a few organic pollutants (i.e. PAHs, PCDD/F and PCBs; see below) (Huygens et al., 2018). Though, Ross et al., (2016) found substantial removal (>90%) of nonylphenol during lab-scale pyrolysis at 300 °C, but 600 °C was required to remove nonylphenol to below the quantification limit in the biochar. Possibly due to the relatively high vapour pressure of NP indicating limited exposure to high temperatures, almost all NP was retrieved in the syngas & biooil effluent. The same fate should be expected for the volatile musk fragrances (HHCB, AHTN, OTNE). Though, Bridle et al., (1990) reported >75% destruction of PCBs at 450°C, presumably due to dehalogenation, PAHs, dioxins and furans (PCDD/F) and PCBs may be formed during the thermochemical processes; PAHs are typically generated at temperatures <600°C, while the generation of PCDD/F and PCBs may occur throughout the whole operating temperature range (300-750°C) if chlorine is present and their adsorption can be favoured by the presence of elemental carbon and soot particles (Bucheli et al., 2015). Hence, the STRUBIAS report authors (Huygens et al., 2018) propose to apply the existing strict quality standards of the European Biochar Certificate (EBC, 2012) to PAH, PCDD/F and PCBs in biochar: <6 mg PAH (16 US EPA congeners)/kg dw, <20 ng WHO toxicity eq. PCDD/F/kg dw and <0.2 mg PCB (PCB 28, 52, 101, 138, 153 and 180)/kg dw. Based on the precautionary principle, Huygens et al., (2018) concludes that **it is justified to exclude highly contaminated feedstocks (e.g. sewage sludge, municipal solid waste, hazardous waste) from the positive input material list to ensure human health and environmental safety**. The positive material list of feedstocks to wet and dry pyrolysis processes includes plant-based materials, bio-waste and certain animal by-products (e.g. bone material and manure).

Important considerations

Extensive (bio)transformation to non-toxic and non-bioaccumulating compounds or, more ideally, complete mineralisation is in many ways the preferred mechanism to minimise the content of these HOCs if already present in the wastewater received by the WWTP, since this will minimise the hazardous waste production.

For biotransformation and complete mineralisation combinations of successive anaerobic and aerobic treatment steps may be beneficial for brominated and chlorinated HOCs. This is valid both for wastewater and sludge treatment processes.

There are indications that it is possible to optimise the biotransformation of the selected HOCs during biological wastewater treatment.

- MBRs may improve the biotransformation of HOCs, but this should be verified for the selected HOCs.
- The potential for nitrifiers and white rot fungi in the biotransformation of the selected HOCs should be tested.

There is generally a lack of data to verify the actual effects on the biotransformation of the selected HOCs during all types of digestions, both during aerobic and anaerobic conditions, and under mesophilic and thermophilic conditions.

- The facilitation of co-metabolism of the HOCs by supplying more readily available substrates could be tested in addition.
- The effects of applying advanced oxidation processes (AOPs) as pre-treatment to the digestion may also be assessed.

It is an option to desorb the HOCs within the sludge line and subsequent resorb the HOC to an adsorbent or apply other types of treatment.

- Treatment of the reject water (liquid from the dewatering of the digested sludge) may be feasible due to potentially much higher concentrations of HOCs, higher temperature and lower flows.
- The effect of alkaline stabilisation and the use of anionic surfactants on the sorption of the selected HOCs in the sludge should be assessed.
- Super-critical fluid extraction of the selected HOCs directly from the sludge should also be assessed.

Enhanced evaporation and subsequent sorption/condensation to isolate and additional treatment to transform/destroy the more volatile HOCs (i.e. HHCB, AHTN, OTNE and NP) may also be possible.

It is of high interest to harvest and reuse as much of the valuable contents (e.g. energy, organic matter and nutrients) of sewage sludge as possible. Some of these may be harvested by other routes than are currently applied:

- If phosphorous (and nitrogen) is precipitated as salts, some of the HOCs may be co-precipitated together with the organic matter. The organic carbon level in the precipitate may possibly be used as a proxy for the purity of the precipitated phosphate salt (3% organic C is suggested as a limit). This should, however, be verified.
- The temperatures typically applied in incineration processes (thermal oxidation under aerobic conditions) are generally sufficient for the destruction of PFOS and PFOA, probably also their precursors, as well as PCB. This should, however, be verified for all the selected HOCs.

Thermal processes under oxygen-limited conditions (e.g. pyrolysis) produce C-rich (>50%) biochar. The knowledge base regarding the proportional removal of specific organic pollutants is limited and primarily restricted to a few organic pollutants. Importantly, generation of PCDD/F and PCBs may occur throughout the whole operating temperature range (300-750°C) if chlorine is present and their adsorption can be favoured by the presence of elemental carbon and soot particles. Based on the precautionary principle, the STRUBIAS project, concludes that it is justified to exclude highly contaminated feedstocks (e.g. sewage sludge, municipal solid waste and hazardous waste) from the positive input material list to ensure human health and environmental safety. The positive material list of feedstocks to wet and dry pyrolysis processes includes plant-based materials, bio-waste and certain animal by-products (e.g. bone material and manure).

References

- Aasen, R., Bøen, A., Bergersen, O. (2006) Effect of increased porosity on short organic acids, bacterial counts and respiration rate during composting of acidic organic waste. *Orbit* 2006. Part 1 119-124.
- Aasen, R., Eggen, T., Sørheim, R., Hartnik, T., Næs, K., Amundsen, C.E. 2009. Screening of polyfluorinated organic compounds at four fire training facilities in Norway. Miljødirektorat rapport TA-2444.
- Abramowicz, D.A. (1990) Aerobic and Anaerobic Biodegradation of PCBs: A Review, *Critical Reviews in Biotechnology*, 10:3, 241-251, DOI: 10.3109/07388559009038210
- Altenburger, R., Nendza, M., Schüürmann G. (2003) Mixture toxicity and its modeling by quantitative structure-activity relationships. *Environ. Toxic. Chem.* 22:1900-1915.
- Amir, S., Hafidi, M., Merlina, G., Hamdi, H., Jouraiphy, A., Gharous, M.E., Revel, J.C. (2005) Fate of phthalic acid esters during composting of both lagooning and activated sludges. *Process. Biochem.* 40: 2183–2204.
- An, X., Xiao, B., Di, X., Dong, H., Tang, H. (2017) Research progress on aging of organic pollutants in geosorbents: A review. *Acta Geochim*, 36:27-43.
- Andersen, S., Gudbransen, M., Haugstad, Hartnik, T. (2012) Noen miljøfarlige stoffer i avløpsslam – forekomst og miljørisiko, TA 3005/2012.
- Andrade, N.A., McConnell, L.L., Anderson, M.O., Torrents, A., Ramirez, M. (2017) Polybrominated diphenyl ethers: Residence time in soils receiving biosolids application. *Env. Poll.* 222:412-422.
- Arp, H.P., Sørmo, E., Breedveld, G. (2017) Uppdating of background data and proposals for new quality standards (normverdier) for contaminated soil. NGI report 20160648-03-R. Rev.no. 0/2017-01-02.
- Arvaniti, O.S., Ventouri, E.I., Stasinakis, A.S., Thomaidis, N.S. (2012) Occurrence of different classes of perfluorinated compounds in Greek wastewater treatment plants and determination of their solid–water distribution coefficients. *J. Hazardous Materials*, 239-240:24-31.
- Arvin E., Henze, M. (2002) Treatment plants for phosphorous removal from wastewater. In: M. Henze, P. Harremöes, J.C. Jansen., E. Arvin (Eds) *Wastewater Treatment – Biological and Chemical Processes*, 3rd ed. Springer-Verlag, Berlin Heidelberg, ISBN 3-540-42228-5, p. 327-367.
- Australian Government (2019) Indirect precursors to perfluorooctanesulfonate (PFOS): Environment tier II assessment. Australian Government, Department of Health, March 8 2019.
- Avendaño, S.M., Zhong, G., Liu, J. (2015) Comment on “biodegradation of perfluorooctanesulfonate (PFOS) as an emerging contaminant”. *Chemosphere* 138:1037–1038.
- Backhaus, T., Faust, M. (2012) Predictive Environmental Risk Assessment of Chemical Mixtures: A Conceptual Framework. *Environ. Sci. Technol.* 46(5):2564–2573.
- Backhaus, T., Karlsson, M. (2014) Screening level mixture risk assessment of pharmaceuticals in STP effluents. *Water Res.* 49:157-65.
- Barret M., Patureau D., Latrille E., Carrère H. (2010a) A three-compartment model for micropollutants sorption in sludge: Methodological approach and insights. *Wat. Res.* 44:616-624.
- Barret M., Carrère H., Latrille E., Wisniewski C., Patureau D. (2010b) Micropollutant and Sludge Characterization for Modeling Sorption Equilibria. *Environ. Sci. Technol.* 2010, 44:1100–1106.
- Batt, A.L., Kim, S., Aga, D.S. (2006) Enhanced Biodegradation of Iopromide and Trimethoprim in Nitrifying Activated Sludge. *Environ. Sci. Technol.* 40:7367-7373.

- Barret, M., Delgadillo-Mirquez, L., Trably, E., Delgenes, N., Braun, F., Cea-Barcia, G., Steyer, J.P., Patureau, D. (2012) Anaerobic removal of trace organic contaminants in sewage sludge: 15 years of experience, *Pedosphere*. 22:508–517.
- Bechmann, M., Deelstra, J., Eggestad H.O., Kløve, B., Stålnacke, P., Vandsemb S., Øygarden, L. (2000) Erosjon og næringsstofftap fra jordbruksarealer. Resultater fra program for Jordsmonnovervåking 1999/00. Jordforsk rapport 106/00.
- Beck-Friis, B., Smårs, S., Jönsson, H., Kirchmann, H. (2001) Gaseous emissions of carbon dioxide, ammonia and nitrous oxide from organic household waste in a compost reactor under different temperature regimes. *J. Agricultural Engineering Research*. 78(4):423-430.
- Beck-Friis, B., Smårs, S., Jönsson, H., Eklind, Y., Kirchmann, H. (2003) Composting of source-separated household organics at different oxygen levels: gaining an understanding of the emission dynamics. *Compost Science & Utilization*. 11(1):41-50.
- Beigel, L., Flaws, J., Hirshfeld, A., O'Conner, J., Elliott, G., Ladies, G., Silbergeld, E., Van Pelt, C., Hurtt, M., Cook, J., and Frame, S. (1998) 90-day feeding and one-generation reproductive study in Crl:CD BR rats with 17-beta-Estradiol. *Toxicological Sciences*, 44: 116-142.
- Bergersen, O., Bøen, A., Sørheim, R. (2009) Strategies to reduce short-chain organic acids and synchronously establish high-rate composting in acidic household waste. *Bioresource Technology*. 100:521-526.
- Berset, J.D., Kupper, T., Ettter, R., Tarradellas, J. (2004) Considerations about the enantioselective transformation of polycyclic musks in wastewater, treated wastewater and sewage sludge and analysis of their fate in a sequence batch reactor plant. *Chemosphere* 57:987–996.
- Bester, K. (2004) Retention characteristics and balance assessment for two polycyclic musk fragrances (HHCb and AHTN) in a typical German sewage treatment plant. *Chemosphere*. 57:863-870.
- Blytt, L.D., Henninge, L.B., Stang, P., Neidel, T.L. (2018) Report 1 - Limit values for organic pollutants in fertilisers based on organic waste origin. Report for Miljødirektoratet.
- Blytt, L.D., Henninge, L.B., Stang, P., Neidel, T.L. (2018b). REPORT 2 – Consequences of introducing requirements for content of organic pollutants in organic fertilisers based on waste covered by the Norwegian Fertiliser Regulations. Report for Miljødirektoratet.
- Blytt, L.D., Bruskeland, A.B., Stang, P. (2013) Organiske miljøgifter i norsk avløpsslam - Resultater fra undersøkelsen i 2012/13. *Norsk Vann – rapport nr. 198- 2013*.
- Blytt, L.D. (2010) Undersøkelse av miljøgifter ved fire norske renseanlegg PFOA, Bisfenol A, Triklosan, Siloksan (D5), Dodecylfenol og 2,4,6-Tri-tert butylfenol. *Aquateam AS rapport. TA-2636/2010 Rapport nr 10-015*.
- Blytt L.D., Stang, P., (2019) Organiske miljøgifter i norsk avløpsslam – Resultater fra undersøkelsen i 2017/18 (Organic pollutants in Norwegian wastewater sludge – Results from the survey in 2017/18). *Norwegian Water BA (Norsk Vann BA) Report 242/2018, 136 pages (in Norwegian)*
- Bossi, R., Strand, J., Sortkjær, O., Larsen, M.M., (2008) Perfluoroalkyl compounds in Danish wastewater treatment plants and aquatic environments. *Environ. International* 34:443-450.
- Bouchiat, R., Veignie, E., Grizard, D., Soebert, C., Vigier, M., Rafin, C., 2016. Ability of filamentous fungi to degrade four emergent water priority pollutants. *Desalination Water Treat.* 57:6740-6746.
- Brambilla, G., Abate, V., Battacone, G., De Filippis, S.P., Esposito, M., Esposito, V., Miniero, R. (2016) Potential impact on food safety and food security from persistent organic pollutants in top soil improvers on Mediterranean pasture. *Sci. Tot. Environ.* 543:581–590.

- Brändli R. (2002) The fate of Polycyclic musks in a waste water treatment plant. Part 1 Water. Diplom Thesis ETH Zürich.
- Briggs, G.G., Bromilow, R.H., Evans, A.A. (1982) Relationship between lipophilicity and root uptake and translocation of non-ionized chemicals by barley. *Pest. Sci.* 13:495–504.
- Brock T.D., Madigan M.T. (1991) *Biology of microorganisms*, 6th ed. Prentice-Hall International Eds. ISBN 0-13-086604-0.
- Brown, S., Devin-Clarke, D., Doubrava, M., & O'Connor, G. (2009) Fate of 4-nonylphenol in a biosolids amended soil. *Chemosphere*, 75(4):549-554.
- Brändli, R.C., Kupper, T., Bucheli, T., Zennegg, M., Huber, S., Ortelli, D., Müller, J., Schaffner, C., Iozza, S., Schmid, P., Berger, U., Edder, P., Oehme, M., Stadelmann, F.X., Tarradellas, J. 2007. Organic pollutants in compost and digestate. Part 2. Polychlorinated dibenzo-p-dioxins, and -furans, dioxin-like polychlorinated biphenyls, brominated flame retardants, perfluorinated alkyl substances, pesticides, and other compounds. *J. Environ. Monit.* 9: 465-472.
- Butt., C. M., Muir, D.C., Mabury, S.A. (2014) Biotransformation pathways of fluorotelomer-based polyfluoroalkyl substances: A review. *Environ.l Toxicol. Chem.* 33(2): 243-267.
- Bøen, A., Aasen, R., Bergersen, O. (2006). Strategies for quick establishment of high-rate composting in acidic household waste. *Orbit 2006*. Part 1: 31-35.
- Cai, Q.-Y., Mo, C.-H., Lü, H., Zeng, Q.-Y., Wu, Q.-T., Li, Y.-W. (2012) Effect of composting on the removal of semivolatile organic chemicals (SVOCs) from sewage sludge. *Bioresource Technology* 126:453–457.
- Cajthaml, T., K resinov a, Z., Svobodov a, K., M€oder, M., 2009. Biodegradation of endocrine-disrupting compounds and suppression of estrogenic activity by ligninolytic fungi. *Chemosphere* 75:745-750.
- Carballa, M., Omil, F., Alder, A.C., Lema, J.M. (2006) Comparison between the conventional anaerobic digestion of sewage sludge and its combination with a chemical or thermal pre-treatment concerning the removal of pharmaceuticals and personal care products. *Wat. Sci. Technol.* 53:109-117.
- Carballa, M., Manterola, G., Larrea, L., Ternes, T., Omil, F., Lema, J.M. (2007a) Influence of ozone pre-treatment on sludge anaerobic digestion: Removal of pharmaceutical and personal care products. *Chemosphere*. 67:1444–1452.
- Carballa, M., Omil, F., Ternes, T., Lema, J.M., (2007b) Fate of pharmaceutical and personal care products (PPCPs) during anaerobic digestion of sewage sludge. *Water Res.* 41:2139–2150.
- Carballa, M., Omil, F., Lema, J.M., Llompert, M., Garcí a-Jares, C., Rodri ´ guez, I., Go ´ mez, M., Ternes, T. (2004) Behavior of pharmaceuticals, cosmetics and hormones in a sewage treatment plant. *Water Res.* 38:2918–2926.
- Cartwright, C.D., Owen, S.A., Thompson, I.A., Burns, R.G. (2000). Biodegradation of diethyl phthalate in soil by a novel pathway. *FEMS Microbiol. Lett.* 186:27–34.
- CEC (Council of the European Communities), 1986. Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC). *Official Journal of the European Communities* No. L 181/6-12.
- Chen, H.Y., Liao, W., Wu, B.Z., Nian, H., Chiu, K., Yak, H.K. (2012) Removing perfluorooctane sulfonate and perfluorooctanoic acid from solid matrices, paper, fabrics, and sand by mineral acid suppression and supercritical carbon dioxide extraction. *Chemosphere*. 89:179-184.

- Chen, F., Ying, G.-G., Chen, Z.-F. (2014b) Field dissipation of four personal care products in biosolids-amended soils in North China. *Environ. Toxicol. Chem.* **33**, pp 2413-2421.
- Chen, X., Pauly, U., Rehfus, S., Bester, K. (2009). Personal care compounds in a reed bed sludge treatment system. *Chemosphere.* **76**(8):1094-1101.
- Choi, M.H., Park, Y. H. (1998) The influence of yeast on thermophilic composting of food waste. *Letters in Applied Microbiology.* **26**, pp 175-178.
- Choi, Y.J., Lazcano, R.K., Yousefi, P., Trim, H., Lee, L.S. (2019) Perfluoroalkyl Acid Characterization in U.S. Municipal Organic Solid Waste Composts. *Environ. Sci. Technol. Lett.* **6**(6):372-377.
- Cicek, N., Franco, J.P., Suidan, M.T., Urbain, V., Manem, J. (1999) Characterisation and comparison of membrane bioreactor and a conventional activated sludge system in the treatment of wastewater containing high molecular weight compounds. *Wat. Envir. Res.* **71**(1): 64-70.
- Cirja M., Ivashechkin P., Schäffer A., Corvini P.F.X. (2008) Factors affecting the removal of organic micropollutants from wastewater in conventional treatment plants (CTP) and membrane bioreactors (MBR). *Rev Environ Sci Biotechnol*, 7:61–78.
- Clara, M., Kreuzinger, N., Strenn, B., Gans, O., Kroiss, H. (2005) The solids retention time – a suitable design parameter to evaluate the capacity of wastewater treatment plants to remove micropollutants. *Water Res* **39**(1):97–106.
- Clarke, B.O., Smith, S.R. (2011) Review of ‘emerging’ organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids. *Environ. Int.* **37**: 226–247.
- Cruz-Morató C., Ferrando-Climent L., Rodríguez-Mozaz S., Barceló D., Marco-Urrea E., Vicent T., Sarrà M. (2013) Degradation of pharmaceuticals in non-sterile urban wastewater by *Trametes versicolor* in a fluidized bed bioreactor. *Water Res.* **47**(14):5200-10. doi: 10.1016/j.watres.2013.06.007.
- Dasu, K., Lee, L. S., Turco, R.F., Nies, L. F. (2013) Aerobic biodegradation of 8: 2 fluorotelomer stearate monoester and 8: 2 fluorotelomer citrate triester in forest soil. *Chemosphere.* **91**(3): 399-405.
- Datta S., Christena L.R. and Rajaram Y.R.S. (2013) Enzyme immobilization: an overview on techniques and support materials. *3 Biotech* (2013) **3**(1):1–9 DOI 10.1007/s13205-012-0071-7.
- Delgadillo-Mirquez L., Lardon L., Steyer J.-P., Patureau D. (2011) A new dynamic model for bioavailability and cometabolism of micropollutants during anaerobic digestion. *Wat Res.* **45**:4511-4521.
- Díaz-Cruz, García-Galán, M.J., Guerra, P., Jelic A., Postigo C., Eljarrat E., Farré M., López de Alda M.J., Petrovic M., Barceló, D. (2009) Analysis of selected emerging contaminants in sewage sludge. *Trends in Anal. Chem.* **28**(11):1263-1275.
- DiFrancesco, A.M., Chiu, P.C., Standley, L.J. Allen, H.E., Salvito D.T. (2004) Dissipation of fragrance materials in sludge-amended soils. *Environ. Sci. Technol.* **38**:194-201.
- Dionisi, D., Bertin, L., Bornoroni, L., Capodicasa, S., Papini, M.P., Fava, F. (2006) Removal of organic xenobiotics in activated sludges under aerobic conditions and anaerobic digestion of the adsorbed species. *J. Chem. Technol. Biotechnol.*, **81**:1496-1505.
- Dubroca, J., Brault, A., Kollmann, A., Touton, I., Jolival, C, Kerhoas, L., Mougin, C. (2005) Biotransformation of Nonylphenol Surfactants in Soils Amended with Contaminated Sewage Sludges. In: Lichtfouse E., Schwarzbauer J., Robert D. (eds) *Environmental Chemistry*. Springer, Berlin, Heidelberg.

- EC (2008) European Commission EC No 2008/98. The Waste Framework Directive. EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives. 12/12/2008.
- EC (2008) European Commission EC No 889/2008/ of 5 September 2008 laying down detailed rules for the implementation of Council Regulation (EC) No 834/2007 on organic production and labelling of organic products with regard to organic production, labelling and control.
- EC (2009) European Commission EC No 1069/2009. REGULATION OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 21 October 2009 laying down health rules as regards animal by-products and derived products not intended for human consumption and repealing Regulation (EC) No 1774/2002 (Animal by-products Regulation).
- EC (2013) European Commission EC 2013/39. Water Framework Directive 2013/39/EU of the European Parliament and of the Council amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy.
- ECHA (2016). Guidance on information requirements and Chemical Safety Assessment Chapter R.16: Environmental exposure assessment. Version 3.0. February 2016. ECHA-16-G-03-EN. ISBN: 978-92-9247-775-2.
- ECHA (2013). Substance Name: Pentadecafluorooctanoic Acid (PFOA). EC Number: 206-397-9. As a substance of very high concern because of its CMR1 and PBT2 properties. Adopted on 14 June 2013.
- ECHA (2018) SUBSTANCE EVALUATION CONCLUSION as required by REACH Article 48 and EVALUATION REPORT for Nonylphenol, branched, ethoxylated. EC No 500-209-1, 81 pages.
- ECHA (2008). Guidance on information requirements and chemical safety assessment Chapter R.10: Characterisation of dose [concentration]-response for environment. Guidance for the implementation of REACH. May 2008.
- ECHA RAC (2015) Opinion on an Annex XV dossier proposing restrictions on Perfluorooctanoic acid (PFOA), its salts and PFOA-related substances, European Chemical Agency Committee for Risk Assessment, ECHA/RAC/RES-O-0000006229-70-02/F, adopted 8 September 2015.
- ECHA RAC SEAC (2014) Background document to the Opinion on the Annex XV dossier proposing restrictions on NONYLPHENOL and NONYLPHENOL ETHOXYLATES. Committee for Risk Assessment (RAC) ECHA/RAC/ RES-O-0000005317-74-01/F and Committee for Socio-economic Analysis (SEAC) ECHA/SEAC/ RES-O-0000005317-74-02/F, 09.09.2014, 441 pages.
- EEC (1986) European Economic Community EEC 1986 86/278/ Council Directive on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture. Official Journal of the European Communities. No. L 181/6-12, Brussels.
- Engen, T., Heimstad, E.S., Heimstad, Stuanes, A.O., Norli, H.R. (2013) Uptake and translocation of organophosphates and other emerging contaminants in food and forage crops. *Environmental Science Pollution Research* 20:4520-4531.
- Engen, T., Lillo, C. (2012) Antidiabetic II Drug Metformin in Plants: Uptake and Translocation to Edible Parts of Cereals, Oily Seeds, Beans, Tomato, Squash, Carrots, and Potatoes. *J. Agricult. Food Chemistry* 60:6929-35.
- Eljarrat, E., Marsh, G., Labandeira, A., Barcelo, D. (2008) Effect of sewage sludges contaminated with polybrominated diphenylethers on agricultural soils. *Chemosphere* 71: 1079–1086.
- Ellis, D. A., Martin, J. W., Muir, D. C., Mabury, S. A. (2003) The use of ¹⁹F NMR and mass spectrometry for the elucidation of novel fluorinated acids and atmospheric fluoroacid precursors evolved in the thermolysis of fluoropolymers. *Analyst*. 128(6):756-764.

- Ellis, D. A., Martin, J. W., De Silva, A. O., Mabury, S. A., Hurley, M. D., Sulbaek Andersen, M. P., Wallington, T. J. (2004) Degradation of fluorotelomer alcohols: a likely atmospheric source of perfluorinated carboxylic acids. *Environ. Sci. Technol.* 38(12):3316-3321.
- Epstein, E. (1997) *The Science of Composting*. Technomic Publishing Company, Inc., Pennsylvania, U.S.A., pp. 19-22.
- Eriksson, U., Haglund, P., Kärrman, A. (2017) Contribution of precursor compounds to the release of per- and polyfluoroalkyl substances (PFASs) from waste water treatment plants (WWTPs). *J. Environ. Sci.* 61:80-90.
- EU (2019) European Union EU 2019/1009. Regulation (EU) 2019/1009 of the European Parliament and of the Council of 5 June 2019 laying down rules on the making available on the market of EU fertilising products.
- EU RAR (2008a) European Union Risk Assessment Report 1,3,4,6,7,8-HEXAHYDRO-4,6,6,7,8,8-EXAMETHYLCYCLOPENTA- γ -2-BENZOPYRAN(1,3,4,6,7,8-HEXAHYDRO-4,6,6,7,8,8-HEXAMETHYLIN-DENO[5,6-C]PYRAN - HHCB), Final version, May 2008.
- EU RAR (2008b) European Union Risk Assessment Report 1-(5,6,7,8-TETRAHYDRO-3,5,5,6,8,8-HEXAMETHYL-2-NAPHTHYL)ETHAN-1-ONE (AHTN), Final version, May 2008.
- EU RAR (2008c) European Union Risk Assessment Report Alkanes, C10-13, chloro, European Commission Directorate-General Joint Research Centre, EUR 23384 EN, 588 pages.
- EU RAR (2008d) European Union Risk Assessment Report bis(2-ethylhexyl)phthalate (DEHP), European Commission Directorate-General Joint Research Centre, EUR 23396 EN. Final report August 2007.
- Eurofin DIN 38414-14 (2011) German standard methods for the examination of water, waste water and sludge - Sludge and sediments (group S) - Part 14: Determination of selected polyfluorinated compounds (PFC) in sludge, compost and soil - Method using high performance liquid chromatography and mass spectrometric detection (HPLC-MS/MS) (S 14)
- Fahlenkamp, H., Nöthe, T., Hannich, B.C. (2005) *Organische Spurenstoffe – Bilanz moderner Kläranlagen unter Berücksichtigung des Wasser- und Schlammwegs I. Organische Spurenstoffe im Wasser und Schlammweg von zwei grossen Kläranlagen*. Gewässerschutz, Wasser, Abwasser 198, 67/1-67/20.
- Falås, P., Baillon-Dhumez, A., Andersen, H.R., Ledin, A. La Cour Jansen, J. (2012) Suspended biofilm carrier and activated sludge removal of acidic pharmaceuticals, *Water Res.* 46: 1167–1175.
- Feng, C., Guang-Guo, Y., Yi-Bing, M., Zhi-Feng, C., Hua-Jie, L., Feng-Jiao, P. (2014) Field dissipation and risk assessment of typical personal care products TCC, TCS, AHTN and HHCB in biosolid-amended soil. *Sci. Tot. Environ.* 470–471: 1078–1086.
- Fernandez-Fontaina, E., Omil, F., Lema, J.M., Carballa, M. (2012) Influence of nitrifying conditions on the biodegradation and sorption of emerging micropollutants. *Water Res.* 46 (16), 5434–5444.
- FOR-2003-07-04-951. Forskrift om gjødselvarer mv. av organisk opphav.
<https://lovdata.no/dokument/SF/forskrift/2003-07-04-951?q=gj%C3%B8dselvarer>
- Frömel, T., Knepper, T.P. (2010) Fluorotelomer ethoxylates: sources of highly fluorinated environmental contaminants part I: biotransformation. *Chemosphere.* 80(11):1387-1392.
- Gan, D.R., Berthouex, P. M. (1994) Disappearance and crop uptake of PCBs from sludge-amended farmland. *Water Environment Research*, 66(1):54-69.
- Gauthier, S.A., Mabury, S.A. (2005) Aqueous photolysis of 8: 2 fluorotelomer alcohol. *Environmental toxicology and chemistry*, 24(8):1837-1846.

- Gerecke, A., Giger, W., Hartmann, P., Heeb, N.V., Kohler, H.E., Schmid, P., Zennegg, M. and Kohler, M. (2006) Anaerobic degradation of brominated flame retardants in sewage sludge. *Chemosphere*, 64:311-317.
- González, M.M., Martín, J., Camacho-Muñoz, D., SanLiutos, J.L., Aparicio, I., Alonso, E. (2012) Degradation and environmental risk of surfactants after the application of compost sludge to the soil. *Waste Management*, 32(7):1324-1331.
- Gonzalez-Gil, L., M. Papa, D., Feretti, E., Ceretti, G., Mazzoleni, N., Steimberg, R., Pedrazzani, G., Bertanza, J.M., Lema, M., Carballa (2016) Is anaerobic digestion effective for the removal of organic micropollutants and biological activities from sewage sludge? *Water Research* 102:211-220.
- Göbel, A., Mc Ardell, C.S., Joss, A., Siegrist, H., Giger, W. (2007) Fate of sulfonamides, macrolides, and trimethoprim in different wastewater treatment technologies, *Sci. Tot. Environ.* 372:361-371.
- Graham P.R. (1973) Phthalate Ester Plasticizers-Why and How They Are Used. *Environmental Health Perspectives*, January 1973:3-12.
- Grandclément C., Seyssiecq I., Piram A., Wong-Wah-Chung P., Vanot G., Tiliacos N., Roche N., Doumenq P. (2017) From the conventional biological wastewater treatment to hybrid processes, the evaluation of organic micropollutant removal: A review. *Wat. Res.* 111:297-317.
- Hai, F.I., Nghiem, L.D., Khan, S.J., Price, W.E., Yamamoto, K. (2014) Wastewater reuse: removal of emerging trace organic contaminants by membrane bioreactors. In: F.I. Hai, K. Yamamoto, C.H. Lee (Eds.), *Membrane Biological Reactors*, IWA Publishing, UK, pp. 165–205.
- Haug, R.T. (1993) *The Practical Handbook of Compost Engineering*. Lewis publishers, Florida, U.S.A., pp 21-93.
- He, L., Fan, S., Müller, K., Wang, H., Che, L., Xu, S., Song, Z., Yuan, G., Rinklebe, J., Tsangl, D.C.W., Ok, Y.S., Bolan, N.S. (2018) Comparative analysis biochar and compost-induced degradation of di-(2-ethylhexyl) phthalate in soils. *Sci. Tot. Environ.* 625: 987–993.
- He, J., Robrock, K.R., Alvarez-Cohen, L. (2006) Microbial Reductive Debromination of Polybrominated Diphenyl Ethers (PBDEs). *Environ. Sci. Technol.* 40:4429-4434.
- Huang, M., Li Y, Gu G. (2008) The effects of hydraulic retention time and sludge retention time on the fate of di-(2-ethylhexyl) phthalate in a laboratory-scale anaerobic–anoxic–aerobic activated sludge system. *Bioresour. Technol.* 99:8107–11.
- Hernández-Leal L, Temmink H, Zeeman G, Buisman CJN. Removal of micropollutants from aerobically treated grey water via ozone and activated carbon. *Water Res* 2011;45: 2887–96.
- Hernandez-Raquet, G., Soef, A., Delgenès, N., Balaguer, P. (2007) Removal of the endocrine disrupter nonylphenol and its estrogenic activity in sludge treatment processes. *Wat. Res.* 41:2643-51.
- Houtz, E.F., Sedlak D.L. (2012) Oxidative Conversion as a Means of Detecting Precursors to Perfluoroalkyl Acids in Urban Runoff. *Environ. Sci. Technol.* 46(17): 9342–9349.
- Hulscher, T.E.M., Cornelissen, G. (1996) Effect of temperature on sorption equilibrium and sorption kinetics of organic micropollutants – a review. *Chemosphere.* 32: 609–626.
- IPEN (2015) Submission of information specified in Annex E to the Stockholm Convention pursuant to Article 8 of the Convention.
- IPTS (2013) Study report on End-of-waste criteria for Biodegradable waste subjected to biological treatment. Draft final report. IPTS, Seville, Spain. July 2013.
- ISO 18635 (2016) Water quality - Determination of short-chain polychlorinated alkanes (SCCPs) in sediment, sewage sludge and suspended (particulate) matter - Method using gas chromatography-mass spectrometry (GC-MS) and electron capture negative ionization (ECNI).

- ISO 13913:2014. Soil quality - Determination of selected phthalates using capillary gas chromatography with mass spectrometric detection (GC/MS).
- ISO/TS 13907:2012. Soil quality - Determination of nonylphenols (NP) and nonylphenol-mono- and diethoxylates - Method by gas chromatography with mass selective detection (GC-MS).
- ISO 22032:2006. Water quality - Determination of selected polybrominated diphenyl ethers in sediment and sewage sludge - Method using extraction and gas chromatography/mass spectrometry.
- JRC (2013) Occurrence and levels of selected compounds in European compost and digestate samples. Results of a Pan European Screening exercise FATE-COMES. JR technical reports. Report EUR 26164 EN. ISBN 978-92-79-33195-4 (pdf).
- Jackson, D. A., Wallington, T. J., Mabury, S. A. (2013) Atmospheric oxidation of polyfluorinated amides: historical source of perfluorinated carboxylic acids to the environment. *Environ. Sci. Technol.* 47(9): 4317-4324.
- Kapanen, A., Vikman, M., Rajasarkka, J., Virta, M., Itavaara, M. (2013) Biotests for environmental quality assessment of composted sewage sludge. *Waste Management* 33:1451–1460.
- Kim, D., Cui, R., Moon, J., Kwak, J. I., An, Y. J. (2019) Soil ecotoxicity study of DEHP with respect to multiple soil species. *Chemosphere*, 216: 387-395.
- Klaschka U., von der Ohe P.C., Bschorer A., Krezmer S., Sengl M., Letzel M. (2013) Occurrences and potential risks of 16 fragrances in five German sewage treatment plants and their receiving waters. *Environ Sci Pollut Res* (2013) 20:2456–2471. DOI 10.1007/s11356-012-1120-9.
- Konieczny, R.M., Horvath, A., Lyngstad, E., Dalen, H., Blytt, L.D., Henninge, L.B., Ferenčík, M., Nilan, M.S., Bergqvist, P-A., Grabic, R., Haukelidsæter S., Randall, S. (2017) Screening programme 2016: Suspected PBT compounds, Environmental monitoring report M-806|2017
- Konieczny, R.M., Henninge, L.B., Dalen, H., Grabic, R., Ferenčík, M., Bergqvist, P-A., Lyngstad, E., Berger, J., Haukelidsæter S., Randall, S. (2018) Screening programme 2017: Suspected PBT compounds, Environmental monitoring report M-1063|2018
- Kortenkamp, A., Martin, O., Evans, R., Faust, Backhaus, T. (2014) Risk of combination effects between decabromodiphenyl ether and other polybrominated diphenyl ethers. Report from Norwegian Environment Agency. M233-2014.
- Krogseth, I.S., Breivik, K., Arnot, J.A., Wania, F., Borgen, A.R., Schlabach, M. (2013) Evaluating the Environmental Fate of Short-Chain Chlorinated Paraffins (SCCPs) in the Nordic Environment Using a Dynamic Multimedia Model. *Environ. Sci. Processes Impacts*, 15: 2240.
- Kupper T., Plagellat C., Brändli R.C., de Alencastro L.F., Grandjean D., Tarradellas J. (2006) Fate and removal of polycyclic musks, UV filters and biocides during wastewater treatment. *Wat. Res.* 40:2603-2612.
- Langdon, K., Warne, M., Kookana, R. (2010) Aquatic Hazard Assessment for Pharmaceuticals, Personal Care Products, and Endocrine-Disrupting Compounds from Biosolids-Amended Land. *Integrated Environmental Assessment and Management*, 6(4): 663.
- Lashermes, G., Houot, S., Barriuso, E. (2010) Sorption and mineralization of organic pollutants during different stages of composting. *Chemosphere*. 79: 455–462.
- Lee, D.M., Langworthy, D.E., Federle, T.W. (2001) Biotransformation of AHTN (Tonalid) in activated sludge. Poster presented at SETAC Conference, Madrid, Spain, May 2001.
- Liu, J., Avendaño, S. M. (2013) Microbial degradation of polyfluoroalkyl chemicals in the environment: A review., *Env. Int.* 61: 98–114.

- Macherius, A., Eggen, T., Lorenz, W., Möder, M., Ondruschka, J., Reemtsma, T. (2012) Metabolization of Bacteriostatic Personal Care Products in Edible Plants and its Consequences for Plant Uptake. *Environ. Sci. Technol.* 46:10797-10804.
- Macherzyński, B., Włodarczyk-Makula, A., Nowacka, A. 2014. Desorption of PAHs from solid phase into liquid phase during co-fermentation of municipal and coke sewage sludge. *Desalin. Water Treat.* 52:3859-3870.
- Mailler R., Gasperi J., Chebbo G., Rocher V. (2014) Priority and emerging pollutants in sewage sludge and fate during sludge treatment. *Waste Management.* 34:1217–1226.
- Matthies, M., Beulke, S. (2017) Considerations of temperature in the context of the persistence classification in the EU *Environ Sci Eur.* 29:15.
- Mattilsynet (2012) Veileder for prøvetaking av slam, kompost og andre avfallsbaserte gjødselvarer. 22 pages.
- Miljøstyrelsen (2018). Miljø- og Fødevareministeriet. Nabolik av reglene om spildevandsslam. Sverige, Norge, Finland, Tyskland og England. Miljøprosjekt nr. 1989. Marts 2018.
- Moermond C.T.A., Verbruggen E.M.J. and Smit C.E. (2010) Environmental risk limits for PFOS – A proposal for water quality standards in accordance with the Water Framework Directive. RIVM Report 601714013/2010, 70 pages.
- Mohapatra, D.P., Cledód, M., Brar, S.K., Surampalli, R.Y. (2016) Application of Wastewater and Biosolids in Soil: Occurrence and Fate of Emerging Contaminants. *Water Air Soil Pollut.* 227:77.
- Naturvårdsverket. 2019. Vägledning om att riskbedöma och åtgärda PFAS-föreningar inom förorenade områden. Naturvårdsverket Rapport 6871 Januari 2019. ISBN 978-91-620-6871 - 4. <http://www.naturvardsverket.se/Documents/publikationer6400/978-91-620-6871-4.pdf?pid=24088>
- NICNAS (2019) Indirect precursors to perfluorooctanesulfonate (PFOS): Environment tier II assessment, Australian Government, Department of Health, National Industrial Chemicals Notification and Assessment Scheme, 8 March 2019.
- Nielsen C.J. (2014) Potential PFOA Precursors - Literature study and theoretical assessment of abiotic degradation pathways leading to PFOA. The Norwegian Environment Agency Report M-231, 46 pages.
- NS-EN ISO (2009) Water quality – Sampling – Part 15: Guidance on preservation and handling of sludge and sediment samples.
- Ødegaard H., Rusten B., Storhaug R. and Paulsrud B. (2009) Veiledning for dimensjonering av avløpsrensning. Norsk Vann Report 168-2009, 57 pages.
- OECD (2007) List of PFOS, PFAS, PFOA, PFCA, related compounds and chemicals that may degrade to PFCA. Environment Directorate Joint Meeting of the Chemicals Committee and the Working Party on Chemicals, Pesticides and Biotechnology, Series on Risk Management No. 21, 2007. ENV/JM/MONO(2006)15.
- O’Grady D.P., Howard P.H., Werner A.F. (1985) Activated sludge biodegradation of 12 commercial phthalate esters *Appl. Environ. Microbiol.*, 49(2):443-445.
- Ozaki, N., Nakazato, A., Nakashima, K., Kindaichi, T., Ohashi, A. (2017) Loading and removal of PAHs, fragrance compounds, triclosan and toxicity by composting process from sewage sludge. *Sci. Tot. Environ.* 605–606:860–866.

- Pan, G., Jia, C., Zhao, D., You, C., Chen, H., Jiang, G. (2009) Effect of cationic and anionic surfactants on the sorption and desorption of perfluorooctane sulfonate (PFOS) on natural sediments. *Environ. Pollut.* 157:325-330.
- Parker, W.J., Monteith, H.D., Melcer, H. (1994) Estimation of anaerobic biodegradation rates for toxic organic compounds in municipal sludge digestion. *Water Res.* 28: 1779–1789.
- Paterakis, N., Chiu, T.Y., Koh, Y.K.K., Lester, J.N., McAdam, E.J., Scrimshaw, M.D., Soares, A., Cartmell, E. (2012) The effectiveness of anaerobic digestion in removing estrogens and nonylphenol ethoxylates. *J. Hazard. Mater.* 199–200: 88-95.
- Paterakis, N., Chiu, T.Y., Koh, Y.K.K., Lester, J.N., McAdam, E.J., Scrimshaw, M.D., Soares, A., Cartmell, E. (2012) The effectiveness of anaerobic digestion in removing estrogens and nonylphenol ethoxylates. *J. Hazard. Mater.* 199–200: 88-95.
- Pham T.T.H., Tyagi R.D., Brar S.K., Surampalli R.Y. (2011) Effect of ultrasonication and Fenton oxidation on biodegradation of bis(2-ethylhexyl) phthalate (DEHP) in wastewater sludge. *Chemosphere* 82:923–928.
- Phan, H.V., Hai, F.I., Kang, J., Dam, H.K., Zhang, R., Price, W.E., Broeckmann, A., Nghiem, L.D. (2014) Simultaneous nitrification/denitrification and trace organiccontaminant (TrOC) removal by an anoxic–aerobic membrane bioreactor (MBR), *Bioresour. Technol.* 165: 96–104.
- Pollice, A., Laera, G. (2005) Effects of complete sludge retention on biomass build-up in a membrane bioreactor. *Water Sci. Technol.* 52(10–11):369–375.
- Poulsen, T.G., Bester, K. (2010) Organic micro pollutant degradation in sewage sludge during composting under thermophilic conditions. *Environ. Sci. Tech.* 44 (13):5086–91.
- Pulkrabová, J., Cemý, J., Száková, J., Svarcová, A., Gramblička, T., Hajšlová, J., Balík, J., Tlustoš, P. (2019) Is the long-term application of sewage sludge turning soil into a sink for organic pollutants? evidence from field studies in the Czech Republic. *J. Soils and Sediments.*
- Qiang, Z., Nie, Y., Ben, W., Qu, J., Zhang, H. (2013) Degradation of endocrine-disrupting chemicals during activated sludge reduction by ozone. *Chemosphere.* 91: 366-373.
- Rivier, P.-A., Havranek, I., Coutris, C., Norli, H.R., Joner, E. (2019) Transfer of organic pollutants from sewage sludge to earthworms and barley under field conditions. *Chemosphere.* 222: 954-960.
- Rankin, K., Lee, H., Tseng, P. J., Mabury, S. A. (2014) Investigating the biodegradability of a fluorotelomer-based acrylate polymer in a soil–plant microcosm by indirect and direct analysis. *Environ. Sci. Technol.* 48(21):2783-12790.
- Rayne, S., Forest, K. (2010) Modelling the hydrolysis of perfluorinated compounds containing carboxylic and phosphoric acid ester functions and sulfonamide groups. *J. Environ. Sci. Health Part A*, 45(4):432-446.
- Reinhardt, T. (2002) Organic acids as a decisive limitation to process dynamics during composting of organic matter. In: Insam, H., Riddech, N and Klammer, S. (Eds.) *Microbiology of Composting.* Springer-Verlag, Berlin Heidelberg, pp.177-188.
- Renner, R. (2008) EPA finds record PFOS, PFOA levels in Alabama grazing fields.
- Rigby, H., Dowding, A., Fernandes, A., Humphries, D., Petch, R.G., Reynolds, C.K., Martin, Rose, M., Smith, S.R. (2015) Organic Contaminant Content and Physico-Chemical Characteristics of Waste Materials Recycled in Agriculture. *Agriculture* 5: 1289-1328.
- Robrock, K.R., Korytár, P., Alvarez-Cohen, L. (2008) Pathways for the anaerobic microbial debromination of polybrominated diphenyl ethers. *Environ. Sci. Technol.* 42: 2845–2852.

- Romijn, C.F.A.M., Luttik, R., Canton, J.H., (1994). Presentation of a general algorithm to include effect assessment on secondary poisoning in the derivation of environmental quality criteria. Part 2. Terrestrial food chains. *Ecotox. Environ. Saf.* 27: 107-127.
- Ruiz-Aguilar, G., 2002. Degradation by white-rot fungi of high concentrations of PCB extracted from a contaminated soil. *Adv. Environ. Res.* 6, 559-568.
- Sadef, Y., Poulsen, T.G., Bester, K. (2014) Impact of compost process temperature on organic micro-pollutant degradation. *Sci. Tot. Environ.* 494-495:306-312.
- Saeger V.W., Tucker E.S. (1976) Biodegradation of Phthalic Acid Esters in River Water and Activated Sludge. *Applied and Environmental Microbiology*, 31(1):29-34.
- Scaglia, B., Tambone, F., Corno, L., Orzi, V., Lazzarini, Y., Garuti, G., Adani, F. (2018) Potential agronomic and environmental properties of thermophilic anaerobically digested municipal sewage sludge measured by an unsupervised and a supervised chemometric approach. *Science of the Total Environment* 637-638:791-802.
- Schlabach, M. (2011) Brominated flame retardants (BFR) in the Nordic environment. Nordic Council of Ministers.
- Scow, K.M., Gan, S., Johnson, C., Ma, G.M. (1995) *Environ. Health Perspectives*, Vol. 103, Supplement 5. June 1995.
- Semblante G.U., Hai F.I., Huang X., Ball A.S. Price W.E. (2015) Review: Trace organic contaminants in biosolids: Impact of conventional wastewater and sludge processing technologies and emerging alternatives. *J. Hazard. Mat.*, 300: 1-17.
- Simonich, S.L., Federle, T.M., Eckhoff, W.S., Rottiers, A., Webb, S., Sabaliunas, D., de Wolf, D. (2002) Removal of fragrance materials during U.S. and European wastewater treatment. *Environ. Sci. Technol.* 36:2839-2847.
- Smyth S.A., Lishman L.A., McBean E.A., Kleywegt S., Yang J.-J., Svoboda M.L., Lee H-B., Seto P. (2007) Fate of Polycyclic and Nitro Musks during Aerobic and Anaerobic Sludge Digestion. *Proceedings of the International Water Association Specialist Conference on Moving Forward Biosolids Sustainability*, Moncton NB, Canada:223-230.
- Smårs, S., Beck-Friis, B., Jönsson, H., Kirchmann, H. (2001) An advanced experimental composting reactor for systematic simulation studies. *Journal of agricultural Engineering Research.*, 78:415-422.
- Stadler, L.B., Su, L., Moline, C.J., Ernstoff, A.S., Aga, D.S., Love, N.G. (2015) Effect of redox conditions on pharmaceutical loss during biological wastewater treatment using sequencing batch reactors, *J. Hazard. Mater.* 282:106-115.
- Staples, C. A., Naylor, C. G., Williams, J. B., Gledhill, W. E. (2001) Ultimate biodegradation of alkylphenol ethoxylate surfactants and their biodegradation intermediates. *Environ. Toxicol. Chemistry.* 20(11):2450-2455.
- Staples, C.A., Peterson, D.R., Parkerton, T.F., Adams, W.J. (1997) The environmental fate of phthalate esters: A literature review. *Chemosphere* 35:667-749.
- Stenstrom, MK., Cardinal, L., Libra, L. (1989) Treatment of hazardous substances in wastewater treatment plants. *Environ Progr* 8(2):107-112.
- Sternbeck, J., Blytt, L.D., Gustavson, K., Frankki, S., Bjergström, M. (2011) Using sludge on arable land – effect based levels and longterm accumulation for certain organic pollutants. Report to the Nordic Council of Ministers. *TemaNord* 2011: 506.

- Suárez S., Lema J.M., Omil F. (2009) Pre-treatment of hospital wastewater by coagulation–flocculation and flotation. *Bioresour. Technol.* 100:2138–46.
- Suarez, S., Lema, J.M., Omil, F. (2010) Removal of Pharmaceutical and Personal Care Products (PPCPs) under nitrifying and denitrifying conditions, *Water Res.* 44:3214–3224.
- Sundberg, C., Jonsson, H. (2005) Process inhibition due to organic acids in fed - batch composting of food waste – influence of starting culture. *Biodegradation* 16(3):205-213.
- Suominen, K., Verta, M., Marttinen, S. (2014) Hazardous organic compounds in biogas plant end products—Soil burden and risk to food safety. *Sci. Tot. Environ.* 491–492:192–199.
- SWECO (2010) Screening of musk substances and metabolites. Malmö 2010-06-01.
- TA-1629/99. Statens forurensningstilsyn. Veiledning om risikovurdering av forurenset grunn (VEILEDNING 99:01a).
- Tadkaew N., Sivakumar M., Khan S.J., McDonald J.A., Nghiem L.D. (2010) Effect of mixed liquor pH on the removal of trace organic contaminants in a membrane bioreactor. *Bioresource Technology*, 101:1494-1500
- TemaNord (2019) Screening of Poly- and Perfluroalkyl Substances (PFASs) and extractable organic Fluorine (EOF) in the Nordic Environment. Kärrman, A., Wang, T., Kallenborn with co-authors. TemaNord 2019:515. ISBN 978-92-893-6062-3.
- Ternes, T.A, Herrmann, N., Bonerz, M., Knacker, T., Siegrist, H., Joss, A. (2004a) *Water Res.* 38: 4075-4084.
- Ternes, T.A., Joss, A., Siegrist, H. (2004b) Peer Reviewed: Scrutinizing Pharmaceuticals and Personal Care Products in Wastewater Treatment. *Environ. Sci. Technol.* 38:393A-399A.
- Terzaghi, E., Zanardini, E., Morosini, C., Raspa, G., Borin, S., Mapelli, F., Vergani, L., Guardo, A.D. (2018) Rhizoremediation half-lives of PCBs: Role of congener composition, organic carbon forms, bioavailability, microbial activity, plant species and soil conditions, on the prediction of fate and persistence in soil. *Sci. Tot. Environ.* 612: 544–560.
- Thomas, K.V., Langford K.H., Muthanna T., Schlabach M., Enge E.K., Borgen A., Ghebremeskel M., Gundersen H., Leknes H., Uggerud H., Haglund P, Liao Z., Liltved H. (2011) Occurrence of selected organic micropollutants and silver at wastewater treatment plants in Norway. KLIF TA 2784/2011.
- Tokarz, J.A., Ahn, M.Y., Leng, J., Filley, T.R., Nies, L. (2008). Reductive debromination of polybrominated diphenyl ethers in anaerobic sediment and a biomimetic system. *Environ. Sci. Technol.* 42:1157-64.
- Tran, N.H., Urase, T., Ngo, H.H., Hu, J., Ong, S.L. (2013) Insight into metabolic and cometabolic activities of autotrophic and heterotrophic microorganisms in the biodegradation of emerging trace organic contaminants. *Bioresour Technol.* 146:721–731.
- Trapp, S. (2000) Modelling uptake into roots and subsequent translocation of neutral and ionisable organic compounds. *Pest. Manag. Sci.* 56:767–778.
- Trapp, S., Legind ,C. (2011) Uptake of organic contaminants from soil into vegetation. In: Swartjes F (Ed.) *Dealing with contaminated sites from theory towards practical application.* Springer, Dordrecht.
- US EPA (2010) Nonylphenol (NP) and Nonylphenol Ethoxylates (NPEs) Action Plan [RIN 2070-ZA09], 8/18/2010, 13 pages.
- Van Leeuwen, S.P.J., Karrman, A., Van Bavel, B., De Boer, J., Lindstrom, G. (2006) *Environ. Sci. Technol.* 40:7854.

- Venkatesan, A.K., Halden, R.U. (2013) National inventory of alkylphenol ethoxylate compounds in US sewage sludges and chemical fate in outdoor soil mesocosms. *Environ. Pollut.* 174:189-193.
- Venkatesan, A. K., Halden, R.U. (2014) Loss and in situ production of perfluoroalkyl chemicals in outdoor biosolids–soil mesocosms. *Environ. Res.* 132:321-327.
- Verlicchi, P., Verlicchi, Al Aukidy, M., Zambello, E. (2012) Occurrence of pharmaceutical compounds in urban wastewater: removal, mass load and environmental risk after a secondary treatment--a review. *Sci. Tot. Environ.* 429: 123-155.
- VKM (2014) Zinc and copper in pig and poultry production – fate and effects in the food chain and the environment. Opinion of the Panel on Animal Feed of the Norwegian Scientific Committee for Food Safety. VKM Report 13-603. ISBN: 978-82-8259-093-8.
- VKM (2009) Risk assessment of contaminants in sewage sludge applied on Norwegian soils. Opinion of the Panel on Contaminants of the Norwegian Scientific Committee for Food Safety, ISBN 978-82-8082-337-3.
<https://vkm.no/download/18.645b840415d03a2fe8f1293/1501260413588/2ae7f1b4e3.pdf>
- Xia, K., Bhandari, A., Das, K., Pillar, G. (2005) Occurrence and Fate of Pharmaceuticals and Personal Care Products (PPCPs) in Biosolids. *J. Environ. Qual.* 34:91–104.
- Xiao, F., Simcik, M.F., Halbach, T.R., Gulliver, J.S. (2015) Perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in soils and groundwater of a U.S. metropolitan area: migration and implications for human exposure. *Water Res* 72:64–74
- Wang, J., Huang, C.P., Allen, H.E. (2000) Surface physical–chemical characteristics of sludge particulates, *Water Environ. Res.* 72 545–553.
- Wang, N., Szostek, B., Buck, R. C., Folsom, P.W., Sulecki, L. M., Capka, V., Berti, W.R., Gannon, J.T. (2005a). Fluorotelomer alcohol biodegradation direct evidence that perfluorinated carbon chains breakdown. *Environ. Sci. Technol.* 39(19): 7516-7528.
- Wang, N., Szostek, B., Folsom, P.W., Sulecki, L.M., Capka, V., Buck, R. C., Berti, W.R., Gannon, J. T. (2005b) Aerobic biotransformation of ¹⁴C-labeled 8-2 telomer B alcohol by activated sludge from a domestic sewage treatment plant. *Environ. Sci. Technol.* 39(2): 531-538.
- Wang, Y., Luo, C., Li, J., Yin, H., Li, X., Zhang, G. (2011) Characterization of PBDEs in soils and vegetations near an e-waste recycling site in South China. *Environ. Pollut.* 159:2443–2448.
- Wang L., Wijekoon K.C., Nghiem L.D., Khan S.J. (2014) Removal of polycyclic musks by anaerobic membrane bioreactor: Biodegradation, biosorption, and enantioselectivity. *Chemosphere* 117:722–729.
- Washington, J. W., Ellington, J. J., Jenkins, T. M., Evans, J. J., Yoo, H., Hafner, S. C., (2009). Degradability of an acrylate-linked, fluorotelomer polymer in soil. *Environ. Sci. Technol.* 43(17):6617-6623.
- Washington, J. W., Jenkins, T. M., Rankin, K., Naile, J. E., (2015). Decades-scale degradation of commercial, side-chain, fluorotelomer-based polymers in soils and water. *Environ. Sci. Technol.* 49(2): 915-923.
- Weber, W., Huang, Q. (2003) Inclusion of Persistent Organic Pollutants in Humification Processes: Direct Chemical Incorporation of Phenanthrene via Oxidative Coupling. *Environ. Sci. Technol.* 37: 4221-4227.
- Weiss, S., Reemtsma, T. (2008) Membrane bioreactors for municipal wastewater treatment – A viable option to reduce the amount of polar pollutants discharged into surface waters? *Water Res.*, 42:3837-3847.

- Yoshida H., Christensen T.H., Guildal T. and Scheutz C. (2015) A comprehensive substance flow analysis of a municipal wastewater and sludge treatment plant. *Chemosphere* 138:874-882.
- Young, C. J., Mabury, S. A. (2010) Atmospheric perfluorinated acid precursors: chemistry, occurrence, and impacts. In *Reviews of Environmental Contamination and Toxicology Volume 208*. pp. 1-109. Springer, New York, NY.
- Yu, J., Hu, J.Y., Tanaka, S., Fujii, S. (2009) Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in sewage treatment plants. *Water Res.* 43:2399–2408.
- Zhang, C., Yan, H., Li, F., Hu, X., Zhou, Q. (2013) Sorption of short-and long-chain perfluoroalkyl surfactants on sewage sludges. *J. Hazard. Mater.* 260:689-699.
- Zhang, B., Yamamoto, K., Ohgaki, S., Kamiko, N. (1997) Floc size distribution and bacterial activities in membrane separation activated sludge processes for small-scale wastewater treatment/reclamation. *Water Sci Technol.* 35(6):37–44.
- Zeng L.Z., Li H., Wang T., Gao Y., iao K., Du Y., Wang Y. and Jiang G. (2012) Behavior, Fate, and Mass Loading of Short Chain Chlorinated Paraffins in an Advanced Municipal Sewage Treatment Plant. | *Environ. Sci. Technol.*
- Zhao, L.Y., Yang, J.Y., Zhan, S.E., Jiang, J.H., Yang, S. (2016). Bioremediation of sediment contaminated with decabronodiphenyl ether (BDE-209) by composting. *Fres. Environ. Bulletin* 25:3700-3708.
- Zhou, Q., Deng, S., Zhang, Q., Fan, Q., Huang, J., Yu, G. (2010) Sorption of perfluorooctane sulfonate and perfluorooctanoate on activated sludge, *Chemosphere*. 81:453–458.
- Zhuang, Y., Jin, L., Luthy, R.G. (2012) Kinetics and Pathways for the Debromination of Polybrominated Diphenyl Ethers by Bimetallic and Nanoscale Zerovalent Iron: Effects of Particle Properties and Catalyst. *Chemosphere*. 89:426-432.
- Österas, A.H., Allmyr, M., Sternbeck, J. (2015) Screening of organic pollutants in sewage sludge amended arable soils. Swedish Environmental Protection Agency.

Appendix A

Waste sources currently and potentially used as feedstocks for fertilizer production

- 1) Food industry waste
 - a) Slaughterhouse waste
 - b) Canning waste
 - c) Potato industry waste
- 2) Farming waste
 - a) Manure
 - b) Agricultural waste
- 3) Food waste from institutional households
- 4) Household waste
- 5) Fish farm waste
- 6) Fishery waste
- 7) Waste from wood processing industry
- 8) Other industrial sludge/-waste
- 9) Garden waste
- 10) Sludge from wastewater treatment plants
- 11) Sludge from drinking water treatment plants
- 12) Mixes of two or more from the list 1-11

Fra Gjødelsvareforskriften (veilederen):

Opphavsmateriale	Gronnsaker / poteter	Eng/Beite	Annet jordbruk	Grøntarealer
Avløpsslam	÷ (2)	÷ (1)	+	+
Animalske biprodukter	+	(3)	+	+
Potetindustriavfall	Ikke på potetarealer eller arealer hvor det er planer om slik dyrking de nærmeste 20 årene (4).			

+ tillatt, ÷ ikke tillatt

(1) Bruk på eng eller beite ikke er tillatt, mens bruk på gjenlegg med høsting eller beiting neste sesong er tillatt

(2) Etter spredning av avløpsslam, kan grønnsaker eller poteter ikke dyrkes de tre neste kalenderårene.

(3) Animaliebiproduktforskriften kan gi begrensninger for bruk på beitearealer og arealer hvor det dyrkes fôrvekster.

(4) I en klagesak i 2017 ble det gjort en tolkning av kravet i gjødelsvareforskriftens vedlegg 4 nr. 1c om at det ikke kan brukes gjødelsvare basert på potetindustriavfall på arealer hvor det er planer om potetdyrking de nærmeste 20 årene. Det er de faktiske planene på spredetidspunktet som er avgjørende, det vil si om brukeren på spredetidspunktet har faktiske planer om å dyrke poteter i løpet av de neste 20 årene. Dette medfører ikke unntak fra kravet om hygienisering (§ 10 punkt 3) som skal sikre at gjødselproduktet ikke utgjør en fare for overføring av sykdomssmitte. Avfall fra potetindustri kan ha høy risiko for å inneholde karanteneskadegjørere og dette er viktig å ta hensyn til ved behandling og bruk. Les mer om risiko og mulige risikoreduserende tiltak i VKMs vurdering av karanteneskadegjørere i potet- og grønnsakspakkerier [her](#).

Tabell 1

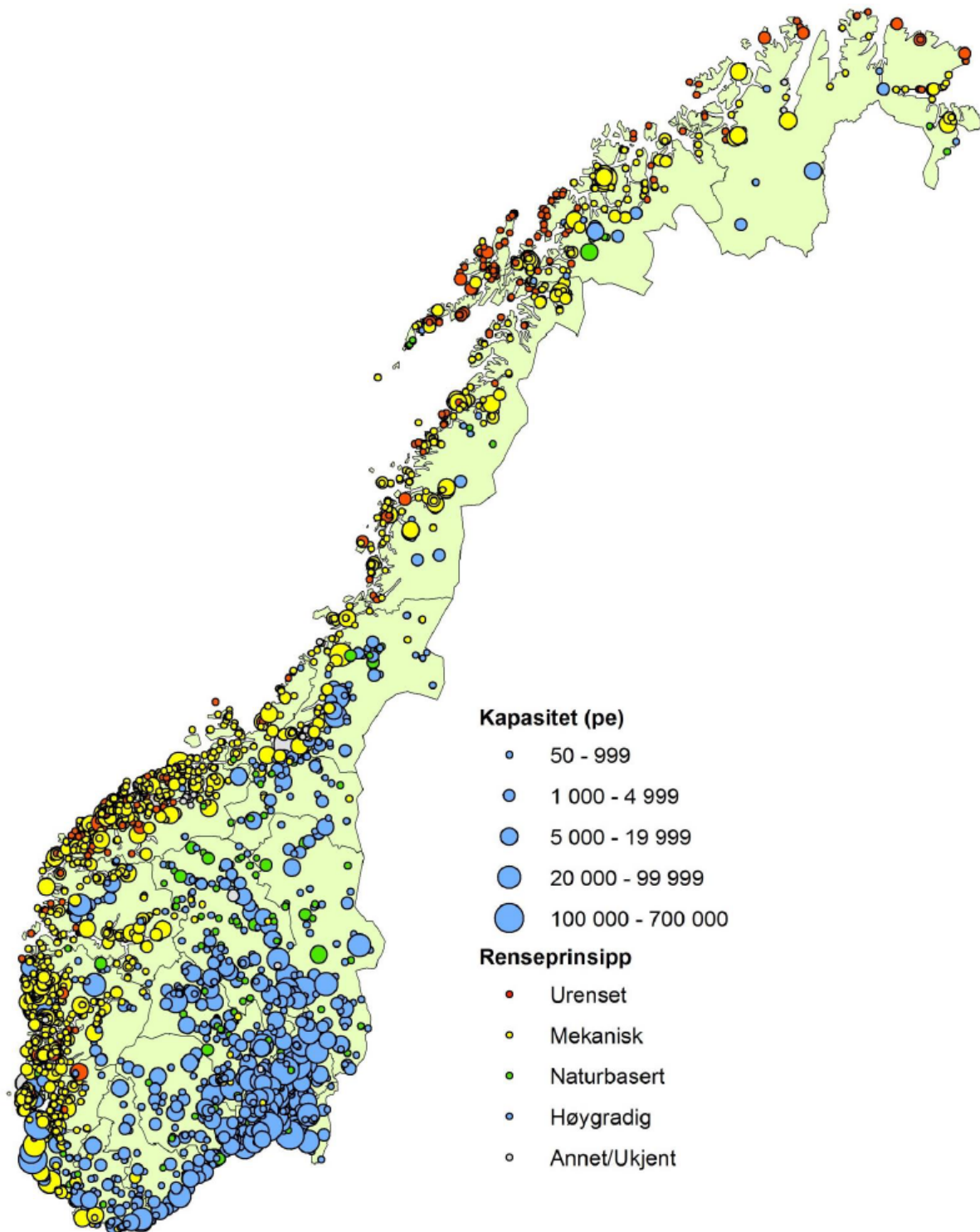
Produkter som omfattes av forskriften og eventuelle krav til registrering av produktet hos Mattilsynet.

Varetyper mv.	Omfattet av forskrift	Registreringsplikt
Spagnumtorv (inkl. gjødsla og kalka spagnumtorv)	+	+
Minerogen myratorv	+	+
Kompost (ikke råvarer i forskriftens vedlegg 4) ¹	+	+
Kompost (råvarer i forskriftens vedlegg 4)	+	+
Hage- og parkavfallskompost	+	+
Hjemmekompost	-	-
Anaerobt omsatt biomasse (råvarer i forskriftens vedlegg 4)	+	+
Tørka biomasse (råvarer i forskriftens vedlegg 4)	+	+
Kalkstabilisert slam (råvarer i forskriftens vedlegg 4)	+	+
Bark	+	-
Bark – kompostert	+	-
Cellulosefiber	+	-
Treflis	+	-
Halm	+	-
Kokosfiber	+	-
Kakaoflis	+	-
Lignit	+	-
Husdyrgjødsel – ubehandlet for omsetning	+	-
Husdyrgjødsel – til eget / leid areal	+	-
Sand	+	-
Silt	+	-
Leir	+	-
Pimpstein	+	-
Mineralull (revet/ikke-revet)	+	-
Ekspanderte mineralprodukter	+	-
Organiske polymerer	+	-
Matter, folier og lignende til jorddekking	-	-
Blandet dyrkingsmedium/ jordforbedrings-/jorddekkingsmidler av kun ikke-registreringspliktige komponenter.	+	-
Marin plantebiomasse	+	-
Gjødsla og kalka produkter registreringspliktige/ikke-registreringspliktige komponenter (utenom gjødsla og kalka spagnumtorv)	+	-
Blandet jordforbedringsmiddel/ dyrkingsmedium/ jorddekkingsmiddel		
1) som inneholder registreringspliktige råvarer	+	+
2) som kun består av ikke-registreringspliktige råvarer	+	-
Anleggsjord uten innhold av organisk avfall og avløpsslam med innhold av produkt basert på organisk avfall og avløpsslam	+	-
Andre produkter	+	+
	Kontakt Mattilsynet	Kontakt Mattilsynet

¹ gjelder ikke bark – kompostert

Appendix B

Location of WWTPs serving >50 PE in Norway (Berge og Sæther, 2018)



Appendix C

Available analysis of the selected HOCs in Norwegian sewage sludge have been collected, and minimum, median, mean, maximum values are summarized in Table AC-1. These numbers include in addition to the Norwegian WWTP campaign (Blytt and Stang, 2019), also data from other Norwegian and Nordic surveys; Blytt 2010, Blytt et al., 2013 and 2019), Thomas et al., 2011; Naturvårdsverket, 2019; Konieczny et al., 2017; Konieczny et al., 2018; Schlabach et al., 2011.

The number of samples for each of the contaminants vary between 60 (PFOA) to 193 (BDE-209). The concentration range varied highly, and the measured maximum concentration was approximately from 10 times higher than the measured minimum concentration (DEHP and NP+NPE) to up to 2000 times higher for SCCP. A maximum concentration of 12258 µg/kg from a study in 2011 (Thomas et al., 2011), was much higher than median at 416 µg/kg dw, and influence this high variation. In general, the median concentration is lower than the mean, except for DEHP and NP+NPE where these values were rather similar.

95th percentiles are typically used as a worst-case exposure assessment, even there are also examples on use of 90th percentiles and mean values. It is a general wish to keep a conservatism in risk assessments in a screening and in the light of a precautionary principle ensuring low possibilities for false decisions. A refinement of the regulation can be performed when more data and knowledge is available.

Since it is not proposed new limit values for the selected HOCs in fertilisers in this project, the following calculated and predicted amount of sewage sludge which not will be applicable with proposed limit values from 2018 (Blytt et al., 2018), but only examples.

Table AC-1. Measured concentrations of the selected HOCs in Norwegian sewages sludge from several studies published from 2010 to 2019, given in µg/kg dw.

Substrate	Min	Max	Mean	Median	n=
DEHP	5200	58000	27601	26700	175
PFOS	1,1	180	13,5	5,2	135
PFOA	0,12	9,20	1,3	0,82	60
SCCP	5,9	12 258	572	204	146
HHCB	370	42000	9979	7950	180
AHTN	49	22 000	2499	1400	170
OTNE	11000	18000	14000	13500	6
BDE-209	4,1	3400	406	250	193
PCB7	1,0	77,0	17	12,0	91,0
NP+NPE	900	11400	4127	3850	95

Data from: Blytt 2010; Blytt et al., 2013; Blytt and Stang, 2019; Naturvågsverket, 2019; Konieczny et al., 2017 and 2018; Schlabach et al., 2011; Thomas et al., 2011.

Total amount of sewage sludge is 121508 tonnes dw, numbers from 2017. The 10 to 95% percentile of contaminants in sludge is shown in Table AC-2.

Based on proposed limit values, it is calculated 10 to 95% percentiles of sewage sludge and the same for each of the selected HOCs. By comparing this, approximately (the closest) percent of sewage sludge with concentration above the proposed limit values are shown for each of the contaminants in Table AC-3.

In general, the median concentrations were lower than the mean, except for DEHP and NP+NPE where these values were rather similar.

Table AC-2. Calculated 10 – 95 % percentiles, minimum, maximum, mean, median, number of samples and non-adjusted and adjusted MLs is shown. The concentrations are shown in µg/kg dw. OTNE is excluded due to few analyses. The percentiles closest to and higher than the non-adjusted MLs are in red. For the non-adjusted MLs for HHCB based on normative values by COWT's approach, 500 µg/kg dw, is lower than 10% percentile sludge.

Substrate	Min	Max	Mean	Median	10 % pers.	20 % pers.	30 % pers.	40 % pers.	50 % pers.	60 % pers.	70 % pers.	80 % pers.	90 % pers.	95 % pers.	n=	Non-adjusted /adjusted MLs (µg/kg dw)*
DEHP	5200	58000	27601	26700	10200	15540	19000	22340	25400	28340	32500	36660	48580	57100	175	50000
PFOS	1,1	180	14	5	2	3	4	5	5	6	9	11	28	60	135	100
PFOA	0,1	9	1,3	0,8	0,1	0,1	0,2	0,8	0,8	1,1	1,4	2,0	4	5	60	100
SCCP	6	12258	572	204	46	68	101	158	204	282	487	800	1260	2432	146	900/2000
HHCB	370	42000	9979	7950	2507	4105	5500	6600	7950	9801	12942	16000	19000	22000	180	500/10000
AHTN	49	22000	2499	1400	511	767	1001	1200	1400	1700	2670	3200	4500	9945	170	500/10000
BDE-209	4	3400	406	250	62	100	130	193	250	320	380	564	990	1500	193	500
PCB7	1,0	77	17	12	3	6	8	11	12	14	17	21	47	56	91	4/20
NP+NPE	900	114000	4127	3850	1700	2342	2588	2972	3850	4672	5048	5696	6600	8456	95	4000/10000

Norsk institutt for bioøkonomi (NIBIO) ble opprettet 1. juli 2015 som en fusjon av Bioforsk, Norsk institutt for landbruksøkonomisk forskning (NILF) og Norsk institutt for skog og landskap.

Bioøkonomi baserer seg på utnyttelse og forvaltning av biologiske ressurser fra jord og hav, fremfor en fossil økonomi som er basert på kull, olje og gass. NIBIO skal være nasjonalt ledende for utvikling av kunnskap om bioøkonomi.

Gjennom forskning og kunnskapsproduksjon skal instituttet bidra til matsikkerhet, bærekraftig ressursforvaltning, innovasjon og verdiskaping innenfor verdikjedene for mat, skog og andre biobaserte næringer. Instituttet skal levere forskning, forvaltningsstøtte og kunnskap til anvendelse i nasjonal beredskap, forvaltning, næringsliv og samfunnet for øvrig.

NIBIO er eid av Landbruks- og matdepartementet som et forvaltningsorgan med særskilte fullmakter og eget styre. Hovedkontoret er på Ås. Instituttet har flere regionale enheter og et avdelingskontor i Oslo.