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International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems

Sirpa Kleemola and Martin Forsius (eds.)

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wge Working Group on Effects of the Convention on Long-range Transboundary Air Pollution

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Abstract

The Integrated Monitoring Programme (ICP IM) is part of the effect-oriented activities under the 1979 Convention on Long-range Transboundary Air Pollution, which covers the region of the United Nations Economic Commission for Europe (UNECE). The main aim of ICP IM is to provide a framework to observe and understand the complex changes occurring in natural/semi natural ecosystems.

This report summarizes the work carried out by the ICP IM Programme Centre and several collaborating institutes. The emphasis of the report is in the work done during the programme year 2020/2021 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
- A report on the recovery of epiphytic lichen communities in Sweden over 20 years of rapid air pollution decline
- A report on long-term impacts of air pollution and climate change at Finnish ICP IM sites
- National Reports on ICP IM activities are presented as annexes.

Keywords: Integrated Monitoring, ecosystems, small catchments, air pollution

Tiivistelmä

Ympäristön yhdennetyn seurannan ohjelma (ICP IM) kuuluu kansainvälisen ilman epäpuhtauksien kaukokulkeutumista koskevan yleissopimuksen "Convention on Long-range Transboundary Air Pollution" (1979) alaisiin seurantaohjelmiin. Yhdennetyn seurannan ohjelmassa selvitetään kaukokulkeutuvien saasteiden ja muiden ympäristömuutosten vaikutuksia elinympäristöömme. Muutosten seurantaa ja ennusteita muutosten laajuudesta ja nopeudesta tehdään yleensä pienillä metsäisillä valuma-alueilla, mutta verkostoon kuuluu myös muita alueita.

Tämä julkaisu on kooste ohjelmakeskuksen ja yhteistyölaitosten toiminnasta kaudella 2020/2021. Julkaisu sisältää:

- Lyhyen yhteenvedon ohjelmassa aiemmin tehdyistä arvioinneista
- Kuvauksen ICP IM ohjelman toiminnasta ja ohjelman seurantaverkosta
- Raportin epifyyttisten jäkäläyhteisöjen elpymisestä Ruotsissa 20 vuoden aikana ilmansaasteiden vähennyttyä nopeasti
- Selvityksen ilmansaasteiden ja ilmastonmuutoksen pitkäaikaisista vaikutuksista Suomen ICP IM alueilla
- Kuvauksia kansallisesta ICP IM toiminnasta eri maissa liitteenä.

Asiasanat: Yhdennetty ympäristön seuranta, ekosysteemit, pienet valuma-alueet, ilmansaasteet

Sammandrag

Programmet för Integrerad övervakning av miljötillståndet (ICP IM) är en del av monitoringstrategin under UNECE:s luftvårdskonvention (LRTAP). Syftet med ICP IM är att utvärdera komplexa miljöförändringar på avrinningsområden.

Rapporten sammanfattar de utvärderingar som gjorts av ICP IM Programme Centre och de samarbetande instituten under programåret 2020/2021. Rapporten innehåller:

- En sammanfattning av programmets nuvarande omfattning och databasens innehåll
- En syntes av tidigare utvärderingar av data från programmet
- En rapport om återhämtningen av epifytiska lavar i Sverige under 20 år av snabb minskning av luftföroreningar
- En rapport om långtida effekter av luftföroreningar och klimatförändring på finska ICP IM områden
- Beskrivning av nationella ICP IM aktiviteter.

Nyckelord: Integrerad miljöövervakning, ekosystem, små avrinningsområden, luftföroreningar

Abbreviations

AMAP	Arctic Monitoring and Assessment Programme
ANC	Acid neutralising capacity
CCE	Coordination Center for Effects
CDM	Centre for Dynamic Modelling (previously JEG DM), a body under ICP M&M
CL	Critical Load
CNTER	Carbon-nitrogen interactions in forest ecosystems
ECE	Economic Commission for Europe
eLTER RI	European Research Infrastructure that LTER Europe is building after being adopted by the 2018 ESFRI Roadmap. The RI is built by the two Horizon 2020 projects “eLTER PPP” (Preparatory Phase Project) and “eLTER PLUS” (Advanced Community project)
EMEP	Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
EU	European Union
EU LIFE	EU’s financial instrument supporting environmental and nature conservation projects throughout the EU
Horizon 2020	H2020, EU Research and Innovation programme
ICP	International Cooperative Programme
ICP Forests	International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests
ICP IM	International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems
ICP Materials	International Cooperative Programme on Effects on Materials
ICP M&M	ICP Modelling and Mapping, International Cooperative Programme on Modelling and Mapping of Critical Loads and Levels and Air Pollution Effects, Risks and Trends
ICP Waters	International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes
ICP Vegetation	International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops
ILTER	International Long Term Ecological Research Network
IM	Integrated Monitoring
JEG	JEG DM, Joint Expert Group on Dynamic Modelling. Now under the acronym CDM
LRTAP Convention	Convention on Long-range Transboundary Air Pollution
LTER Europe	European Long-Term Ecosystem Research Network
LTER Network	Long Term Ecological Research Network
NFP	National Focal Point
TF	Task Force
Task Force on Health	Joint Task Force on the Health Aspects of Air Pollution
UNECE	United Nations Economic Commission for Europe
WGE	Working Group on Effects

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Summary

Background and objectives of ICP IM

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 subprogrammes which are linked by the use of the same parameters (cross-media flux approach) and/or same or close stations (cause-effect approach).

The International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM, www.syke.fi/nature/icpim) is part of the Effects Monitoring Strategy under the Convention on Long-range Transboundary Air Pollution (LRTAP Convention). The main objectives of the ICP IM are:

- To monitor the biological, chemical and physical state of ecosystems (catchments/plots) over time in order to provide an explanation of changes in terms of causative environmental factors, including natural changes, air pollution and climate change, with the aim to provide a scientific basis for emission control.
- To develop and validate models for the simulation of ecosystem responses and use them (a) to estimate responses to actual or predicted changes in pollution stress, and (b) in concert with survey data to make regional assessments.
- To carry out biomonitoring to detect natural changes, in particular to assess effects of air pollutants and climate change.

The full implementation of the ICP IM will allow ecological effects of heavy metals, persistent organic substances and tropospheric ozone to be determined. A primary concern is the provision of scientific and statistically reliable data that can be used in modelling and decision making.

The ICP IM sites (mostly forested catchments) are located in undisturbed areas, such as nature reserves or comparable areas. The ICP IM network presently covers forty-eight sites from fifteen countries. The international Programme Centre is located at the Finnish Environment Institute in Helsinki. The present status of the monitoring activities is described in detail in Chapter 1 of this report.

A manual detailing the protocols for monitoring each of the necessary physical, chemical and biological parameters is applied throughout the programme (Manual for Integrated Monitoring 1998, and updated web version).

Assessment activities within the ICP IM

Assessment of data collected in the ICP IM framework is carried out at both national and international levels. Key tasks regarding international ICP IM data have been:

- Input-output and proton budgets
- Trend analysis of bulk and throughfall deposition and runoff water chemistry
- Assessment of responses using biological data
- Dynamic modelling and assessment of the effects of different emission / deposition scenarios, including confounding effects of climate change processes
- Assessment of concentrations, pools and fluxes of heavy metals
- Calculation of critical loads for sulphur and nitrogen compounds, and assessment of critical load exceedance, as well as links between critical load exceedance and empirical impact indicators
- Evaluation and reporting of ICP IM data in relation to the revision process of the Gothenburg Protocol

Conclusions from international studies using ICP IM data

Input-output and proton budgets, C/N interactions

Ion mass budgets have proved to be useful for evaluating the importance of various biogeochemical processes that regulate the buffering properties in ecosystems. Long-term monitoring of mass balances and ion ratios in catchments/plots can also serve as an early warning system to identify the ecological effects of different anthropogenically derived pollutants, and to verify the effects of emission reductions.

The most recent results from ICP IM studies are available from the study of Vuorenmaa et al. (2017). Site-specific annual input-output budgets were calculated for sulphate (SO_4) and total inorganic nitrogen (TIN = NO_3-N + NH_4-N) for 17 European ICP IM sites in 1990–2012. Temporal trends for input (deposition) and output (runoff water) fluxes and net retention/net release of SO_4 and TIN were also analysed. Large spatial variability in the input and output fluxes of SO_4 and TIN reflects important gradients of air pollution effects in Europe, with the highest deposition and runoff water fluxes in southern Scandinavia, Central and Eastern Europe and the lowest fluxes at more remote sites in northern European regions. A significant decrease in the total (wet + dry) non-marine SO_4 deposition and bulk deposition of TIN was found at 90% and 65% of the sites, respectively. Output fluxes of non-marine SO_4 in runoff decreased significantly at 65% of the sites, indicating positive effects of international emission abatement actions in Europe during the last 25 years. Catchments retained SO_4 in the early and mid-1990s, but this shifted towards a net release in the late 1990s, which may be due to the mobilisation of legacy S pools accumulated during times of high atmospheric SO_4 deposition. Despite decreased deposition, TIN output fluxes and retention rates showed a mixed response with both decreasing (9 sites) and increasing (8 sites) trend slopes, but trends were rarely significant. In general, TIN was strongly retained in the catchments not affected by natural disturbances. The long-term annual variation in net releases for SO_4 was explained by variations in runoff and SO_4 concentrations in deposition, while a variation in TIN concentrations in runoff was mostly associated with a variation of the TIN retention rate in catchments. Net losses of SO_4 may lead to a slower recovery of surface waters than those predicted by the decrease in SO_4 deposition. Continued enrichment of N in catchment soils poses a threat to terrestrial biodiversity and may ultimately lead to higher TIN runoff through N saturation or climate change. Continued monitoring and further evaluations of mass balance budgets are thus needed.

Earlier results from ICP IM studies are summarised below.

The first results of input-output and proton budget calculations were presented in the 4th Annual Synoptic Report (ICP IM Programme Centre 1995) and the updated results regarding the effects of N deposition were presented in Forsius et al. (1996). Data from selected ICP IM sites were also included in European studies for evaluating soil organic horizon C/N-ratio as an indicator of nitrate leaching (Dise et al. 1998, MacDonald et al. 2002). Results regarding the calculation of fluxes and trends of S and N compounds were presented in a scientific paper prepared for the Acid Rain Conference, Japan, December 2000 (Forsius et al. 2001). A scientific paper regarding calculations of proton budgets was published in 2005 (Forsius et al. 2005).

The budget calculations showed that there was a large difference between the sites regarding the relative importance of the various processes involved in the transfer of acidity. These differences reflected both the gradients in deposition inputs and the differences in site characteristics. The proton budget calculations showed a clear relationship between the net acidifying effect of nitrogen processes and the amount of N deposition. When the deposition increases also N processes become increasingly important as net sources of acidity.

A critical deposition threshold of about 8–10 kg N ha^{-1} yr^{-1}, indicated by several previous assessments, was confirmed by the input-output calculations with the ICP IM data (Forsius et al. 2001). The output flux of nitrogen was strongly correlated with key ecosystem variables like N deposition, N concentration in organic matter and current year needles, and N flux in litterfall (Forsius et al. 1996). Soil organic horizon C/N-ratio seems to give a reasonable estimate of the annual export flux of N for European forested sites receiving throughfall deposition of N up to about 30 kg N ha^{-1} yr^{-1}. When stratifying data based on C/N ratios less than or equal to 25 and greater than 25, highly significant relationships were observed between N input and nitrate leached (Dise et al. 1998, MacDonald et al. 2002, Gundersen et al. 2006). Such statistical relationships from intensively studied sites can be efficiently used in conjugation with regional monitoring data (e.g. ICP Forests and ICP Waters data) in order to link process level data with regional-scale questions.

An assessment on changes in the retention and release of S and N compounds at the ICP IM sites was prepared for the 21st Annual Report (Vuorenmaa et al. 2012). Updated and revised data were included in the continuation of the work in the 22nd and 23rdAnnual Reports (Vuorenmaa et al. 2013, 2014). The relationship between N deposition and organic N loss and the role of organic nitrogen in the total nitrogen output fluxes were derived in Vuorenmaa et al. (2013).

Sulphur budgets calculations indicated a net release of S from many ICP IM sites, indicating that the soils are releasing previously accumulated S. Similar results have been obtained in other European plot and catchment studies.

The reduction in deposition of S and N compounds at the ICP IM sites, as a result of the implementation of the “Protocol to Abate Acidification, Eutrophication and Ground-level Ozone” of the LRTAP Convention (“Gothenburg protocol”), was estimated for the year 2010 using transfer matrices and official emissions. Continued implementation of the protocol will further decrease the deposition of S and N at the ICP IM sites in western and north western parts of Europe, but in more eastern parts the decrease will be smaller (Forsius et al. 2001).

Results from the ICP IM sites were also summarised in an assessment report prepared by the Working Group on Effects of the LRTAP Convention (WGE) (Sliggers & Kakebeeke 2004, Working Group on Effects 2004).

ICP IM contributed to an assessment report on reactive nitrogen (N_r) of the WGE. This report was prepared for submission to the TF on Reactive Nitrogen and other bodies of the LRTAP Convention to show what relevant information has been collected by the ICP programmes under the aegis of the WGE to allow a better understanding of N_r effects in the ECE region. The report contributed relevant information for the revision of the Gothenburg Protocol. A revised Gothenburg Protocol was successfully finalised in 2012. A new revision process of the Protocol is currently ongoing in 2021-2023, and ICP IM is again contributing.

It should also be recognised that there are important links between N deposition and the sequestration of C in the ecosystems (and thus direct links to climate change processes). These questions were studied in the CNTER-project in which data from both the ICP IM and EU/Intensive Monitoring sites were used (Gundersen et al. 2006). A summary report of the CNTER-results on C/N -interactions and nitrogen effects in European forest ecosystems was prepared for the WGE meeting 2007 (ECE/EB.AIR/WG.1/2007/10).

Trend assessments

Empirical evidence on the development of environmental effects is of central importance for the assessment of success of international emission reduction policy. 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. Vuorenmaa et al. (2018) evaluated long-term trends (1990–2015) for deposition and runoff water chemistry and fluxes, and climatic variables at 25 ICP IM sites in Europe that commonly belong also to the LTER Europe/ILTER networks. The trend assessment was published in a special issue in Science of the Total Environment with the title: "International Long-Term Ecological Research (ILTER) network". The recent results from trend assessment at IM sites 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. Concentrations and deposition fluxes of xSO_4, and consequently acidity in precipitation, have substantially decreased in IM areas. Inorganic N (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. However, 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. 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 TIN 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 TIN exports in the catchments.

Vuorenmaa et al. (2018) reported that the IM sites are located in areas with very different N deposition, and it is obvious that not all potential drivers were included in the empirical model in the study, and further analysis with specific landscape and soil data is needed to elucidate the variation in inorganic N concentrations in runoff at IM sites.

Vuorenmaa et al. (2020) carried out a first analysis on the impact of internal catchment N-related parameters on TIN leaching. A significant negative correlation was found between the annual change of TIN concentrations and fluxes in runoff, and mean TIN fluxes in throughfall, tot N concentrations and N/P-ratios in foliage and litterfall, and tot N concentrations and fluxes in soil water. A significant positive correlation was found between the mean concentrations and fluxes of TIN in runoff and mean TIN deposition in throughfall and mean tot N concentrations and N/P-ratios in foliage and litterfall. Using multiple regression analysis, the annual change in TIN concentrations and fluxes and mean TIN concentrations and fluxes in runoff were dominantly explained by mean tot N concentrations in foliage (R-squares 0.88–0.97). Discriminant analysis was applied with sites having significant decrease in TIN concentrations in runoff and sites having no significant decrease as the dependent dichotomy variable (classes). The foliage N/P-ratio distinguished between two trend classes, and the sites with no significant decrease exhibited higher N/P-ratio than the sites with significant decrease. Since majority of sites showed downward trend slope in TIN concentrations (76%) and fluxes (69%), these results mean that the most N-affected sites with the highest N deposition to the forest floor and highest N concentrations in foliage, litterfall, runoff water and soil water, showed the most pronounced decreases of TIN in runoff. Decrease of TIN in concentrations and fluxes in runoff was also pronounced at sites where decreasing trend of TIN in bulk deposition was highest. Work on this issue is continuing.

Earlier work is summarised below.

First results from a trend analysis of monthly ICP IM data on bulk and throughfall deposition as well as runoff water chemistry were presented in Vuorenmaa (1997). ICP IM data on water chemistry were also used for a trend analysis carried out by the ICP Waters and results were presented in the Nine Year Report of that programme (Lükewille et al. 1997).

Calculations on the trends of N and S compounds, base cations and hydrogen ions were made for 22 ICP IM sites with available data across Europe (Forsius et al. 2001). The site-specific trends were calculated for deposition and runoff water fluxes using monthly data and non-parametric methods. Statistically significant downward trends of SO_4, NO_3 and NH_4 bulk deposition (fluxes or concentrations) were observed at 50% of the ICP IM sites. Sites with higher N deposition and lower C/N-ratios clearly showed higher N output fluxes, and the results were consistent with previous observations from European forested ecosystems. Decreasing SO_4 and base cation trends in runoff waters were commonly observed at the ICP IM sites. At some sites in the Nordic countries decreasing NO_3 and H^+ trends (increasing pH) were also observed. The results partly confirmed the effective implementation of emission reduction policy in Europe. However, clear responses were not observed at all sites, showing that recovery at many sensitive sites can be slow and that the response at individual sites may vary greatly.

Data from ICP IM sites were also used in a study of the long-term changes and recovery at nine calibrated catchments in Norway, Sweden and Finland (Moldan et al. 2001, RECOVER: 2010 project). Runoff responses to the decreasing deposition trends were rapid and clear at the nine catchments. Trends at all catchments showed the same general picture as from small lakes in Scandinavia.

It was agreed at the ICP IM Task Force meeting in 2004 that a new trend analysis should be carried out. The preliminary results were presented in Kleemola (2005) and the updated results in the 15th Annual Report (Kleemola & Forsius 2006). Statistically significant decreases in SO_4 concentrations were observed at a majority of sites in both deposition and runoff/soil water quality. Increases in ANC (acid neutralising capacity) were also commonly observed. For NO_3 the situation was more complex, with fewer decreasing trends in deposition and even some increasing trends in runoff/soil water.

Results from several ICPs and EMEP were used in an assessment report on acidifying pollutants, arctic haze and acidification in the arctic region prepared for the Arctic Monitoring and Assessment Programme (AMAP, Forsius & Nyman 2006, www.amap.no). Sulphate concentrations in air showed generally decreasing trends since the 1990s. In contrast, levels of nitrate aerosol were increasing during the arctic haze season at two stations in the Canadian arctic and Alaska, indicating a decoupling between the trends in sulphur and nitrogen. Chemical monitoring data showed that lakes in the Euro-Arctic Barents region are showing regional scale recovery. Direct effects of sulphur dioxide emissions on trees, dwarf shrubs and epiphytic lichens were observed close to large smelter point sources.

The recent trend assessment using monthly ICP IM data (Vuorenmaa et al. 2018) was preceded by corresponding trend evaluations for the periods 1993–2006 and 1990–2013 (Vuorenmaa et al. 2009, 2016, respectively). Moreover, trends for annual input and output fluxes of SO_4 and TIN were evaluated for the period 1990–2012 (Vuorenmaa et al. 2017). These results clearly showed the regional-scale decreasing trends of SO_4 in deposition and runoff/soil water, and suggested that IM catchments have increasingly responded to the decreases in S emissions and depositions of SO_4 since the early 1990s. Decreased nitrogen emissions also resulted in decrease of inorganic N deposition, but to a lesser extent than that of SO_4, and trends in TIN fluxes in runoff were highly variable due to complex processes in terrestrial catchment that are not yet fully understood. Besides, the net release of SO_4 in forested catchments fueled by the mobilisation of legacy S pools, accumulated during times of high atmospheric sulphur deposition, may delay the recovery from acidification. The more efficient retention of inorganic N than SO_4 results in generally higher leaching fluxes of SO_4 than those of inorganic N in European forested ecosystems. SO_4 thus remains the dominant source of actual soil acidification despite the generally lower input of SO_4 than inorganic N. Critical load calculations for Europe also indicated exceedances of the N critical loads over large areas. Long-term trends for deposition and runoff variables were for the first time evaluated together with climatic variables (precipitation, runoff water volume and air temperature) at IM sites by Vuorenmaa et al. (2016). Many study sites exhibited long-term seasonal trends with a significant increase in air temperature, precipitation and runoff particularly in spring and autumn, but annual trends were rarely significant. It was concluded that the sulphur and nitrogen problem thus clearly requires continued attention as a European air pollution issue, and further long-term monitoring and trend assessments of different ecosystem compartments and climatic variables are needed to evaluate the effects, not only of emission reduction policies, but also of changing climate.

An assessment on changes in the retention and release of S and N compounds at the ICP IM sites was prepared for the 21st Annual Report (Vuorenmaa et al. 2012). Updated and revised data were included in the continuation of the work in the 22nd and 23rd Annual Reports. The role of organic nitrogen in mass balance budget was derived and trends of S and N in fluxes were analysed (Vuorenmaa et al. 2013, 2014).

Detected responses in biological data

The effect of pollutant deposition on natural vegetation, including both trees and understorey vegetation, is one of the central concerns in the impact assessment and prediction. The most recent ICP IM study on dose-response relationships is presented in Section 2 of this report (recovery of epiphytic lichen communities, Weldon & Grandin). Previous work was published by Dirnböck et al. (2014). This study utilised a new ICP IM database for biological data and focussed on effects on forest floor vegetation from elevated nitrogen deposition. Results on dynamic modelling of vegetation responses have also recently been published (Dirnböck et al. 2018, see below)

In many European countries airborne nitrogen coming from agriculture and fossil fuel burning exceeds critical thresholds and threatens the functioning of ecosystems. One effect is that high levels of nitrogen stimulate the growth of only a few plants that outcompete other, often rare, species. As a consequence, biodiversity declines. Though this is known to happen in natural and semi-natural grasslands, it has never been shown in forest ecosystems where management is a strong, mostly overriding determinant of biodiversity. Dirnböck et al. (2014) utilised long-term monitoring data from 28 Integrated Monitoring sites to analyse temporal trends in plant species cover and diversity. At sites where nitrogen deposition exceeded the critical load, the cover of forest plant species preferring nutrient-poor soils (oligotrophic species) significantly decreased whereas plant species preferring nutrient-rich soils (eutrophic species) showed – though weak – an opposite trend. These results show that airborne nitrogen has changed the structure and composition of forest floor vegetation in Europe. Plant species diversity did not decrease significantly within the observed period, but the majority of newly established species was found to be eutrophic. Hence it was hypothesised that without reducing nitrogen deposition below the critical load forest biodiversity will decline in the future.

Earlier work on biological data is summarised below.

The first assessment of vegetation monitoring data at ICP IM sites with regards to N and S deposition was carried out by Liu (1996). Vegetation monitoring was found useful in reflecting the effects of atmospheric deposition and soil water chemistry, especially regarding sulphur and nitrogen. The results suggested that plants respond to N deposition more directly than to S deposition with respect to vegetation indices.

De Zwart (1998) carried out an exploratory analysis of possible causes underlying the aspect of forest damage at ICP IM sites, using multivariate statistics. These results suggested that coniferous defoliation, discolouration and lifespan of needles in the diverse phenomena of forest damage are for respectively 18%, 42% and 55% explained by the combined action of ozone and acidifying sulphur and nitrogen compounds in air.

As a separate exercise, the epiphytic lichen flora of 25 European ICP IM monitoring sites, all situated in areas remote from local air pollution sources, was statistically related to measured levels of SO_2 in air, NH_4^+, NO_3^- and SO_4^{2-} in precipitation, annual bulk precipitation, and annual average temperature (van Herk et al. 2003, de Zwart et al. 2003). It was concluded that long distance transport of nitrogen air pollution is important in determining the occurrence of acidophytic lichen species and constitutes a threat to natural populations that is strongly underestimated so far.

In 2010, the Task Force meeting decided upon a new reporting format for biological data. The new format was based on primary raw data, and not aggregated mean values as before. All countries were encouraged to re-report old data in the new format. This was successful and as a result, the full potential of the biological data from the ICP Integrated Monitoring network could be utilised to raise and answer research question that the old database could not.

Dynamic modelling and assessment of the effects of emission/deposition scenarios

In a policy-oriented framework, dynamic models are needed to explore the temporal aspect of ecosystem protection and recovery. The critical load concept, used for defining the environmental protection levels, does not reveal the time scales of recovery. Priority in the ICP IM work is given to site-specific modelling. The role of ICP IM is to provide detailed and consistent physical and chemical data and a long time-series of observations for key sites against which model performance can be assessed and key uncertainties identified (see Jenkins et al. 2003). ICP IM participates also in the work of the Joint Expert Group on Dynamic Modelling (JEG) of the WGE. Since September 2019, this expert group has reorganised into an international designated centre under the International Cooperative Programme on Modelling and Mapping, under the name Centre for Dynamic Modelling (CDM).

Dynamic vegetation modelling at ICP IM sites has been conducted with contributions from ICP M&M, ICP Forests, and the LTER Europe network. The VSD+ model was applied to simulate soil chemistry at 26 sites in ten countries throughout Europe (Holmberg & Dirnböck 2015, 2016, Dirnböck et al. 2018a, 2018b, Holmberg et al. 2018). Simulated future soil conditions improved under projected decrease in deposition and current climate conditions: higher pH, BS and C:N at 21, 16 and 12 of the sites, respectively. Dirnböck et al. (2018b) found, however, that a release from eutrophication is not expected to result from the decrease in N deposition under current legislation emission (CLE) reduction targets until 2030.

Dynamic models have also previously been developed and used for the emission/deposition and climate change scenario assessment at several selected ICP IM sites (e.g. Forsius et al. 1997, 1998a, 1998b, Posch et al. 1997, Jenkins et al. 2003, Futter et al. 2008, 2009). These models are flexible and can be adjusted for the assessment of alternative scenarios of policy importance. The modelling studies have shown that the recovery of soil and water quality of the ecosystems is determined by both the amount and the time of implementation of emission reductions. According to the models, the timing of emission reductions determines the state of recovery over a short time scale (up to 30 years). The quicker the target level of reductions is achieved, the more rapidly the surface water and soil status recover. For the long-term response (> 30 years), the magnitude of emission reductions is more important than the timing of the reduction. The model simulations also indicate that N emission controls are very important to enable the maximum recovery in response to S emission reductions. Increased nitrogen leaching has the potential to not only offset the recovery predicted in response to S emission reduction, but further to promote substantial deterioration in pH status of freshwaters and other N pollution problems in some areas of Europe.

Work has also been conducted to predict potential climate change impacts on air pollution related processes at the sites. The large EU-project Euro-limpacs (2004–2009) studied the global change impacts on freshwater ecosystems. The institutes involved in the project used data collected at ICP IM and ICP Waters sites as key datasets for the modelling, time-series and experimental work of the project. A modelling assessment on the global change impacts on acidification recovery was carried out in the project (Wright et al. 2006). The results showed that climate/global change induced changes may clearly have a large impact on future acidification recovery patterns and need to be addressed if reliable future predictions are wanted (decadal time scale). However, the relative significance of the different scenarios was to a large extent determined by site-specific characteristics. For example, changes in sea-salt deposition were only important at coastal sites and changes in decomposition of organic matter at sites which are already nitrogen saturated.

In response to environmental concerns, the use of biomass energy has become an important mitigation strategy against climate change. A summary report on links between climate change and air pollution effects, based on results of the Euro-limpacs project, was prepared for the WGE meeting 2008 (ECE/EB.AIR/WG.1/2008/10). It was concluded that the increased use of forest harvest residues for biofuel production is predicted to have a significant negative influence on the base cation budgets causing re-acidification at the study catchments. Sustainable forestry management policies would need to consider the combined impact of air pollution and harvesting practices.

Pools and fluxes of heavy metals

The work to assess spatial and temporal trends on concentrations, stores and fluxes of heavy metals at ICP IM sites is led by Sweden. In 26th Annual Report data on Pb, Cd, Hg, Cu and Zn from countries in the ICP IM were presented (Åkerblom & Lundin 2017). These data will be used for establishment of background heavy metal concentrations in forested compartments and risk assessments of heavy metals.

The results presented by Eklöf et al. (2020) showed that declining metal deposition and/or recovery from acidification have resulted in decreasing Cd and Pb concentrations in runoff at many of the European ICP IM sites during the last 30 years. In contrast, the Hg concentrations in runoff did only show one statistically ($p<0.05$) significant decreasing trend. Catchment Cd, Pb and Hg input-output budgets were also calculated for the four ICP IM sites in Sweden. At catchment level, the mass-balances for Cd and Pb showed that the exports via runoff (RW) could account for only 13–70% and 21–56%, respectively, of the total inputs (TF+LF). These results are in agreement with other studies, indicating metal accumulation in the soils.

In many national studies on ICP IM sites, detailed site-specific budget calculations of heavy metals (including Hg) have improved the scientific understanding of ecosystem processes, retention times and critical thresholds. ICP IM sites are also used for dynamic model development of these compounds. For the future evaluation of emission reductions of heavy metals to the atmosphere site-specific long-term trends for fluxes of heavy metals (primarily for Cd, Pb, and Hg and depending on availability of data, also Cu and Zn) will be analysed in deposition (input) and runoff (output), using available long-term monthly data collected across ICP IM sites in Europe. This will be done to see if fluxes of heavy metals in deposition and runoff respond to changes in emission reductions in Europe. Reduction in heavy metal emissions is hypothesised to be reflected in decreasing heavy metal concentrations (Åkerblom & Lundin 2015), taking into account climatic variation over time and between regions also in decreasing heavy metal fluxes. Temporal trend analysis in heavy metal fluxes will provide a detailed understanding of responses in heavy metal mass balances to emission reductions and give indication on possible change in retention of heavy metals in catchments over time. This overview will also provide an estimate on the significance in heavy metal mass balances over time and identify uncertainties in the mass balances and needs for improvements.

Input-output budgets of Hg help to explain the increase or no change in Hg concentrations in the upper-most forest soil mor-layer in spite of the general decrease in atmospheric deposition (Åkerblom & Lundin 2015). One process that is not accounted for in ICP IM programme is the land-atmosphere exchange of Hg. The phenomenon of land-atmosphere exchange has been known for a long time, but it has been quantified only recently due to the development of micrometerological systems for continuous measurements (Osterwalder et al. 2016). In the case of mass balance calculations for Hg new evidence has shown that land-atmosphere exchange during a 2-year study over a peatland can be more than double the flux in stream runoff (Osterwalder et al. 2017). Based on natural Hg stable isotope studies in podzols and histosols, significant Hg re-emission from organic soil horizons occurred (Jiskra et al. 2015). These novel observations and knowledge about processes that govern land-atmosphere exchange of Hg calls for methods and approaches to account for this important flux in the catchment cycle of Hg within ICP IM.

The objective of the aluminium (Al) contribution of Krám and Kleemola in the 28th Annual Report (2019) was to collect and present recently available information about Al fractions from the Integrated

Monitoring (IM) database and stimulate the IM National Focal Points to checkout and add not yet reported Al fractions data to the IM database for a publication in peer-reviewed journal. Aluminium (Al) does not belong to the group of so-called heavy metals and is not transferred in large quantities by atmospheric deposition to forest catchments like most of the heavy metals. However, elevated inputs of strong acids from the anthropogenic atmospheric deposition to sensitive sites could mobilize Al from soils and stream sediments in a form of potentially toxic Al fractions to surface waters (Gensemer & Playle 1999). Different fractions of aqueous Al have very different toxicity levels for aquatic biota. Modified methods of the original Al fractionation procedure of Driscoll (1984) were applied and reported from fourteen IM catchments. Total monomeric Al (Al_m) and organic monomeric Al (Al_o, sometimes called non-labile Al) were measured in surface water by a colorimetry method. The Al_o was separated using a strong cation exchange resin, the method utilized charge exclusion by ion exchange. Potentially toxic inorganic monomeric Al (Al_i, sometimes called labile Al) was calculated as the difference between Al_m and Al_o. The ICP IM database contains relevant data about Al fractions in surface runoff from fourteen catchments so far. These catchments belong to seven countries: Finland (5), Norway (3), United Kingdom (2), Czech Republic (1), Estonia (1), Sweden (1) and Switzerland (1). Distinct patterns were evident in runoff waters of these catchments. The highest Al_i values were detected at CZ02 (median 340 μg L^{-1}) and at SE04 (median 210 μg L^{-1}). Very high Al_i concentrations were measured at NO01 and NO03 (median 170 μg L^{-1} and 130 μg L^{-1}, respectively). Slightly elevated Al_i values were documented at GB02, EE02, FI01 and FI02. The remaining IM catchments (GB01, FI03, FI04, FI05, NO02 and CH02) showed very low Al_i concentrations in runoff water. Fast additions of missing Al_i values from catchments with available, but not reported Al_i data to the IM database is advisable (Krám & Kleemola 2019).

Previous work on heavy metals is summarised below.

Preliminary results on concentrations, fluxes and catchment retention were reported to the Working Group on Effects in 2001 (document EB.AIR/WG.1/2001/10). The main findings on heavy metals budgets and critical loads at ICP IM sites were presented by Bringmark (2011). Input/output budgets and catchment retention for Cd, Pb and Hg in the years 1997–2011 were determined for 14 ICP IM catchments across Europe (Bringmark et al. 2013). Litterfall plus throughfall was taken as a measure of the total deposition of Pb and Hg (wet + dry) on the basis of evidence suggesting that, for these metals, internal circulation is negligible. The same is not true for Cd. Excluding a few sites with high discharge, between 74 and 94% of the input, Pb was retained within the catchments; significant Cd retention was also observed. High losses of Pb (>1.4 mg m^{-2} yr^{-1}) and Cd (>0.15 mg m^{-2} yr^{-1}) were observed in two mountainous Central European sites with high water discharge. All other sites had outputs below or equal to 0.36 and 0.06 mg m^{-2} yr^{-1}, respectively, for the two metals. Almost complete retention of Hg, 86–99% of input, was reported in the Swedish sites. These high levels of metal retention were maintained even in the face of recent dramatic reductions in pollutant loads. In the Progress report on heavy metal trends at ICP IM sites (Åkerblom & Lundin 2015) temporal trends were seen in forest floor with decreasing concentrations for Cd and Pb while Hg did not change. An increase in heavy metal concentrations was also seen in deeper mineral soil horizon indicating a translocation of heavy metals from upper to deeper soil horizons.

Calculation of critical loads and their exceedance, relationships to effect indicators

The critical load (CL) methodology has been a key science-based tool for assessing the environmental consequences of air pollution. Critical loads are deposition thresholds used to describe the sensitivity of ecosystems to atmospheric deposition. Critical loads for eutrophication and acidification were computed using a long-term dataset of intensively studied forested ecosystem ICP Integrated Monitoring sites (n = 17) in northern and central Europe (Forsius et al. 2021). The sites belong to the ICP Integrated

Monitoring and eLTER networks. The link between the site-specific calculations and time-series of CL exceedances and measured site data was evaluated using long-term measurements (1990–2017) for bulk deposition, throughfall and runoff water chemistry. Novel techniques for presenting exceedances of CLs and their temporal development were also developed. Concentrations and fluxes of sulphate, total inorganic nitrogen (TIN) and acidity in deposition substantially decreased at the sites. Decreases in sulphur (S) deposition resulted in statistically significant decreased concentrations and fluxes of sulphate in runoff and decreasing trends of TIN in runoff were more common than increasing trends. The temporal developments of the exceedance of the CLs indicated the more effective reductions of S deposition compared to nitrogen (N) at the sites. There was a relation between calculated exceedance of the CLs and measured runoff water concentrations and fluxes, and most sites with higher CL exceedances showed larger decreases in both TIN and H^+ concentrations and fluxes. Sites with higher cumulative exceedance of eutrophication CLs (averaged over 3 and 30 years) generally showed higher TIN concentrations in runoff. The results provided evidence on the link between CL exceedances and empirical impacts, increasing confidence in the methodology used for the European-scale CL calculations. The results also confirm that emission abatement actions are having their intended effects on CL exceedances and ecosystem impacts.

In Holmberg et al. (2013) empirical impact indicators of acidification and eutrophication were determined from stream water chemistry and runoff observations at ICP IM catchments. The indicators were compared with exceedances of critical loads of acidification and eutrophication obtained with deposition estimates for the year 2000. Empirical impact indicators agreed well with the calculated exceedances. Annual mean fluxes and concentrations of acid neutralising capacity (ANC) were negatively correlated with the exceedance of critical loads of acidification. Observed leaching of nitrogen was positively correlated with the exceedances of critical loads (Holmberg et al. 2013). This study was revisited with new data on N concentrations and fluxes (Holmberg et al. 2017). For most sites, there was an improvement visible as a shift towards less exceedance and lower concentrations of total inorganic nitrogen (TIN) in runoff. At the majority of the sites both the input and the output flux of TIN decreased between the two observation periods 2000–2002 and 2013–2015. Data from the ICP IM provide evidence of a connection between modelled critical loads and empirical monitoring results for acidification parameters and nutrient nitrogen.

Planned activities

- Maintenance and development of central ICP IM database at Swedish Agricultural University (SLU) since January 2021.
- Continued assessment of the long-term effects of air pollutants to support the implementation of emission reduction protocols, including:
    - Assessment of trends.
    - Calculation of ecosystem budgets, empirical deposition thresholds and site-specific critical loads.
    - Dynamic modelling and scenario assessment.
    - Comparison of calculated critical load exceedances with observed ecosystem effects.
- Calculation of pools and fluxes of heavy metals at selected sites.
- Assessment of cause-effect relationships for biological data, particularly vegetation.
- Coordination of work and cooperation with other ICPs, particularly regarding dynamic modelling (all ICPs), cause-effect relationships in terrestrial systems (ICP Forests, ICP Vegetation), and surface waters (ICP Waters).
- Participation in the development of the European LTER network (Long Term Ecosystem Research Network, www.lter-europe.net) and eLTER RI (European Research Infrastructure) after being adopted by the 2018 ESFRI Roadmap. The RI is built by the two Horizon 2020 projects “eLTER PPP” (Preparatory Phase Project) and “eLTER PLUS” (Advanced Community project)

- Cooperation with other external organisations and programmes, particularly the International Long Term Ecological Research Network (ILTER, www.ilter.network, Mirtl et al. 2018).
- Participation in projects with a global change perspective.

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1 ICP IM activities, monitoring sites and available data

1.1 **Review of the ICP IM activities from June 2020 to June 2021**

Meetings

- Co-Chair Ulf Grandin represented ICP IM in a meeting organised by the European Commission about the NEC Directive, held on-line 4 June 2020.
- Ulf Grandin represented ICP IM and gave a presentation about current activities at the ICP Forests Task Force meeting, held on-line 11–12 June 2020.
- ICP IM Programme Manager Martin Forsius and Co-Chair Ulf Grandin represented ICP IM and Co-Chair Salar Valinia represented Sweden in the Fifth Joint Session of the Working Group on Effects and the Steering Body to EMEP, held on-line 14–17 September 2020.
- Martin Forsius, Ulf Grandin and Maria Holmberg participated in the eLTER PPP and PLUS project meeting, held on-line 13–16 October 2020.
- Martin Forsius, Ulf Grandin and Salar Valinia represented ICP IM at the videoconference of the Joint meeting of EMEP Steering Body and Working Group on Effects Extended Bureau, 1–3 March 2021.
- Martin Forsius, Ulf Grandin and Maria Holmberg took part in the eLTER PPP and PLUS project meeting, held on-line 7–16 April 2021.
- The twenty-ninth meeting of the Programme Task Force on ICP Integrated Monitoring was held on-line 13–14 April 2021.
- Maria Holmberg represented ICP IM and gave a presentation about current activities, at the ICP Modelling and Mapping Task Force meeting, held on-line 20–21 April 2021.
- Ulf Grandin represented ICP IM and gave a presentation about current activities, at the ICP Waters Task Force meeting, held on-line 28–29 April 2021.
- Ulf Grandin represented ICP IM and gave a presentation about current activities, at the ICP Forests Task Force meeting, held on-line 10–11 June 2021.

Data issues

After November 1st, 2020 the National Focal Points (NFPs) reported their 2019 results to the ICP IM Programme Centre. The Programme Centre carried out standard check-up of the results and incorporated them into the IM database.

Scientific work and activities in priority topics

- A scientific paper ’Weak recovery of epiphytic lichen communities in Sweden over 20 years of rapid air pollution decline’ (Weldon & Grandin) was published. A summary of this paper is included in the present Annual Report.
- A scientific paper ‘Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe’ (Forsius et al. 2021) was published.
- A scientific paper on effects of nitrogen enrichment on forest vegetation (Weldon et al.) was submitted. This paper was prepared in co-operation with ICP Forests.
- A scientific paper on Hg and HM trends in concentrations and fluxes across ICP Integrated Monitoring sites in Europe (Eklöf et al.) is planned for 2021/2022.
- A scientific paper on impacts of internal catchment-related nitrogen parameters to total inorganic nutrient nitrogen (TIN) leaching (Vuorenmaa et al.) will be finalised in 2021.

- The Programme Centre prepared the ICP IM contribution to the 2020 Joint Progress Report on policy relevant scientific findings for the WGE (ECE/EB.AIR/GE.1/2020/3–ECE/EB.AIR/WG.1/2020/3).
- The Programme Centre and Task Force Chairs contributed to the WGE report on the revision of the Gothenburg protocol.
- Task Force Chairs together with the Programme Centre and a group of experts from the IM community developed the "IM light" concept.

1.2 **Activities and tasks planned for 2021–2023**

Activities/tasks related to the programme's present objectives, carried out in close collaboration with other ICPs/ Task Forces

According to the WGE work plan 2020–2021, ICP IM will produce the following reports/papers:
- Scientific paper on effects of nitrogen enrichment on forest vegetation. A cooperation between ICP IM & ICP Forests. (WGE item 1.1.1.17) manuscript submitted.
- Scientific paper on Hg and HM trends in concentrations and fluxes across ICP Integrated Monitoring sites in Europe (WGE item 1.1.1.1.6), report published in AR29, paper in preparation.
- Scientific paper on the impacts of catchment characteristics, climate and hydrology on N processes (WGE item 1.1.1.15, listed as: Scientific paper on impacts of internal catchment related nitrogen parameters to total inorganic nutrient nitrogen (TIN) leaching), report published in AR29, paper in preparation.

Suggested ICP IM activities for the WGE 2022–23 work plan:
- 2022: Operationalise and advertise “IM light” as an attractive monitoring protocol, aiming at adding more ecosystem types in the ICP IM monitoring
- 2022: Continued work on the trends in ecosystems connected to the review of the Gothenburg Protocol and questions asked from the policy groups
- 2023: Scientific paper on modelling and assessment of biodiversity and ecosystem impacts, in co-operation with e.g. Centre for Dynamic Modelling (CDM).
- Contribution to the revision process of the Gothenburg Protocol, in a coordinated process of the WGE.

Other activities

- Maintenance and development of central ICP IM database at Swedish Agricultural University (SLU) since January 2021.
- Arrangement of the 30th Task Force meeting (2022)
- Preparation of the 31st ICP IM Annual Report (2022)
- Preparation of the ICP IM contribution to assessment reports of the WGE
- Participation in meetings of the WGE and other ICPs

Activities/tasks aimed at further development of the programme

- Participation in the development of the European LTER network (Long Term Ecosystem Research Network, www.lter-europe.net) and eLTER-RI (European Research Infrastructure) after being adopted by the 2018 ESFRI Roadmap. The RI is built by the two Horizon 2020 projects “eLTER PPP” (Preparatory Phase Project) and “eLTER PLUS” (Advanced Community project)

- Participation in the activities of other external organisations, particularly the International Long Term Ecological Research Network (ILTER, www.ilter.network)

1.3 Published reports and articles 2020–2021

Evaluations of international ICP IM data and related publications

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S., Váňa, M. 2021. Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe. Science of the Total Environment 753.

Eklöf, K. et al. 2020. Temporal trends and input-output budgets of heavy metals in ICP IM catchments. In: Kleemola, S. & Forsius, M. eds. 29th Annual Report 2020: Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 31/2020, pp. 31–35.

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Evaluations of national ICP IM data and publications of ICP IM representatives

Andronikov, A.V., Novák, M., Krám, P., Šebek, O., Andronikova, I.E., Efremenko, N.A., Borodulina, G.S., Subetto, D.A., Stepanova, M., Antalova, E., Levichev, M.A., Zobkova, M.V. & Chesalina, G.L. 2020. Behaviour of Cr in runoff from two catchments underlain by felsic bedrock. Hydrological Sciences Journal 65(16): 2765–2782.

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Dirnböck, T., Kraus, D., Grote, R., Klatt, S., Kobler, J., Schindlbacher, A. et al. 2020. Substantial understory contribution to the C sink of a European temperate mountain forest landscape. Landscape Ecol 35 (2): 483–499. DOI: 10.1007/s10980-019-00960-2.

Hashemi, F., Pohle, I., Pullens, J. W. M., Tornbjerg, H., Kyllmar, K., Marttila, H. Lepistö, A., Kløve, B., Futter, M. & Kronvang, B. 2020. Conceptual mini-catchment typologies for testing dominant controls of nutrient dynamics in three Nordic countries. Water 12(6):1776. DOI: 10.3390/w12061776.

Hood-Nowotny, R., Schmittner, K., Ziss, E., Leitner, S., Watzinger, A., Gorfer, M. et al. 2021. Functional response of an Austrian forest soil to N addition. Environ. Res. Commun. 3 (2), 025001. DOI: 10.1088/2515-7620/abe8c7.

Kaste, Ø., Austnes, K. & de Wit, H. A. 2020. Streamwater responses to reduced nitrogen deposition at four small upland catchments in Norway. Ambio, 49(11): 1759-1770.

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Kyllönen, K., Vestenius, M., Anttila, P., Makkonen, U., Aurela, M., Wängberg, I., Nerentorp Mastromonaco, M., Hakola, H. 2020 Trends and source apportionment of atmospheric heavy metals at a subarctic site during 1996–2018, Atmos. Env., 236, 117644. DOI: 10.1016/j.atmosenv.2020.117644.

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Löfgren S. (Ed.). 2020. Integrated monitoring of the environmental status in Swedish forest ecosystems – IM. Annual report for 2019. Dept. Aquatic Sciences and Assessment, SLU Report 2020:6, 28 pp + appendix. In Swedish. English summary. https://pub.epsilon.slu.se/21730/1/lofgren_s_210119.pdf

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1.4 **Monitoring sites and data**

The following countries have continued data submission to the ICP IM database during the period 2016–2020: Austria, Belarus, the Czech Republic, Estonia, Finland, Germany, Ireland, Italy, Lithuania, Norway, Poland, the Russian Federation, Spain, Sweden and Switzerland.

The number of sites with on-going data submission for at least part of the data years 2015–2019 is 48 from fifteen countries. Sites from Canada, Latvia and United Kingdom only contain older data.

An overview of the data reported internationally to the ICP IM database is given in Table 1.1. Additional earlier reported data are available from sites outside those presented in Table 1.1. and Fig. 1.1. Locations of the ICP IM monitoring sites are shown in Fig. 1.1.

Figure 1.1 Geographical location of ICP IM sites.

Table 1.1. Internationally reported data from ICP IM sites (- subprogramme not possible to carry out, * or forest health parameters in former Forest stands/Trees).

AREA	SUBPROGRAMME															*								
	AM	AC	PC	MC	TF	SF	SC	SW	GW	RW	LC	FC	LF	RB	LB	FD	VG	BI	VS	EP	AL	MB	BB	BV
	meteorology	air chemistry	precipitation chemistry	moss chemistry	throughfall	stemflow	soil chemistry	soil water chemistry	groundwater chemistry	runoff water chemistry	lake water chemistry	foliage chemistry	litterfall	hydrobiology of streams	hydrobiology of lakes	forest damage	vegetation	bioelements	vegetation structure	trunk epiphytes	aerial green algae	microbial decomposition	bird inventory	vegetation inventory
AT01 ZÖBELBODEN	95-19	95-19	93-19		93-19	99-04	04	93-19		95-19	-	92-17	93-19				93			93-98				
BY02 BEREZINA BR	89-15	89-15	89-15				95-98			95-15														
CH02 LAGO NERO	15-19	15-19	15-19				18	17-19		15-19	15-16						17							
CZ01 ANENSKE POVODI	89-19	89-19	89-19	89	89-19		02-15	07-19	08-19	89-19	-			07	-									
CZ02 LYSINA	67-18	93-96	90-18		91-18		93	90-18	89-18	89-18	91-18	94	08	07	11		15	94			14-15		10	
DE01 FORELLENBACH	90-19	90-19	90-19	90	90-19	90-05	90-11	90-19	88-19	90-19	-	90-19	90-19		-	90-14	90-08		00	92-95		94-19	91-02	90-95
DE02 NEUGLOBSOW	67-19	98-19	98-19		98-19	04-19	04-16	98-19	98-19		98-19	06-19	04-19				04-17							
EE01 VILSANDI	95-19	94-19	94-19	94-15	94-19	94-19	94-15	94-18	95-96	-	-	94-19	94-19	-	-	94-17	94-97			94-04		94-19		94
EE02 SAAREJÄRVE	94-19	98-19	94-19	94-16	94-19	94-19	94-15	95-18	95-14	94-19	96	94-19	94-19			96-17	96-12	12		94-15	94-17	96-19	98-14	
ES02 BERTIZ	08-17	08-19	07-19		07-19	08-19	10-15	07-18		07-19		08-18	08-19			07-12	07		07					
FI01 VALKEA-KOTINEN	88-19	94-19	88-19	88-96	89-17	89-99	88-89	89-17		88-19	87-19	88-17	90-16		90-93	88-91	88-09			88-97		90	87-89	87
FI03 HIETAJÄRVI	88-19	93-00	88-19	89-96	89-17	89-99	88	89-17		88-19	87-19	88-17	90-16		90	88-91	90-09			90-97		90-91	87-89	
FI06 PALLASJÄRVI	13-19		14-19		16-17			02-17		04-19	04-19	95-17	07-16										88-89	
IE01 BRACKLOON WOOD			91-16		91-11	92-97		91-16			-	91-96	91-98	-	-									
IT01 RENON-RITTEN	90-19	93-19	93-14		93-13	93-13	93-11	93-13		00-13	-	93-10	00	-	-	92-13	09		05-09	92		93-11		
IT03 PASSO LAVAZE	92-08	93-19	92-13		94-13	94-00	93-95	95-07		01-13	-	93-05	94	-	-	93-19	95-09		99-09	92				
IT05 SELVA PIANA	97-19	97-19	97-19		97-19	97-19	95	02-08		-	-	97-19		-	-	97-19	09		99-09					
IT06 PIANO LIMINA	99-19	97-16	97-19		97-19	97-19	95	19		-	-	97-19		-	-	97-19	09		99-09					
IT07 CARREGA	97-19	97-19	97-19		97-19	97-00	95	19		98-13	-	97-19		-	-	97-19	09		99-09					
IT09 MONTE RUFENO	97-19	97-19	97-19		97-19	97-00	95	02-08		97-14	-	97-19		-	-	97-19	09		99-09					
IT10 VAL MASINO	97-19	00-15	97-15		97-15		95	05-07		-	-	97-05		-	-	97-09	09		99-09					
IT12 COLOGNOLE	97-01	97-15	97-15		97-15	97-00	95			-	-	97-05		-	-	97-09	09		99-09					
IT13 LA THUILE	97-19	97-15	09-15		09-15		95			-	-	97-05		-	-	97-09			99-08					
LT01 AUKSTAITIJA	93-13	93-19	93-19	93-10	93-19		93-05	94-19	93-19	93-19		06-19	99-19	12		00-19	93-18		02-15	93-19	93-19			93
LT03 ZEMAITIJA	90-13	95-19	95-19	06-10	95-19		94-05	95-19	95-19	95-19		06-19	99-19	95-12		00-19	94-18		02-15	94-19	94-19			94
NO01 BIRKENES	87-19	87-19	87-19	92	89-19		87-11	86-19	87-88	87-19	-	86-17	87-02		-	91-18	86-18			86				
NO02 KÅRVATN	87-91	87-19	87-19	88	89-11		89-13	89-10		87-19	-	89-09	89-02		-	92-10	89-09							
NO03 LANGTJERN		87-97	77-19		86-03		91-13	91-03		87-19		86-03	87-02											
PL01 PUSZCZA BORECKA	06-19	16-19	16-19		16-19		17	10-19			16-19		06-19				16							
PL05 WIGRY	06-19	16-19	16-19		16-19		19	06-19		16-19			05-19				16							
PL06 PARSENTA	10-19	16-19	94-19		96-19			10-19		94-19			10-19											
PL07 POJEZIERZE CHELMINSKIE	16-19	16-18	16-19				18			16-19														
PL08 KAMPINOS	09-19	16-19	16-19		16-19		16	12-19		16-19			10-19				16							
PL09 LYSOGORY	05-19	16-19	16-19		16-19			05-19		16-19			05-19				16							
PL10 BESKIDY	94-19	16-19	94-19		02-19			11-19		94-19			09-19				16							
PL11 WOLIN	16-19	16-19	16-19		17-19			16-19		16-19			16-19											
PL12 ROZTOCZE	16-19	16-19	16-19		16-19			16-19		16-19			16-19				16							
RU03 CAUCASUS BR	89-94	89-19	89-98																					
RU04 OKA-TERRACE BR	89-06	89-19	89-98	90										93-99		93-19	93-02			93		94-96		
RU12 ASTRAKHAN BR	93-94	93-19	93-94																					
RU13 CENTRAL FOREST BR	93	93-94	93													09-19	18							
RU14 VORONEZH BR	94	94-19	94-98																					
RU16 VELIKIY ISLAND				89-90			89	89	89						93-99	93-19	91-94			89-94	93	94-95		91
RU47 KURSK																18-19								
SE04 GÅRDSJÖN F1	87-19	88-19	87-19	95	87-19		95-10	87-19	79-19	87-19	-	99-19	96-19		-	97-01	95-19	91-15	91-15	96-16	92-19	95-19		
SE14 ANEBODA	96-19	96-19	96-19	95	96-19		96-11	95-19	96-19	96-19	-	99-19	95-19		-	97-01	82-19	96-16	06-16	97-17	97-19	95-19		
SE15 KINDLA	97-19	96-19	96-19		96-19		97-12	95-19	97-19	96-19	-	97-19	95-19		-	98-01	96-19	98-18	98-18	98-18	97-19	95-19		
SE16 GAMMTRATTEN	99-19	99-19	99-19		99-19		00-18	00-19	00-19	99-19		99-19	00-19			00-01	99-19	99-19	99-19	00-15	00-19	00-19		

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2 Weak recovery of epiphytic lichen communities in Sweden over 20 years of rapid air pollution decline

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2.1 Introduction

Many lichen species are sensitive to air pollution and react rapidly, especially to sulphur and nitrogen (Skye 1979). This sensitivity is augmented by a slow growth rate, growth on substrates often exposed to air pollution and an ability to absorb more sulphur dioxide, SO_2, at a given concentration than most vascular plants (Nash & Gries 1991). As a consequence, many species of lichens were adversely affected by the widespread high levels of sulphur deposition originating in the burning of fossil fuels in industrialised areas, which also spread further afield via long range atmospheric transport (Gilbert 1986, Richardson 1988).

Although there are numerous studies showing the suitability of lichens as indicators of declining air quality, there are relatively few studies investigating the recovery of the lichen community as an effect of improved quality. Long term monitoring data have shown a recovery of SO_2 sensitive species in areas where SO_2 deposition levels have trended strongly downwards in recent decades (Pescott et al. 2015), but failures in recolonisation have also been recorded in some species while others increased (Bates et al. 2001).

Attempts to reduce levels of nitrogen deposition (principally nitrogen oxides NO_x from the combustion of fossil fuels and ammonium NH_4 from agriculture) have met with less success than SO_2 controls (Sutton et al. 2011). One result of this is that changes seen in recent decades in air pollution patterns do not always have a straightforward effect on lichens, due to the much steeper decline generally seen in deposition of SO_2 than in N. Areas previously heavily affected by acidification can show a recovery of acid sensitive species of epiphytic lichens, but simultaneously a eutrophication effect due to persistently high deposition of nitrogen while sulphur deposition decreased (Friedel & Müller 2004).

Cover of epiphytic lichens on mature living tree trunks has been monitored every fifth year since 1997 at four IM sites in Sweden (Fig. 2.1), along with monthly measurements of deposition levels of both nitrogen and sulphur compounds. Together with the strong gradient in deposition levels between the sites, from high levels on the south west coast to very low levels in the north, this dataset provides a basis for investigating the long-term impacts of atmospheric deposition of pollutants on lichen communities. The monitoring sites (Fig. 2.1) are located in long-term protected spruce forest nature reserves. Avoiding the confounding effects of forestry is particularly important for lichen studies as forest continuity is important for many species, due to the higher substrate quality found there and sufficient time available for slow colonisation processes to occur (Fritz et al. 2008).

In this study, we use a 20-year time series on epiphytic lichens collected along a depositional gradient with decreasing deposition over time, to assess to what extent the lichen community has recovered due to decreasing levels of air pollution.

2.2 Methods

The monitoring sites (Fig. 2.1) vary in size from 3.7 to 45 ha. The lichen monitoring follows a repeated measure design with inventories of the same trees every fifth year, resulting in four inventories at each site, as of 2020. However, due to severe storm and bark beetle disturbance in 2005–2008 at one of the sites, Aneboda (Weldon & Grandin, 2019) the trees selected for lichen monitoring are restricted to those monitoring plots that still have any living trees left.

On each selected trunk, all epiphytic lichens are recorded using the point frequency method. A 40 × 40 cm transparent film with 400 regularly spaced holes is attached to the trunk at 120 cm above the ground, and the number of holes covering each lichen species under the film are counted.

As the selection of trees is randomised, all kinds of tree species may be selected. However, as Norway spruce (*Picea abies* (L.) H. Karst) is the most common species, this is also the dominant tree in the monitoring programme.

Figure 2.1. Location of the study sites in Sweden.

2.2.1 Statistical methods

For each tree, a lichen community air pollution sensitivity index was calculated as a weighted community mean value of the air pollution sensitivity values provided by Hultengren et al. (1991) – the higher value the less tolerant to acidity, i.e. the better air quality. Similarly, species preference for nitrogen was calculated based on the values given in Wirth (2010), with values ranging from oligotrophic (1) to eutrophic (9). Shannon diversity index values for each site/year combination were calculated and beta diversity between the communities on sample trees was investigated within each site (Baselga et al. 2018). Changes in beta diversity are decomposed into a turnover and a nestedness component, quantifying the extent to which changes are driven by species replacement or community homogenisation (Baselga 2010). The analysis is based on Sørensen's dissimilarity index, using presence/absence data. We resample the multiple plot dissimilarities 100 times and calculate the probability of a higher/lower value of each beta diversity measure in survey 4 relative to survey 1, yielding a p-value (Baselga 2010, Baselga et al. 2015). We also applied a complementary analysis of the pair-wise matrices generated, using a Mantel test to test for similarity between the results for the survey 1 and survey 4. Note that as this is a test for similarity, we would generally expect opposite results from the resampling test for differences.

Changes in S and N sensitive species were analysed, using the following definitions. N sensitive species are those in the 1st quartile in the distribution of the N preference values of all species found across all sites. S sensitive species are those in the 4th quartile in the distribution of the S sensitivity values of all species found across all sites.

Temporal trends in diversity and the lichen community indices were assessed using a mixed model (lme in R package nlme (Pinheiro et al. 2016)) to account for the nested nature of observations (i.e. tree nested within plot as a random factor) and a first order autocorrelation structure to compensate for repeated observations by assigning time as a continuous covariate (corCAR1 in nlme). Trends in sulphur and nitrogen deposition were tested for significance using simple linear regression.

2.3 **Results**

2.3.1 Deposition

Gårdsjön, while still having the highest deposition levels of N and S, has seen steep drops in the concentrations of airborne pollutants found in throughfall monitoring over the 20 years included in the dataset. Concentrations of SO_4, NH_4 and NO_3 all declined significantly ($p < 0.05$, Fig 2.2). Kindla and Aneboda have also shown significant reductions in S concentrations. NH_4 and NO_3 concentrations declined significantly at Kindla, but at Aneboda NO_3 levels remained flat and there was an increase in NH_4. Gammtratten has had low levels of both N and S deposition throughout the monitoring period, but even here concentration levels of SO_4 and NO_3 have declined significantly.

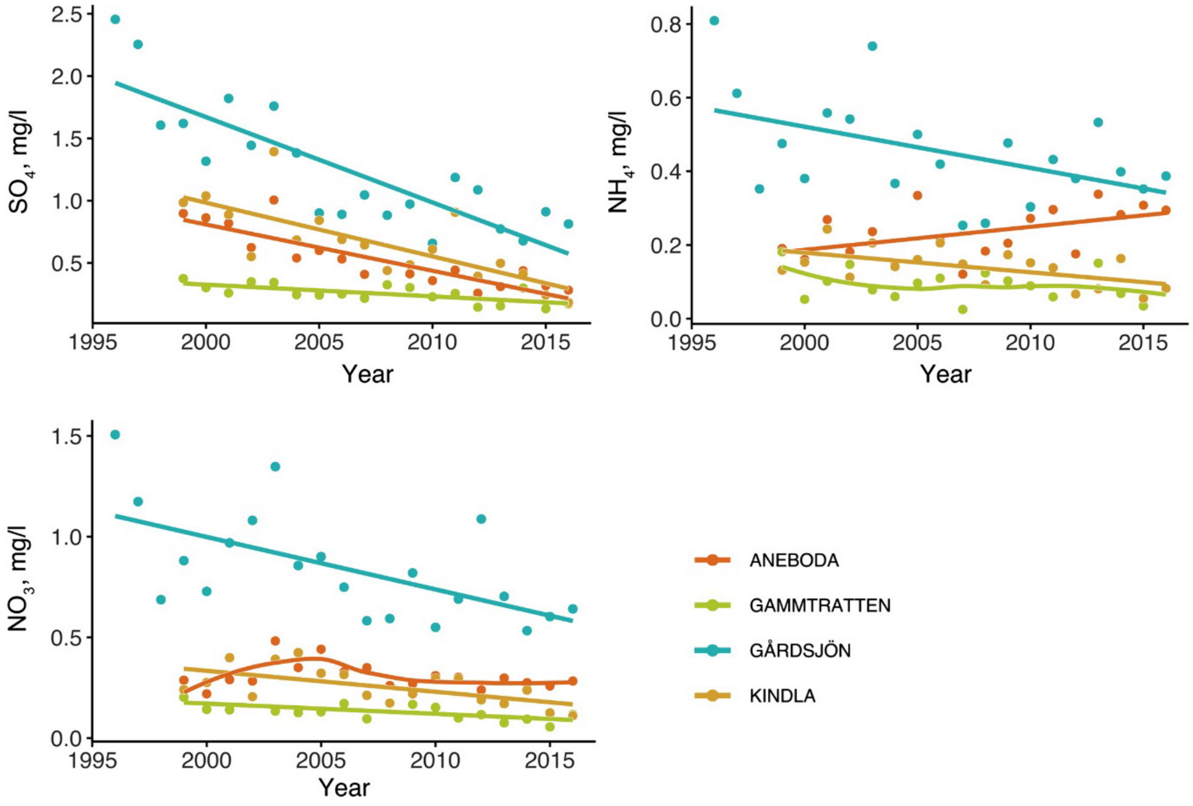

Figure 2.1. Mean annual concentrations in throughfall deposition of atmospheric pollutants at the Swedish IM sites located along a north-south depositional gradient. Smoothed lines indicate a non-significant trend.

2.3.2 Sensitivity index

Hultengren sensitivity index increased at Gårdsjön (p = 0.009) and decreased at Kindla (p = 0.03) and Gammtratten (p = 0.0004) (Fig. 2.3, Table 2.1). At Gammtratten the decline in sensitivity takes the form of an initial drop followed by no clear trend. Although the change at Aneboda is not significant, there is a clear change in the distribution of results following the disturbances there beginning in 2005 (Fig. 2.3).

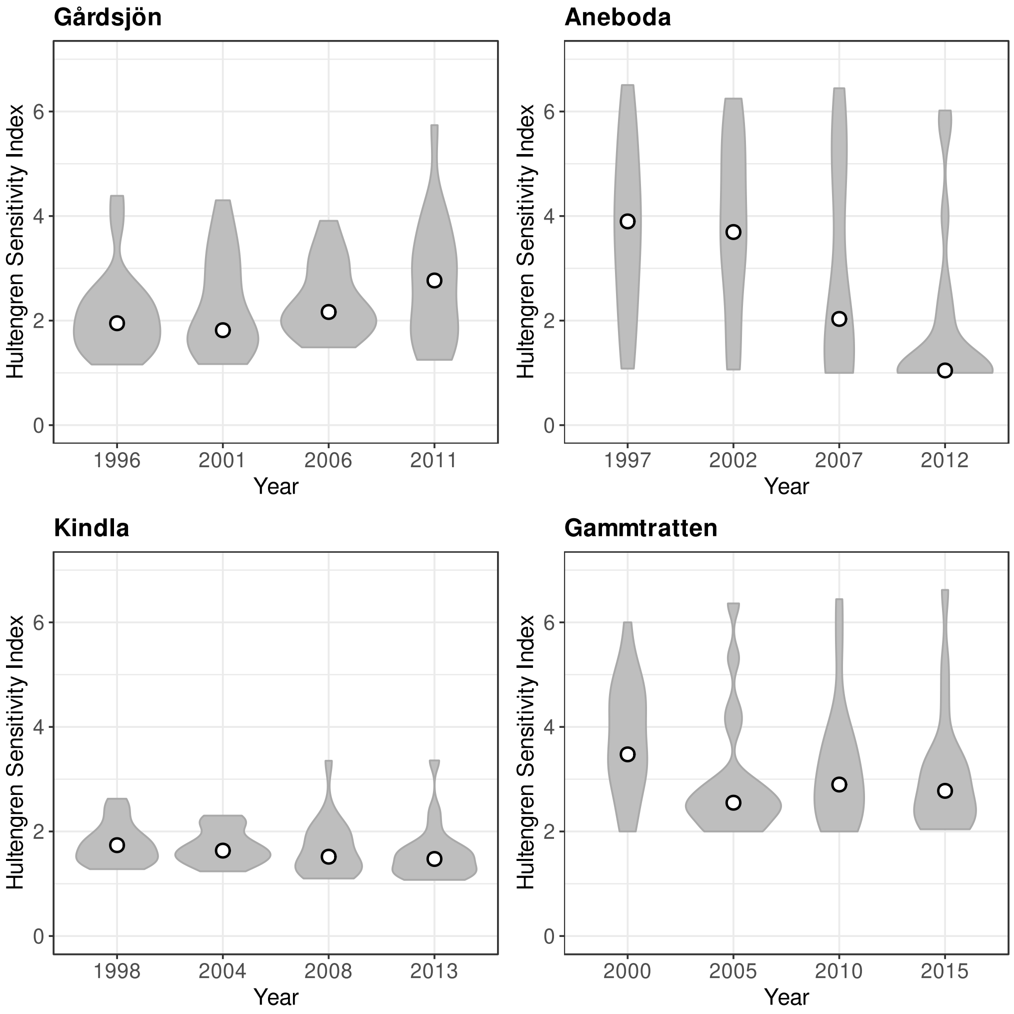

Figure 2.3. Changes over time in the Hultengren S sensitivity index for epiphytic lichens at the Swedish IM monitoring sites situated along a north-south depositional gradient. Dots indicate median values and the shaded area shows the distribution of data.

2.3.3 Nitrogen preference

The mean Wirth nitrogen index decreased at Gårdsjön (p=0.049), increased at Kindla (p=0.0026) and showed no significant change at Gammtratten and Aneboda (Fig. 2.4, Table 2.1).

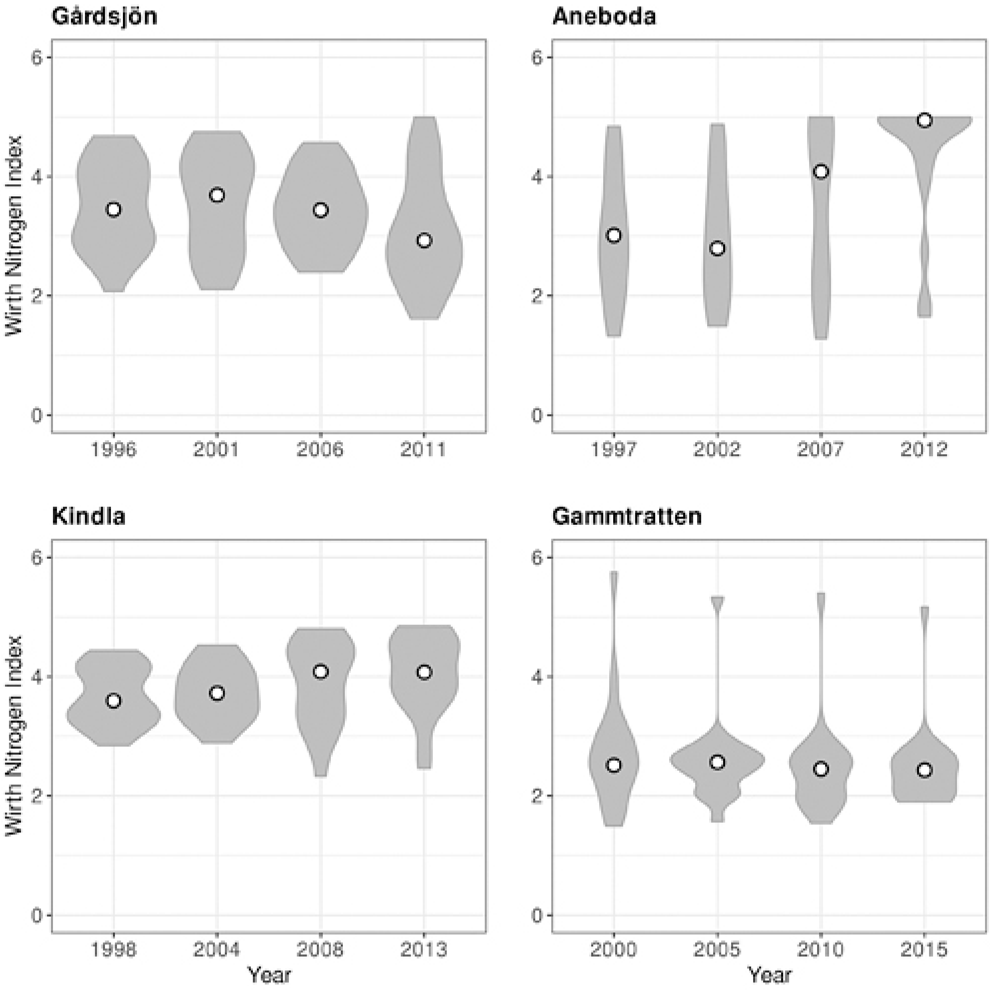

Figure 2.4. Changes over time in the Wirth nitrogen index, at the Swedish IM monitoring sites situated along a north-south depositional gradient. Dots indicate median values and the shaded area shows the distribution of data.

2.3.4 Species richness and diversity

Lichen species richness decreased at Gårdsjön (p=0.0033) and Aneboda (p=0.040) and showed no significant change at Kindla and Gammtratten (Fig. 2.5, Table 2.1).

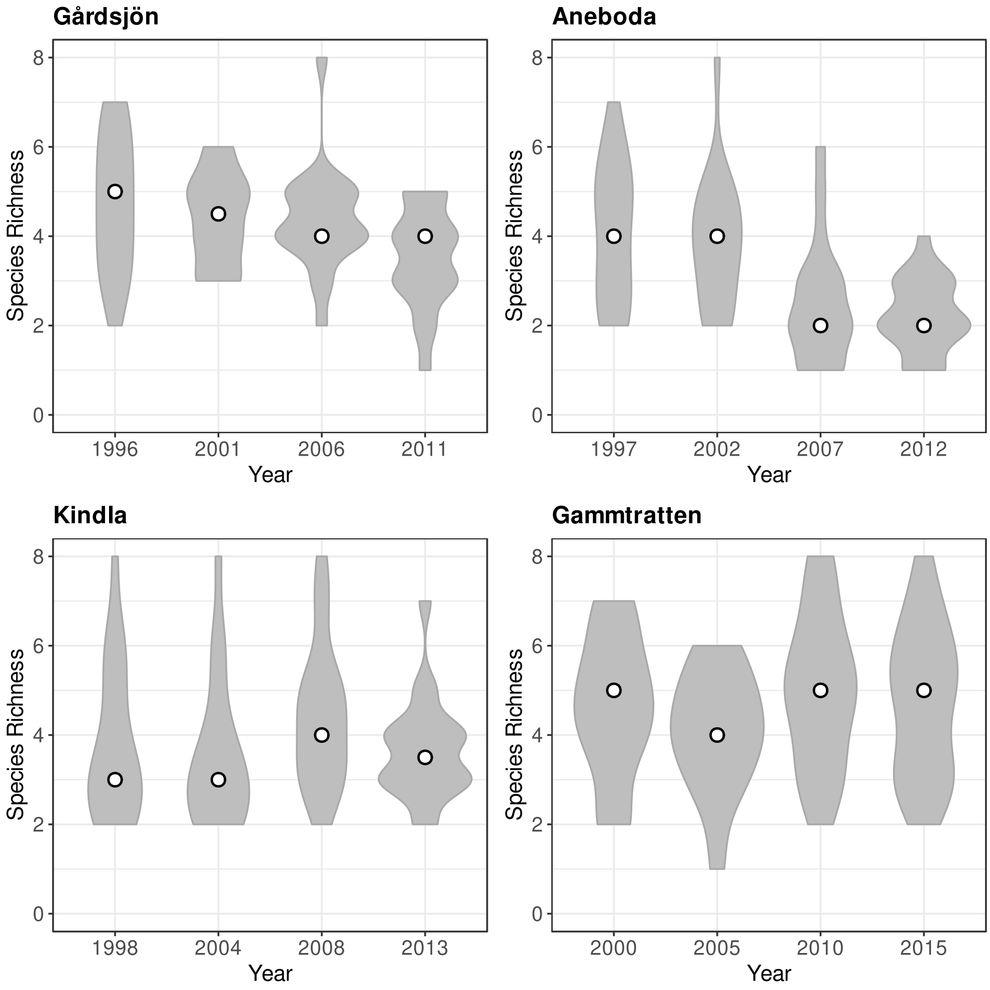

Figure 2.5. Changes over time in species richness at the Swedish IM monitoring sites situated along a north-south depositional gradient. Dots indicate median values and the shaded area shows the distribution of data.

Lichen species Shannon diversity index decreased at Gårdsjön (0.0090) and Aneboda (p=0.0062) and showed no significant change at Kindla and Gammtratten (Table 2.1). Patterns of change over time at the different sites are very similar to those seen in species richness (Fig. 2.4) and the plot is therefore omitted here.

2.3.5 Richness of S and N sensitive species only

The richness of S sensitive species decreased at Gammtratten (p=0.028) and Aneboda (p=0.0078) and showed no significant change at Kindla and Gårdsjön (Table 2.1). The richness of N sensitive species decreased at Aneboda (p=0.0041) and showed no significant change elsewhere (Table 2.1).

2.3.6 Summary of mixed model results

The tested response variables showed mainly decreases at the southern sites Gårdsjön and Aneboda, while the two less affected sites Kindla and Gammtratten showed few significant changes over time (Table 2.1).

Table 2.1. Summary of mixed model results.

	Gårdsjön	**Aneboda**	**Kindla**	**Gammtratten**
Hultengren sensitivity	**+***	n.s	**-****	**-***
S sensitive species	n.s	**-****	n.s	**-***
Nitrogen preference	**-***	n.s	**+***	n.s
N sensitive species	n.s	**-****	n.s	n.s
Richness	**-****	**-***	n.s	n.s
Shannon Diversity	**-***	**-****	n.s	n.s

∗ = $p < 0.05$, ∗∗ = $p < 0.01$. Minus sign indicates a significant decrease, while a plus sign indicates a significant increase. “n.s” indicates a non-significant change.

2.3.7 Beta Diversity

Further insight into community shifts can be gained from analysing the changes over time in beta diversity, taking the communities present on individual trees as the unit of comparison (See Table 2 in published article for detailed results).

At Gammtratten, the northern “pristine” site, all measures of beta diversity were stable, with no significant changes found between the first and last surveys. Kindla was also largely stable, but with a small decline in overall beta diversity on the resampling test (although also significant similarity on the Mantel test). At Aneboda however, there was an increase in turnover, a decrease in nestedness and an increase in overall beta diversity. At Gårdsjön, the most polluted site, there was no overall change in beta diversity but this masks a decrease in turnover and an increase in nestedness.

2.4 Discussion

We hypothesised that the lichen community in the more polluted area would show a recovery during the period of the monitoring programme, while the lichen community in the low pollution northern area would not change significantly, and that mean S sensitivity would increase at the polluted sites while the mean nitrogen preference would show more limited or no decreases. The most polluted site (Gårdsjön) did show an increase in mean S sensitivity and a decline in mean N preference but also declines in species richness and Shannon diversity. The two sites with intermediate pollution levels (Aneboda and Kindla) demonstrated no improvements and declines on some measures, while even the supposedly pristine far northern site (Gammtratten) showed a decline in S sensitivity.

The Gårdsjön site, while still showing the highest deposition levels, has seen the largest drops in both S and N deposition. Despite this, the recovery of the lichen flora is weak. Overall species richness and diversity have continued to decline over the monitoring period, which suggests that recovery of the lichen community is still far from complete after 20 years. The northern "pristine" site, Gammtratten, has as expected the lowest levels of both SO_2. and N deposition throughout the monitoring period but despite this, Hultengren sensitivity index has declined, a change also seen in the decline in richness of the most sensitive species. We can see (Fig. 2.3) that this can be characterised as a failure to recover from losses early in the monitoring period. No change in N preference at Gammtratten was found, indicating an absence of eutrophication effects, and no clear pattern of change can be seen in the measures of diversity, supporting our assumption that Gammtratten is to a large extent a pristine site with low previous and present deposition and hence minimal dynamics in the epiphytic lichen community. Nevertheless, the changes in S sensitive species are concerning.

In the 20-year period of this study we can see changes in the lichen communities at the studied sites but the hypothesised recovery of sensitive species with improvements in pollution levels is only partially found. While both nitrogen and sulphur deposition have declined strongly, levels of both remain above estimates of pre-industrial levels and may still be sufficient to negatively impact the most sensitive species (Engardt et al. 2017).

Another potential explanation is that disturbances over wide geographic scales and long periods can result in a regional species pool which is depleted of sensitive species (Cornell & Harrison 2014). A disturbance such as acid rain clearly does not have an influence only on a monitoring site but also on a much wider surrounding area. While improved conditions imply that sensitive species should recolonise, this requires that those sensitive species are still sufficiently abundant in the regional species pool after this long period of elevated deposition of atmospheric pollutants, i.e. that they actually have somewhere to recolonise from. If the regional species pool is depleted, recolonisation must occur across much longer distances than a strictly local disturbance would involve, and the colonisation of new sites is often a slow process in lichens (Buckley 2011, Dettki et al. 2000, Sillett et al. 2000). Another factor potentially limiting recolonisation is that surrounding forested areas are mostly actively managed for timber production. This results in a much larger proportion of young forest than would naturally be found, which restricts the colonisation of those species which are more commonly (or even exclusively) found in old growth forests (Lie et al. 2009). The combined effect of air pollution and forestry can result in the survival of isolated populations of sensitive lichens which are vulnerable to local extinction with the nearest population source too far away for recolonisation to occur.

2.5 Conclusion

While we expected to see a clear improvement in indicators of lichen community health over a period of declining depositions, our results show a recovery that is at best partial, alongside further declines in Hultengren sensitivity index at two sites and some indications of eutrophication and community homogenisation. There are two likely explanations for this, the first being continued/cumulative deposition adversely affecting some sensitive species. A second factor which is likely important in explaining the limited recovery seen where conditions have improved is that of dispersal limitation from a depleted regional species pool adversely affecting recolonisation. With continued declines in emissions, an eventual recovery is to be expected. However, given the wide geographic and temporal impact of the disturbance and the dispersal limitation of epiphytic lichen species, a full recovery of the pre-disturbance lichen community could take many decades. Our results confirm that lichens are sensitive to air pollution. However, the use of lichens as indicators during recovery from air pollution may be less reliable in cases where the regional species pool has been depleted (whether by pollution, management practices or other factors). Lichens may be good indicators of recovery from point source pollutions, but not necessarily in cases of large-scale transboundary air pollution, where the potential impact of regional species pools should also be taken into consideration.

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3 Long-term impacts of air pollution and climate change at Finnish ICP IM sites

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Full reports are available on ICP IM webpage

3.1 **Introduction**

The long-term time series of physical, chemical and biological data collected within the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) offer possibilities to assess the vulnerability and ecosystem impacts of global change drivers – climate change and long-range transported air pollutants – at sensitive ecosystems. In addition to the work of the Convention on Long-range Transboundary Air Pollution (CLRTAP), the ICP IM monitoring aims to support EU legislation, such as Priority Actions Framework (PAF) for Natura 2000, which is mainly designed "to maintain and restore, at a favourable conservation status, natural habitats and species of EU importance, whilst taking account of economic, social and cultural requirements and regional and local characteristics". In this report, the vulnerability and ecosystem impact of global change drivers on sensitive Natura 2000 areas were assessed at two Finnish ICP IM sites located in South and Eastern Finland with different deposition and climate gradients using long-term ecosystem data (1987–2019). These sites belong to Natura 2000 network and were demonstration sites in EU FRESHABIT LIFE IP project, in which the effects of global pressures for Natura 2000 areas were assessed.

The effect of long-term changes in air pollution and climate, the change of DOC concentration from atmosphere through the terrestrial area to streams and lakes, and the changes connected to the extreme or altered weather events were assessed using long-term ecosystem monitoring data. The concept of critical loads (CLs) is the basis for air pollution control policies in Europe (Amann et al. 2011), and critical loads were determined for the Valkea-Kotinen and Iso Hietajärvi demonstration sites with respect to the acidification of surface waters, and eutrophication of the habitat. Furthermore, long-term changes in understorey vegetation were also studied.

3.2 **Material and methods**

3.2.1 Demonstration sites

Impacts of global change drivers were demonstrated in intensively monitored ICP IM sites Lake Valkea-Kotinen and Lake Iso Hietajärvi and their catchments. These sites are located in protected Natura 2000 areas in South and Eastern Finland and represent key habitats of these regions. Intensive data from different compartments in terrestrial and aquatic ecosystems have been collected since the late 1980s, providing unique data sets for assessing the long-term impacts of climate change and air pollutants (www.syke.fi/nature/icpim, ICP IM network).

The first demonstration area, L. Valkea-Kotinen, is a boreal, small (A=0.042 km^2) headwater lake with a small pristine forested catchment (A=0.22 km^2) in South Finland. The forest dominantly consists of old-growth Norway spruce (*Picea abies*), with Scots pine (*Pinus sylvestris*), aspen (*Populus tremula*) and birch (*Betula* spp.). The forest and peatland cover 86% and 19% of the total catchment area, respectively. The bedrock is acid-sensitive, dominated by slow-weathering granitoids and gneisses. Another demonstration area, L. Iso Hietajärvi, is a boreal, small (A=0.83 km^2) headwater lake with a small pristine forested catchment (A=4.64 km^2) in Eastern Finland. The forest dominantly consists of Scots pine (*Pinus sylvestris*), Norway spruce (*Picea abies*), with aspen (*Populus tremula*) and birch (*Betula* spp.). The forest, peatland and lakes cover 55%, 35% and 23% of the total catchment area, respectively. The bedrock is acid-sensitive, dominated by slow-weathering granitoids and gneisses. The catchments of the demonstration sites are located inside a conservation area and have been intact for over 50-100 years, and therefore long-range transported air pollutants and climate change are the only external disturbances.

3.2.2 Data

The data collected within ICP IM included bulk deposition chemistry (PC), throughfall (TF), litterfall and needles (LF), lake water chemistry (LC), runoff water chemistry and hydrology (RW) and soil water chemistry (SW). Understorey vegetation was also studied including measurements of plant cover and structure of tree stand and climatic variables, nitrogen deposition and nitrogen content of plant species. Extreme weather events were identified from the modelled weather data. Critical loads of acidification and eutrophication and their exceedances for Valkea-Kotinen and Iso Hietajärvi were also determined using observed S and N deposition. More information on the sampling and analyses of PC, TF, LF, LC, RW, SW, understorey vegetation and calculation of the critical loads are given in EU FRESHABIT LIFE IP assessment reports.

3.3 **Results and discussion**

3.3.1 Runoff and air and water temperature

In 1990–2019, the mean annual runoff in Valkea-Kotinen and Iso Hietajärvi catchments was 191 mm yr^{-1} and 384 mm yr^{-1}, respectively, without any significant trend neither on annual nor monthly basis. There were significant increases ($p < 0.05$) in air temperature in January–December in both areas from 1990 to 2019. The increase in South Finland was pronounced in autumn with a significant increase in September and November (0.08 °C yr^{-1} and 0.12 °C yr^{-1}, respectively) on a monthly basis. In Eastern Finland, a significant increase was detected in spring (May, 0.11 °C yr^{-1}), late summer (August, 0.07 °C yr^{-1}) and autumn (September, 0.07 °C yr^{-1} and November, 0.19 °C yr^{-1}). While air temperature has increased in Valkea-Kotinen area, lake water temperature has not correspondingly increased in 1990–2019. However, significant increases in epilimnetic (1 m) temperatures in March, and hypolimnetic (5

m) temperatures in May were detected. In the same period 1990–2019, the annual water temperature of the surface layer (0–1 m) has significantly increased in L. Iso Hietajärvi. The trend was most pronounced in September during the autumn overturn, when water temperature significantly increased in all water layers.

3.3.2 Acidification parameters and trace (heavy) metals

In 1990s, L. Valkea-Kotinen exhibited acidic conditions, and alkalinity and pH values were commonly < 0 µeq l^{-1} and ≤ 5, respectively. Due to the decreased acid deposition, sulphate concentration has significantly decreased by about 50% during the study period 1990–2019, and the acidification reversal was also recorded in L. Valkea-Kotinen. Base cations also declined in the lake, but to a lesser extent than sulphate, indicating the improved acid-base status of soils, and led to significant increase in buffering capacity (alkalinity and ANC) and pH in lake water. Lake Iso Hietajärvi exhibits neutral or only weak acidic conditions with the mean pH 6.7 (range 5.9–7.3) between 1990 and 2019. Lake Iso Hietajärvi is located in low SO_4 deposition area, and airborne acidification has not markedly taken place in the lake, although the lake can be considered sensitive to acidification with low buffering capacity (mean alkalinity 88 µeq l^{-1} in 1990–2019) and has been affected to some extent to acid deposition. Due to the decreased acid S deposition, sulphate concentration has significantly decreased by about 60% during the study period 1990–2019, and the acidification reversal with a significant increase in buffering capacity (alkalinity and ANC) was also recorded in L. Iso Hietajärvi.

In L. Valkea-Kotinen, the decrease in xSO_4 concentration was slower than expected, based on the clear decrease of total xSO_4 deposition (80–90%). This indicates a delayed response in SO_4 output in the catchment to decreased deposition. At the beginning of the 1990s, the L. Valkea-Kotinen catchment retained 30% of sulphate deposition due to strong retention of SO_4 in peatlands and in peaty soils, but after the mid-1990s the catchment shifted from retention to net release (output > input) of sulphate. The recovery of forested catchments from SO_4 deposition – in terms of SO_4 net release – appeared to be most pronounced in catchments with the highest SO_4 deposition level and also having the strongest decrease in the SO_4 deposition load, but this process has taken place also in low SO_4 deposition areas (Vuorenmaa et al. 2017). In the low SO_4 deposition area in L. Iso Hietajärvi, SO_4 was mainly retained in the catchment (input > output), but the retention rate declined over the study period, and at the end of the study period, the catchment shifted towards a net release (output > input). These results show that forest soils are now recovering from acid deposition by releasing of stored airborne sulphur that had accumulated in the past (e.g. De Vries et al. 2001).

Along with decreased acidifying emissions, emissions of trace (heavy) metals, particularly of Hg, Cd and Pb, substantially decreased in Europe (Travnikov et al. 2012), and in line with this, the deposition of the trace (heavy) metals in the Valkea-Kotinen catchment clearly declined over the past decades (Ruoho-Airola et al. 2014). The general decrease of trace metals deposition was reflected to some extent in lake water concentrations. Total aluminium (Al tot) increased significantly, but a significant decreasing trend was detected for arsenic (As), lead (Pb) and nickel (Ni) between 1994 and 2019. For zinc (Zn), copper (Cu) and chromium (Cr) the maximum concentrations, indicated by 90% percentiles, decreased between the 1990s and 2010s. It is obvious that decrease of long-range transported trace metals deposition has also taken place in remote Iso Hietajärvi region. The general decrease of trace metals deposition was reflected to some extent in lake water concentrations. A significant decreasing trend was detected for arsenic (As), chromium (Cr), lead (Pb) and nickel (Ni) between 1994 and 2019. For copper (Cu) the median value and maximum concentrations, indicated by 90% percentiles, decreased between the 1990s and 2010s.

3.3.3 Total organic carbon and water colour

During the past 30 years, L. Valkea-Kotinen showed further brownification with a significant increase in both total organic carbon (TOC) concentration and water colour. From 1990 to 2019, TOC concentration has increased approximately by 4 mg l^{-1} and water colour 50 mg Pt l^{-1}. Lake Iso Hietajärvi can be considered as a clear water lake (mean water colour and total organic carbon concentrations (TOC) 29 mg Pt l^{-1} and 4.7 mg l^{-1}, respectively, in 1990–2019), but during the past 30 years, L. Iso Hietajärvi has also shown further brownification with a significant increase in both TOC concentration and water colour. Browning in the 1990s and early 2000s has been attributed dominantly to improved air chemistry i.e. substantially decreased acid sulphate deposition and variations in sea-salt deposition, acting through chemically-controlled organic matter solubility in catchment soils (e.g. Monteith et al. 2007). Recently, changes in climatic conditions, such as increased precipitation and discharge, are exerting greater influence on variation and increasing TOC concentrations in surface waters (e.g. de Wit et al. 2016).

Increased TOC concentration and water colour, and the consequent decrease in light penetration into the lake may have large ecological impacts on L. Valkea-Kotinen, such as decreasing primary production (Arvola et al. 2014) and decreasing feeding efficiency and growth of perch (Rask et al. 2014). Increased TOC and water colour may lead to heat absorption in shallower water layers and is reported to strengthen the thermal stratification within couple of day after ice-off in L. Valkea-Kotinen, causing incomplete spring overturn and deteriorated oxygen conditions in the lower part of the water column (Vuorenmaa et al. 2014). In anoxic conditions, it is likely that phosphorus stored in the sediment will be released into the water, causing eutrophication. Oxygen deficiency in the hypolimnion of L. Valkea-Kotinen has also caused production of methyl-Hg (Verta et al. 2010) and its accumulation in fish (Rask et al. 2010).

3.3.4 Nitrogen, phosphorus and oxygen

During the period 1990–2019, there was no significant long-term trend in total inorganic nitrogen concentrations (TIN=NO_3-N+NH_4-N) in L. Valkea-Kotinen on an annual basis. However, a seasonal pattern was detected: TIN concentration decreased significantly in the epilimnion in winter. Instead, in L. Iso Hietajärvi there was a significant decreasing trend in TIN concentrations on an annual basis. Seasonally, the decrease in nitrate (NO_3-N) concentration was pronounced in summer. At both catchments, the trend slopes were generally decreasing rather than increasing, which is in agreement with declined TIN deposition in the regions, and annual deposition amounts. The trend slopes of TIN concentrations in surface waters have been generally decreasing rather than increasing also in other undisturbed forested catchments elsewhere in Europe (Vuorenmaa et al. 2018).

Studies from European forested ecosystems have shown that nitrate leaching mainly occurs when the inorganic N deposition input is above a critical deposition threshold of ca. 10 kg ha^{-1} yr^{-1} (e.g. Dise and Wright 1995). During the period 2010–2017, the mean annual TIN deposition in the regions was $\leq$ 2.5 kg ha^{-1} yr^{-1} (source: Finnish Meteorological Institute), which was clearly below the critical deposition threshold, which should mean low deposition-driven risk of N leaching. Moreover, the input-output budgets of inorganic nitrogen for the Valkea-Kotinen and Iso Hietajärvi catchments showed high net retention (> 95%) of inorganic nitrogen (Vuorenmaa et al. 2017). Total nitrogen concentration in both lakes did not exhibit any long-term trend in 1990–2019.

Lake Valkea-Kotinen is a humic, dystrophic lake (mean tot P in 1990–2019 17 µg l^{-1}, range 10–42 µg l^{-1}). No consistent significant trend was found in total phosphorus (tot P) concentration in the epilimnion in 1990–2019, but short-term patterns were evident. Concentration of tot P in 1990–1999 decreased (-0.62 µg l^{-1} yr^{-1}), and in 2000–2019 it increased (0.31 µg l^{-1} yr^{-1}). One possible reason for the variability in tot P concentration might be deteriorated oxygen conditions in the hypolimnion, and enhanced release of phosphorus from the sediment in the 2000s. The incomplete spring overturn seems to be a significant factor influencing tot P concentration in the L. Valkea-Kotinen (Arvola et al. 2014, Vuorenmaa et al. 2014). Lake Iso Hietajärvi is a nutrient poor, oligotrophic lake with mean total phosphorus (tot P) concentration 5.7 µg l^{-1} (range 2–14 µg l^{-1}) in 1990–2019. During the same period, tot P concentration showed significant decreasing trend. Seasonally, the decreasing trend was pronounced in early summer (June). In the same period, N:P-ratio exhibited weak increasing trend, indicating the increasing importance of phosphorus as a limiting nutrient, which has been detected in other small boreal lakes in Finland (Arvola et al. 2014). Lake Iso Hietajärvi has not suffered anoxic conditions in the hypolimnion (bottom layer), although long-term records show gradual decrease in the bottom layer oxygen concentrations. This may be due to increased organic matter in the lake and consequent accelerated decomposition in the bottom layer. Increased leaching of dissolved organic matter accompanied with organic phosphorus has not increased tot P concentrations in L. Iso Hietajärvi.

3.3.5 Sulphur and nitrogen deposition and exceedances of critical loads

The acidity critical load function at Valkea-Kotinen is determined by the values $CL_{max}N$=2060 eq ha^{-1} yr^{-1}, and $CL_{max}S$=373 eq ha^{-1} yr^{-1}, using the critical ANC concentration of 20 µeq l^{-1}. Correspondingly, the acidity critical load function at Iso Hietajärvi site is determined by the values $CL_{max}N$=2960 eq ha^{-1} yr^{-1}, and $CL_{max}S$=653 eq ha^{-1} yr^{-1}. In the beginning of the period 1990–2017, the acidity critical load was exceeded for some years at Valkea-Kotinen, whereas the acidity critical load was not exceeded at Iso Hietäjärvi during the observation period. The critical ANC concentration of 20 µeq l^{-1} was not violated neither at Valkea-Kotinen nor at Iso Hietajärvi during the observation period.

The empirical critical load of eutrophication at Valkea-Kotinen and Iso Hietajärvi was estimated to 5–8(10) kg N ha^{-1} yr^{-1}, using the range for Picea taiga woodland (G3.A) and mixed taiga woodland with Betula (G4.2) suggested by Bobbink et al. (2010) for habitats classified according to the EUNIS (European Nature Information System) habitat system for Europe (Davies et al. 2004). The mass balance critical load of eutrophication at Valkea-Kotinen and Iso Hietajärvi was determined as 3.3 kg N ha^{-1} yr^{-1} and 5 kg N ha^{-1} yr^{-1}, respectively. These values were lower or equaling the minimum value of the empirical critical load, thus describing the CL of eutrophication at sites.

The critical loads at Valkea-Kotinen were exceeded in the beginning of the period 1990–2017, whereas at Iso Hietajärvi no exceedance occurred. The observed TIN concentrations in runoff were, however, lower than the acceptable concentration of N 1.3 mg l^{-1} during the whole period.

The critical concentrations (ANC 20 µeq l^{-1}, TIN 1.3 mg l^{-1}) at both sites were not violated at any time during the observation period, and the ANC concentrations have increased, and the TIN concentrations decreased over the observation period. For eutrophication at Valkea-Kotinen and Iso Hietajärvi, there is a pattern that low TIN concentrations are coupled to low exceedance values (more negative Ex_{eut}).

3.4 **Long-term changes in terrestrial part of the catchment**

3.4.1 Long-term temporal trends in bulk precipitation, throughfall, soil water, stream water, foliage and litterfall

Successful emission reduction measures in Europe over the past 30–40 years have led to a declining deposition of air pollutants in Europe, as shown at Valkea-Kotinen and Iso Hietajärvi demonstration sites. The emission control programmes have been particularly successful for sulphur (S), and sulphur concentrations have significantly decreased in bulk deposition (PC) and throughfall (TF). Total deposition of S has decreased in the Valkea-Kotinen and Iso Hietajärvi area by 70–90% since 1990. In addition to deposition, sulphur concentrations have significantly decreased in green needles and litterfall (LF) and in runoff water (RW).

At Valkea-Kotinen catchment, SO_4-S concentrations have also decreased significantly in soil water (SW) at the depth of 20 cm. At Iso Hietajärvi, the corresponding decrease in SW was found at the depth of 5 cm. Therefore, the reduced SO_2 emissions have resulted not only in reduced deposition loads to the forest canopy and litterfall, but the reduction has also taken place in the soil and in the runoff water. Decreasing trend in SO_4-S concentrations obviously reflected also to strong acid anions, which had significant decreasing trend in all liquid samples, excluding soil water under organic layer (Valkea-Kotinen) and in mineral soil (Iso Hietajärvi).

Sum of base cation concentrations in PC was slightly decreasing at both sites, but not significantly. The slightly decreasing trend in PC is probably due to the fact that in 1990s emissions from the oil shale power plants in Estonia increased the base cation deposition in the South and south eastern Finland, but due to the application of dust removal technology, the recent base cation emissions of the region have decreased (Ruoho-Airola et al. 2003). At Valkea-Kotinen, base cations in TF increased significantly, while in SW at depth of 20 cm and in RW a significant decreasing trend was detected. Because of a decreasing trend of base cations in PC, it is obvious that the detected significant increase in base cation concentrations in TF is related to the intercepted dry deposition on the tree canopy, and the consequent wash-off by precipitation. In addition to dry deposition, wash-off and leaching from the above ground biomass and tree litter are the most relevant sources, which may increase base cation concentrations in TF. At Iso Hietajärvi, all other aquatic solutions, except RW showed slightly decreasing trend in base cations, which probably in TF reflects the decreased base cation concentrations in PC, while in SW, the decline may be related to the decline of SO_4-S concentrations.

At Valkea-Kotinen, there was a decreasing trend in litterfall (needles and other litterfall fraction) in Mg and K as well as in green needles. In general base cation concentrations are usually high in needles, however, due to cation exchange reaction hydrogen ions in TF can replace adsorbed cations, especially K, which is seen also in our results. The base cation concentrations decreased significantly ($p<0.05$) in SW at the 20 cm depth, but had an increasing trend at 5 cm depth. Increasing trends in TF base cations may have had an effect also on soil solution under organic soil layer (humus), but a decline in soil water at 20 cm is probably due to efficient uptake of base cations by roots. On the other hand, the decline is also related to the decline in SO_4-S concentrations. According to Singh et al. (1980) a reduction in the amount of SO_4 anions as required to accompany cations would lead to a reduction in base cation concentrations in the soil solution. At Iso Hietajärvi, there was no significant trends in base cations (Ca, Mg, K) in green needles and litterfall, except in Mg, which significantly decreased in pine LF needles. Slightly decreasing trend in Mg concentrations was also seen in green needles and other LF fractions, but in aquatic samples, for example, in PC and TF, there was no change. Probably there has been less uptake of Mg from soil, which is seen as decreased green needle and LF concentrations. Traditionally nutrient content of needles is used as an indicator of the tree vitality and nutrition level in soil, in addition as an indicator of air pollution. However, despite decreased Mg concentration in green needles, nutrient concentrations were in balance in living needles.

The combined effects of changes in SO_4-S, strong acids and base cation concentrations reflected at both sites as an increasing trend in ANC value in the studied aquatic solutions, excluding SW at the depth of 20 cm, which had slightly decreasing trend. In view of the general decrease of SO_4-S concentrations in PC, TF, SW and RW, the decrease in ANC in SW at the depth of 20 cm is somewhat contradictory. However, the slight decrease in base cation concentrations can be the main cause for the decreasing trend in ANC in mineral soil layer and may be related, for example, to a natural succession of the forest ecosystem. When trees and other vegetation take up base cations from the soil, protons are released into the soil. In a mature natural forest ecosystem, the increase in acidity is neutralized by nutrient release through decomposition and mineralization of litter. However, it is possible that in an old forest, there can be an imbalance between base cation uptake and release from litter mineralization (Bérden et al. 1987).

At Valkea-Kotinen, DOC concentrations in PC significantly decreased, while a significant increase of DOC was found in TF. At Iso Hietajärvi, a similar pattern was detected, while trends were not significant. At Valkea-Kotinen, DOC in TF was positively correlated with air temperature, which is in agreement with other studies (Kalbitz et al. 2000, Solinger 2001, Ukonmaanaho et al. 2014), and this was expected because in the growing season DOC concentrations were usually higher compared to winter conditions. Obviously, the increased temperature and precipitation in study regions were reflected as longer growing seasons, which in turn led to a higher net primary production and a higher foliar litter production rate. At Iso Hietajärvi, increased DOC concentration in SW in organic soil layer (5 cm) and mineral soil layer (20 cm) indicates that decomposition of organic material has increased in soil. Since the senescing needle mass is the primary source of DOC-producing substrate, a higher input rate of litter may reflect a higher DOC production in the canopy and soils, and subsequently an increased DOC concentration in TF and SW (Fröberg et al. 2006). At Valkea-Kotinen, DOC concentration trend was significantly decreasing under mineral soil layer (20 cm) and slightly increasing under organic soil layer (5 cm). Lush vegetation in old growth forest and lot of decaying material explain slightly increasing DOC trend under organic soil layer, but this effect did not last to the 20 cm depth, where lack of decaying material has led to slightly decreasing DOC trend. Other option is that with declining SO_4-S inputs the DOC competes more efficiently with SO_4-S from adsorption sites in the soil and therefore a decrease in DOC concentration in mineral soil horizons can be expected with decreasing concentration of SO_4-S (Gobran and Nilsson 1988). There was no significant increase in litterfall amount at study sites during the study period, although amount of Pine LF needle fraction at Iso Hietajärvi, and amount of other litterfall fraction at Valkea-Kotinen, had a slight increasing trend. It is obvious, however, that the biomass of ground vegetation has increased, and decaying material from there has increased decomposition and release of DOC.

In Valkea-Kotinen and Iso Hietajärvi region, total inorganic nitrogen (TIN=NO_3-N + NH_4-N) bulk deposition ranges from 2.1 to 2.5 kg ha^{-1} yr^{-1}, respectively. European nitrogen (N) emissions have also decreased and have resulted in a decrease of TIN deposition, as shown at Valkea-Kotinen and Iso Hietajärvi demonstration sites, where total N and NO_3-N concentrations in deposition significantly decreased between 1990 and 2018. However, the decrease of N deposition has been generally smaller than that of S. European N emissions have decreased less than those of S, and the bulk deposition of N has generally exceeded S deposition on an equivalent basis since the late 1990s.

A decreasing TIN trend in forest at both sites was also found, excluding SW under the mineral soil (20 cm). Trend is obviously related to the decreased TIN deposition trend. In Finland TIN deposition has not been critically high (> 10 kg ha^{-1} yr^{-1}, see Dise and Wright, 1995) during the study period, although the study period started already in the 1990s, when elevated nitrogen concentrations were common and still are, for example, in central Europe. Similar decreasing trend in N concentration was seen also in litterfall (both foliar and other fraction) but not in green needles, which had significant increasing trend, which can be explained by more efficient nitrogen retranslocation from senescing needles to the younger needles. Plants take up part of the nitrogen directly through precipitation, therefore decreased concentration in green needles would have been more expected due to the decreased deposition. Earlier higher N deposition may still be in internal N flux of forest ecosystem and can be expressed as elevated concentration of needles. In soil solution TIN concentrations were often below analytical detection limit, and therefore results are only indicative. Similarly, decreasing trend was also seen in total nitrogen concentration in different solutions, which is an indication that N emission have decreased during the study period.

3.4.2 Do extreme weather events change DOC export from terrestrial to aquatic ecosystems?

In Valkea-Kotinen and Iso Hietajärvi areas, the mean annual precipitation in 1990–2018 was 588 mm yr^{-1} and 617 mm yr^{-1}, and annual mean temperature 4.3 °C and 2.7 °C, respectively. From the study period 1990–2018 three most warm, cold, dry and wet years were chosen, when annual temperature was over or less than average as well as years when annual precipitation was lower or greater than average. The effect of single extreme weather event (heavy storm/rainfall) was also assessed. In addition, three years were chosen when DOC/TOC concentrations in TF and RW was highest (DOC/TOC^{high}) or lowest (DOC/TOC^{low}). At Iso Hietajärvi, the highest (DOC/TOC^{high}) or lowest (DOC/TOC^{low}) was also determined for SW (20 cm).

At Valkea-Kotinen site, DOC/TOC^{high} concentration in TF was from 27 to 58% higher than on average, and DOC/TOC^{low} concentration from 36 to 38% smaller than on average. Correspondingly, DOC/TOC^{high} in RW was from 22 to 27% higher than on average and DOC/TOC^{low} was from 14 to 17% lower than on average. Results indicated that during the three driest years, the DOC/TOC concentration was highest in TF, while a similar pattern was not found in RW. In general, there was an indication that both dry and wet periods have an effect on DOC/TOC concentrations, although the effect of wet season was not clear. At Iso Hietajärvi site, DOC/TOC^{high} concentration in TF was from 30 to 51% higher than on average, and DOC/TOC^{low} concentration from 22 to 27% smaller than on average. Correspondingly, DOC/TOC^{high} in RW was from 25 to 51% higher than on average and DOC/TOC^{low} was from 20 to 34% lower than on average. In SW (20 cm) differences were greater, being for DOC/TOC^{high} concentration nearly 300% higher in 2008, which was also one of the years, when precipitation sum peaked (> 800 mm a^{-1}). Although some maximum/minimum temperature/precipitation years coincided with DOC/TOC^{high} and DOC/TOC^{low} concentrations, no clear pattern between DOC/TOC concentrations and temperature/precipitation was identified, and there was no strong correlation between temperature or precipitation and DOC/TOC concentrations in different water samples. In addition, no clear storm/heavy rainfall effect on concentrations was observed. Therefore, it seems that up until now the extreme weather events, which are expected to be more frequent in a future, have not had a strong impact on the study areas. However, it should be taken into account that study areas are pristine protected areas, with old-growth forest, high amount of biomass, high biodiversity in vegetation and other biota and the forest structure is diverse compared to managed forest, all these characteristics are supposed to increase resilience against the effects of extreme weather events.

3.5 **Long-term changes in vegetation**

3.5.1 Stand characteristics

The Valkea-Kotinen forest monitoring site represents an old-growth Norway spruce (*Picea abies*) dominated *Oxalis-Myrtillus* forest type. In addition to Norway spruce (63% from the total living volume), birch (*Betula* spp., 32%) and Scots pine (*Pinus sylvestris*, 5%) grew as mixture. The Iso Hietajärvi forest monitoring site characterizes an old-growth Scots pine dominated *Empetrum-Vaccinium* (EVT) forest type of middle boreal zone. In addition to Scots pine (93% from the total living volume) some birches (*Betula pendula* and *B. pubescens*) and Norway spruces were present, but in general, the whole study site represents very homogenous dryish Scots pine heath forest. Based on the visual estimation, the canopy cover at Valkea-Kotinen and Iso Hietajärvi was 74% and 27% in 2014 and 64% and 21% in 2019, respectively. This indicates that the light level has increased probably due to defoliation of old-tree crowns or falling of dead trees.

3.5.2 Changes in species number and cover

The total percentage cover of the understorey vegetation has increased in the Valkea-Kotinen monitoring plot during the period 1998–2019. The sampling design changed after 1998, and therefore results concerning comparisons between 1998 and 2003–2019 should be interpreted with caution. Anyway, both the cover of bryophytes (mainly *Hylocomium splendens*) and dwarf shrubs (mainly blueberry, *Vaccinium myrtillus*) have increased at least 10% and the total cover more than 20% during 2003–2019. If the year 1998 is included to the comparison, the increasing trend is much higher: 30% for bryophytes and 40% for the total cover. Herbs also showed a slight increase during 2009–2019. The number of vascular species in different years remained relatively stable, the highest variation was found in the number of herbs. In the Iso Hietajärvi monitoring plot, the total percentage cover of the understorey vegetation stayed very stable during the period 1998–2019. The temporal species turnover was very low in all species groups, which is typical for old succession stages of boreal forests. The cover of cowberry (*Vaccinium vitis-idaea*) increased approximately 8–10% during 2003–2019. The cover of blueberry (*Vaccinium myrtillus*), instead, did not change. The herb *Melampyrum pratense* showed yearly variation in occurrence.

The variation in climatic factors, such as annual temperature sum and annual precipitation, largely regulated the abundance relationships of the plant species. At Valkea-Kotinen, the cover of bilberry showed an increasing trend, but its cover decreased in dry years. The cover of bilberry was slightly lower in the years with high temperature sum and low precipitation sum which indicated dry growing conditions. At Iso Hietajärvi, the cover of cowberry showed an increasing trend, and obviously it was regulated by climatic variation. There was a slight covariation in the cover % of bilberry and cowberry with the annual temperature sum. When the temperature sum was high, the cover of dwarf shrubs increased. In the years with high precipitation sum, the temperature sum was low, and this kind of weather correlated with lower cover of bilberry. Furthermore, the lower the annual average minimum temperature was, the lower was the cover of bilberry and cowberry. On the other hand, *Melampyrum pratense* had the highest cover in the rainy years with low temperature sum. In addition to nutrient availability and climatic factors, also the amount of light regulates the abundance of plant species in forests habitats (Verhayen et al. 2012, Tonteri et al. 2016). It is likely that particularly dwarf shrubs and herbs have benefited from the increase of light level due to decrease in canopy cover.

3.5.3 Nitrogen deposition and nitrogen concentration of plants

The total nitrogen (tot N) deposition in the Valkea-Kotinen study area in 2006–2009 was at the same level in PC (3.12 kg ha^{-1} yr^{-1}) and TF (3.34 kg ha^{-1} yr^{-1}). The total inorganic N deposition (TIN=NH_4-N+NO_3-N) was higher in PC (2.63 kg ha^{-1} yr^{-1}) than in TF (1.57 kg ha^{-1} yr^{-1}) indicating tree crowns (or their epiphytes) N uptake when it passes through the canopy. On the other hand, dissolved organic N (DON) deposition was higher in TF (1.77 kg ha^{-1} yr^{-1}) than in PC (0.49 kg ha^{-1} yr^{-1}) indicating that DON was leached from needles or epiphytic organisms. Thus, the amount of DON and TIN deposition were at the same level in the forest habitat. At Iso Hietajärvi, tot N deposition in the year 2009 was also at the same level in PC (2.53 kg ha^{-1} yr^{-1}) and TF (2.40 kg ha^{-1} yr^{-1}), while TIN deposition was slightly higher in PC (1.98 kg ha^{-1} yr^{-1}) than in TF (1.71 kg ha^{-1} yr^{-1}), and DON deposition was higher in TF (0.69 kg ha^{-1} yr^{-1}) than in PC (0.55 kg ha^{-1} yr^{-1})

DON has been shown to serve as an important source of N for boreal bryophytes (Forsum et al. 2009, Salemaa et al. 2020), which is likely also in the Valkea-Kotinen forest. The tot N concentration of the upper part of the bryophyte species in 2009 were as follows: *Hylocomium splendens* 1.70%, *Pleurozium schreberi* 1.58%, *Dicranum* spp. 2.11% and other species 1.71%. The occurrence of nitrophilic bryophytes (*Sciuro-hypnum*, *Brachythecium* and *Plagiothecium* genera) indicates relatively high nitrogen level of the site. N concentration of bryophytes, herbs, grasses and bilberry leaves were at the same level (1.7–2.0%). The TF tot N deposition level of 3.3 kg ha^{-1} and bryophyte N concentration of 1.48%

have been found to cause inhibition of biological N_2 fixation by cyanobacteria living on *H. splendens* (Salemaa et al. 2019). The Valkea-Kotinen monitoring plot showed corresponding tot N deposition level and higher N concentration of bryophytes (1.7%). It is possible that bryophytes were approaching N saturated stage already at this N level in forest. Although TIN in PC has showed a decreasing trend in Valkea-Kotinen study area, the earlier higher N deposition might have been accumulated in the forest ecosystem, and now it appeared as high N concentration in bryophytes. The results emphasize the importance of bryophyte layer in accumulating different N forms and affecting internal N flux of the forest ecosystem. This should be considered when evaluating the critical N loads for the most sensitive organisms of boreal forests.

At Iso Hietajärvi, the N concentration of all bryophyte species as well as evergreen dwarf shrubs were relatively low. The total N concentration of the upper part of the bryophyte species in 2009 were as follows: *Hylocomium splendens* 0.87%, *Pleurozium schreberi* 1.02%, *Dicranum* spp. 1.13% and other bryophyte species 1.04%. In the lichen species (*Cetraria islandica* and *Cladina* spp.) the average N concentration was 0.4%. These N concentrations were so low that it can be concluded that bryophytes had not reached the N saturated stage in their tissues. In fact, according to earlier studies TF deposition level should be over 3.3 kg ha^{-1} and bryophyte N concentration over 1.4% to cause inhibition of N_2 fixation by cyanobacteria living on *H. splendens* and *P. schreberi* (Salemaa et al. 2019). The corresponding values of these variables were lower in the Hietajärvi data, indicating that bryophyte growth was here N limited. Similarly, the N concentration of dwarf shrubs was low (mostly < 1%). Only leaves of bilberry and *Melampyrum pratense* had little higher concentration, 1.4 and 1.9%, respectively. All these biological indicators suggest that the N deposition level in the Iso Hietajärvi forest area is under the critical N load.

3.6 **Conclusions**

Valkea-Kotinen and Iso Hietajärvi demonstration sites are pristine, sensitive Natura 2000 areas in South and Eastern Finland, respectively. The catchments are located inside a conservation area, and therefore are not affected by direct human disturbance. The results verify that even remote pristine ecosystems, such as protected Natura 2000 areas, are susceptible to harmful environmental changes due to global pressures. On the other hand, the ecosystems have resilience to recover, if impacts of global change drivers are decreasing.

The international emission abatement actions for air pollutants have led to a recovery from acidification, and to a lesser extent, a decrease in trace metal loadings. However, processes regulating sulphur retention and release in the catchment are still not fully understood. The increase in dissolved organic matter and organic carbon concentration and consequent brownification of lake waters may have large ecological impacts on lake ecosystems and changing carbon dynamics in the lakes is one the key challenges in the future. These processes – net release of sulphate and browning of lakes – have been mainly driven by changed acid deposition, but climate-driven changes in hydrological conditions are becoming increasingly important, as atmospheric SO_4 input has declined. Another challenge is the enrichment in nutrients due to changing in-lake processes and climate-driven conditions, which may play an important role in affecting the processes in pristine lakes.

The calculated critical loads of acidity and eutrophication were not exceeded at the catchment area of Iso Hietajärvi and are no longer exceeded at the catchment area of Valkea-Kotinen. In concert with decreasing eutrophication critical loads, also the inorganic nitrogen concentrations have decreased.

Precipitation is strongly modified before it enters from the terrestrial part of the Valkea-Kotinen and Iso Hietajärvi catchments to the surface water. For example, when precipitation passed through the canopy to the soil, concentrations of inorganic nitrogen decreased due to the uptake of tree canopy and other vegetation, while for example base cation concentrations mainly increased due to leaching and wash-off nutrients from the canopy. Long-term trends in concentrations confirmed that observed decrease in SO_4-S deposition was seen also in different parts of the forest ecosystems. Instead, increase in DOC concentration, as has been observed in surface waters throughout Europe and North America, was not noticeable in the forest part of the Valkea-Kotinen and Iso Hietajärvi regions, only DOC concentration in throughfall and soil water at depth of 20 cm increased significantly over studied period, respectively. Clear effects of extreme weather events to the forest ecosystems were not found, which probably indicates the resilience of the old-growth forests.

Over 20 years of monitoring has produced valuable knowledge of the succession of undisturbed old-growth forests in Valkea-Kotinen and Iso Hietajärvi. However, many natural factors are intermingled with the climatic factors regulating the cover changes of plant species in boreal forests, and it is difficult to draw conclusions whether climate change already has affected the understorey vegetation. At Valkea-Kotinen, the cover of bilberry showed an increasing trend during the monitoring period 1998–2019, but its cover decreased in dry years. It is probable that bilberry benefits from a slight increase of temperature, but it suffers if extreme dry periods in summers are yearly repeated and stay long time. Shading trees act against the effects of climate change, because canopy prevents heating of the forest floor. The canopy cover of the study plot decreased during the monitoring period possibly due to crown defoliation and falling of old and decayed trees. In the forest area of the Valkea-Kotinen catchment the N concentration of bryophytes is still relatively high (1.7%). This may indicate that earlier higher N deposition has accumulated in the forest ecosystem, and it now appears as high N concentration in bryophytes and possible progression towards N saturation.Therefore, it is possible that slow changes in the forest ecosystem, e.g. in the structure of bryophyte communities take place already at N deposition level < 5 kg ha^{-1} yr^{-1}. This should be taken into account when evaluating the critical N loads for the most sensitive organisms of boreal forests. If the excess of nitrogen in TF deposition is available for other organisms and plants, and amount of light increases because of tree falling, eutrophication of plant communities is possible (Verheyen et al. 2012, Dirnböck et al. 2014). Therefore, nitrogen economy of bryophytes has many effects on the function of boreal forest ecosystem. At Iso Hietajärvi, the cover % of cowberry showed an increasing trend during the study period 1998–2019, and there was a slight covariation in the cover % and the annual temperature sum. On the other hand, the cover % of bilberry has changed only little. If cowberry intensify its biomass production due to increased temperature, this may cause changes in the other plant community and affect even nutrient fluxes in the forest ecosystem. The low N deposition and low N concentrations of bryophytes and vascular species indicated that biomass production of terrestrial plants was N limited and the deposition level was under the critical N load in the studied forest ecosystem.

Our assessment strongly emphasizes the importance of the integrated aquatic and terrestrial long-term monitoring on the effects of air pollution, climate change and their interactions, due to the complex processes involved. Ecological monitoring under international agreements and legislation, such as United Nations Economic Commission for Europe Convention on Long-Range Transboundary Air Pollution (UNECE CLRTAP) and National Emission Ceilings Directive (NECD), are key activities set up to evaluate the effects, not only of emission reduction policies, but also of the changing climate.

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Annex I

Report on National ICP IM activities in Austria

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The only ICP Integrated Monitoring station in Austria, Zöbelboden, is located in the northern part of the National Park Kalkalpen. Its altitude ranges from 550 to 956 m.a.s.l. and its area is 5.7 km^2 (Fig. 1). Mean monthly temperature varies from 1°C in January to 15.5°C in August. The average temperature is 7.2 (at 900 m.a.s.l.). Annual precipitation ranges from 1500 to 1800 mm and snow accumulates commonly between October and May with an average duration of about 4 months. Due to the dominance of dolomite, the catchment is not as heavily karstified as limestone karst systems but shows typical karst features such as conduits and sink holes. The site can be split into steep slopes (30–70°, 550–850 m.a.s.l.) and a plateau (850–950 m.a.s.l.). Chromic Cambisols and Hydromorphic Stagnosols with an average thickness of 50 cm and Lithic and Rendzic Leptosols with an average thickness of 12 cm can be found at the plateau and the slopes, respectively. Both the plateau and the slopes are mainly covered by forest. At the plateau Norway spruce (*Picea abies* (L.) Karst.) interspersed with beech (*Fagus sylvatica* L.) was planted after a clear cut around the year 1910. The vegetation at the slopes is dominated by semi-natural mixed mountain forest with beech as the dominant species, Norway spruce, maple (*Acer pseudoplatanus*), and ash (*Fraxinus excelsior*). At the slopes no forest management has been conducted since the establishment of the National Park in the year 1997.

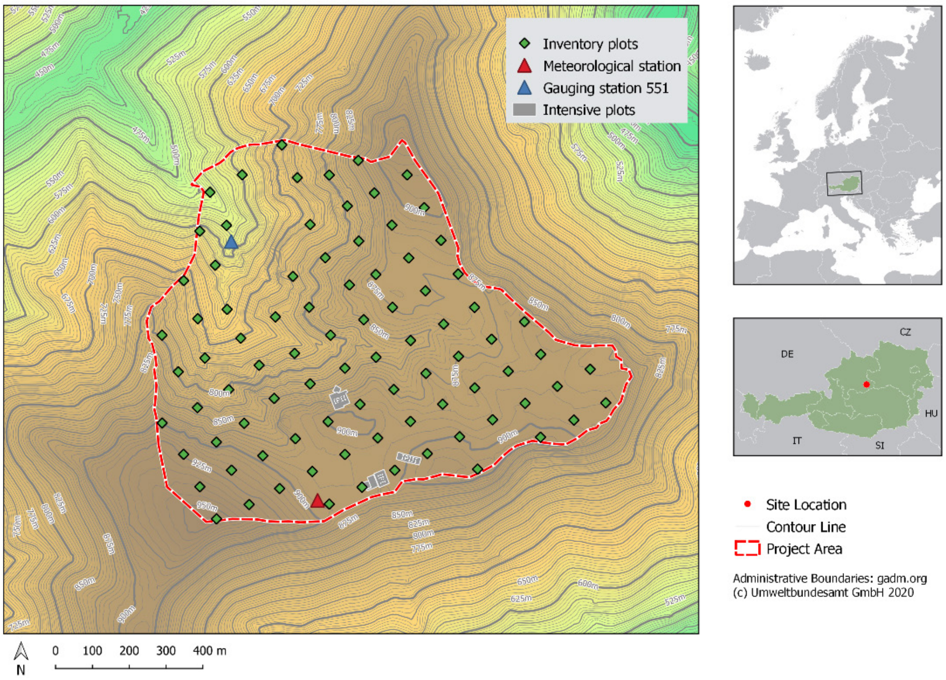

Figure 2. Location of LTER Zöbelboden with the main monitoring installations. See https://deims.org/8eda49e9-1f4e-4f3e-b58e-e0bb25dc32a6 for more details about the site, the measurement parameter, and the available data.

Measurements according to the ICP Integrated Monitoring Programme started in the year 1992. The site also hosts air pollution monitoring in the framework of EMEP and is part of the Austrian EU NEC directive sites network. Apart from reporting to the ICP Integrated Monitoring Programme, the data and metadata is publicly available at the DEIMS-SDR portal (https://deims.org/8eda49e9-1f4e-4f3e-b58e-e0bb25dc32a6). Since 2006 Zöbelboden is part of Long-term Ecological Research (LTER Austria) and serves as a research station for a number of universities and research institutes within Austria and beyond. This development led to additional instrumentation such as a CO_2 flux tower and soil chamber measurements, sapflow and automated dendrometers, and a high-resolution runoff probe for NO_3, DOC, and TOC (https://www.lter-austria.at/en/cwn-project/). Recently, automated biodiversity monitoring started in the framework of the global LifePlan project (https://www2.helsinki.fi/en/projects/lifeplan).

Here we present our newest findings regarding N deposition at Zöbelboden and its long-term effects on the ecosystem, lichens, and plants.

High ecosystem N retention

Recently we published a synthesis study using the 27 years of measurements of N deposition and effects together with a hydrological model and climatic scenarios. In this study, we used the newest findings from research carried out at Zöbelboden (Leitner et al. 2020, Hood-Nowotny et al. 2021). The Zöbelboden area was exposed to increasing N deposition during the 20th century (up to 30 to 35 kg N ha^{-1} yr^{-1}), which are still at levels of 25.5±3.6 and 19.9±4.2 kg N ha^{-1} yr^{-1} in the spruce and the mixed deciduous forests, respectively. Albeit N deposition exceeded critical loads for several decades, 70-83% of the inorganic N retained in the catchment from 2000 to 2018 and NO_3^--N concentrations in the runoff stayed < 10 mg l^{-1} unless high-flow events occurred or forest stand replacing disturbances. We identified tree growth as the main sink for inorganic N, which might, together with lower runoff, increase retention of only weakly decreasing N deposition in the future. However, since recurring forest stand replacement is predicted in the future as a result of a combination of climatically driven disturbance agents, pulses of elevated nitrate concentrations in the catchment runoff will likely add to groundwater pollution (Dirnböck et al. 2020). This result corroborates the findings from ICP's cross site analyses, where our site data has been incorporated (Vuorenmaa et al. 2018, Vuorenmaa et al. 2017) and helped evaluating critical loads (Forsius et al. 2021).

N deposition affects biological indicators

Long-term decreasing trends of epiphytic lichen diversity at Zöbelboden could be related to elevated N deposition (Mayer et al. 2013). This temporal trajectory is in line with broader scale trends and proves that lichens, though were recovering from former acidifying S deposition for some time, are now strongly affected by airborne N pollution. Vascular plants in the forest understorey are another effect related indicator for biodiversity. A trend analysis of data between 1993 and 2014 showed that N deposition, which had an effect until the year 2005, became less important relative to disturbance and climate change (Helm et al. 2017). Data from Zöbelboden were also part of a European cross-site analysis showing that N deposition has had its effect on the pauperization of forest understory in Europe (Staude et al. 2020) and that currently legislated N deposition reductions will unlikely render the expected results (Dirnböck et al. 2018).

Using the Zöbelboden for ecosystem research

Besides ICP Integrated Monitoring, the station Zöbelboden is also part of LTER Austria which is pursuing an integration in the Europe-wide eLTER Research Infrastructure in accordance with the Austrian roadmap for research infrastructures. Owing to its excellent instrumentation and long-term data, the

Zöbelboden was and is included in numerous national, European, and international research projects (FWF DICE, ÖAW C-Alps, ACRP CCN-Adapt, ACRP CentForCSink, ACRP WoodNClimate, EU Live+ EnvEurope, EU SEE Orientgate, EU ExpeER, EU eLTER, EU Horizon2020 EcoPotential, ACRP EXAFOR, EU Horizon2020 eLTER PLUS, etc.).

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Annex II

Report on National ICP IM activities in Sweden 2019

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Introduction

The Swedish integrated monitoring programme is run on four sites distributed from south central Sweden (SE14 Aneboda), over the middle part (SE15 Kindla), to a northerly site (SE16 Gammtratten). The long-term monitoring site SE04 Gårdsjön F1 is complementary on the inland of the west coast and has been influenced by long-term high deposition loads. The sites are well-defined catchments with mainly coniferous forest stands dominated by bilberry spruce forests on glacial till deposited above the highest coastline. Hence, there has been no water sorting of the soil material. Both climate and deposition gradients coincide with the distribution of the sites from south to north (Table 1).

Table 1. Geographic location and long-term climate and hydrology at the Swedish IM sites (long-term average values, 1961–1990).

Site name	SE04 Gårdsjön F1	SE14 Aneboda	SE15 Kindla	SE16 Gammtratten
Latitude; Longitude	N 58° 03´; E 12° 01´	N 57° 05´; E 14° 32´	N 59° 45´; E 14° 54´	N 63° 51´; E 18° 06´
Altitude, m	114–140	210–240	312–415	410–545
Area, ha	3.7	18.9	20.4	45
Mean annual temperature, °C	+6.7	+5.8	+4.2	+1.2
Mean annual precipitation, mm	1000	750	900	750
Mean annual evapotransporation, mm	480	470	450	370
Mean annual runoff, mm	520	280	450	380

The forest stands are mainly over 100 years old and at least three of them have several hundred years of natural continuity. Until the 1950's, the woodlands were lightly grazed in restricted areas. In early 2005, a heavy storm struck the IM site SE14 Aneboda. Compared with other forests in the region, however, this site managed rather well and roughly 20–30% of the trees in the area were storm-felled. In 1996, the total number of large woody debris in the form of logs was 317 in the surveyed plots, which decreased to 257 in 2001. In 2006, after the storm, the number of logs increased to 433, corresponding to 2711 logs in the whole catchment. In later years, 2007–2010, bark beetle (*Ips typographus*) infestation has almost totally erased the old spruce trees. In 2011 more than 80% of the trees with a diameter at breast height over 35 cm were dead (Löfgren et al. 2014) and currently almost all spruce trees with diameter of ≥20 cm are dead. Also at SE04 Gårdsjön F1, considerable natural processes have influenced the forest stand conditions during later years, with increasing number of dead trees due to both storm felling and bark beetle infestation. Occasionally, access to the site is hampered due to fallen trees, creating a need for chain saw cleaning of foot paths. Also in SE15 Kindla, an increasing number of fallen trees and logs exert perturbation, forming gaps in the forest.

In the following, presented results mainly relate to 2019 and include climate, hydrology and water chemistry as well as some ongoing work at the four Swedish IM sites (Löfgren 2020).

Climate and Hydrology in 2019

Based on long-term (1961–1990) mean values from the Swedish Meteorological and Hydrological Institute (SMHI), and measured data from climate monitoring in the IM sites, the 2019 annual mean temperatures were 1.1–1.3°C higher for all four sites. Largest deviation occurred at the southern site SE14 Aneboda. Compared with the on-site measured time series, 19 years at site SE16 Gammtratten and 23 years at the other sites, the temperatures in 2019 were somewhat higher at three sites with 0.2 and 1.3°C. SE15 Kindla had lowest difference. The most northern site SE16 Gammtratten showed a 0.3°C lower mean temperature in 2019 compared to the long-term series. The annual mean values were only slightly lower compared to the period 2014–2016 when temperatures were the highest observed for the whole measurement period with exception for SE15 Kindla where the temperature was slightly higher in the years 1999 and 2000. The variations between years have been considerable, especially for the last nine years, over 3°C at three of the sites. Smaller variations, only 1.4°C, were found at the central site SE15 Kindla. Low temperatures were observed in the years 2010 and 2012, 1.7–2.1°C below the 23 years average at three sites, while SE15 Kindla only deviated with 0.9°C below the series mean. Higher temperatures were observed for the months January to April and December for all sites. Similar conditions were observed in 2018. However, the winter temperatures at the two northern sites were still below 0°C.

Compared to the SMHI long-term average values (1961–1990), the precipitation amounts in 2019 were higher in the two southern sites, 20% and 29% at SE04 Gårdsjön and SE14 Aneboda, respectively. In SE15 Kindla the amount was exactly the long-term mean, while SE16 Gammtratten showed 20% lower precipitation compared with mean. This deviated from 2018 when all sites had lower precipitation amounts compared to the long-term mean. Distribution over months showed low values in summer apart from SE14 Aneboda with slightly more rainfall than usual. Apart from SE16 Gammtratten with lower values, the autumn months September to December exhibited larger precipitation compared with the long-term averages.

The characteristic annual hydrological patterns of the southern catchments are high groundwater levels during winter and lower levels in summer and early autumn. At the northern locations, the general picture is low groundwater levels in winter when precipitation is stored as snow, raising levels at spring snowmelt followed by lower levels in summer due to evapotranspiration and groundwater outflow. However, depending on rainfall events in summer and/or autumn, the groundwater levels could occasionally be elevated also during these periods. In 2019, the two sites SE14 Aneboda and SE16 Gammtratten started the year on low levels while site SE15 Kindla had fairly high groundwater levels. Spring snowmelt and rain at SE14 Aneboda elevated the groundwater levels in May-June. For SE16 Gammtratten evapotranspiration together with groundwater flow lowered the levels in late summer. Autumn rains turned the groundwater levels close to ground surface at one of the two stations in

SE14Aneboda and also at SE15 Kindla. The upslope station located in a well-drained slope at SE14 Aneboda had the same groundwater level pattern, but this level never reached above one meter below ground surface. At SE15 Kindla, the groundwater level generally varies in the range 0.2–0.8 m below ground surface, but the soils and piezometer location are sensitive to rain causing larger temporal variations. This deviates especially from SE16 Gammtratten piezometer site, where smooth groundwater level variations dominate at depths of 2–3 m below ground surface. The 1m depth is only reached during snowmelt, while the autumn showed small elevation dependent on few rainfall events and later also as precipitation accumulated as snow until next spring snowmelt. Compared with 2018, the groundwater levels were in general higher at all sites. Only SE15 Kindla showed almost normal levels, but the lowest groundwater level stayed at 0.8 m which was considerably higher compared with 1.5 m in 2018. The groundwater levels in 2019 were closer to those observed in 2016–2017.

The groundwater levels were reflected in the stream water discharge patterns (Fig. 1).

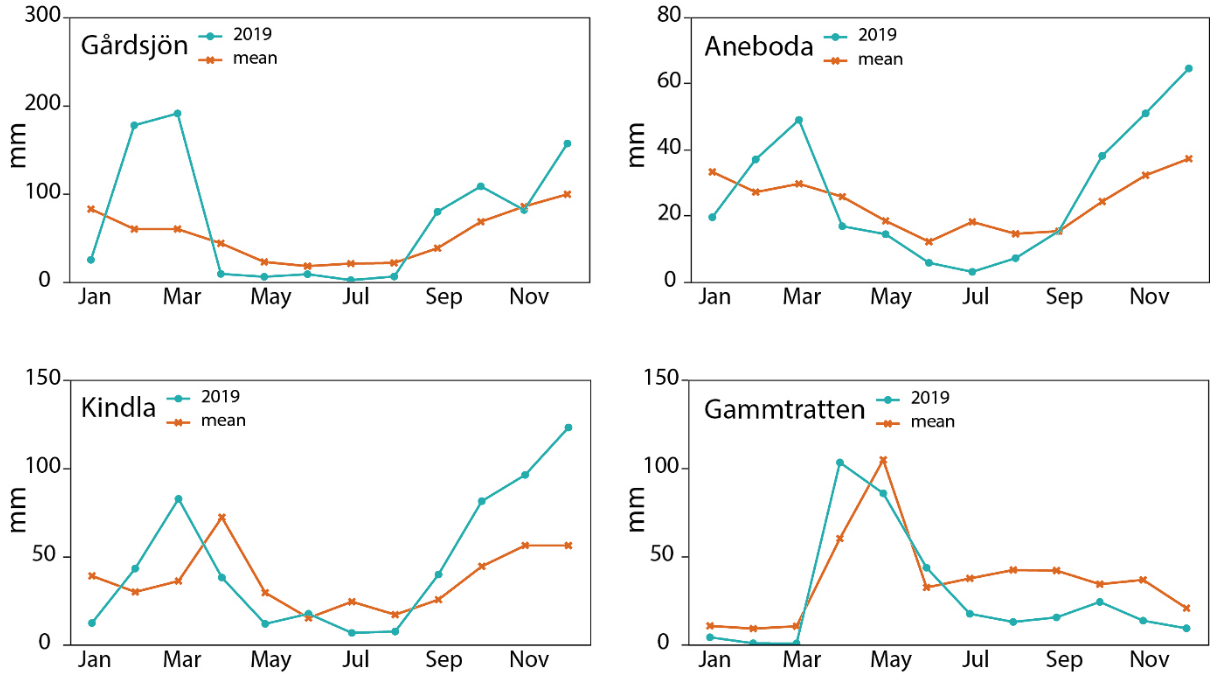

Figure 1. Monthly discharges at the four Swedish IM sites in 2019 compared with the monthly averages for the period 1996–2019 (mean). Note the different scales on the Y-axis.

Precipitation, evapotranspiration and groundwater levels affect the runoff patterns. The runoff pattern for SE16 Gammtratten 2019 was fairly typical with a snowmelt peak in April and May and lower discharges in summer and autumn, but with a small peak in October before low temperatures caused snowfall and snow accumulation. An earlier snowmelt peak was noticed compared with 2018 and the long-term pattern. At SE04 Gårdsjön, the pattern deviated from the long-term mean with high discharge in February-March not observed in 2018. In summer and autumn 2019 the discharges were similar to mean values with increasing discharges from September to December. Runoff at SE15 Kindla showed an early snowmelt peak in March and comparably low summer runoff. Autumn rains increased the discharges from September to December, reaching higher levels than usual at the end of the year. In 2019, SE14 Aneboda showed similar runoff pattern as SE15 Kindla with a peak runoff in March, low discharge in summer with no runoff during a short period in July, whereafter runoff increased (Fig. 1).

At the two northern sites, generally, snow accumulates during winter, resulting in low groundwater levels and low stream water discharge. However, warm winter periods with temperatures above 0°C have during a number of years contributed to snowmelt and excess runoff also during this season. In December 2019, only the most northern site SE16 Gammtratten had low flows while the other sites showed high runoff (Fig. 1). In January, runoff was low at the two most northern sites, but the flow started to increase already in February at SE15 Kindla. The 2018 runoff patterns deviated somewhat from this pattern with low runoff throughout the year and especially during summer and autumn. In spring, normal levels with snowmelt peaks occurred.

In 2019, the annual runoff made up 44–69% of the annual precipitation (Table 2), which is similar compared to the normal 40–60%. The highest value belongs to SE04 Gårdsjön, which had similar values in 2018 and 2017 when the proportion was 64% and 63%, respectively. The reason is high runoff at the end of the year when evapotranspiration is low. At SE04 Gårdsjön, it is quite normal that runoff constitutes almost 2/3 of the precipitation. At SE14 Aneboda, storm felling, followed by bark beetle attacks, have reduced the forest canopy cover, inducing low interception. For 2019, the total evapotranspiration was estimated to 409 mm, which is within the range of previous years, 477 mm in 2017 and 349 mm in 2016. In 2018, the evapotranspiration was much lower, only 179 mm. Low precipitation and dry conditions contributed to this low evapotranspiration 2018. In 2016 and 2018, the annual runoff range was wider than usual and made up 31–83% of the annual precipitation at the four sites.

Table 2. Compilation of the 2019 water balances for the four Swedish IM sites. P – Precipitation, TF – Throughfall, I – Interception, R – Water runoff.

	Gårdsjön SE04		**Aneboda SE14**		**Kindla SE15**		**Gammtratten SE16**	
	mm	**% of P**	**mm**	**% of P**	**mm**	**% of P**	**mm**	**% of P**
Bulk precipitation, P	1240	100	731	100	1072	100	618	100
Throughfall, TF	991	80	715	98	1052	98	581	94
Interception, P-TF	248	20	16	2	20	2	37	6
Runoff, R	851	69	322	44	559	52	330	53
P-R	389	31	409	56	513	48	287	47

At SE15 Kindla, the 2019 water balance was influenced by relatively high precipitation, resulting in normal distribution between evapotranspiration and runoff, 48% and 52%, respectively. In 2018, evapotranspiration was 56% and runoff 44%. At the northern site SE16 Gammtratten, throughfall and bulk precipitation were fairly similar with 94% throughfall. Similar pattern was observed during several years. Presumably, snow deposition infers large uncertainties, resulting in erroneous estimates of especially bulk precipitation. A nearby SMHI station reports higher precipitation, generating more realistic evapotranspiration. However, both evapotranspiration and runoff were reasonable related to precipitation in 2019. In summary and based on the estimated evapotranspiration (P-R), it seems that the very dry summer 2018 furnished low evapotranspiration at all four sites. Year 2019 showed more reasonable evapotranspiration (Table 2).

Water chemistry in 2019

Low ion concentrations in bulk deposition (electrolytical conductivity 1–2 mS m^{-1}) characterise all four Swedish IM sites. The concentrations of ions in throughfall, including dry deposition, were higher at the three most southern sites. At the northern site SE16 Gammtratten, the conductivity in throughfall (0.8 mS m^{-1}) was almost the same as in bulk deposition indicating very low sea salt deposition and uptake of ions by the trees. At the two most southern sites, sea salt deposition provides tangibly higher ion concentrations, especially at the west coast SE04 Gårdsjön site (4.0 mS m^{-1} in throughfall).

The groundwater pathways are fairly short and shallow in the catchments, providing rapid soil solution flow paths from infiltration to surface water runoff. However, the conductivity in soil water was higher compared to throughfall showing influences from evapotranspiration and soil chemical processes. The deposition acidity has during the last 10 years been rather similar at all sites with somewhat higher pH values (0–0.3 units) in throughfall compared with bulk deposition. In 2019, however, SE04 Gårdsjön, SE15 Kindla and SE16 Gammtratten had similar pH (5.1–5.3) in both bulk deposition and throughfall. SE14 Aneboda had 0.3 pH-units higher pH in throughfall compared with bulk deposition (Table 3).

Table 3. Mean deposition chemistry values 2019 at the four Swedish IM sites. S and N in kg ha^{-1} yr^{-1}.

	SE04	SE14	SE15	SE16
pH, bulk deposition	5.0	5.1	5.3	5.1
pH, throughfall	5.2	5.4	5.3	5.2
S, bulk deposition	3.4	1.8	2.4	0.8
N, bulk deposition	8.7	5.7	5.6	2.0

During the soil solution passage through the catchment soils, organic acids were added and leached on its way to stream runoff. In the upslope recharge areas, pH in soil water in the upper soil layers (E-horizon) was mainly lower than in throughfall with about 0.6 pH-units for the three southern sites. SE16 Gammtratten had the lowest pH-value 3.8 in soil water, being 1.4 units lower compared with throughfall. However, in the organic rich discharge areas at SE15 Kindla and SE16 Gammtratten, pH was higher in soil solution compared with throughfall, while the opposite occurred at SE14 Aneboda and SE04 Gårdsjön.

In the recharge areas, the buffering capacity in soil water and groundwater varied between negative and positive values, but values were most frequently on the negative side, especially for SE14 Aneboda, -0.08 mEq L^{-1}. This may be an effect of nitrification. In the discharge areas, the buffering capacity in groundwater was considerable, 0,07 mEq L^{-1}. In groundwater at SE15 Kindla, ANC was high, 0.21 mEq L^{-1}, while SE04 Gårdsjön and SE16 Gammtratten showed lower values 0.03 and 0.05 mEq L^{-1}, respectively and were lower compared with SE14 Aneboda. Bicarbonate (HCO_3) occurred at SE15 Kindla and SE16 Gammtratten, but not at SE14 Aneboda and possibly not at SE04 Gårdsjön. The latter is not measured but indicated by the very low pH of 4.4.

The stream waters were acidic with pH values below 4.7 at all sites except SE16 Gammtratten having a pH of 5.6. The stream water buffer capacity was positive at all sites (ANC $\geq$ 0.035 mEq L^{-1}), however close to zero at SE15 Kindla (ANC $\approx$ 0.001 mEq L^{-1}). Anions of weak organic acids and bicarbonate alkalinity contributed to the positive ANC (0.1 mEq L^{-1}) at SE16 Gammtratten. At SE15 Kindla and SE04 Gårdsjön, the stream waters were more acidic compared with the other two sites probably due to oxidation of organically bound sulphur related to the legacy from earlier sulphur deposition.

The share of major anions in bulk deposition was similar for sulphate, chloride and nitrate at three of the sites, while chloride dominated at SE04 Gårdsjön due to the proximity to the sea. Sea salt showed clear influences on throughfall at SE04 Gårdsjön and also at SE14 Aneboda indicating effects of dry deposition. In throughfall, organic anions contributed significantly at all four sites. The chemical composition changed along the flow paths through the catchment soils and e.g. sulphate concentrations were higher in stream water compared with deposition, indicating desorption or mineralization of previously accumulated sulphur in the soils. For Aneboda, nitrification has contributed to relatively high nitrate values in the recharge area soil water (0.05 mEq L^{-1}), however, values turning rather low in 2019 with concentration only 0.012 mEq L^{-1}. Lower concentrations occurred in the discharge areas, probably due to nitrogen uptake and denitrification.

At SE16 Gammtratten site in the north, sulphate concentrations in soil water and stream water were higher compared to throughfall, indicating release from the soil pool. Organic anions and HCO_3^- dominated the stream water anion flow (2/3 of the anions) to be compared with 1/3 at SE15 Kindla and 1/6 at SE14 Aneboda and SE04 Gårdsjön.

Base cation relations indicate soil and soil water processes. In deposition, Na^+ dominated the base cations for all sites. This was valid also in stream water except for at the northern site SE16 Gammtratten where Ca^{2+} showed the highest concentrations. At sites SE04 Gårdsjön and SE14 Aneboda, Cl^- concentrations were similar with Na^+ while Na^+ dominated at the other two sites. A higher outflow of Na^+ compared to Cl^- in stream water indicates weathering of minerals and release of base cations. Mg^{2+} was the second highest base cation in runoff water at SE04 Gårdsjön, also a result of the marine influence, while Mg^{2+} and Ca^{2+} were quite equal at the other three sites.

Besides effects on ANC and pH, the stream water chemistry was to a considerable extent influenced by organic matter. At SE14 Aneboda, the DOC concentration was high, 28 mg L^{-1}, while SE04 Gårdsjön showed 17 mg DOC L^{-1} and the two northern sites had ca 10 mg DOC L^{-1}. High DOC concentrations create prerequisites for metal complexation and transport as well as high fluxes of organic nitrogen. This was the dominating nitrogen fraction in all stream waters, ranging from 0.21 to 0.67 mg N_{org} L^{-1}. The shares of N_{org}/N_{tot} were 88–97%, SE16 Gammtratten having the highest share and the other sites very similar values, 88–89%. Inorganic N concentrations were low (≤0.04 N_{inorg} L^{-1}) at three sites at SE14 Aneboda considerably higher (0.08 N_{inorg} L^{-1}). However, the inorganic nitrogen flux was higher at the two southern sites compared to the northern ones with 0.26 and 0.46 kg N_{inorg} ha^{-1} yr^{-1}, in the north 0.02–0.12 kg N_{inorg} ha^{-1} yr^{-1}. The high concentrations in stream water at SE14 Aneboda, probably reflect the poor forest stand condition. Compared with 2012, however, when the mean inorganic N concentration exceeded 1100 µg L^{-1}, a considerable decrease has occurred.

Total phosphorus concentrations (P_{tot}) in bulk deposition varied between 5 and 36 µg L^{-1} with the highest value at SE14 Aneboda and lowest at SE16 Gammtratten. In stream water, SE14 Aneboda showed the highest P_{tot} (26 µg L^{-1}) as well as DOC concentrations. The other sites had average P_{tot} concentrations between 3 and 9 µg L^{-1} with the lowest value at SE15 Kindla.

Inorganic aluminum (Al_i), toxic to fish and other gill-breathing organisms, is analysed in soil solution, groundwater and surface waters at the IM sites. Relatively high total Al concentrations occurred in the soil solution (0.5–1.6 mg L^{-1}), and fairly high concentrations (1.1 and 0.7 mg L^{-1}, respectively) occurred also in groundwater in the recharge areas at SE04 Gårdsjön and SE14 Aneboda. The stream water Al_{tot}-concentrations were between 0.5 and 0.7 mg L^{-1} at the three sites with low pH (4.5–4.7). The Al_{tot} concentrations were lower, approximately 0.24 mg L^{-1} at the northern site SE16 Gammtratten with a pH of 5.6. Inorganic Al made up 15–58% of the total Al with the highest levels at low pH at SE15 Kindla and the lowest at SE04 Gårdsjön, corresponding to 0.05–0.30 mg Al_i L^{-1}. According to the SEPA classification system, the Al_i concentrations at SE14 Aneboda and SE15 Kindla are considered *extremely high*, but *moderate* at SE04 Gårdsjön and SE16 Gammtratten.

The priority heavy metals Pb, Cd and Hg were still accumulating in the SE14 Aneboda catchment soils, while the stream concentrations were low compared with the levels causing biological effects. Only Pb had somewhat higher concentrations compared with the established limits for ecological effects. However, methyl mercury (Hg_{Me}), only measured at SE14 Aneboda, was still relatively high creating prerequisites for bioaccumulation. In stream water, the mean Hg_{tot} and Hg_{Me} concentrations were

10.5 ng L^{-1} and 1.5 ng L^{-1}, respectively. Half of the Hg_{tot} deposition was accumulated in the catchment soil and higher concentrations of Hg_{Me} in stream water compared with values in throughfall, indicate ongoing methylation. Heavy metal analyses were financed by SITES.

In summary, the four Swedish IM sites show low ion concentrations and permanently acidic conditions in the aquatic media. In stream water, only the northern site SE16 Gammtratten had buffering capacity related to bicarbonate alkalinity. Organic matter has an impact on the water quality with respect to colour, metal complexation, and phosphorus concentrations at all sites, but less at SE15 Kindla, where rapid soil water flow paths provide relatively low DOC concentrations but acidic waters. At SE14 Aneboda, the forest dieback provides a relatively high share of runoff as well as high nitrate concentrations compared with the other three sites. At SE04 Gårdsjön, deposition is strongly influenced by the sea.

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