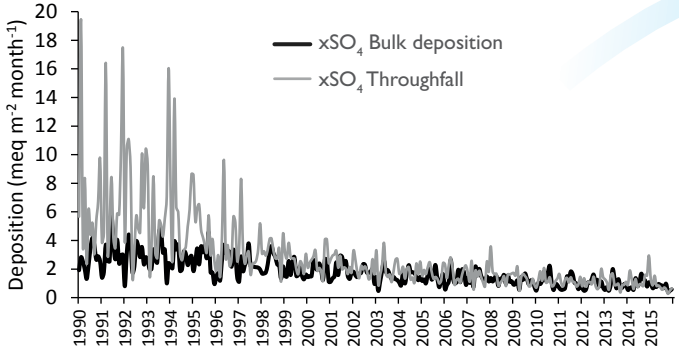
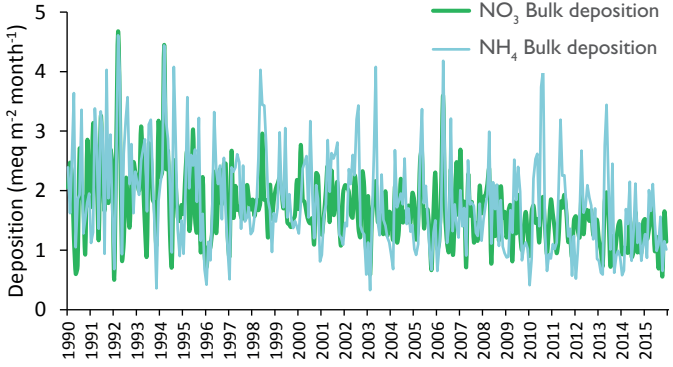
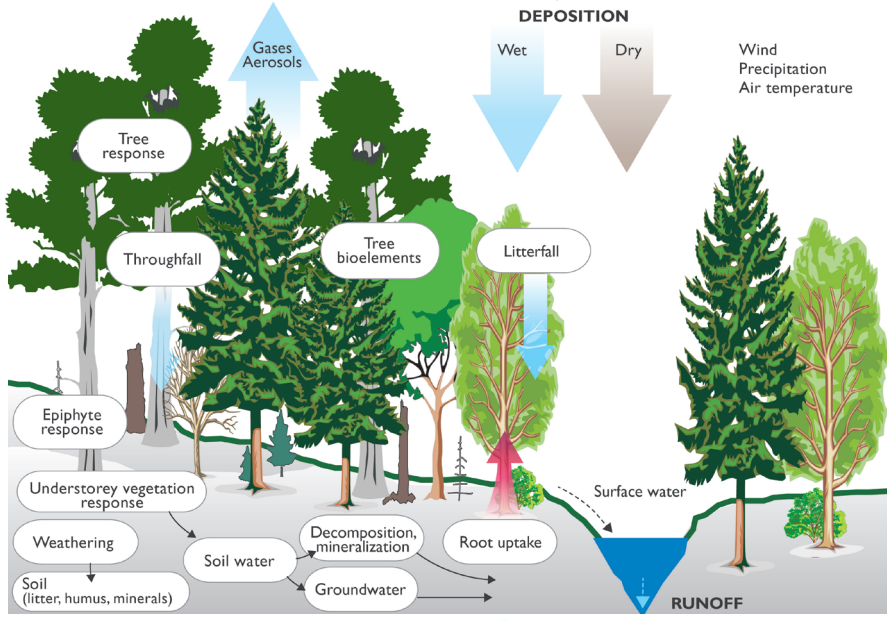
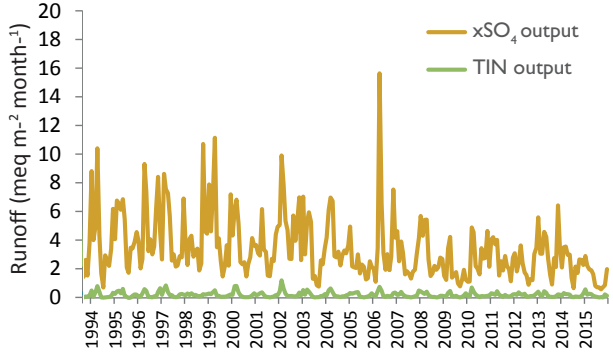
***Graphical Abstract**









ILTER/STOTEN

Vuorenmaa, J. et al.: Long-term changes (1990-2015) in atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in Europe in relation to changes in emissions and hydrometeorological conditions

Highlights

- Trends in runoff fluxes of SO_4 have increasingly responded to the decrease in S emissions
- Trends in NO_3 concentrations in deposition and runoff are predominantly decreasing
- Trends in inorganic N output fluxes are still highly variable
- Variation of SO_4 in runoff was most powerfully explained by deposition pattern
- No clear signs of a consistent climate-driven increase in inorganic N loss in forest catchments

Ms. Ref. No.: STOTEN-D-17-05394R1

Long-term changes (1990-2015) in the atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in Europe in relation to changes in emissions and hydrometeorological conditions

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Abstract

The international Long-Term Ecological Research Network (ILTER) encompasses hundreds of long-term research/monitoring sites located in a wide array of ecosystems that can help us understand environmental change across the globe. We evaluated long-term trends (1990–2015) for bulk deposition, throughfall and runoff water chemistry and fluxes, and climatic variables in 25 forested catchments in Europe belonging to the UNECE International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM). Many of the IM sites form part of the monitoring infrastructures of this larger ILTER network. Trends were evaluated for monthly concentrations of non-marine (anthropogenic fraction, denoted as x) sulphate (xSO_4) and base cations x(Ca + Mg), hydrogen ion (H^+), inorganic N (NO_3 and NH_4) and ANC (Acid Neutralising Capacity) and their respective fluxes into and out of the catchments and for monthly precipitation, runoff and air temperature. A significant decrease of xSO_4 deposition resulted in decreases in concentrations and fluxes of xSO_4 in runoff, being significant at 90% and 60% of the sites, respectively. Bulk deposition of NO_3 and NH_4 decreased significantly at 60–80% (concentrations) and 40–60% (fluxes) of the sites. Concentrations and fluxes of NO_3 in runoff decreased at 73% and 63% of the sites, respectively, and NO_3 concentrations decreased significantly at 50% of the sites. Thus, the LTER/ICP IM network confirms the positive effects of the emission reductions in Europe. Air temperature increased significantly at 61% of the sites, while trends for precipitation and runoff were rarely significant. The site-specific variation of xSO_4 concentrations in runoff was most strongly explained by deposition. Climatic variables and deposition explained the variation of inorganic N concentrations in runoff at single sites poorly, and as yet there are no clear signs of a consistent deposition-driven or climate-driven increase in inorganic N exports in the catchments.

Keywords: Sulphur, nitrogen, climate, trends, monitoring, LTER

Introduction

Increased emissions of air pollutants and greenhouse gases into the atmosphere since the 1950s have escalated environmental problems from the local to the global scale. The long-range transport of sulphur (SO_2) and nitrogen compounds (NO_x, NH_x) has caused widespread acidification of acid-sensitive aquatic ecosystems in Europe and North America (e.g. Leivestad and Muniz, 1976; Rodhe et al., 1995; Schindler, 1988; Ulrich et al., 1980; Wright et al., 2005). A sustained accumulation of deposited inorganic N in forest soil and vegetation also poses a threat to ecosystems through nutrient enrichment and nutrient imbalance (Bergström et al., 2005; Bergström and Jansson, 2006; Lepori and Keck, 2012; Stevens et al., 2011) and deteriorated tree mineral nutrition (Jonard et al., 2014). It also poses a threat to biodiversity, as a consequence of the eutrophication of sensitive ecosystems, as shown by the results of the international networks of forested sites from both ICP IM (International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems) and ICP Forests (International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests) sites under the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP) (Dirnböck et al., 2014) and other studies (Bleeker et al., 2011; Bobbink et al., 2010; MEA, 2005; Sala et al., 2000). At the same time, emissions of greenhouse gases into the atmosphere are causing global warming, and consequent climate change affects freshwater and terrestrial ecosystems. There is growing evidence that, for example, lakes throughout the world, particularly in northern Europe and North America have been subject to climate change-driven warming (Hook et al., 2012; Schneider and Hook, 2010), and a substantial body of research demonstrates the sensitivity of lakes to the climate and shows that physical, chemical and biological lake properties respond rapidly to climate-related changes (e.g. Adrian et al., 2009; Jeppesen et al., 2012; Rosenzweig et al., 2007; Shimoda et al., 2011). Many of the retention and release processes for sulphate and inorganic N in catchment soil are sensitive to climatic variables, and would, therefore, be affected by climate change (e.g. Dirnböck et al., 2016; Mitchell et al., 2013; Moore et al., 2010; Templer et al., 2012; Wright and Jenkins, 2001). Inter-annual variations in water chemistry related to variations in the deposition of air pollutants and climate are greater than the expected improvement in water chemical status in 2020. The effects of climate variability

and change are expected to offset and delay chemical and biological recovery of acid-sensitive waters, for example (de Wit et al., 2015).

Observed detrimental effects of transboundary air pollution led to international negotiations on emission reductions under the CLRTAP, signed in 1979 under the UNECE (UNECE, 1996). Since the 1980s, environmental regulations have led to declining emissions of air pollutants in Europe, and overall emissions of SO_2 and NO_x declined by ca. 60% and ca. 45%, respectively, between 1990 and 2014 (Fagerli et al., 2016), resulting in a declining deposition of air pollutants. Emission reduction measures have been less successful for nitrogen than sulphur, and the decrease in inorganic N deposition has not been observed as strongly as for SO_4 (e.g. Waldner et al., 2014). Emissions of NH_3 decreased by ca. 20%, but they stabilised or even increased slightly between 2000 and 2014 (Fagerli et al., 2016).

In order to assess the impacts of air pollution and climate change in the environment, a long-term integrated monitoring approach in remote unmanaged areas including physical, chemical and biological variables is needed. The multidisciplinary International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) is one of the activities set up under the UNECE CLTRAP to develop the necessary international co-operation in the assessment of the air pollutant effects and ecosystem impacts of climate change. In addition to ICP IM, the Long-Term Ecosystem Research (LTER) infrastructures are mainly focused on ecological phenomena that could be investigated at the local level (site-level) in natural or semi-natural ecosystems, but support the interpretation of larger scale processes. The concepts of LTER and ICP IM are closely related, and therefore many of the ICP IM sites form part of the monitoring infrastructures of these larger LTER sites.

The ultimate goals of air pollution emission abatement actions are the improvement and recovery of damaged terrestrial and aquatic ecosystems, and the protection of threatened or affected ecosystems has increasingly received considerable attention (de Wit et al., 2015). Successful reductions in air pollution emissions over the past 30 years in Europe have led to substantial improvements in ecosystems, e.g. substantially decreased SO_4 deposition has led to widespread recovery from the acidification of sensitive

freshwater ecosystems in Europe and North America (de Wit et al, 2015; Garmo et al., 2014; Helliwell et al., 2014). Implementing air pollution reduction policy is costly. For example, integrated assessment model studies estimated a total cost of approximately EUR 59 billion per year to further reduce European S, N and VOC emissions to below 1990 levels by 2010 (Amann et al., 2000). The Clean Air Policy Package and its main legislative instrument, the National Emission Ceilings Directive, set binding national reduction objectives for six air pollutants (SO_2, NO_x, NMVOCs, NH_3, PM2.5 and CH_4) to be met by 2020 and 2030. It also implements the UNECE CLRTAP 1999 Gothenburg/Multi-effect Protocol to Abate Acidification, Eutrophication and Ground-level Ozone as amended in 2012. The European Commission estimates that the costs of pollution abatement to implement the EU Clean Air Package are expected to reach EUR 3.4 billion per year in 2030 (Maas and Grenfelt, 2016). It is, therefore, essential that empirical evidence is available for assessing and documenting the ecosystem responses of costly emission reduction investments. In this paper, we analysed site-specific long-term trends for concentrations of acidifying and eutrophying air pollutants in deposition (input) and runoff (output) and their fluxes, using available long-term monthly data (with the longest time series being 1990–2015) collected in the international ICP IM network of forested research catchments in Europe. In addition, the long-term trends for climatic variables using monthly data were also analysed. The main aims of the present study are: (i) to evaluate whether concentrations and fluxes of air pollutants in deposition and runoff have changed during the course of successful emission reductions in different regions in Europe, and (ii) to assess the changes in concentrations and fluxes in the context of emission and deposition reduction responses and climatic variation. We hypothesise that fluxes and concentrations of SO_4 and inorganic N show decreasing temporal trends in unmanaged forested catchments across Europe due to international emission reduction measures. We further hypothesise that climate and hydrology (precipitation, runoff, air temperature) are additional factors that explain temporal patterns of S and N output fluxes.

2. Materials and methods

2.1 Site description

The long-term data used in this study relies on the monitoring of unmanaged and calibrated forest catchments belonging to the ICP IM network. Many of the sites also belong to the LTER-Europe and international LTER (ILTER) networks for long-term ecosystem research. Long-term trends of air pollution effects and climatic variables were evaluated at a selection of 25 IM sites in 11 countries in Europe between 1990 and 2015 (Fig. 1, Table 1). The selection was guided by the availability of deposition (bulk and throughfall) data, runoff chemistry data and runoff volume data in the ICP IM database.

The LTER/IM catchments are located in nature conservation areas or semi-natural areas with minimum direct human disturbance. Many of the catchments have been relatively intact for as much as over 100 years, and are therefore suitable for the monitoring of air pollution and climate change effects on ecosystems (Manual for Integrated Monitoring, 1998). The multidisciplinary ICP IM under the CLRTAP has been conducted since the late 1980s, enabling a cause-effect approach for studying the long-term effects of air pollution and climate change on ecosystems in forested catchments across Europe with different deposition, climate and acidification and eutrophication potential. The ICP IM network provides the only data set that uses consistent and simultaneous physical, chemical and biological measurements over time from atmosphere, terrestrial and aquatic ecosystems across Europe.

The dominant vegetation in the monitored catchments mainly consists of northern and central European coniferous and broadleaf forests (Table 1). The type of bedrock and soil within the catchment areas varies widely; some consist of sorted sediments on sedimentary bedrock, others are till soils on igneous and metamorphic bedrock and some sites contain extensive wetlands and lakes. The soils in Fennoscandia (Finland, Sweden and Norway) within northern Europe are thin and young glacial or supra-aquatic acid-sensitive soils with underlying granite bedrock. Unglaciated but thin and acid-sensitive soils also characterise the catchment of CZ02. The soils in DE01 and CZ01 are thin to medium-deep soils with underlying acidic granitic or paragneiss bedrock, respectively, while the other catchments in the Baltic

States and in many parts of central, eastern and southern Europe are located in areas with medium-deep to thick surface deposits and sandstone and limestone present with dolomite bedrock. Mineral soils dominate most of the catchments, but some of the catchments include considerable areas of peaty soils.

2.2 Sampling

Methods for the collection, storage and analysis of bulk deposition and throughfall samples are described in the programme manual (Manual for Integrated Monitoring, 1998). Samples for bulk deposition (largely wet deposition but also including some dry deposition), including the precipitation amount and chemistry of bulk precipitation, were collected in an open area within or adjacent to each catchment, using continuously open HDPE (high-density polyethylene) plastic funnel collectors. At some sites, the precipitation amount for the deposition estimate was obtained from meteorological rain gauges situated within the catchment or from the closest climate station nearby. At sites with regular winter conditions (snow cover), cylindrical HDPE collectors or purpose-made plastic bags were used to collect the winter snowfall. The minimum number of samplers for precipitation amount (meteorology) and bulk precipitation chemistry (deposition) is one sampler per site.

Precipitation which passes through the canopy to the forest floor (throughfall) was also sampled. It is well known that precipitation under the forest canopy differs in quality and quantity from that of precipitation collected in an open area due to the wash-off of dry deposition and strong canopy interactions, such as e.g. leachates produced by the canopy, and uptake of N by plant tissue and through stomata (e.g. Draaijers and Erisman, 1995). Throughfall samples were collected using funnel-shaped collectors, which were placed randomly or systematically around the plot or in a grid under the canopy. During winter, at the sites with snow cover, snow collectors (a plastic ring and attached plastic bag) were used to collect snowfall under the canopy. The number of throughfall samplers usually ranges from 10 (minimum) to 20 per site.

The bulk deposition samples are collected weekly, and analysed as a monthly composite sample. Throughfall sampling is made monthly, weekly or at a time interval between the two, e.g. every two or three weeks, depending mainly on the climate and the method used. Throughfall samples from a number of collectors are pooled to a composite sample representative for a certain stand. Weekly samples can be analysed or mixed with monthly samples before analyses. All the deposition samples are stored at 4 °C before analysis.

Samples for runoff water chemistry were collected, usually weekly or fortnightly, at the catchment outlets, where water levels are also continuously recorded to calculate stream discharge. At some sites, the sampling of runoff water chemistry was carried out monthly during the base-flow period in winter and summer. As the quantitative calculation of the runoff at site AT01 is impeded by the karstified geology, runoff at the weir and in the extended catchment was modelled by a process-based semi-distributed karst model (Hartmann et al., 2016). The IM catchments DE02 and EE01 have no measurements of surface runoff water volume and chemistry, but monitoring of soil water chemistry is carried out at these sites, and therefore trend results only for soil water concentrations are presented in this study.
Methods for the collection, storage and analysis of runoff and soil water samples are described in more detail in the ICP IM programme manual (Manual for Integrated Monitoring, 1998).

2.3 Parameters and data preparation

The integrated monitoring of ecosystems means physical, chemical and biological measurements over time of different ecosystem compartments simultaneously at the same location. In practice, monitoring is divided into a number of compartmental sub-programmes, which are linked by the use of the same parameters (cross-media flux approach) and/or the same or nearby stations (cause-effect approach). Therefore the experimental unit of our study/analyses is a well-defined calibrated forest catchment in which deposition fluxes (input) to the defined area and runoff water fluxes (output) from the defined area were measured.

225 Trends for deposition and runoff were evaluated for monthly concentrations (μeq l^{-1}) and fluxes (meq m^{-2}
226 $month^{-1}$) of non-marine (x denotes non-marine fraction) sulphate (xSO_4), base cations (xCa + xMg),
227 hydrogen ion (H^+), nitrate (NO_3-N), ammonium (NH_4-N) and ANC (Acid Neutralising Capacity). To
228 distinguish changes in anthropogenic SO_4 and base cations (Ca + Mg) from climate-related variations in
229 sea salt, trends for deposition and runoff chemistry and fluxes for SO_4 and base cations were calculated
230 using non-marine fractions. The sea salt-corrected fractions were calculated by subtracting the marine
231 contribution estimated from the ratio of the ion to Cl in seawater (Lyman and Fleming, 1940). ANC was
232 calculated as Σ(base cations) – Σ(strong acid anions) equal to (Ca + Mg + Na + K) – (SO_4 + NO_3 + Cl),
233 and trends for ANC were analysed using concentrations (μeq l^{-1}). Monthly deposition (both for bulk
234 deposition and throughfall) fluxes were calculated as the product of the respective volume-weighted ion
235 concentration and monthly precipitation sum. Output fluxes were calculated as the product of monthly
236 runoff and volume-weighted monthly mean concentration (weekly of fortnightly sampling) or single
237 sample solute concentration (monthly sampling). Chemical input and output fluxes are expressed as meq
238 m^{-2} $month^{-1}$.
239
240 Hydrometeorological variables such as precipitation amount, runoff volume and air temperature are
241 regularly measured as part of the ICP Integrated Monitoring programme. Monthly sum of precipitation
242 and runoff volume (mm $month^{-1}$) and mean monthly air temperature (°C) were examined for long-term
243 trends of climatic variables.
244
245 **2.4 Statistical analysis**
246
247 The Seasonal Kendall test (SKT) (Gilbert, 1987; Helsel and Hirch, 1995; Hirsch et al., 1982) was used for
248 detecting long-term monotonic trends in chemical concentrations and fluxes and climatic variables for
249 each of the study sites, and SKT was applied to monthly data. SKT is an extension of the Mann-Kendall
250 test, and SKT is widely used in detecting monotonic trends in water chemistry records because it is not
251 particularly sensitive to missing data and outliers, and is robust with respect to non-normality and serial
252 character (e.g. seasonal changes). A Visual Basic program for a multivariate and conditional Mann-

Kendall test of monotonic trends was used for trend detection, and a multivariate technique, in which correction for covariates and trend detection are carried out simultaneously, was applied (Libiseller and Grimvall, 2002). The magnitude of trend slope was estimated by the Theil-Sen slope estimation method (Sen, 1968), and was expressed as μeq l^{-1} yr^{-1} for chemical concentrations, meq m^{-2} yr^{-1} for chemical fluxes, mm yr^{-1} for precipitation and runoff and °C yr^{-1} for air temperature. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis, i.e. providing at least 95% confidence that the detected trend was significantly different from a zero.

Statistical models to explain monthly variation of xSO_4 and NO_3 concentrations in runoff for each of the study sites between 1990 and 2015 were built using stepwise multiple regression analysis.
The explanatory variables were monthly precipitation and runoff volume, mean monthly air temperature and monthly concentration and flux of xSO_4 and sum of inorganic N ($TIN=NO_3+NH_4$) in bulk deposition and throughfall. A stepwise regression procedure was applied for 15 catchments which had a complete data set of explanatory variables covering precipitation, runoff volume, air temperature, deposition (both bulk deposition and throughfall) and runoff chemistry (Table 2). Stepwise regression analysis used forward and backward selection, and only explanatory variables having a significance of $p < 0.05$ were included in the model. Statistical analyses were performed by using SAS Enterprise Guide version 5.1 for Windows.

3. Results

3.1 Gradients and trends in precipitation, air temperature and deposition

The studied IM areas exhibit a great range of precipitation amounts. Mean annual precipitation exceeding 900–1000 mm yr^{-1} occurred generally in stations near the coast in the vicinity of the North Atlantic Ocean in Norway (NO01, NO02, NO03) and in the south-western part of Sweden (SE04), and in high altitude regions in central Europe (AT01, CZ02, DE01, IT01, IT03 and IT09) (Table 2). A number of IM sites are

280 located in lowland areas (e.g. BY02, DE02, EE01, EE02, FI01, FI03, LT01 and PL06) and have relatively
281 low precipitation (600–700 mm yr^{-1}). The long-term annual (January–December) precipitation records
282 showed decreasing trends at 10 sites (40%) and increasing trends at 15 sites (60%) (Fig. 2), but trends
283 were rarely significant. Significant increasing trends were detected, but only at three sites (DE02, EE01,
284 NO02) (Table S1, Supplementary material). Precipitation records of individual months showed almost
285 equally decreasing (149 out of the 300 monthly records) and increasing (151 out of the 300 monthly
286 records) trends, but only 4–5% of the trends were significant. The few significant trends were mostly
287 observed for winter and spring months (January–May) (Fig. 2, Table S1, Supplementary material).
288
289 Annual (January–December) air temperature records in 1990–2015 showed predominantly increasing
290 trends (17 out of the 18 sites), with a significant increase at 11(61%) sites located both in central and
291 northern parts of Europe (Fig. 2, Table S2, Supplementary material). Air temperature records of
292 individual months showed increasing trends in 152 out of the 216 monthly records (70%), and 28 out of
293 the 216 monthly records (13%) increased significantly. The significant increasing monthly trends were
294 detected mostly during spring (April–May, 36% of the significant monthly trends) and late autumn
295 (November, 32% of the significant monthly trends) (Fig. 2, Table S2, Supplementary material).
296
297 The deposition of xSO_4 and inorganic N (TIN) showed large differences between the sites, with the
298 highest values at sites located in parts of central, eastern and southern Europe and the lowest values at
299 sites in northern regions. The sites in south-western Fennoscandia (NO01, SE04) were also exposed to
300 high xSO_4 and TIN depositions (Table 2). The throughfall (surrogate to dry deposition) of xSO_4 was
301 higher than the bulk deposition of xSO_4 at the majority of the IM sites, indicating the importance of dry
302 deposition fraction of xSO_4 for total deposition (e.g. Vuorenmaa et al., 2017).
303
304 The study sites that have been exposed to the highest xSO_4 and TIN deposition during the period 1990–
305 2015 (Table 2) also showed the strongest reductions in the deposition. The bulk deposition of xSO_4
306 decreased significantly at all study sites within the study period, and xSO_4 in throughfall exhibited a
307 significant decrease in 1990–2015 as well (Figs. 3 and 4, Table S3, Supplementary material, Fig S1,

Supplementary material). Concentrations and fluxes of xSO_4 in throughfall (mean slopes –3.70 μeq l^{-1} yr^{-1} and –0.15 meq m^{-2} yr^{-1}, respectively) decreased more than those of bulk deposition (mean slopes –1.39 μeq l^{-1} yr^{-1} and –0.08 meq m^{-2} yr^{-1}, respectively) (Table 3).

The IM sites showed dominantly negative trend slopes in NO_3 and NH_4 concentrations in bulk deposition (> 90% of the sites), and a decrease of NO_3 and NH_4 concentrations in bulk deposition was significant at 20 (80%) and 16 (64%) out of the 25 sites, respectively (Fig. 3, Table S3, Supplementary material). The fluxes of inorganic N in bulk deposition also showed largely negative trends (> 80% of the sites) (Figs. 3 and 4, Fig. S1, Supplementary material), with a significant decrease in NO_3 and NH_4 fluxes at 15 (60%) and 11 (44%) of the sites, respectively. Significant increases in inorganic N concentrations and fluxes in bulk deposition were not detected. Concentrations of NO_3 and NH_4 in throughfall also showed predominantly negative trend slopes (91% and 70% out of the 23 sites, respectively), a decrease in NO_3 concentrations was significant at 16 (70%) sites, and NH_4 concentrations decreased significantly at 11 (48%) sites. Fluxes for NO_3 and NH_4 in throughfall decreased at 96% and 74% of the sites, and the decrease was significant at 65% and 22% of the sites, respectively. Three sites (EE01, NO02 and SE14) showed significant increases in NH_4 concentrations and fluxes in throughfall.

Concentrations and fluxes of non-marine base cations (xBC= xCa + xMg) in bulk deposition and throughfall decreased at the majority of the sites (ca. 60–70% of the sites) in 1990–2015, being significant at ca. 30–55% of the sites. Base cation concentrations and fluxes in bulk deposition decreased less than those of xSO_4 in general (Table 3), allowing acid neutralising capacity (ANC) to increase, being significant at ca. 70–80% of the sites in bulk deposition and throughfall (Fig. 3). Along with decreased acid anion (xSO_4 and NO_3) concentrations and increased ANC in precipitation, hydrogen ion (H^+) concentrations, i.e. acidity of precipitation, decreased (increase of pH) in bulk deposition and throughfall, being significant at ca. 70% of the sites (Fig. 3, Table S3, Supplementary material, Fig. S1, Supplementary material).

Following a steeper decrease in the 1990s, concentrations and deposition fluxes for xSO_4, TIN and acidity in precipitation experienced a more gradual decrease during the 2000s. In general, the xBC deposition levelled out or even increased between 2001 and 2015 (Table 3).

3.2 Gradients and trends in runoff volume, chemistry and catchment output fluxes

The runoff volume pattern was in agreement with the precipitation pattern. The highest annual runoff volume occurred at sites located in south-western Scandinavia and in central parts of Europe, and the lowest values occurred generally in low altitude areas, e.g. in the Baltic States and in parts of Sweden and Finland (Table 2). The forest at DE01 consists of ca. 60% young spruce and mixed stands regenerating from a bark beetle attack; at this site, the annual amount of runoff increased due to decreased evapotranspiration (Bernsteinová et al., 2015). Annual runoff records (January–December) showed almost equally positive (10 sites) and negative (9 sites) trends, but trends were rarely significant. Detected significant trends were increasing, but only at four sites (EE02, LT03, NO03, SE04). Runoff volume records for individual months showed slightly less decreasing (102 out of the 226 monthly records, 45%) than increasing (124 out of the 226 monthly records, 55%) trends, but only 5% of them were significant. The significant decreasing trends (12% out of the 102 decreasing trend slopes) were observed mostly in the summer months (June–July, 70%), while significant increasing trends (10% out of the increasing 124 trend slopes) were more evenly distributed throughout the year (Fig. 2, Table S2, Supplementary material).

Similar to the deposition gradients, there were large differences in the annual output fluxes of xSO_4 in runoff between the different sites. The highest mean annual output fluxes of xSO_4 were observed at IM sites located in parts of south-western Scandinavia, central and eastern Europe, where xSO_4 deposition has been elevated, and the lowest fluxes at sites in some remote northern regions (Table 2). Concentrations and fluxes of xSO_4 in runoff decreased significantly at 19 out of the 22 sites (86%) and 12 out of the 19 sites (63%), respectively, between 1990 and 2015 (Figs. 5 and 6, Table S4, Supplementary material, Fig. S2, Supplementary material). Concentrations of H^+ and ANC in runoff decreased and

363 increased significantly at 15 out of the 22 sites (70%) (Fig. 5, Fig. S2, Supplementary material).
364 Concentrations of xSO_4 and H^+ in soil water at IM sites DE02 and EE01 decreased significantly as well,
365 and resulted in an increase of ANC, dependent on the soil depth (Table S5, Supplementary material).
366
367 The highest annual output fluxes of NO_3 were found at sites located in parts of south-western
368 Scandinavia, central and eastern Europe, where TIN deposition was elevated, and output flux rate
369 decreased gradually towards the northern region (Table 2). Nitrate clearly dominated the sum of monthly
370 TIN (NO_3 + NH_4) concentrations (n=4987, mean=78%, median=90%, SD=26) and fluxes (n=4383,
371 mean=79%, median=93%, SD=26) and annual TIN fluxes (Table 2). Trends in NO_3 concentrations were
372 decreasing (16 out of the 22 sites, 73%) rather than increasing, while NH_4 concentrations were decreasing
373 only at 10 out of the 19 sites (53%). Concentrations of NO_3 decreased significantly at 50% of the sites,
374 but increased significantly at only three sites (AT01, BY02, SE14) and NH_4 concentrations increased
375 significantly at two sites (PL10, SE14). Trends in fluxes of inorganic N in runoff showed a more mixed
376 response with both decreasing and increasing trends. Output fluxes of NO_3 were decreasing at 12 out of
377 the 19 sites (63%), being significant at four sites (21%) (Figs. 5 and 6, Fig. S2, Supplementary material).
378 A significant increase in output fluxes of NO_3 was detected for two catchments (SE04, SE14).
379 Concentrations of NO_3 in soil water at site DE02 predominantly decreased, while NH_4 increased at all soil
380 depths. Concentrations of NO_3 and NH_4 in soil water at site EE01 tended to increase at all soil depths
381 (Table S5, Supplementary material).
382
383 Significant monthly trends for concentrations of NO_3 occurred commonly in spring, early summer and
384 autumn, while corresponding trends for fluxes occurred generally between spring and autumn. Monthly
385 concentrations of xSO_4 decreased most significantly in June, October and November, but concentrations
386 decreased generally more steadily throughout the year compared to the xSO_4 fluxes, in which significant
387 downward trends occurred most commonly in spring (Fig. 7).
388
389 The monthly variation of xSO_4 concentrations in runoff (xSO_4 *rwc*) was explained by variations in air
390 temperature (xSO_4 *at*), runoff volume (xSO_4 *rw*) and deposition (xSO_4 *tfc*, *tff*, *bdc*, *bdf*) (Fig. 8, Table S6,

391 Supplementary material). Air temperature and runoff were selected predictors ($p < 0.05$) at 11 (73%) and
392 9 (60%) out of the 15 sites, respectively. Decreasing concentrations and fluxes in bulk deposition (xSO_4
393 *bdc* and xSO_4 *bdf*, respectively) and throughfall (xSO_4 *tfc* and xSO_4 *tff*, respectively) were predictor
394 variables at ca. 30–50% of the sites, but the variation of xSO_4 deposition (concentration or flux in bulk
395 deposition and throughfall) was the first predictor variable at 10 sites, and the model gave highest partial
396 *R*-squares for deposition from 0.03 to 0.42. The variation of xSO_4 concentrations in throughfall (xSO_4 *tfc*)
397 had the highest predictive ability among the explaining deposition variables. The model generally
398 explained the variation of xSO_4 *rwc* from 16% to 58% between the sites. Combining the results for all
399 studied IM catchments, the variation of xSO_4 *rwc* was best explained by xSO_4 *tfc*. The variation of TIN
400 concentrations in runoff (TIN *rwc*) was also mostly associated with a variation in air temperature, and
401 temperature was the first predictor in 11 IM catchments. The variations in the runoff volume (TIN *rw*)
402 and concentrations and fluxes in bulk (TIN *bdc* and TIN *bdf*, respectively) or throughfall (TIN *tfc* and
403 TIN *tff*, respectively) were predictors only at 1 to 4 sites (Fig. 8, Table S6, Supplementary material). The
404 model generally explained the variation of TIN *rwc* from 4% to 39% between the sites, and similar to
405 variation in xSO_4 *rwc*, the variation in throughfall (TIN *tfc*) was the first predictor explaining variation in
406 TIN *rwc* in the whole data.
407
408 **4. Discussion**
409
410 **4.1 Changes in deposition chemistry and fluxes**
411
412 The spatial differences in xSO_4 and TIN deposition in IM areas reflect well-known emission and
413 deposition gradients of air pollutants in Europe (Lövblad et al., 2004; Vuorenmaa et al., 2017; Waldner et
414 al., 2014). Central and eastern parts of Europe were historically large sources of emissions, and thus sites
415 in the region (e.g. CZ01, CZ02, LT03, DE01, AT01, PL06, PL10) received the highest anthropogenic
416 xSO_4 and TIN deposition, while the long-range transport and deposition of S and N decrease gradually
417 towards northern remote regions. At the IM sites that received the highest deposition, SO_4 deposition has
418 substantially decreased from a level of 150–250 meq m^{-2} yr^{-1} to < 50 meq m^{-2} yr^{-1} between 1990 and the

present time (Vuorenmaa et al., 2017). The high xSO_4 and TIN deposition at sites in southern Scandinavia (NO01, SE04) was due to the elevated long-range transport and can also be explained, at least partly, by high amounts of precipitation.

Successful emission reduction measures in Europe over the past 30–40 years have led to a declining deposition of air pollutants (Colette et al., 2016), as shown at IM sites throughout Europe. The emission control programmes have been particularly successful for S, and the deposition of xSO_4 decreased at studied IM sites located in the historically high S emission and deposition regions in central-eastern Europe by 70–90% and in the northern remote regions by 60–80% between 1990 and 2015. The dry deposition of xSO_4 decreased more than the bulk deposition (ΔThroughfall > Δ Bulk deposition), which is in agreement with previous studies for a number of European forested catchments (e.g. Prechtel et al., 2001; Waldner et al., 2014). SO_4 concentrations in throughfall are influenced by interception deposition, where the relative decrease has been even more pronounced, because improved emission control techniques and fuel-switching away from high sulphur-containing solid and liquid fuels to low sulphur fuels have markedly reduced S-containing gases and particles in emissions and ambient air concentrations in Europe (Amann et al., 2013). Decreased N emissions have resulted in a decrease of NO_3 and NH_4 depositions at the majority of the IM sites in 1990–2015, but the decrease of TIN deposition has been generally smaller than that of xSO_4. European N emissions in 1990–2015 have decreased less than those of S, and the bulk deposition of TIN has generally exceeded xSO_4 deposition on an equivalent basis since the late 1990s (e.g. Forsius et al., 2005). Like for xSO_4, a significant decrease of TIN in throughfall at many of the IM sites may indicate the pronounced effect of declining dry deposition as well, or increased canopy uptake. The acid anion (xSO_4 and NO_3) concentrations in precipitation have decreased, while trends for base cation concentrations exhibited only a gradual change during the 2000s. This has generally resulted in an increase of acid neutralising capacity (ANC) and a decrease of H^+ (increase of pH) in precipitation.

Changes in emission reductions and emission reduction responses on deposition chemistry in Europe were more pronounced in the 1990s than 2000s. Sulphur emissions decreased substantially from 1990

until the early 2000s, and after that emissions exhibited a more gradual decrease. Following a steeper decrease from 1990, emissions of NO_x also experienced a more gradual decrease since the early 2000s (Colette et al., 2016). These emission patterns were reflected by a steeper decrease in concentrations and deposition fluxes of SO_4 and TIN, and in acidity of precipitation as well, in the 1990s compared to the 2000s (Aas and Vet, 2011), as also shown at IM sites.

4.2 Changes in runoff water chemistry and catchment output fluxes of SO_4

The substantial decrease of xSO_4 deposition has evidently resulted in a decrease of xSO_4 concentrations and output fluxes in forested IM catchments in large parts of Europe between 1990 and 2015. Although the runoff volume records in 1990–2015 showed almost equally increasing and decreasing trend slopes, our results showed that 63% of the IM sites exhibited a significant decrease in output fluxes. The previous trend assessment for monthly concentrations and fluxes at IM sites in 1993–2006 showed that xSO_4 output fluxes in catchments used in the present study decreased significantly at 40% of the sites (Vuorenmaa et al., 2009). This suggests that IM catchments have increasingly responded to the decreases in S emissions and the deposition of SO_4. A much larger proportion of the sites (86%) showed significant decreasing trends in xSO_4 concentrations between 1990 and 2015. The short-term inter-annual fluctuations in runoff volume, which may largely modify the output fluxes of SO_4, can mask long-term changes in matter dynamics in ecosystems (e.g. Prechtel et al., 2001). Long-term mass balance budgets from IM catchments have shown that variation in the annual retention and net release of SO_4 from soils can be partly explained by variation in annual runoff, thus also masking long-term trends in output fluxes (Vuorenmaa et al. 2017). Nevertheless, our results are consistent with the recent regional trend analysis of surface water chemistry in Europe as part of the UNECE ICP Waters programme (Garmo et al., 2014) and another European assessment of surface water SO_4 concentrations (Helliwell et al., 2014), which have also shown clear decreases of xSO_4 concentrations in surface waters that eventually resulted from decreased xSO_4 fluxes into the water courses. Sulphur emissions have substantially reduced in North America as well, which have resulted in a widespread decline of SO_4 deposition, a consequent decline of

SO_4 concentrations and an increase of ANC in acid-sensitive surface waters (e.g. Garmo et al., 2014; Kahl et al., 2004; Stoddard et al., 1999).

Concentrations of xSO_4 and H^+ in soil water at IM sites EE01 and DE02 decreased significantly as well, showing that the declined S emissions and deposition loads have resulted not only in decreased xSO_4 concentrations and fluxes in surface runoff water, but a similar trend (and recovery from acidification) also proceeds in the soil at these sites. Several studies throughout Europe have documented decreasing trends in SO_4 concentrations in soil water in forested catchments (e.g. Karlsson et al., 2011; Kvaalen et al., 2002; Löfgren et al., 2011; Sawicka et al., 2016; Ukonmaanaho et al., 2014).

The different emission and deposition patterns between the 1990s and 2000s likely reflected the trends in runoff concentrations and fluxes at IM sites. Decrease in concentrations and output fluxes for xSO_4, TIN and H^+ was steeper in the period 1990–2000 than in the period 2001–2015. Garmo et al. (2014) also reported that the decrease in xSO_4 concentrations in acid-sensitive surface waters in Europe was stronger in the 1990s than in the 2000s, and also trends in concentrations of other indicators of recovery from acidification tended to be less pronounced during the 2000s, suggesting that the rate of improvement of water quality has slowed. The more gradual decrease in concentrations and fluxes of SO_4 in IM catchments in the 2000s compared to the 1990s may also be due to an increased net release of SO_4. The IM catchments generally retained SO_4 (input > output) in the early 1990s, but since the late 1990s, they commonly shifted towards net release (output > input) (Vuorenmaa et al., 2017). Many other studies on forested catchments in Europe and North America have also shown an increased net release of SO_4 fuelled by the mobilisation of legacy S pools accumulated during times of high atmospheric SO_4 deposition (Augustaitis et al., 2010; De Vries et al., 2003, 2001; Forsius et al., 2005; Löfgren et al., 2001; Mitchell et al., 2013, 2011; Prechtel et al., 2001; Watmough et al., 2005).

The studied IM catchments vary in their sensitivity to acidification, and the sites in Finland, Sweden and Norway and the Czech site CZ02 are considered to be susceptible to acidification (ANC in runoff commonly < 100 $\mu eq\ l^{-1}$). Although a decreasing trend in the atmospheric acid input has been less

pronounced during the 2000s, the most acid-sensitive IM catchments in the present study are experiencing a recovery from sulphate-driven acidification, indicated by clear increases in pH and ANC in the soil-water ecosystem. Trends in surface water chemistry have shown widespread and consistent recovery from acidification in Europe due to the decreased SO_4 input and loss (de Wit et al., 2015), and progressing recovery from acidification at acid-sensitive IM sites has been documented in more detail for CZ02 (Krám et al., 2012), FI01 (Ukonmaanaho et al., 2014; Vuorenmaa et al., 2014), NO01 (Wright, 2008) and SE04, SE14, SE15 and SE16 (Löfgren et al., 2011).

The xSO_4 deposition (particularly throughfall) was clearly the strongest predictor explaining variation in xSO_4 concentrations in runoff (xSO_4 *rwc*) at the studied IM sites, but the predictive power of xSO_4 deposition was poorer than expected. Median values for the coefficient of determination ranged from 19 to 20% for concentrations and from 4 to 9% for fluxes. Thus, drivers other than deposition are also likely to be regulating present trends in runoff water xSO_4 concentrations. Air temperature and runoff volume explained the variation in xSO_4 *rwc* at the majority of the IM sites, but climatic variables were rarely the first predictor, and their predictive power (coefficient of determination) was clearly poorer than that of deposition. As indicated, the net release of SO_4 due to desorption processes and the excess mineralisation of organic S in soils in response to decreased levels of deposition have been observed in many forested catchments in Europe and North America, which may partly explain the present xSO_4 trend patterns in catchment output at IM sites. It has previously been shown that climate-driven changes in hydrometeorological conditions, such as variations in watershed wetness and runoff, wetting and drying cycles and soil temperature, together with internal SO_4 sources, can largely regulate SO_4 loss from catchments (Benčoková et al., 2011; Dillon et al., 1997; Mitchell et al., 2013; Rice et al., 2014; Wright, 1998; Wright and Jenkins, 2001). The effects of climatic drivers on S-cycling in catchment soils are expected to become increasingly important, as atmospheric SO_4 input has declined (e.g. Mitchell et al., 2013) and climate change continues.

4.3 Changes in runoff water chemistry and catchment output fluxes of inorganic N

Enhanced leaching of NO_3 from IM catchments can be associated with high deposition inputs of TIN (Holmberg et al., 2013, Vuorenmaa et al., 2017). An elevated N deposition has been found to be related to elevated TIN concentrations in soil water and TIN leaching in many areas in Europe (Gundersen, 1995; Iost et al., 2012; Waldner et al., 2015). Nitrate leaching mainly occurs when TIN deposition is above a critical deposition threshold of ca. 10 kg ha^{-1} yr^{-1} (ca. 70 meq m^{-2} yr^{-1}) (Dise and Wright, 1995; Kaste et al., 2007; MacDonald et al., 2002; Stoddard et al., 2001; Wright et al., 2001). Dise et al. (2009) have also determined that N in throughfall over 8 kg ha^{-1} yr^{-1} (ca. 60 meq m^{-2} yr^{-1}) is necessary for N leaching to occur. The mean annual TIN deposition (NO_3 + NH_4) in 1990–2015 in IM catchments AT01, CZ01, CZ02, NO01, PL06 and PL10 (with no substantial forest disturbance) equalled or exceeded most clearly these deposition thresholds, and at these sites the output fluxes of TIN were also elevated compared to the other sites (Table 2). Likewise, Holmberg et al. (2013) found that at IM sites where the critical loads of nutrient nitrogen were exceeded, they also showed higher TIN concentrations and fluxes in runoff. Elevated leaching of NO_3 was found at DE01, but high TIN output was related to widespread and substantial forest dieback of Norway spruce (70% of the catchment area) and consequent excess N mineralisation due to bark beetle infestation (1997–2007), although – along with the recovery of forests – leaching of NO_3 started to decrease after 2007 (Beudert et al., 2014; Vuorenmaa et al., 2017). Concentrations of NH_4 in runoff in forested catchments are usually very low due to effective microbial immobilisation in the soil (e.g. Booth et al., 2005; Corre et al., 2007) and uptake by plants (i.e. trees), and in the majority of catchments NO_3 clearly dominated the TIN loss. Concentrations of NH_4 in runoff in the Norwegian IM catchments NO01, NO02 and NO03 are known to be negligible, which is why NH_4 was not included in the chemical analysis in the runoff water chemistry monitoring program (H. de Wit, pers.comm.). In Finnish and Swedish IM catchments, the flux of NH_4 was larger than that of NO_3, although it was comparatively small, or the contribution of NH_4 to the TIN fluxes was proportionally important (FI01, FI03, SE04, SE15, SE16). This is likely due to catchment characteristics, such as hydrological flow paths, elevation gradients and proportions of organic soils. These forest ecosystems are likely still N limited and therefore there is no significant nitrification of NH_4.

557 The present trend of TIN deposition at IM sites is decreasing, which should generally lead to decreased
558 NO_3 concentrations in runoff (Forsius et al., 2005; Holmberg et al., 2013; Wright et al., 2001). Trends for
559 NO_3 and NH_4 in runoff showed a mixed response with both positive and negative trend slopes, but at
560 more than 60% of the sites TIN concentrations and fluxes were decreasing, and NO_3 concentrations
561 decreased even at 73% of the sites, with a significant decrease at 50% of the sites. The previous trend
562 assessment (1993–2006) for monthly concentrations and fluxes at IM sites (Vuorenmaa et al., 2009)
563 showed decreasing trends for NO_3 concentrations and fluxes in runoff at 48% and 42% of the sites,
564 respectively, with a significant decrease both in concentrations and fluxes at 20% of the sites. Thus, the
565 present trend in NO_3 concentrations and output fluxes is decreasing at the majority of the sites, and a
566 decreasing trend has strengthened. Vuorenmaa et al. (2017) reported long-term (1990–2012) annual
567 input-output budgets of inorganic N for 17 IM catchments located in low or intermediate N deposition
568 areas, and they found that deposited inorganic N was, in general, effectively retained in undisturbed
569 catchments. As yet there are no widespread signs of a consistent increase in NO_3 concentrations or
570 exports in sensitive undisturbed freshwater, i.e. no widespread signs of N saturation in Europe and North
571 America (Garmo et al., 2014; Helliwell et al., 2014; Mitchell, 2011; Watmough et al., 2005; Wright et al.,
572 2001). However, contrary to the status and trends in Europe and North America, the signs of elevated
573 NO_3 leaching from N-saturated ecosystems have been documented from Asia (Duan et al., 2016a, see
574 section 4.4)
575
576 The trends for the concentrations and output fluxes of TIN at IM sites are, however, still variable,
577 indicating that surface water-watershed nitrogen dynamics are inherently complex, as nitrogen is strongly
578 affected by biological processes and hydrological conditions, and nitrate concentrations in surface waters
579 may fluctuate greatly by season and spatially across ecosystems (e.g. Aber et al., 2003). Moreover, the
580 short- and long-term variations in the climate and forest disturbance may mask long-term trends caused
581 by N deposition (Dale et al., 2001; Wright et al., 2001). One might infer that the risk of N saturation is
582 decreasing at IM sites, because of the somewhat decreasing trend in NO_3 leaching. Nitrogen saturation of
583 terrestrial ecosystems may occur when N input and available inorganic N exceeds biotic demand, and
584 may result in excess NO_3 leaching into surface waters. An elevated NO_3 loss from catchments can be

associated with a high N deposition, e.g. as shown at IM sites by Holmberg et al. (2013), and has reached elevated levels in forested areas which are prone to chronic N deposition (Corre et al., 2007; Kiese et al., 2011; Thimonier et al., 2010). However, the elevated leaching of NO_3 is only one signal of nitrogen saturation and may not be indicative in all sites (Lovett and Goodale, 2011). It should be noted that studied IM catchments are rarely located in very high N deposition areas. In recent decades TIN deposition in these areas rarely exceeded 100 meq m^{-2} yr^{-1} (ca 15 kg ha^{-1} yr^{-1}), which can be considered an intermediate N deposition level that is documented to increase the deposition-driven risk of elevated NO_3 leaching (e.g. Dise and Wright, 1995). It should also be noted that large forest areas in Europe, or at least in central Europe, were subjected to former/ancient forest and soil exploitation. Periodic 'resetting' of the N accumulation clock through e.g. harvesting and fire could maintain the baseline N accumulation over long time periods (Dise et al., 2009), and N storage dynamics should be taken into account when interpreting decreasing TIN behaviour in light of expected movement towards saturation. Several recent global studies showing that the unbalanced inputs of C and N relative to P induced significant changes in organism stoichiometry, resulting in profound and uncertain consequences on the structure, functioning and diversity of terrestrial and aquatic ecosystems (Peñuelas et al., 2013, 2012; Sardans et al., 2012). Jonard et al. (2015) reported deteriorated tree mineral nutrition (mainly phosphorus) in forests in Europe due to the elevated N deposition. Surveys covering lakes in Europe and North America in low and high N deposition regions (Bergström et al., 2005; Bergström and Jansson, 2006; Elser et al., 2009) suggested that the atmospheric deposition of N in excess of natural levels has increased inorganic nitrogen concentrations, which is likely to have caused a shift from natural phytoplankton N limitation to P limitation. The shift from N or N+P limitation towards to P limitation was observed to be most pronounced in oligotrophic lakes at a relatively low N deposition level, from 2 to 5 kg N ha^{-1} yr^{-1} (Bergström et al., 2005; Bergström and Jansson, 2006). Our focus in this paper was on large-scale spatial and temporal trends in deposition (input) and runoff (output), and we did not study the negative effects of N (and S) deposition on ecosystems in detail at the study sites, but these alarming findings call for further studies at IM sites.

We detected a significant long-term (1990–2015) increase of TIN concentrations and/or fluxes at five sites (AT01, BY02, PL10, SE04 and SE14), but trends were not likely to be linked to the direct N deposition effects. Site AT01 is a leaky karst catchment, where high TIN deposition causes a high NO_3 loss, even if the forests are not N-saturated. The catchment has a fast runoff dynamic, and snowmelt periods and heavy rain events cause a strong throughflow, dictating not only annual but also long-term N budgets (Jost et al., 2011). Site AT01 was also affected by storm-driven forest disturbance causing elevated NO_3 leaching during the period 2007–2010 (T. Dirnböck, pers.comm). If this period is masked from the trend analysis, the trend for NO_3 concentrations remained increasing and significant (0.97 µeq l^{-1} yr^{-1}, p=0.048). In January 2005, site SE14 was hit by a severe storm, causing substantial damage to the forest by windthrow followed by a bark beetle infestation (~50% of trees killed/seriously affected in 2009), which substantially increased the variability and mean of the annual TIN output from 0.5–3.5 meq m^{-2} yr^{-1} (mean 1.3 meq m^{-2} yr^{-1}) to 1.8–8.3 meq m^{-2} yr^{-1} (mean 5.0 meq m^{-2} yr^{-1}) between the periods 1997–2006 and 2007–2015, respectively. The disturbance regime caused increased TIN concentrations and NO_3 output flux at site SE14 from 2007 on (Löfgren et al., 2011). We did not detect any significant increases in inorganic N concentrations for site SE04 in 1990–2015, and therefore the increase in NO_3 flux may be partly related to increased runoff. The storm in 2005 also hit this site, but caused much less direct damage and bark beetle infestation than at site SE14 (Löfgren et al., 2011). Precipitation increased – although not significantly – at site SE04, and a strong relationship between runoff and precipitation (R^2=0.65, $p < 0.0001$, data not shown) may indicate a precipitation-driven increase in runoff. Significant increasing trends in TIN concentrations in the large semi-natural IM catchments BY02 (A=1780 km^2) and PL10 (A=13 km^2) may be partly due to the direct human influence, such as agricultural leaching, from the catchment.

Air temperature and throughfall of TIN explained the variation in TIN concentrations in runoff (TIN *rwc*) at most of the IM sites, and air temperature was the first predictor at ca. 70% of the sites. Globally increasing trends in surface air temperature are widely documented, and were also detected ($p < 0.05$) at ca. 60% of the IM sites in 1990–2015. The predictive power of air temperature, however, was poor

639 (coefficient of determination ranged between 3% and 22%). Unlike in xSO_4 *rwc*, the site-specific
640 variation of TIN *rwc* was rarely explained by runoff volume. The model generally explained the variation
641 of TIN *rwc* from 4% to 39% between the sites. In contrast to site-specific variation, the variation of TIN
642 *rwc* in the combined data, however, was best explained by TIN *tfc*. Dise et al. (2009) found that in forest
643 ecosystems with chronically elevated N deposition, the throughfall flux of inorganic N was the strongest
644 predictor of N leaching, and N leaching from these ecosystems is primarily driven by the flux of N
645 through deposition and canopy interception rather than any intrinsic attributes of the sites themselves,
646 including climate, topography, hydrology, vegetation or soil properties. As indicated, the IM sites are
647 located in areas with very different N deposition gradients, and it is obvious that not all potential drivers
648 (see e.g. Rothwell et al., 2008) were included in the empirical model in this study, and further analysis
649 with specific landscape and soil data is needed to elucidate the variation in inorganic N concentrations at
650 IM sites. Elevated leaching losses of TIN are generally linked to high N deposition, but losses and trends
651 of NO_3 may be highly variable between sites exposed to relatively similar levels of N deposition
652 (Bringmark and Kvarnäs, 1995; Rothwell et al., 2008), and also other factors than TIN deposition may
653 largely modify TIN losses and trends from forested catchments (Lovett and Goodale, 2011). These factors
654 would include e.g. site characteristics (Brumme and Khanna, 2008; Gundersen et al., 1998), acid
655 deposition (Kopáček et al., 2013; Oulehle et al., 2011), denitrification (Wexler et al., 2014), soil organic
656 N mineralisation and nitrification (Kreutzer et al., 2009), immobilisation (Booth et al., 2005; Corre et al.,
657 2007), disturbance legacies (Bernal et al., 2012; Dale et al., 2001), climatic variables (Brookshire et al.,
658 2011; de Wit et al., 2008; Monteith et al., 2000; Wright and Jenkins, 2001) and changes in tree
659 composition (Crowley and Lovett, 2017). De Wit et al. (2008) reported increasing trends in NO_3 fluxes in
660 runoff during the period 1973/1978–2005 at sites NO01 and NO02, which are located in high and low N
661 deposition areas, respectively, but these trends were likely related to climatic variables, such as changes
662 in snow depth, winter discharge and air temperature. Our model included air temperature at the majority
663 of the study sites which was negatively related to TIN *rwc*. This negative relationship can be at least
664 partly related to the efficient biological uptake of available nitrogen compounds through plants and soil
665 microbes (e.g. Tamm, 1991), soil immobilisation and nutrient uptake by aquatic biota (e.g. algae and
666 bryophytes) (e.g. Mulholland, 2004), which is why NO_3 concentrations in surface waters are usually at a

low level during the summer growing season, and peak in the dormant season/snowmelt in winter and spring. It should be noted that ultimately soil temperature controls N-cycling in catchments, but soil temperature, particularly in the presence of snow, is not a linear function of air temperature (see e.g. de Wit et al., 2008). Nevertheless, the present trend in TIN concentrations and fluxes in runoff is decreasing – particularly for NO_3 – at the majority of the sites, and the influence of long-term variation of climatic variables on TIN *rwc* trends did not strongly arise from this data set and analysis. While a continued decrease in N deposition is anticipated at the ICP IM sites in the future (Forsius et al., 2005; Holmberg et al., 2013), nitrogen continues to accumulate in catchment soils and vegetation, which may ultimately lead to biodiversity losses, decreased soil capacity to retain N and an increased leaching of TIN. Enhanced TIN leaching may be superimposed by climate change, e.g. through increased mineralisation and nitrification rates in the soils due to increased temperature (Beier et al., 2008; Rustad et al., 2001; Wright and Jenkins, 2001), but also an absence of a response has been observed (Beier et al., 2008). Dirnböck et al. (2017) have also suggested that expected future climate change will likely increase ecosystem N retention through increasing N immobilisation in tree biomass and soil organic matter (SOM). Our knowledge on the combined effects of changing climate and a rise in atmospheric CO_2 is also still limited (Norby et al., 2010). In addition to inorganic N, organic N in IM catchments also needs further study, because climate change impacts on the production and mineralisation of organic nitrogen and leaching of organic matter, and the potential risk of an elevated N loss from watersheds to surface waters may also be anticipated in the future.

4.4 New hotspot regions of global S and N emissions and deposition

While a recovery in acid-sensitive surface waters has taken place in Europe and North America due to the substantial reductions in S and N emissions and deposition over the past 20–30 years, many countries in South America, Africa and Asia have experienced an increase in industrialisation and S and N emissions during the past decades (Smith et al., 2011). Therefore, further expansion of acidifying and eutrophying deposition in these regions in recent decades would warrant the collection of new long-term monitoring data on the ecosystem effects of S and N deposition. At present, Asia, particularly East Asia, has become

a global hotspot of S and N deposition (Smith et al., 2015; Vet et al., 2014). Driven by a dramatic economic development, Asian SO_2, NO_x and NH_3 emissions have increased rapidly over recent decades, and for all of the three acidifying precursors (SO_2, NO_x, and NH_3), more than 35% of the global emissions were contributed by Asia in 2005, mainly by China (Smith et al., 2011). Emissions of SO_2 and NO_x in China increased rapidly until 2005 and 2011, respectively, but subsequent emission abatement actions have resulted in a decline in emissions and deposition, although decreases were more evident for SO_4 than inorganic N (Duan et al., 2016a).

High S and N emissions have resulted in elevated SO_4 and NO_3 concentrations in surface water in many parts of East Asia, and has caused surface water acidification in some regions with acid-sensitive soil properties, but generally surface water acidification may not be a serious regional issue across Asia due to the soil properties (S and N sink), good buffering capacity of inland waters and high alkaline Ca deposition (Duan et al., 2016a; Yu et al., 2017). However, high S deposition in China has led to an increasing trend of SO_4 concentrations in rivers and increased riverine output fluxes (Duan et al., 2016b), and has also caused general soil acidification in many regions in East Asia (Duan et al., 2016a). Nitrogen deposition, especially of NH_4, is of increasing concern in Asia due to nitrification and nitrate leaching in N-saturated ecosystems causing acidification of soils and water. Enhanced NO_3 leaching has been observed in China and Japan, and N-derived decreasing pH-values have been reported for some streams (Duan et al., 2016a; Qiao et al., 2014). Although further studies are needed, the acidifying effect of N deposition may be more important than S deposition in well-drained tropical/subtropical soils due to high SO_4 adsorption. The relative importance of N deposition in future acidification may increase, because the role of S as an acidifying agent is likely to decrease, as has occurred in Europe and North America. Excess nitrogen deposition has not only led to acidification, but has also resulted in ecosystem eutrophication in East Asia, shown as changes in N dynamics, plant growth or biodiversity. The decrease in S (and N) deposition has started a recovery from soil acidification, but as with Europe and North America, however, the large stores of adsorbed SO_4 are expected to be desorbed, a process which delays the recovery of the soil from acidification. Thus, how quickly soils respond to decreased deposition in these regions is uncertain (Duan et al., 2016a).

Conclusions

A pattern of S and N emission reduction responses in large areas across Europe is shown by trend analysis from the international ICP IM network of forested research catchments also belonging to the LTER (Long-Term Ecosystem Research) research infrastructure. Concentrations and deposition fluxes of xSO_4, and consequently acidity in precipitation, have substantially decreased in IM areas. TIN deposition has decreased in most of the IM areas, but to a lesser extent than that of xSO_4. Substantially decreased xSO_4 deposition has resulted in decreased concentrations and output fluxes of xSO_4 in runoff, and decreasing trends of TIN concentrations in runoff – particularly for NO_3 – are more prominent than increasing trends. In addition, decreasing trends appeared to strengthen over the course of emission reductions during the last 25 years. TIN concentrations in runoff were mainly decreasing, while trends in output fluxes were more variable, but trend slopes were decreasing rather than increasing. The ICP IM/LTER network covers important deposition gradients in Europe, and these results confirm that emission abatement actions are having their intended effects on precipitation and runoff water chemistry in the course of successful emission reductions in different regions in Europe, even though decreasing trends for S and N emissions and deposition and deposition reduction responses in runoff water chemistry tended to be more gradual since the early 2000s.

At most IM catchments, xSO_4 is on average leached out at the same level as xSO_4 deposition, or output fluxes in runoff have been higher than input fluxes in deposition, while deposited TIN is effectively retained in catchments. Thus, generally higher leaching fluxes of xSO_4 than those of TIN indicate that SO_4 processes are generally the dominant source of actual soil acidification, despite the lower deposition inputs of SO_4, than TIN (De Vries et al., 2007; Forsius et al., 2005).

The effects of climatic drivers on trends of SO_4 losses in catchment soils, together with internal SO_4 sources, are anticipated to become increasingly important as atmospheric SO_4 deposition has declined. The combined effect of climate variability/change and N deposition is also a potential concern, as many

of the retention and release processes of TIN are sensitive to changes in climatic variables. Deposited N continues to accumulate in catchment soils and vegetation, but as of yet there are no clear signs of a consistent climate-driven increase in TIN concentrations or exports in forested IM catchments. Further analysis of processes regulating mobilisation and the release of SO_4 and TIN in terrestrial ecosystems are needed to allow an evaluation of the effects of not only emission reduction policies, but also of the changing climate. This study strongly emphasises the importance of the larger scale integrated long-term monitoring and research of different ecosystem compartments under the LTER infrastructures for detecting the variety of impacts of changing environmental conditions on ecosystems.

Acknowledgements

The authors would like to thank the UNECE LRTAP Convention Trust Fund, the Swedish Environmental Protection Agency and the European Commission through the H2020-project eLTER (grant agreement no. 654359) for financial support for the study. We would also like to express our thanks to the national focal points and many national institutions involved in the ICP IM and LTER work for continued intensive field monitoring and data collection efforts.

Appendix A. Supplementary data

Supplementary data to this article can be found online at ….

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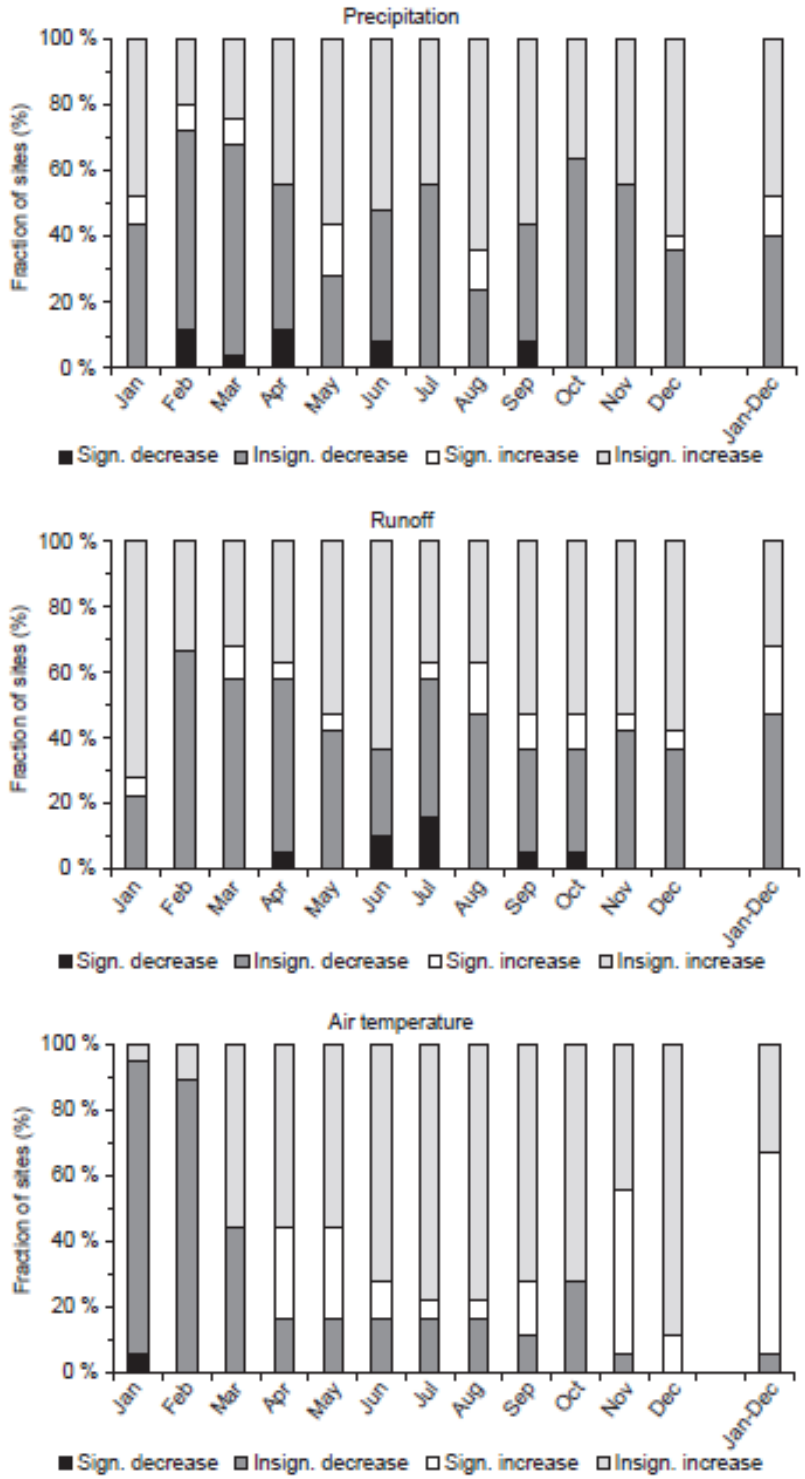
Figure captions

Fig. 1. Location of the 25 ICP Integrated Monitoring sites included in the trend assessment.



1267

1268 Fig. 2. Percentage of Integrated Monitoring sites with a significant decreasing (black), insignificant
1269 decreasing (dark grey), significant increasing (white) and insignificant increasing (light grey) trend in
1270 monthly and annual (Jan–Dec) records of precipitation (top), runoff (middle) and air temperature
1271 (bottom) in 1990–2015.

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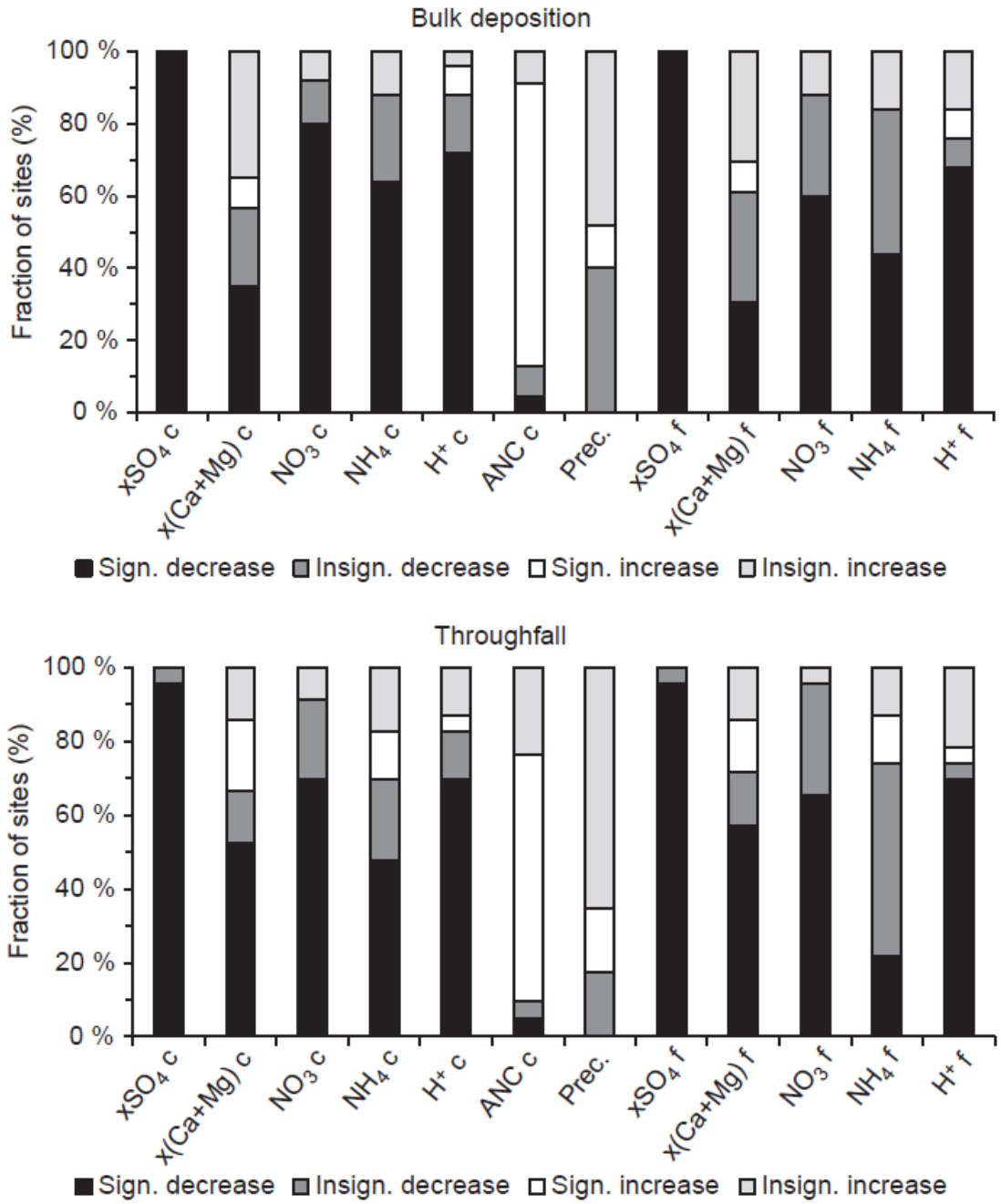


Fig. 3. Percentage of Integrated Monitoring sites with a significant decreasing (black), insignificant decreasing (dark grey), significant increasing (white) and insignificant increasing (light grey) trend in concentrations (denoted as c) and fluxes (denoted as f) of bulk deposition (top) and throughfall (bottom) in 1990–2015.

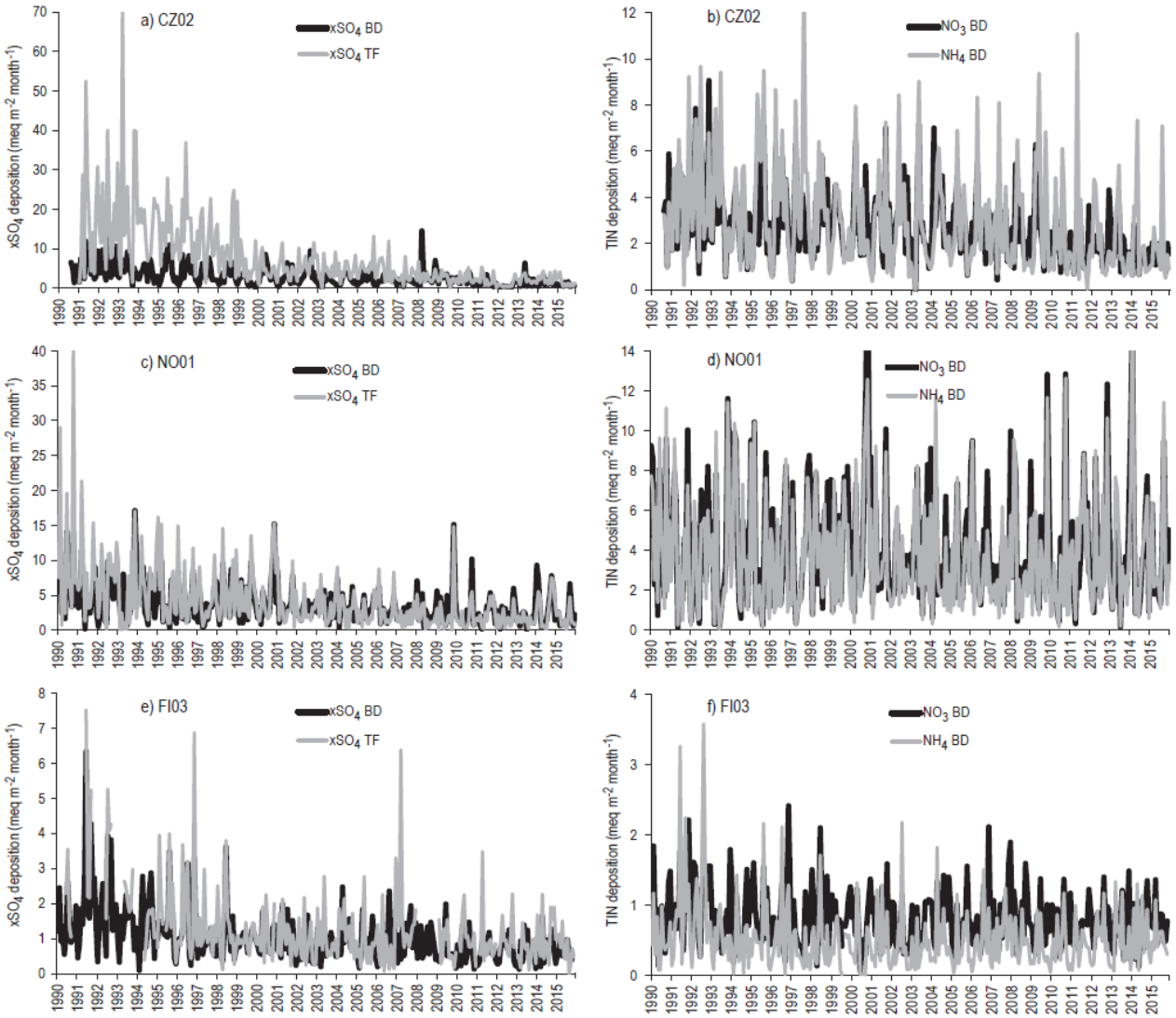


Fig. 4. Monthly bulk (BD) and throughfall (TF) deposition (meq m^{-2} $month^{-1}$) of xSO_4 and monthly bulk deposition (BD) of NO_3 and NH_4 (inorganic N=TIN) (meq m^{-2} $month^{-1}$) in 1990–2015 in catchments CZ02 (Lysina, Czech Republic) (a and b, respectively), NO01 (Birkenes, Norway) (c and d, respectively) and FI03 (Hietajärvi, Finland) (e and f, respectively) reflecting different deposition and hydrometeorological gradients.

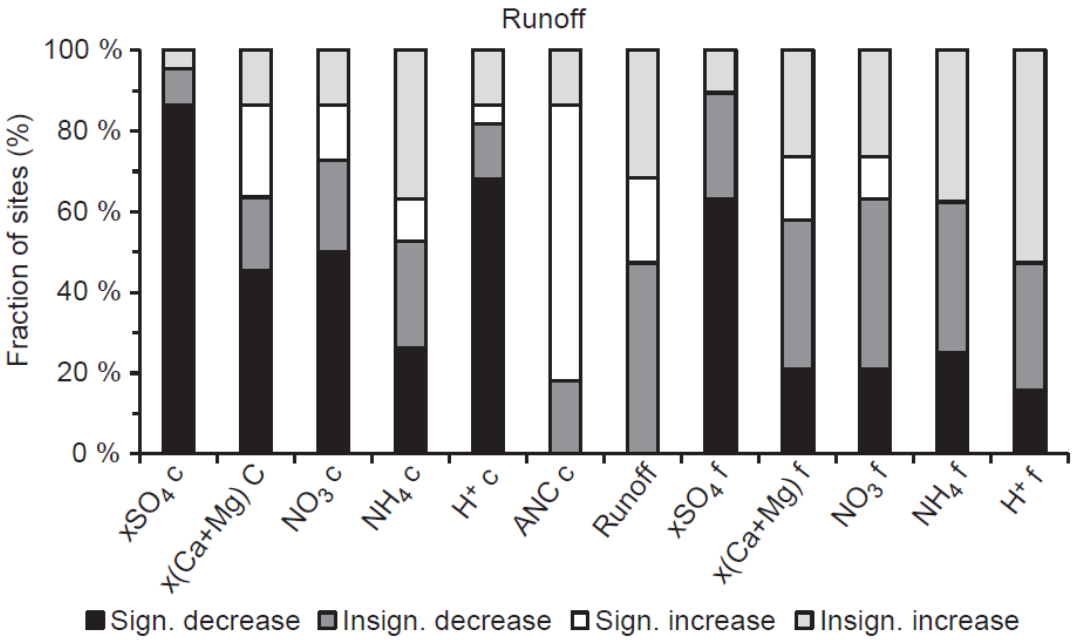


Fig. 5. Percentage of Integrated Monitoring sites with a significant decreasing (black), insignificant decreasing (dark grey), significant increasing (white) and insignificant increasing (light grey) trend in concentrations (denoted as c) and fluxes (denoted as f) of runoff in 1990–2015.

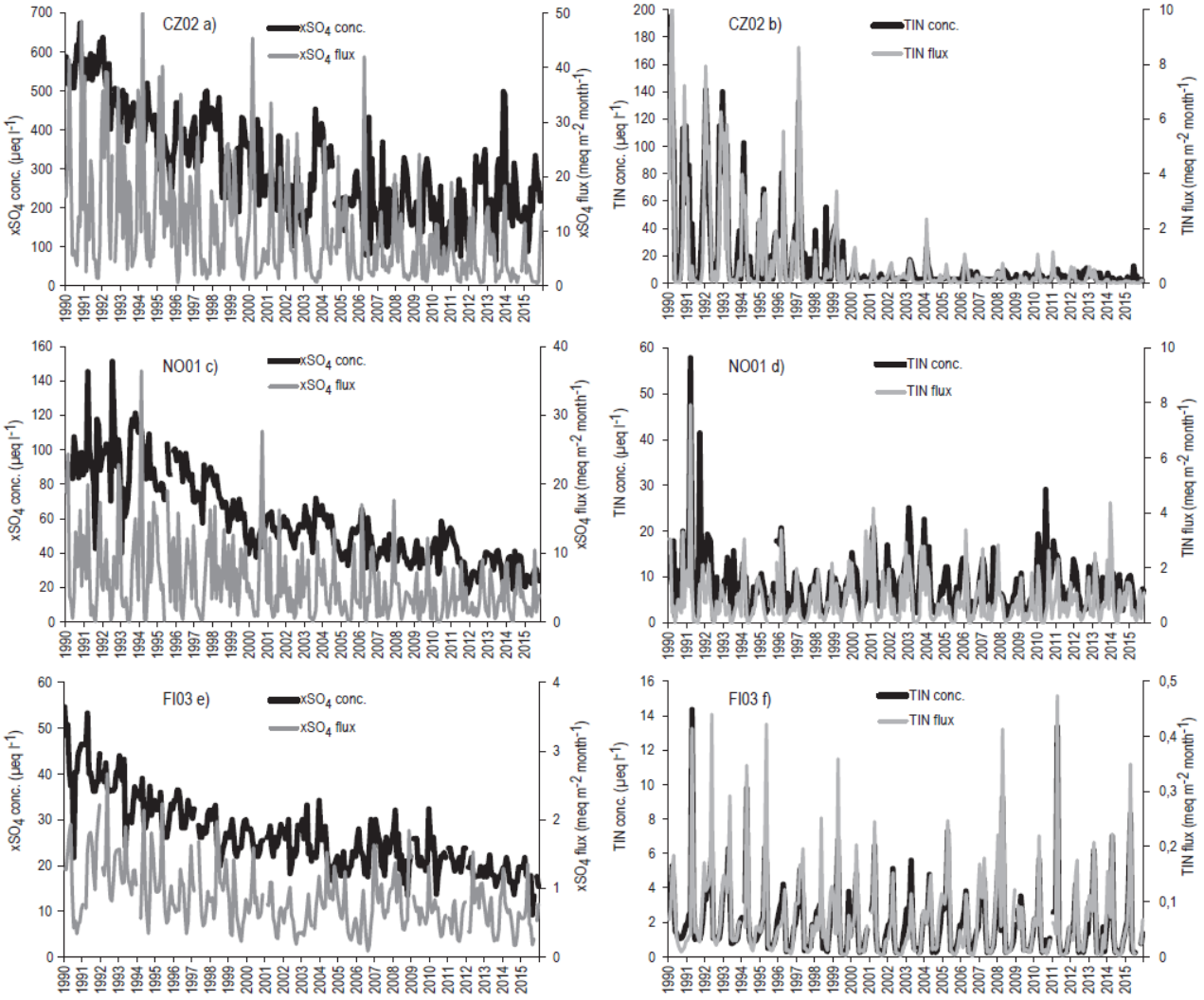


Fig. 6. Monthly runoff water concentrations (left y-axis, µeq l^{-1}) and fluxes (right y-axis, meq m^{-2} $month^{-1}$) of xSO_4 and inorganic N (TIN) in 1990–2015 in catchments CZ02 (Lysina, Czech Republic) (a and b, respectively) NO01 (Birkenes, Norway) (c and d, respectively) and FI03 (Hietajärvi, Finland) (e and f, respectively) reflecting different deposition and hydrometeorological gradients.

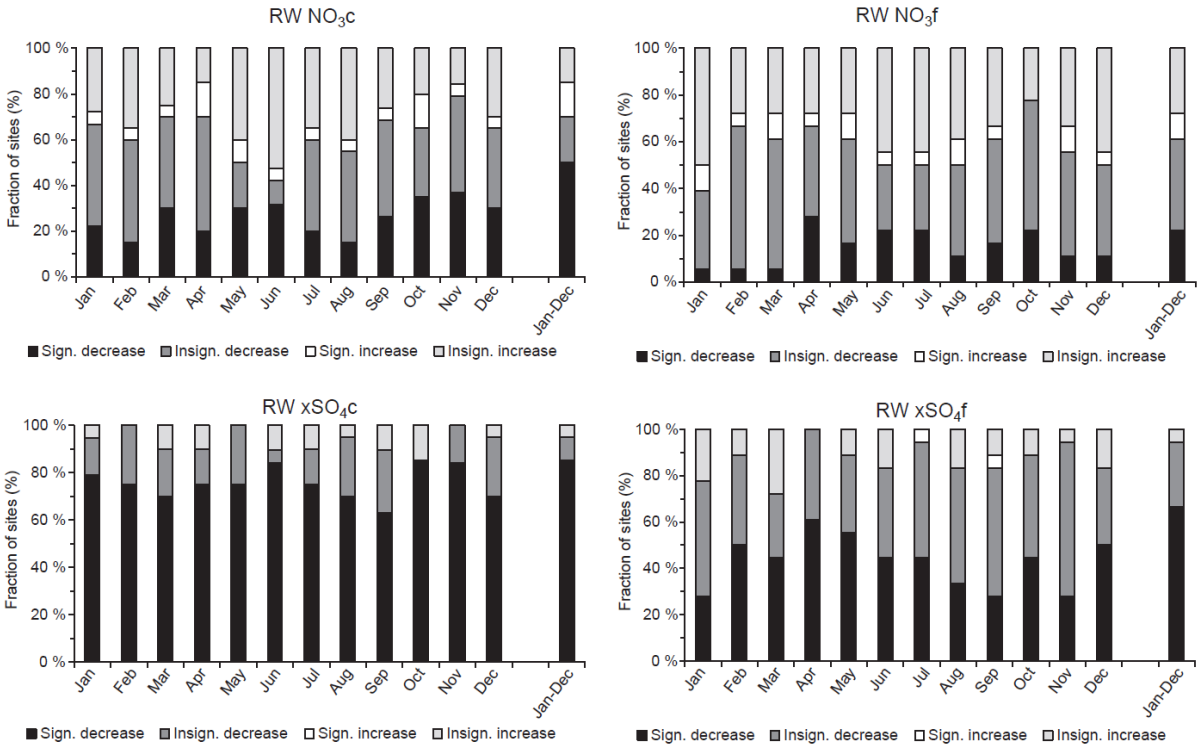
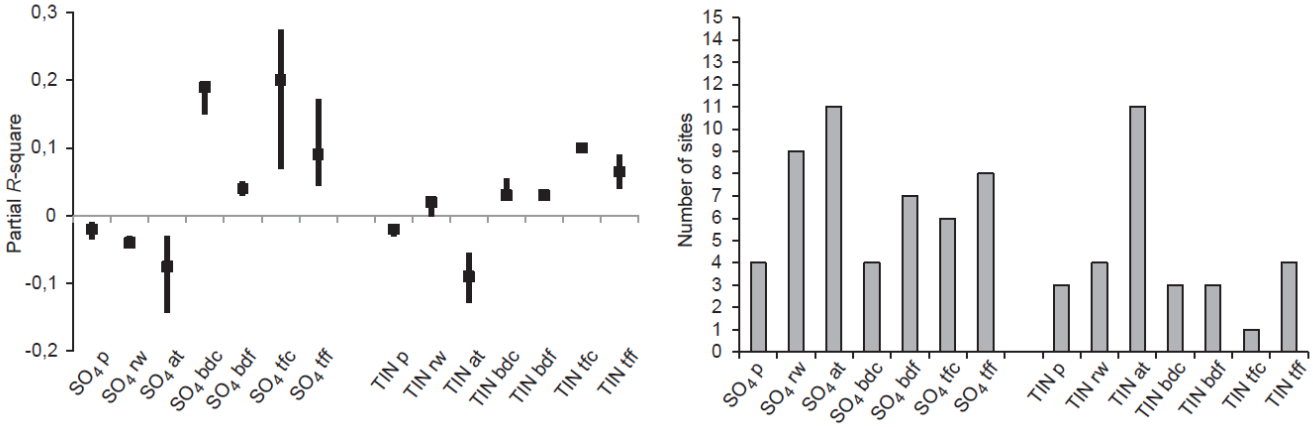


Fig. 7. Percentage of Integrated Monitoring sites with a significant decreasing (black), insignificant decreasing (dark grey), significant increasing (white) and insignificant increasing (light grey) trend in monthly and annual (Jan–Dec) NO_3 and xSO_4 runoff concentrations (denoted as c) and fluxes (denoted as f) in 1990–2015.



1311

1312 Fig. 8. Percentiles (25%, median 50%, 75%) of partial *R*-squares of explanatory variables for variation in
1313 xSO_4 and TIN concentrations in runoff (left), and number of sites in which different explanatory variables
1314 were selected in the model (right). The lower and upper lines indicate 25^{th} and 75^{th} percentiles,
1315 respectively, and a square indicates the median value (p, precipitation; rw, runoff volume; at, air
1316 temperature; xSO_4 *bdc*, xSO_4 concentration in bulk deposition; xSO_4 *bdf*, xSO_4 flux in bulk deposition;
1317 xSO_4 *tfc*, xSO_4 concentration in throughfall; xSO_4 *tff*, xSO_4 flux in throughfall; TIN *bdc*, TIN
1318 concentration in bulk deposition; TIN *bdf*, TIN flux in bulk deposition; TIN *tfc*, TIN concentration in
1319 throughfall; TIN *tff*, TIN flux in throughfall.

Table

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Tables

Table 1. Basic catchment characteristics in the studied Integrated Monitoring catchments.

Site code	Site name	Country	ILTER	Catchment area (km^2)	Altitude (m)	Forest area (%)	Lakes (%)	Peatland (%)	Predominant vegetation	Dominant bedrocks	Soil type
AT01	Zöbelboden	Austria	x	0.90	550–950	100	0	0	Norway spruce, European beech	Calcitic dolomite	Chromic Cambisols, Hydromorphic Stagnosols, Lithic and Rendzic Leptosols
BY02	Berezina	Belarus		1780	155–227	83	2	69	Scots pine, Norway spruce	Sand	Podzols, alluvial soils
CZ01	Anenske Povodi	Czech Republic		0.29	487–543	90	0	0	Norway spruce	Biotitic and sillimanitic-biotitic paragneiss	Dystric Cambisols
CZ02	Lysina	Czech Republic	x	0.27	829–949	100	0	6	Norway spruce	Leucogranite	Podzol, Gleysol
DE01	Forellenbach	Germany	x	0.69	787–1293	95	0	30	Norway spruce, European beech	Granite, gneiss	Dystric and Podzolic Cambisols, Rankers and Lithosols
DE02	Neuglobsow	Germany		14.2	65	55	32	<2	European beech, Scots pine	Pleistocene	Haplic Arenosol
EE01	Vilsandi	Estonia	x	0.008	2–5	95	0	0	Scots pine	Calcitic dolomite	Calcari-Gleyic Leptosol
EE02	Saarejärve	Estonia	x	3.32	44–77	68	8	10	Norway spruce, Scots pine	Sandstone, limestone	Haplic Podzol, glaciofluvial sands
FI01	Valkea-Kotinen	Finland	x	0.30	150–190	86	13	19	Norway spruce, Scots pine	Mica gneiss	Dystric Cambisols, Histols
FI03	Hietajärvi	Finland		4.64	165–214	55	23	35	Scots pine dominated	Porphyritic granodiorites	Fibric Histosols, Podzols
IT01	Renon-Ritten	Italy	x	0.009	1720–1750	100	0	0	Norway spruce, Swiss pine	Quartz-porphyry	Podzol
IT03	Passo Lavazzèe	Italy	x	2.0	1750–1800	69	0	0	Norway spruce, Swiss pine	Crystalline (granite)	Haplic Podzols
IT07	Carrega	Italy	x	0.50	180–200	100	0	0	Sessile oak, Manna ash	Non-consolidated clay	Haplic Luvisols
IT09	Monte Rufeno	Italy	x	0.50	650–690	100	0	0	Austrian oak, European hophornbeam	Flysch (sandstone clay)	Dystric Cambisols
LT01	Aukstaitija	Lithuania	x	1.02	159–189	100	0	10	Norway spruce, Scots pine	Sandstone, limestone	Podzols
LT03	Zemaitija	Lithuania	x	1.47	147–180	100	0	20	Norway spruce, Scots pine	Sandstone, limestone	Podzols
NO01	Birkenes	Norway	x	0.41	200–300	90	0	7	Norway spruce, Scots pine	Granite	Podzols, Histosols, Leptosols
NO02	Kårvatn	Norway	x	25	200–1375	18	4	2	Scots pine, alpine birch	Gneiss, quartzite	Podzols
NO03	Langtjern	Norway	x	4.8	500–710	67	5	25	Scots pine	Granite, gneiss	Podzols
PL06	Storkowo	Poland		74.3	83–203	41	0.3	1.7	Scots pine	Sand, loamy sand	Podzols
PL10	Szymbark	Poland		13	301–753	38	0	0	European beech, fir	Sandstone, shale	Dystric and Eutric Cambisols
SE04	Gårdsjön	Sweden	x	0.04	114–140	95	0	10	Norway spruce	Granite	Podzol, Histosols
SE14	Aneboda	Sweden	x	0.19	210–240	99	0	17	Norway spruce, Scots pine	Granite	Podzol, Gleysols, Histosols
SE15	Kindla	Sweden	x	0.20	312–415	99	0	24	Norway spruce	Granite	Podzol, Histosols
SE16	Gammtratten	Sweden	x	0.45	410–545	99	0	16	Norway spruce, Scots pine	Granite	Podzol, Histosols

Table 2. Annual average values for climatic variables (precipitation, runoff and air temperature) and the deposition and output fluxes of xSO_4, NO_3, NH_4, H^+ and ANC in studied Integrated Monitoring catchments in 1990–2015 (P=precipitation, RW=runoff volume, AT=air temperature, BD= bulk deposition, TF=throughfall, output=runoff water flux, n.d.= no data). Annual average values (mm yr^{-1} for precipitation and runoff, °C yr^{-1} for air temperature and meq m^{-2} yr^{-1} for deposition and output fluxes) were calculated for the period for which data was available.

Site	Data	P	RW	AT	xSO_4 BD	xSO_4 TF	xSO_4 RW	NO_3 BD	NO_3 TF	NO_3 RW	NH_4 BD	NH_4 TF	NH_4 RW	H^+ BD	H^+ TF	H^+ RW	ANC BD	ANC TF	ANC RW
		mm yr^{-1}		°C yr^{-1}	meq m^{-2} yr^{-1}												µeq l^{-1} yr^{-1}		
AT01	1993–2015	1623	407	7.4	35	38	22	44	67	46	70	64	0.15	17	15	0.01	–22	–9.6	3747
BY02	1990–2015	699	n.d.	6.3	26	n.d.	n.d.	20	n.d.	n.d.	28	n.d.	n.d.	5.8	n.d.	n.d.	–58	n.d.	3054
CZ01	1990–2015	652	49	8.0	33	83	60	30	47	4.2	37	56	0.13	13	19	0.01	–73	–179	460
CZ02	1990–2015	987	432	6.3	36	89	119	32	31	7.5	35	27	0.34	26	36	33	–61	–69	–61
DE01	1991–2015	1229	993	6.4	28	40	67	35	37	75	36	23	3.6	22	25	0.90	–35	14	162
DE02	1998–2015	599	n.d.	9.2	17	21	n.d.	21	25	n.d.	23	23	n.d.	8.3	7.2	n.d.	–39	27	n.d.
EE01	1994–2015	574	n.d.	7.8	16	29	n.d.	14	22	n.d.	18	18	n.d.	11	8.5	1.9	–17	170	1016
EE02	1994–2015	662	165	5.8	24	36	42	13	8.0	9.0	15	11	0.78	3.1	2.9	0.19	33	126	2928
FI01	1990–2015	633	191	4.4	14	26	25	12	6.9	0.51	8.8	4.0	0.81	15	8.5	6.2	–39	50	83
FI03	1990–2015	629	381	2.8	12	17	10	9.6	6.7	0.53	6.2	6.9	0.26	13	15	0.25	–34	–9.4	118
IT01	1993–2014	981	78	4.3	20	24	8.6	25	28	0.59	29	17	0.09	4.4	3.5	0.01	–14	71	355
IT03	1997–2013	1065	n.d.	n.d.	19	15	n.d.	21	16	n.d.	26	13	n.d.	2.3	3.7	n.d.	11	45	264
IT07	1997–2015	869	n.d.	n.d.	29	42	n.d.	36	62	n.d.	50	79	n.d.	2.9	1.5	n.d.	–24	63	n.d.
IT09	1997–2015	1017	n.d.	n.d.	28	30	n.d.	26	34	n.d.	19	14	n.d.	4.1	2.9	n.d.	26	118	5642
LT01	1993–2015	666	128	6.6	21	21	138	15	12	2.0	22	11	0.22	8.1	6.3	0.01	–25	53	2698
LT03	1995–2015	859	164	7.0	24	62	108	25	26	2.0	29	23	0.41	11	6.3	0.02	–29	163	1249
NO01	1990–2015	1623	1139	6.0	43	50	67	49	32	10	44	28	n.d.	40	31	27	–48	–15	–20
NO02[1)]	1990–2015	1492	1850	n.d.	7.7	7.9	13	6.4	5.0	2.2	11	8.0	n.d.	8.4	8.5	1.3	–6.2	–2.4	34
NO03[2)]	1990–2015	979	636	n.d.	16	n.d.	18	18	n.d.	0.8	19	n.d.	n.d.	12	n.d.	8.0	–26	n.d.	41
PL06	1995–2015	700	260	n.d.	22	40	178	20	22	34	29	61	5.8	12	13	0.00	–37	19	3393
PL10[3)]	1995–2015	870	403	n.d.	51	85	316	41	64	46	48	35	8.5	17	22	0.01	–54	37	2718
SE04	1990–2015	1166	620	7.4	30	44	61	35	41	1.3	32	22	1.0	24	21	34	–44	–15	–35
SE14	1996–2015	838	311	5.9	19	16	39	25	11	2.6	22	8.9	0.46	18	6.1	9.1	–44	32	84
SE15	1996–2015	913	491	5.0	17	20	48	18	9.5	0.33	16	5.8	0.25	16	7.3	14	–31	47	–13
SE16	1999–2015	693	447	1.9	9.8	7.4	15	9.0	4.9	0.23	7.8	3.6	0.21	8.9	6.5	1.8	–18	2.3	91

[1)]TF data for NO02 1990–2011, [2)]PC data for NO03 1998–2015, [3)]TF data for PL10 2002–2015

Table 3. Annual changes of concentrations (µeq l^{-1} yr^{-1}, denoted as *c*), precipitation/runoff (P/RW, mm yr^{-1}) and fluxes (meq m^{-2} yr^{-1}, denoted as *f*) for xSO_4, xBC, NO_3, NH_4, H^+ and ANC in bulk deposition (BD), throughfall (TF) and runoff (RW) in the periods 1990–2000, 2001–2015 and 1990–2015 at the studied Integrated Monitoring sites.

Programme	Period		xSO_4 *c*	xBC *c*	NO_3 *c*	NH_4 *c*	H^+ *c*	ANC	P/RW	xSO_4 *f*	xBC *f*	NO_3 *f*	NH_4 *f*	H^+ *f*
			µeq l^{-1} yr^{-1}						mm yr^{-1}	meq m^{-2} yr^{-1}				
BD	1990–2000	Mean	–3.96	–0.13	–0.88	–2.09	–1.10	4.26	1.52	–0.15	0.03	–0.01	–0.07	–0.05
		Median	–3.15	–0.07	–0.83	–0.89	–1.38	4.16	0.40	–0.18	0.02	–0.02	–0.03	–0.03
	2001– 2015	Mean	–1.01	0.15	–0.49	–0.20	–0.42	1.93	–0.17	–0.06	0.00	–0.04	–0.01	–0.03
		Median	–0.97	0.05	–0.26	–0.14	–0.45	1.22	–0.25	–0.06	0.00	–0.03	0.00	–0.02
	1990–2015	Mean	–1.45	–0.07	–0.46	–0.52	–0.59	1.94	0.08	–0.08	–0.01	–0.03	–0.03	–0.04
		Median	–1.34	–0.02	–0.43	–0.41	–0.66	1.57	0.00	–0.10	0.00	–0.04	–0.03	–0.04
TF	1990–2000	Mean	–12.3	–5.89	–1.26	–2.68	–4.70	6.39	1.42	–0.48	–0.17	–0.06	–0.08	–0.17
		Median	–9.26	–3.60	–1.07	–0.80	–3.10	5.42	1.33	–0.41	–0.10	–0.04	–0.01	–0.11
	2001– 2015	Mean	–2.41	–0.08	–0.79	–0.18	–0.32	3.51	0.15	–0.10	–0.02	–0.04	–0.02	–0.02
		Median	–1.81	–0.78	–0.69	–0.28	–0.27	1.00	0.27	–0.09	–0.04	–0.03	–0.02	–0.01
	1990–2015	Mean	–4.17	–0.98	–0.62	–0.17	–1.12	4.52	0.37	–0.17	–0.05	–0.03	–0.01	–0.05
		Median	–2.64	–1.21	–0.60	–0.16	–0.60	2.04	0.30	–0.14	–0.06	–0.02	–0.01	–0.03
RW	1990–2000	Mean	–16.0	–21.3	–0.29	0.18	–0.39	–1.44	0.62	–0.32	–0.30	0.01	–0.01	0.00
		Median	–10.2	–4.02	–0.04	0.00	–0.00	2.19	0.07	–0.05	–0.02	0.00	–0.00	0.00
	2001– 2015	Mean	–10.8	–4.84	–0.72	–0.06	–0.28	7.55	0.14	–0.12	0.05	–0.04	–0.00	–0.00
		Median	–3.87	–0.95	–0.01	–0.01	–0.00	1.97	0.08	–0.07	–0.01	–0.00	0.00	0.00
	1990–2015	Mean	–7.23	–3.93	–0.22	0.01	–0.29	4.21	0.13	–0.12	–0.05	–0.00	0.00	–0.01
		Median	–3.36	–1.18	–0.02	0.00	0.00	2.42	0.00	–0.06	–0.01	0.00	0.00	0.00

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