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29th Annual Report 2020

Convention on Long-range Transboundary Air Pollution

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

Sirpa Kleemola and Martin Forsius (eds.)

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**International Cooperative Programme
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wge Working Group on Effects of the
Convention on Long-range
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Helsinki 2020

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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 summarises 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 2019/2020 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 temporal trends and input - output budgets of heavy metals in ICP IM catchments
- An interim assessment of the impact of internal nitrogen-related parameters and exceedances of critical loads of eutrophication on long-term changes in the inorganic nitrogen output in European ICP Integrated Monitoring catchments
- National Reports on ICP IM activities are presented as annexes.

Keywords: Integrated Monitoring, ecosystems, small catchments, air pollution, critical loads, dynamic modelling

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 2019/2020, joka sisältää:

- Lyhyen yhteenvedon ohjelmassa aiemmin tehdyistä arvioinneista
- Kuvauksen ICP IM ohjelman toiminnasta ja ohjelman seurantaverkosta
- Katsauksen raskasmetallipitoisuuksien trendeihin ja raskasmetallien massataseisiin ICP IM alueilla
- Väliraportin epäorgaanisen typen huuhtoutumisen pitkän ajan muutoksista ICP IM alueilla ja eri valuma-aluetekijöiden sekä rehevöitymisen kriittisen kuormituksen ylityksen vaikutuksista kuormituksen vaihteluun
- Kuvauksia kansallisesta ICP IM toiminnasta eri maissa liitteenä.

Asiasanat: Yhdennetty ympäristön seuranta, ekosysteemit, pienet valuma-alueet, ilmansaasteet, kriittinen kuormitus, dynaamiset mallit

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 2019/2020. Rapporten innehåller:

- En sammanfattning av programmets nuvarande omfattning och databasens innehåll
- En syntes av tidigare utvärderingar av data från programmet
- En rapport om trender och massbalanser av metaller i ICP IM områden
- En rapport om långsiktiga förändringar i flöden av oorganiskt kväve från ICP IM områden – med beaktande av avrinningsområdets egenskaper och gränsvärden för luftburen kvävebelastning
- Beskrivning av nationella ICP IM aktiviteter.

Nyckelord: Integrerad miljöövervakning, ekosystem, små avrinningsområden, luftföroreningar, kritisk belasting, dynamiska modeller

Contents

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 Road map. 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

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.

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 23rd Annual 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.

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. Thus, the next phase of the work on trend assessment will be an assessment of the role of internal nitrogen parameters (Vuorenmaa et al. in prep.).

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 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.

Previous 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 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. In this report (see Chpter 2) we evaluate if the declining trends in atmospheric deposition of Cd, Pb and Hg during recent decades are reflected in the runoff concentrations from European catchments within the ICP IM network. In addition to the direct effect of reduced deposition of metals during the last decades, less metals may also be mobilised from terrestrial soils to surface waters as a result of recovery from acidification during the same period (Lydersen et al. 2002). In this report, also the catchment Cd, Pb and Hg input-output budgets are calculated for the four ICP IM sites in Sweden.

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 phenomena of land-atmosphere exchange has been known for 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 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 mobilise 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 utilised 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

Empirical impact indicators of acidification and eutrophication were determined from stream water chemistry and runoff observations at ICP IM catchments (Holmberg et al. 2013). 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 a central ICP IM database at the Programme Centre. However, a possible migration of the data base and its maintenance from the Programme Centre to the ICP IM lead country (Sweden) is under investigation.
- 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.

References

Åkerblom, S. & Lundin, L. 2015. Progress report on heavy metal trends at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 24th Annual Report 2015. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 31/2015, pp. 32–36.

Åkerblom, S. & Lundin, L. 2017. Report on concentrations of heavy metals in important forest ecosystem compartments. In: Kleemola, S. & Forsius, M. (Eds.) 26th Annual Report 2017. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 24/2017, pp. 36–42.

Bringmark, L. 2011. Report on updated heavy metal budgets and critical loads. In: Kleemola, S. & Forsius, M. (Eds.) 20th Annual Report 2011. ICP Integrated Monitoring. The Finnish Environment 18/2011, pp. 33–35. Finnish Environment Institute, Helsinki.

Bringmark, L., Lundin, L., Augustaitis, A., Beudert, B., Dieffenbach-Fries, H., Dirnböck, T., Grabner, M-T., Hutchins, M., Krám, P., Lyulko, I., Ruoho-Airola, T. & Váňa, M. 2013. Trace Metal Budgets for Forested Catchments in Europe – Pb, Cd, Hg, Cu and Zn. Water, Air, and Soil Pollution, 224: 1502, 14p.

Dirnböck, T., Grandin, U., Bernhard-Römermann, M., Beudert, B., Canullo, R., Forsius, M., Grabner, M.-T., Holmberg, M., Kleemola, S., Lundin, L., Mirtl, M., Neumann, M., Pompei, E., Salemaa, M., Starlinger, F., Staszewski, T. & Uzięblo, A. K. 2014. Forest floor vegetation response to nitrogen deposition in Europe. Global Change Biology 20: 429–440.

Dirnböck, T., Holmberg, M. & Pröll, G. 2018a. Progress report on dynamic soil-vegetation modelling. In: Kleemola, S. & Forsius, M. (Eds.). 27th Annual Report, International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems. Reports of the Finnish Environment Institute 20/2018: 30–33.

Dirnböck, T., Pröll, G., Austnes, K., Beloica, J., Beudert, B., Canullo, R., De Marco, A., Fornasier, M.A., Futter, M., Goergen, K., Grandin, U., Holmberg, M., Lindroos, A.J., Mirtl, M., Neirynck, J., Pecka, T., Nieminen, T.M., Nordbakken, J.F., Posch, M., Reinds, G.J., Rowe, E., Salemaa, M., Scheuschner, T., Starlinger, F., Uzięblo, A.K., Valinia, S., Weldon, J. Wamelink, W. & Forsius, M. 2018b. Currently legislated decreases in nitrogen deposition will yield only limited plant species recovery in European forests. Environmental Research Letters 13 (2018) 125010.

Dise, N.B, Matzner, E. & Forsius, M. 1998. Evaluation of organic horizon C:N ratio as an indicator of nitrate leaching in conifer forests across Europe. Environmental Pollution 102, S1: 453–456.

Driscoll, C.T. 1984. A procedure for the fractionation of aqueous aluminum in dilute acidic waters. International Journal of Environmental Analytical Chemistry 16: 267–284.

Forsius, M., Kleemola, S. & Vuorenmaa, J. 1996. Assessment of nitrogen processes at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 5th Annual Report 1996. UNECE ICP Integrated Monitoring. The Finnish Environment 27, pp. 25–38. Finnish Environment Institute, Helsinki.

Forsius, M., Alveteg, M., Bak, J., Guardans, R., Holmberg, M., Jenkins, A., Johansson, M., Kleemola, S., Rankinen, K., Renshaw, M., Sverdrup, H. & Syri, S. 1997. Assessment of the Effects of the EU Acidification Strategy: Dynamic modelling on Integrated Monitoring sites. Finnish Environment Institute, Helsinki. 40 p.

Forsius, M., Alveteg, M., Jenkins, A., Johansson, M., Kleemola, S., Lükewille, A., Posch, M., Sverdrup, H. & Walse, C. 1998a. MAGIC, SAFE and SMART model applications at Integrated Monitoring Sites: Effects of emission reduction scenarios. Water, Air, and Soil Pollution 105: 21–30.

Forsius, M., Guardans, R., Jenkins, A., Lundin, L. & Nielsen, K.E. (Eds.) 1998b. Integrated Monitoring: Environmental assessment through model and empirical analysis – Final results from an EU/LIFE-project. The Finnish Environment 218. Finnish Environment Institute, Helsinki, 172 p.

Forsius, M., Kleemola, S., Vuorenmaa, J. & Syri, S. 2001. Fluxes and trends of nitrogen and sulphur compounds at Integrated Monitoring Sites in Europe. Water, Air, and Soil Pollution 130: 1641–1648.

Forsius, M., Kleemola, S. & Starr, M. 2005. Proton budgets for a monitoring network of European forested catchments: impacts of nitrogen and sulphur deposition. Ecological Indicators 5: 73–83.

Forsius, M. & Nyman, M. (Eds.) 2006. AMAP assessment 2006: acidifying pollutants, arctic haze, and acidification in the Arctic. Oslo, Arctic Monitoring and Assessment Program (AMAP). 112 p. www.amap.no.

Futter, M., Starr, M., Forsius, M. & Holmberg, M. 2008. Modelling long-term patterns of dissolved organic carbon concentrations in the surface waters of a boreal catchment. Hydrology and Earth System Sciences 12: 437–447.

Futter, M.N., Forsius, M., Holmberg, M. & Starr, M. 2009. A long-term simulation of the effects of acidic deposition and climate change on surface water dissolved organic carbon concentrations in a boreal catchment. Hydrology Research 40: 291–305.

Gensemer, R.W. & Playle, R.C. 1999. The bioavailability and toxicity of aluminum in aquatic environments. Critical Reviews in Environmental Science and Technology 29: 315–450.

Gundersen, P., Berg, B., Currie, W. S., Dise, N.B., Emmett, B.A., Gauci, V., Holmberg, M., Kjønaas, O.J., Mol-Dijkstra, J., van der Salm, C., Schmidt, I.K., Tietema, A., Wessel, W.W., Vestgarden, L.S., Akselsson, C., De Vries, W., Forsius, M., Kros, H., Matzner, E., Moldan, F., Nadelhoffer, K. J., Nilsson, L.-O., Reinds, G.J., Rosengren, U., Stuanes, A.O. & Wright, R.F. 2006. Carbon-Nitrogen Interactions in Forest Ecosystems – Final Report. Forest & Landscape Working Papers no. 17–2006, Danish Centre for Forest, Landscape and Planning, KVL. 62 p.

van Herk, C. M., Mathijssen-Spiekman, E. A. M. & de Zwart, D. 2003. Long distance nitrogen air pollution effects on lichens in Europe. The Lichenologist 35 (4): 347–359.

Holmberg, M., Vuorenmaa, J., Posch, M., Forsius, M., Lundin, L., Kleemola, S., Augustaitis, A., Beudert, B., de Wit, H.A., Dirnböck, T., Evans, C.D., Frey. J., Grandin, U., Indriksone. I., Krám, P., Pompei, E., Schulte-Bisping, H., Srybny, A. & Vána, M. 2013. Relationship between critical load exceedances and empirical impact indicators at Integrated Monitoring sites across Europe. Ecological Indicators: 24:256–265.

Holmberg, M. & Dirnböck, T. 2015. Progress report on dynamic vegetation modelling at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 24th Annual Report 2015. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 31/2015, pp. 23–27.

Holmberg, M. & Dirnböck, T. 2016. Dynamic vegetation modelling at ecosystem monitoring and research sites. In: Kleemola, S. & Forsius, M. (Eds.) 25th Annual Report 2016. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 29/2016, pp. 27–33.

Holmberg, M., Vuorenmaa, J., Posch, M., Kleemola, S., Augustaitis, A., Beudert , B., de Wit, H.A, Dirnböck, T., Frey, J., Forsius, M., Hakola, H., Kobler, J., Krám, P., Lundin, L. & Váňa, M. 2017. Relationship between critical load exceedances and empirical impact indicators at IM sites - Update 2017. In: Kleemola, S. & Forsius, M. (Eds.) 26th Annual Report 2017. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 24/2017, pp. 29–35.

Holmberg, M., Aherne, J., Austnes, K., Beloica, J., De Marco, A., Dirnböck, T., Fornasier, M.F., Goergen, K., Futter, M., Lindroos, A.J., Krám, P., Neirynck, J., Nieminen, T.M., Pecka, T., Posch, M., Rowe, E.C., Scheuschner, T., Schlutow, A., Valinia, S. & Forsius, M. 2018. Modelling study of soil C, N and pH response to air pollution and climate change using European LTER site observations. Science of the Total Environment 640-641: 387-399.

ICP IM Programme Centre 1995. Assessment of nitrogen processes on ICP IM sites. In: 4th Annual Synoptic Report 1995, UNECE ICP Integrated Monitoring, pp. 19–61. Finnish Environment Agency, Helsinki.

Jenkins, A., Larssen, T., Moldan, F., Hruška, J., Krám, P. & Kleemola, S. 2003. Dynamic modelling at Integrated Monitoring sites – Model testing against observations and uncertainty. The Finnish Environment 636. Finnish Environment Institute, Helsinki. 37 p.

Jiskra, M., Wiederhold, J. G., Skyllberg, U., Kronberg, R. M., Hajdas, I. & Kretzschmar, R. 2015. Mercury deposition and re-emission pathways in boreal forest soils investigated with Hg isotope signatures. Environ. Sci. Technol. 49, (12), 7188-7196.

Kleemola, S. 2005. Trend assessment of bulk deposition, throughfall and runoff water chemistry at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 14th Annual Report 2005. ICP Integrated Monitoring. The Finnish Environment 788, pp. 32–37. Finnish Environment Institute, Helsinki.

Kleemola, S. & Forsius, M. 2006. Trend assessment of bulk deposition, throughfall and runoff water/ soil water chemistry at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 15th Annual Report 2006. ICP Integrated Monitoring. The Finnish Environment 30/2006, pp. 22–48. Finnish Environment Institute, Helsinki.

Krám, P. & Kleemola, S. 2019. Aluminium fractions in surface waters draining catchments of ICP Integrated Monitoring network. In: Kleemola, S. & Forsius, M. (eds.). 28th Annual Report 2019. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 33/2019, pp. 31–36.

Liu, Q. 1996. Vegetation monitoring in the ICP IM programme: Evaluation of data with regard to effects of N and S deposition. In: Kleemola, S. & Forsius, M. (Eds.) 5th Annual Report 1996. UNECE ICP Integrated Monitoring. The Finnish Environment 27, pp. 55–79. Finnish Environment Institute, Helsinki.

Lükewille, A., Jeffries, D., Johannessen, M., Raddum, G., Stoddard, J. & Traaen, T. 1997. The nine year report: Acidification of surface water in Europe and North America. Long-term developments (1980s and 1990s). Norwegian Institute for Water Research, Oslo. NIVA Report 3637–97.

MacDonald, J.A., Dise, N.B., Matzner, E., Armbruster, M., Gundersen, P. & Forsius, M. 2002. Nitrogen input together with ecosystem nitrogen enrichment predict nitrate leaching from European forests. Global Change Biology 8: 1028–1033.

Manual for Integrated Monitoring 1998. Finnish Environment Institute, ICP IM Programme Centre, Helsinki, Finland. Original version (https://helda.helsinki.fi/handle/10138/242414), updated version: www.syke.fi/nature/icpim > Manual for Integrated Monitoring.
Mirtl, M., Borer, E.T., Djukic, I., Forsius, M., Haubold, H., Hugo, W., Jourdan, J., Lindenmayer, D., McDowell, W.H., Muraoka, H, Orenstein. D.E., Pauw, J.C., Peterseil, J., Shibata, H., Wohner, C., Yu, X. & Haase, P. 2018. Genesis, goals and achievements of Long-Term Ecological Research at the global scale: A critical review of ILTER and future directions. Science of the Total Environment 626: 1439–1462.
Moldan, F., Wright, R.F., Löfgren, S., Forsius, M., Ruoho-Airola, T. & Skjelkvåle, B.L. 2001. Long-term changes in acidification and recovery at nine calibrated catchments in Norway, Sweden and Finland. Hydrology and Earth System Sciences 5: 339–349.
Osterwalder, S., Bishop, K., Alewell, C., Fritsche, J., Laudon, H., Åkerblom, S. & Nilsson, M. B. 2017. Mercury evasion from a boreal peatland shortens the timeline for recovery from legacy pollution. Scientific Reports 7, 16022.
Osterwalder, S., Fritsche, J., Alewell, C., Schmutz, M., Nilsson, M. B., Jocher, G., Sommar, J., Rinne, J. & Bishop, K. 2016. A dual-inlet, single detector relaxed eddy accumulation system for long-term measurement of mercury flux. Atmos. Meas. Tech. 9, (2), 509-524.
Posch, M., Johansson, M. & Forsius, M. 1997. Critical loads and dynamic models. In: Kleemola, S. & Forsius, M. (Eds.) 6th Annual Report 1997. UN ECE ICP Integrated Monitoring. The Finnish Environment 116, pp. 13–23. Finnish Environment Institute, Helsinki.
Sliggers, J. & Kakebeeke, W. (Eds.) 2004. Clearing the Air: 25 years of the Convention on Long-range Transboundary Air Pollution. Geneva, United Nations Economic Commission for Europe.167 p.
Vuorenmaa, J. 1997. Trend assessment of bulk and throughfall deposition and runoff water chemistry at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 6th Annual Report 1997. UN ECE ICP Integrated Monitoring. The Finnish Environment 116, pp. 24–42. Finnish Environment Institute, Helsinki.
Vuorenmaa, J., Kleemola, S. & Forsius, M. 2009. Trend assessment of bulk deposition, throughfall and runoff water/soil water chemistry at ICP IM sites In: Kleemola, S. & Forsius, M. (Eds.) 18th Annual Report 2009. ICP Integrated Monitoring. The Finnish Environment 23/2009, pp. 36–63. Finnish Environment Institute, Helsinki.
Vuorenmaa, J. et al. 2012. Interim report: Sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe. In: Kleemola, S. & Forsius, M. (Eds.) 21st Annual Report 2012. ICP Integrated Monitoring. The Finnish Environment 28/2012, pp.23–34. Finnish Environment Institute, Helsinki.
Vuorenmaa, J. et al. 2013. Sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe. In: Kleemola, S. & Forsius, M. (Eds.) 22nd Annual Report 2013. ICP Integrated Monitoring. Reports of the Finnish Environment Institute 25/2013, pp. 35–43.
Vuorenmaa, J. et al. 2014. Sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe in 1990-2012. In: Kleemola, S. & Forsius, M. (Eds.) 23rd Annual Report 2014. ICP Integrated Monitoring. Reports of the Finnish Environment Institute 23/2014, pp. 28–35.
Vuorenmaa, J., Augustaitis A., Beudert, B., Clarke, N., de Wit H., Dirnböck, T., Forsius, M., Frey, J., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin L., Marchetto, A., Ruoho-Airola, T., Schulte-Bisping, H., Srybny, A., Tait, D., Ukonmaanaho, L. & Váňa M. 2016. Trend assessments for deposition and runoff water chemistry concentrations and fluxes and climatic variables at ICP Integrated Monitoring sites in 1990-2013. In: Kleemola, S. & Forsius, M. (Eds.). 25th Annual Report, International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems. Reports of the Finnish Environment Institute 29/2016: 34–51.
Vuorenmaa, J., Augustaitis, A., Beudert, B., Clarke, N., de Wit, H.A., Dirnböck, T., Frey, J., Forsius, M., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L.,Ruoho-Airola, T., Ukonmaanaho, L. & Váňa, M. 2017. Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990-2012). Ecological Indicators 76: 15–29.
Vuorenmaa, J. Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H., Dirnböck, T., Frey, J., Hakola, H., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Löfgren, S., Marchetto, A., Pecka, T., Schulte-Bisping, H., Skotak, K., Srybny, A., Szpikowski, J., Ukonmaanaho, L., Váňa, M., Åkerblom, S. & Forsius, M. 2018. Long-term changes (1990–2015) in the atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in Europe in relation to changes in emissions and hydrometeorological conditions. Science of the Total Environment 625:1129–1145.
Working Group on Effects 2004. Integrated Monitoring of Ecosystems. In: Review and assessment of air pollution effects and their recorded trends. Report of the Working Group on Effects of the Convention on Long-range Transboundary Air Pollution, pp. 30–33. Geneva, United Nations Economic Commission for Europe.
Wright, R.F., Aherne, J., Bishop, K., Camarero, L., Cosby, B.J., Erlandsson, M., Evans, C.D., Forsius, M., Hardekopf, D., Helliwell, R., Hruška, J., Jenkins, A., Kopáček, J., Moldan, F., Posch, M. & Rogora, M. 2006. Modelling the effect of climate change on recovery of acidified freshwaters: Relative sensitivity of individual processes in the MAGIC model. Science of the Total Environment 365: 154–166.
de Zwart, D. 1998. Multivariate gradient analysis applied to relate chemical and biological observations. In: Kleemola, S. & Forsius, M. (Eds.) 7th Annual Report 1998. UN ECE ICP Integrated Monitoring. The Finnish Environment 217, pp. 15–29. Finnish Environment Institute, Helsinki.
de Zwart, D., van Herk, K.C.M. & Mathijssen-Spiekman, L.E.A. 2003. Long distance nitrogen air pollution effects on lichens in Europe. In: Kleemola, S. & Forsius, M. (Eds.) 12th Annual Report 2003. UN ECE ICP Integrated Monitoring. The Finnish Environment 637, pp. 32–37. Finnish Environment Institute, Helsinki.

1 ICP IM activities, monitoring sites and available data

1.1 Review of the ICP IM activities from June 1st, 2019 to June 1st, 2020

Meetings

- The twenty-seventh meeting of the Programme Task Force on ICP Integrated Monitoring was organised as a joint 2019 Task Force Meeting of ICP Waters and ICP Integrated Monitoring in Helsinki, Finland from June 4 to June 6, 2019.
- Sören Wulff (Swedish National Focal Point in ICP Forests) kindly represented ICP IM and gave a presentation on ICP IM activities (prepared by Ulf Grandin) in the 35th Task Force Meeting of ICP Forests in Ankara, Turkey, 13–14 June 2019.
- ICP IM Programme Manager Martin Forsius and Maria Holmberg represented ICP IM in the 2nd ILTER Open Science Meeting in Leipzig, Germany 2–6 September 2019.
- 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 in Geneva, Switzerland, 9–13 September 2019.
- Ulf Grandin took part in the eLTER PPP (Preparatory Phase Project) and PLUS (Advanced Community project) shareholder meeting in Berlin 8–9 October 2019.
- Martin Forsius attended NKL (Nordiska Klimat och Luftgruppen) Workshop in Oslo, Norway, 19–20 November 2019.
- Martin Forsius and Ulf Grandin represented ICP IM and Salar Valinia represented Sweden in the Convention on Long-range Transboundary Air Pollution 40th Anniversary Special Session in Geneva, Switzerland, 11–12 December 2019.
- Martin Forsius took part in the eLTER PPP and PLUS Steering Committee's planning meeting in Berlin, Germany, 18–19 February 2020.
- Martin Forsius participated in the videoconference of the Joint meeting of EMEP Steering Body and Working Group on Effects Extended Bureau, 24–26 March 2020.
- Martin Forsius, Ulf Grandin and Maria Holmberg participated in the online kick-off meeting of the two new eLTER projects - eLTER PLUS and eLTER PPP from 30 March to 3 April 2020.
- Maria Holmberg represented ICP IM in the 36th Modelling & Mapping virtual Task Force Meeting 21–22 April 2020.
- The twenty-eight meeting of the Programme Task Force on ICP Integrated Monitoring to be held in Riga, Latvia, was organised as an on-line meeting on 13–14 May 2020 due to the outbreak of Corona virus.

Projects, data issues

- After December 1st, 2019 the National Focal Points (NFPs) reported their 2018 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 in priority topics

- The Programme Centre prepared the ICP IM contribution to the Joint Report 2019 of the ICPs, TF health and Joint Expert Group on Dynamic Modelling for the WGE (ECE/EB.AIR/GE.1/2019/3– ECE/EB.AIR/WG.1/2019/3).
- A manuscript for the scientific paper 'Assessing critical load exceedances and ecosystem impacts of N and S deposition at calibrated forested catchments in Europe' (Forsius et al.) was prepared.
- Report on Hg and HM trends in concentrations and fluxes across ICP Integrated Monitoring sites in Europe (Eklöf et al.) is included as chapter 2 'Temporal trends and input - output budgets of heavy metals in ICP IM catchments' in the present Annual Report. A scientific paper on this topic is planned for 2021.
- An interim assessment 'Long-term changes in the inorganic nitrogen output in European ICP Integrated Monitoring catchments – an assessment of the impact of internal nitrogen-related parameters and exceedances of critical loads of eutrophication' (Vuorenmaa et al.) is presented as chapter 3 in this Annual Report. A scientific paper will be finalised in 2020.
- ICP IM participates also in the work of 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). Priority in the ICP IM work is given to site-specific modelling activities and development/testing of new methodologies for assessing the connections between air pollution and climate change.

1.2 Activities and tasks planned for 2020–2021

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, ICP IM will produce the following reports/papers:

- 2020: 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)
- 2020: Scientific paper on effects of nitrogen enrichment on forest vegetation. A cooperation between ICP IM & ICP Forests. (WGE item 1.1.1.17)
- 2021: 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)

In addition to the items on the WGE work plan, ICP IM aims at producing the following scientific outputs:

- 2020/21: Scientific paper on the relationship between critical load exceedances and empirical ecosystem impact indicators, manuscript (Forsius et al.) available
- 2020/21: Scientific paper on the recovery in the epiphytic lichen community in the IM catchments, after the decrease in S deposition, manuscript (Weldon & Grandin) submitted
- 2020: Progress report on heavy metal trends in concentrations and fluxes across ICP IM sites in Europe, potentially in cooperation with ICP Waters

Other activities

- Maintenance and development of central ICP IM database
- Arrangement of the 29th Task Force meeting (2021)
- Preparation of the 30th ICP IM Annual Report (2021)
- 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 2019–2020

Evaluations of international ICP IM data and related publications

Grennfelt, P., Engleryd, A., Forsius, M. et al. 2020. Acid rain and air pollution: 50 years of progress in environmental science and policy. Ambio 49, 849–864.

Kleemola, S. & Forsius, M. (eds.) 2019. 28th Annual Report 2019. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 33/2019, Helsinki. 47 p. http://hdl.handle.net/10138/304484

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

Casetou-Gustafson, S., Grip, H., Hillier, S., Linder, S., Olsson, B. A., Simonsson, M. & Stendahl, J. 2020. Current, steady-state and historical weathering rates of base cations at two forest sites in northern and southern Sweden: a comparison of three methods. Biogeosciences 17:281–304.

Dirnböck, T., Kraus, D., Grote, R., Klatt, S., Kobler, J., Schindlbacher, A., Seidl, R., Thom, D. & Kiese, R. 2020. Substantial understory contribution to the C sink of a European temperate mountain forest landscape. Landscape Ecology 35: 483–499.

Douinot, A., Tetzlaff, D., Maneta, M., Kuppel, S., Schulte-Bisping, H. & Soulsby, C. 2019. Ecohydrological modelling with EcH2O-iso to quantify forest and grassland effects on water partitioning and flux ages. Hydrological Processes. 33:2174–2191.

Hayes, N.R., Buss, H.L., Moore, O.L., Krám, P. & Pancost, R.D. 2020. Controls on granitic weathering fronts in contrasting climates. Chemical Geology 535, ArtNo 119450, 1–19.

Leitner, S., Dirnböck, T., Kobler, J. & Zechmeister-Boltenstern, S. 2020. Legacy effects of drought on nitrate leaching in a temperate mixed forest on karst. Journal of Environmental Management 262, 110338.
Löfgren, S. (ed.) 2019. Integrated monitoring of the environmental status in Swedish forest ecosystems – IM. Annual report for 2018. Swedish University of Agricultural Sciences. Department of Aquatic Sciences and Assessment, Report 2019:7. Uppsala Sweden. 34 pp, 23 appendices. (in Swedish with English summary). https://pub.epsilon.slu.se/16519/1/lofgren_s_191217.pdf
Novák, M., Holmden, C., Farkaš, J., Krám, P., Hruška, J., Čuřík, J., Veselovský, F., Štěpánová, M., Kochergina, Y.V., Erban, V., Fottová, D., Šimeček, M., Bohdálková, L., Přechová, E., Voldřichová, P., Černohous, V. 2020. Calcium and strontium isotope dynamics in three polluted forest ecosystems of the Czech Republic, Central Europe. Chemical Geology 536: ArtNo. 119472, 1–12.
Rosenstock, N. P., Stendahl, J., van der Heijden, G., Lundin, L., McGivney, E., Bishop, K. & Löfgren, S. 2019. Base cations in the soil bank: non-exchangeable pools may sustain centuries of net loss to forestry and leaching, SOIL 5:351–366.
Schmitz, A., Sanders, T., Bolte, A., Bussotti, F., Dirnböck, T., Johnson, J., Penuelas, J., Pollastrini, M., Prescher, A.-K., Sardans, J., Verstraeten, A., de Vries, W. 2019. Responses of forest ecosystems in Europe to decreasing nitrogen deposition. Environmental Pollution 244, 980–994.
Staude, I.R., Waller, D.M., Bernhardt-Römermann, M., Bjorkman, A.D., Brunet, J., deFrenne, P., Máliš, F., Verheyen, K., Wulf, W., Pereira, H.M., Hédl R., Jandt U., Chudomelová, M., Decocq, G., Dirnböck T., Durak T., Heinken, T., Jaroszewicz, B., Kopecký, M., Macek, M., Malicki, M., Naaf, T., Nagel, T.A., Petrík, P., Reczynska, K., Van Calster, H., Vild, O., Vangansbeke, P., Ortmann-Ajkai, A., Pielech, R., Berki, I., Schmidt, W., Standovár, T., Swierkosz, K., Teleki, B., and Baeten, L. 2020. Replacements of small- by large-ranged species scale up to diversity loss in Europe's temperate forest biome. Nature Ecology & Evolution 4, 802–808.
Weldon, J, Grandin, U. 2019. Major disturbances test resilience at a long-term boreal forest monitoring site. Ecology and Evolution 9: 4275–4288.
Yu, X., Lamačová, A., Shu, L., Duffy, C., Krám, P., Hruška, J., White, T. & Lin, K. 2020. Data rescue in manuscripts: a hydrologic modelling study example. Hydrological Sciences Journal 65 (5):763–769.

1.4 Monitoring sites and data

The following countries have continued data submission to the ICP IM database during the period 2015–2019: 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 2014–2018 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.

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	hydrobio-logy of lakes	forest damage	vegetation	bioelements	vegetation structure	trunk epiphytes	aerial green algae	microbial de-composition	bird inventory	vegetation inventory
AT01	ZÖBELBODEN	95-18	95-18	93-18		93-18	99-04	04	93-18		95-18	-	92-17	93-18				93			93-98				
BY02	BEREZINA BR	89-15	89-15	89-15				95-98			95-15														
CH02	LAGO NERO	15-18	15-18	15-18				18	17-18		15-18	15-16						17							
CZ01	ANENSKE POVODI	89-18	89-18	89-18	89	89-18		02-15	07-18	08-18	89-18	-			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	90-18	90-18	90-18	90	90-18	90-05	90-11	90-18	88-18	90-18	-	90-18	90-18		-	90-14	90-08		00	92-95		94-18	91-02	90-95
DE02	NEUGLOBSOW	67-18	98-18	98-18		98-18	04-18	04-16	98-18	98-18		98-18	06-18	04-18				04-17							
EE01	VILSANDI	95-18	94-18	94-18	94-15	94-18	94-18	94-15	94-18	95-96	-	-	94-18	94-18	-	-	94-17	94-97			94-04		94-18		94
EE02	SAAREJÄRVE	94-18	98-18	94-18	94-16	94-18	94-18	94-15	95-18	95-14	94-18	96	94-18	94-18			96-17	96-12	12		94-15	94-17	96-18	98-14	
ES02	BERTIZ	08-17	08-17	07-17		07-17	08-17	10-15	07-17		07-17		08-16	08-17			07-12	07		07					
FI01	VALKEA-KOTINEN	88-17	94-17	88-17	88-96	89-17	89-99	88-89	89-17		88-18	87-18	88-17	90-16		90-93	88-91	88-09			88-97		90	87-89	87
FI03	HIETAJÄRVI	88-17	93-00	88-17	89-96	89-17	89-99	88	89-17		88-18	87-18	88-17	90-16		90	88-91	90-09			90-97		90-91	87-89	
FI06	PALLASJÄRVI			14-17		16-17			02-17		04-17	04-17	95-17	07-16										88-89	
IE01	BRACKLOON WOOD			91-16		91-11	92-97		91-16			-	91-96	91-98	-	-									
IT01	RENON-RITTEN	90-18	93-18	93-14		93-13	93-13	93-11	93-13		00-13	-	93-10	00	-	-	92-13	09		05-09	92		93-11		
IT02	MONTICOLO-MONTIGGL	77-13	93	93-14		93-13	93-13	93-10	93-13		-	-	93-01	00	-	-	92-13				92		93-11		
IT05	SELVA PIANA	97-08	97-15	97-18		97-18	97-18	95	02-08		-	-	97-05		-	-	97-09	09		99-09					
IT06	PIANO LIMINA	99-08	97-16	97-18		97-18	97-18	95			-	-	97-05		-	-	97-09	09		99-09					
IT07	CARREGA	97-08	97-16	97-18		97-18	97-00	95			98-13	-	97-05		-	-	97-09	09		99-09					
IT09	MONTE RUFENO	97-08	97-16	97-18		97-18	97-00	95	02-08		97-14	-	97-05		-	-	97-09	09		99-09					
IT10	VAL MASINO	97-08	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	97-08	97-15	09-15		09-15		95			-	-	97-05		-	-	97-09			99-08					
LT01	AUKSTAITIJA	93-13	93-18	93-18	93-10	93-18		93-05	94-18	93-18	93-18		06-18	99-18	12		00-18	93-18		02-15	93-18	93-18			93
LT03	ZEMAITIJA	90-13	95-18	95-18	06-10	95-18		94-05	95-18	95-18	95-18		06-18	99-18	95-12		00-18	94-18		02-15	94-18	94-18			94
NO01	BIRKENES	87-18	87-18	87-18	92	89-18		87-11	86-18	87-88	87-18	-	86-17	87-02		-	91-18	86-18			86				
NO02	KÅRVATN	87-91	87-18	87-18	88	89-11		89-13	89-10		87-18	-	89-09	89-02		-	92-10	89-09							
NO03	LANGTJERN		87-97	77-18		86-03		91-13	91-03		87-18		86-03	87-02											
PL01	PUSZCZA BORECKA	06-18	16-18	16-18		16-18		17	10-18			16-18		06-18				16							
PL05	WIGRY	06-18	16-18	16-18		16-18			06-18		16-18			05-18				16							
PL06	PARSENTA	10-18	16-18	94-18		96-18			10-18		94-18			10-18											
PL07	POJEZIERZE CHELMINSKIE	16-18	16-18	16-18				18			16-18														
PL08	KAMPINOS	09-18	16-18	16-18		16-18		16	12-18		16-18			10-18				16							
PL09	LYSOGORY	05-18	16-18	16-18		16-18			05-18		16-18			05-18				16							
PL10	BESKIDY	94-18	16-18	94-18		02-18			11-18		94-18			09-18				16							
PL11	WOLIN	16-18	16-18	16-18		17-18			16-18		16-18			16-18											
PL12	ROZTOCZE	16-18	16-18	16-18		16-18			16-18		16-18			16-18				16							
RU03	CAUCASUS BR	89-94	89-17	89-98																					
RU04	OKA-TERRACE BR	89-06	89-17	89-98	90										93-99		93-14	93-02			93		94-96		
RU12	ASTRAKHAN BR	93-94	93-17	93-94																					
RU13	CENTRAL FOREST BR	93	93-94	93													09-18	18							
RU14	VORONEZH BR	94	94-17	94-98																					
RU16	VELIKIY ISLAND				89-90			89	89	89						93-99	93-18	91-94			89-94	93	94-95		91
RU47	KURSK																18								
SE04	GÅRDSJÖN F1	87-18	88-18	87-17	95	87-18		95-10	87-18	79-18	87-18	-	99-18	96-18		-	97-01	95-16	91-15	91-15	96-16	92-17	95-18		
SE14	ANEBODA	96-18	96-18	96-18	95	96-18		96-11	95-18	96-18	96-18	-	99-18	95-18		-	97-01	82-16	96-16	06-16	97-17	97-16	95-18		
SE15	KINDLA	97-18	96-18	96-18		96-18		97-12	95-18	97-18	96-18	-	97-18	95-18		-	98-01	96-17	98-18	98-18	98-18	97-17	95-18		
SE16	GAMMTRATTEN	99-18	99-18	99-18		99-18		00-18	00-18	00-18	99-18		99-18	00-18			00-01	99-18	99-14	99-14	00-15	00-17	00-18		

Figure 1.1. Geographical location of ICP IM sites.

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2 Temporal trends and input - output budgets of heavy metals in ICP IM catchments

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2.1 Introduction

The long-term time series on heavy metals collected within the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) offer possibilities to compare trends and catchment budgets of these metals in a wide geographical range of sites within Europe. The environmental monitoring and modelling within ICP IM aim to support the work of the Convention on Long-range Transboundary Air Pollution (CLRTAP) (Sliggers and Kakebeeke 2004). This report focuses on temporal trends and catchment budgets of cadmium (Cd), lead (Pb) and mercury (Hg), which have been identified as particularly harmful in the 1998 Aarhus Protocol on Heavy Metals, an international agreement on pollution control under the CLRTAP (UNECE 2003). These three heavy metals are all defined as long-range atmospheric pollutants that can be deposited far from the emission sources (Johansson et al. 2001).

The emissions of Cd, Pb and Hg to the atmosphere derive from natural and anthropogenic sources, such as energy production, waste incineration and various use of metals (Pacyna et al. 2009). The emissions of anthropogenic Cd, Pb and Hg have increased since the start of the industrial era. Emission controls have resulted in declined heavy metal emissions and depositions during the last decades. In Europe, the emissions of Cd peaked in the 1960s and in the 1970s for Pb (Pacyna et al. 2007). The global emissions of Hg peaked already during the gold rush in the 1890s. In Europe the emissions of Hg have declined since the 1980s (Streets et al. 2011). However, the global Hg emissions are still increasing. European environmental monitoring has shown that Cd and Pb concentrations in precipitation have decreased by 45% between 1990 and 2005 (Pacyna et al. 2009). The decreasing trends of heavy metals in the deposition have resulted in decreasing concentrations of Cd and Pb in mosses between 1990 and 2000 by 42% and 57%, respectively (Harmens et al. 2008). Atmospheric concentrations of elemental mercury (Hg^0) declined annually by 1–2%, with a subsequent decrease in wet deposition of Hg from 1990 to 2014 at sites in North America and Europe (Zhang et al. 2016).

In this report we evaluate if the declining trends in atmospheric deposition of Cd, Pb and Hg during recent decades are reflected in the runoff concentrations from European catchments within the ICP IM network. In addition to the direct effect of reduced deposition of metals during the last decades, less metals may also be mobilised from terrestrial soils to surface waters as a result of recovery from acidification during the same period (Lydersen et al. 2002). In this report, we calculate the catchment Cd, Pb and Hg input-output budgets for the four ICP IM sites in Sweden.

2.2 Methods

The data collected within ICP IM are reported by each country to the ICP IM Programme Centre at the Finnish Environment Institute (SYKE, Helsinki, Finland). The data are divided in a set of subprograms, including bulk deposition (BD), throughfall (TF), litterfall (LF), and runoff water (RW) at 13 ICP IM sites. Data on RW were obtained from the international database for Sweden, Finland, Latvia, Belarus, the Czech Republic and Austria (Fig. 2.1). For the mass balances, we have used national data of annual fluxes of BD, TF, LF and RW from the four Swedish ICP IM sites Gårdsjön (SE04), Aneboda (SE14), Kindla (SE15) and Gammtratten (SE16).

The temporal trends of Cd and Pb concentrations in runoff water were evaluated only if the time-series included at least 10 years of data with more than 100 observations. The trends of Hg concentrations in runoff were evaluated for sites with at least 6 years of data including 100 observations or more. The Cd and Pb trends were evaluated for 13 sites, while the Hg trends were evaluated for 4 sites (1 in Sweden, SE04 and 3 in Finland, FI01, FI03, FI06). The lengths of the Cd and Pb time-series varied between 13 and 30 years, starting in 1988 as earliest and most often ending in 2018. The non-parametric Seasonal-Kendall test, unsensitive to extreme values and taking into account seasonal variation (Loftis et al. 1991), was used for testing the statistical significance ($p<0.05$) of the temporal trends for Cd, Pb and Hg.

Based on data from the Swedish ICP IM sites, input-output budgets were calculated at catchment level for Cd, Pb and Hg. The catchments are well-defined and forest dominated with no or insignificant degrees of forest management for at least 100 years. Wet deposition was collected as bulk deposition in open field with no tree canopies and as throughfall under the tree canopies for determining precipitation amounts and metal concentrations. Litterfall was sampled in mesh sacks under the canopies, followed by drying and weighing at the laboratory. Discharge was continuously registered at weirs in the catchment outlets. Stream water samples were collected in the inlet to these dams. On all samples, the concentrations of Cd, Pb and Hg were analysed. Fluxes were calculated for BD, TF, LF and RW, but as forest cover dominates these catchments, only TF and LF were used as input in the mass-balances.

2.3 Results and discussions

The concentrations of Cd in RW decreased over time at 9 of the 13 sites and increased at one of them (Fig.2.1). For Pb, the concentration trends were decreasing at 4 of the 13 sites and increased at 2 of them. The LV02 catchment that showed increasing concentrations of Cd exhibited a positive trend also for Pb. The fact that declining Cd concentrations in runoff were detected at a majority of the sites suggests that it reflects the general decline in Cd and acid deposition over Europe during the last decades. Declining trends of Cd and Pb in runoff at many of the sites are in agreement with earlier studies from Scandinavian streams in 1984–1995 (Lydersen et al. 2002). For Hg concentrations, 1 (FI06) of the 4 sites showed decreasing trends and no trends were observed at the other 5 sites. Earlier studies have shown almost no Hg trends in Swedish streams and rivers (Eklöf et al. 2012), which support the results from the ICP IM sites.

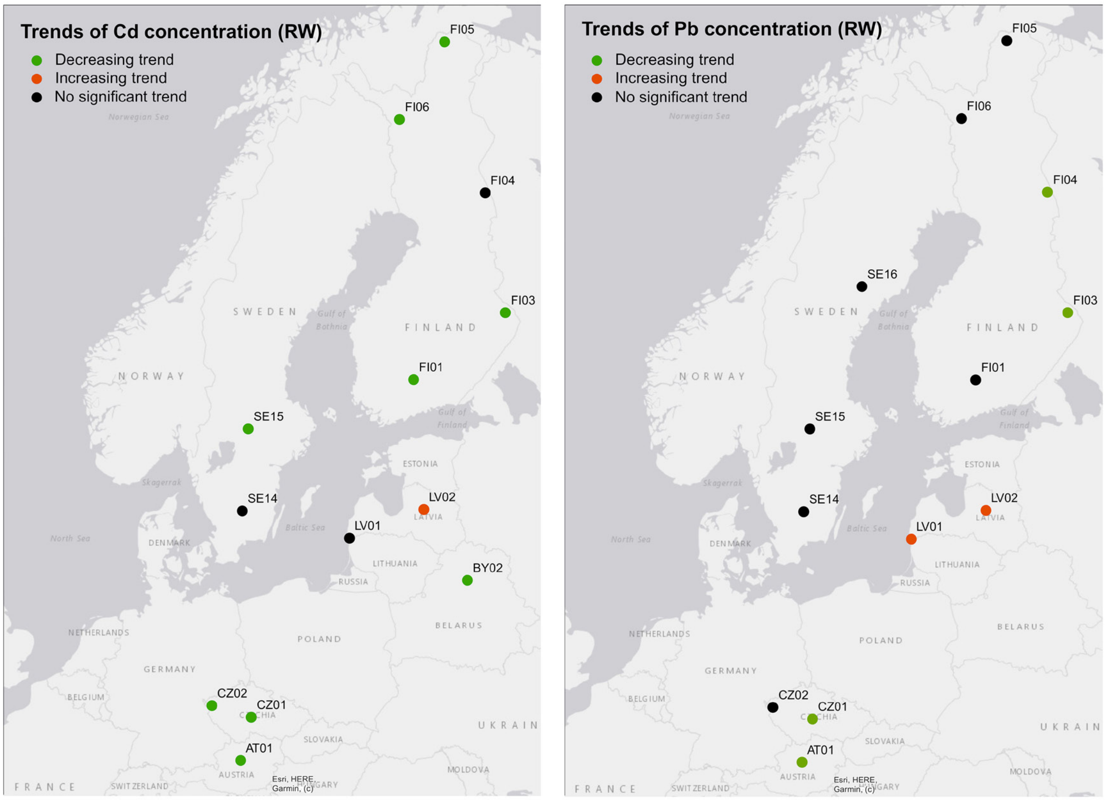

Figure 2.1. Cadmium (left) and lead (right) concentration trends in runoff (RW) at the ICP IM sites between 1988 and 2018. The times series length varies between sites. Statistically significant ($p<0.05$) increased (red dots), decreased (green dots) or no trends (black dots) according to Seasonal-Kendall tests.

At the Swedish ICP IM sites, 13–70% and 21–56% of the annual total deposition (TF+LF) of Cd and Pb, respectively, leave the catchments by runoff (RW, Table 2.1). The Hg budget is not presented, but generally less than 10% of the annual total deposition is accounted for in the RW export at these sites (Eklöf et al. in prep.). Pb and Hg have a stronger association to organic compounds compared to Cd (Lydersen et al. 2002) and are therefore to a higher degree retained in the upper soil horizons than Cd (Johansson et al. 2001). In agreement with this work, earlier studies (Aastrup et al. 1991, Ukonmaanaho et al. 2001, Grigal 2002, Bringmark et al. 2013) have seldom found runoff export to exceed the total input (TF+LF), which indicates that metals are accumulating in the catchments. At least for Hg, the upper soil layer concentrations increase over time at the Swedish ICP IM sites (Åkerblom & Lundin 2015). The Pb and Cd concentrations decreased in the upper and increased in the lower soil layers of some soil types in the Austrian ICP IM site (Kobler et al. 2010). At catchment level, the heavy metal budgets may be influenced by internal sources and fluxes. In forested catchments of eastern Finland e.g., the weathering rates of Pb were found to be an important internal source comparable with dry deposition (Starr et al. 2003). Moreover, internal sources and fluxes as well as volatilization (primarily Hg) complicate the budget and may affect these calculations.

Table 2.1. Mass balance fluxes (mg m^{-2} y^{-1}) for cadmium (Cd) and lead (Pb) based on data from the Swedish ICP IM sites; Aneboda, Gammtratten, Gårdsjön and Kindla. Median values are based on the annual fluxes during the period 1997 to 2018. The number of years (n) in the time series are presented within parentheses.

Metal	Site	Bulk deposition (BD)	Throughfall (TF)	Litterfall (LF)	Runoff (RW)	Total deposition (TF+LF)	% RW of TF+LF
Cd	Aneboda	0.01 (7)	0.02 (7)	0.1 (18)	0.01 (18)	0.1	13
	Gamm-tratten	0.01 (3)	0.01 (3)	0.02 (14)	0.01 (14)	0.03	29
	Gårdsjön	0.03 (13)	0.03 (13)	0.1 (12)	0.03 (12)	0.1	30
	Kindla	0.03 (3)	0.02 (3)	0.02 (18)	0.03 (18)	0.04	70
Pb	Aneboda	0.9 (7)	0.4 (9)	0.5 (18)	0.3 (18)	0.9	33
	Gamm-tratten	0.5 (3)	0.3 (4)	0.2 (14)	0.1 (12)	0.5	20
	Gårdsjön	1.0 (13)	0.4 (4)	0.5 (10)	0.5 (3)	0.9	56
	Kindla	0.6 (3)	0.5 (5)	0.5 (18)	0.2 (11)	1.0	21

2.4 Conclusions

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 1 statistically ($p<0.05$) significant decreasing trend. Decreasing Cd and Pb concentrations have been shown in earlier studies, but the data from ICP IM are unique in the sense that they are both geographically distributed and long-term.

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.

References

Aastrup, M., Johnson, J., Bringmark, E., Bringmark, I. & Iverfeldt, Å. 1991. Occurence and transport of mercury within a small catchment area. Water Air & Soil Pollution 56(1): 155–167.

Åkerblom, S. & Lundin, L. 2015. Progress report on heavy metal trends at ICP IM sites. In: Kleemola, S. & Forsius, M. (Eds.) 24th Annual Report 2015. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 31/2015 pp. 32–36.

Bringmark, L., Lundin, L., Augustaitis, A., Beudert, B., Dieffenbach-Fries, H., Dirnböck, T., Grabner, M.-T., Hutchins, M., Kram, P., Lyulko, I., Ruoho-Airola, T. & Váňa, M. 2013. Trace metal budgets for forested catchments in Europe – Pb, Cd, Hg, Cu and Zn. Water, Air, & Soil Pollution 224(4): 1–14.

Eklöf, K., Fölster, J., Sonesten, L. & Bishop, K. 2012. Spatial and temporal variation of THg concentrations in run-off water from 19 boreal catchments, 2000-2010. Environmental Pollution 164: 102–109.

Grigal, D. F. 2002. Inputs and outputs of mercury from terrestrial watersheds: a review. Environmental Reviews 10: 1–39.

Harmens, H., Norris, D. A., Koerber, G. R., Buse, A., Steinnes, E. & Rühling, Å. 2008. Temporal trends (1990–2000) in the concentration of cadmium, lead and mercury in mosses across Europe. Environmental Pollution 151(2): 368–376.

Johansson, K., Bergbäck, B. & Tyler, G. 2001. Impact of atmospheric long-range transport of lead, mercury and cadmium on the Swedish forest environment. Water, Air and Soil Pollution: Focus 1, 279–296.

Kobler, J., Fitz, J.F., Dirnböck, T. & Mirtl, M. 2010. Soil type affects migration pattern of airborne Pb and Cd under a spruce-beech forest of the UNECE Integrated Monitoring site Zöbelboden, Austria. Environmental Pollution 158: 849–854.

Loftis, J. C., Taylor, C. H., Newell, A. D. & Chapman, P. L. 1991. Multivariate trend testing of lake water quality. Water Resources Bulletin 27(3): 461–473.

Lydersen, E., Löfgren, S. & Arnesen, R. T. 2002. Metals in Scandinavian surface waters: Effects of acidification, liming, and potential reacidification. Critical Reviews in Environmental Science and Technology 32(2-3): 73–295.

Pacyna, E. G., Pacyna, J. M., Fudala, J., Strzelecka-Jastrzab, E., Hlawiczka, S., Panasiuk, D., Nitter, S., Pregger, T., Pfeiffer, H. & Friedrich, R. 2007. Current and future emissions of selected heavy metals to the atmosphere from anthropogenic sources in Europe. Atmospheric Environment 41(38): 8557–8566.

Pacyna, J. M., Pacyna, E. G. & Aas, W. 2009. Changes of emissions and atmospheric deposition of mercury, lead, and cadmium. Atmospheric Environment 43(1): 117–127.

Sliggers, J. & Kakebeeke, W. 2004. Clearing the Air. 25 years of the Convention on Long-range Transboundary Air Pollution. UNECE. New York and Geneva. 167 p.

Starr, M., Lindroos, A.-J., Ukonmaanaho, L., Tarvainen, T. & Tanskanen, H. 2003. Weathering release of heavy metals from soil in comparison to deposition, litterfall and leaching fluxes in a remote, boreal coniferous forest. Applied Geochemistry 18(4): 607–613.

Streets, D. G., Devane, M. K., Lu, Z., Bond, T. C., Sunderland, E. M. & Jacob, D. J. 2011. All-time releases of mercury to the atmosphere from human activities. Environmental Science & Technology 45(24): 10485–10491.

Ukonmaanaho, L., Starr, M., Mannio, J. & Ruoho-Airola, T. 2001. Heavy metal budgets for two headwater forested catchments in background areas of Finland. Environmental Pollution 114(1): 63–75.

UNECE 2003. Convention on Long-range Transboundary Air Pollution on Heavy Metals – The 1998 Aarhus protocol on heavy metals. United Nations Economic Commision for Europe, 33 p.

Zhang, Y., Jacob, D. J., Horowitz, H. M., Chen, L., Amos, H. M., Krabbenhoft, D. P., Slemr, F., St. Louis, V. L. & Sunderland, E. M. 2016. Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions. 113(3): 526–531.

3 Long-term changes in the inorganic nitrogen output in European ICP Integrated Monitoring catchments

– an assessment of the impact of internal nitrogen-related parameters and exceedances of critical loads of eutrophication

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Interim report

3.1 Introduction

Successful emission reduction measures in Europe over the past 30–40 years have led to a declining deposition of nitrogen (N), as shown at ICP Integrated Monitoring (ICP IM) sites throughout Europe (Vuorenmaa et al. 2018). Decreasing trend of total inorganic N (TIN = NO_3 + NH_4) deposition should generally lead to decreased NO_3 concentrations in runoff; and, indeed, decreasing trends of TIN concentrations in runoff – particularly for NO_3 – are more prominent than increasing trends at the IM sites (Vuorenmaa et al. 2018). The trends for the concentrations and output fluxes of TIN are, however, still variable, indicating that surface water-watershed N dynamics are inherently complex, as nitrogen is strongly affected by biological processes and hydrological conditions. Deposited N continues to accumulate in catchment soils and vegetation, but so far TIN is effectively retained in the unmanaged IM forested catchments located in low or intermediate N deposition areas (Vuorenmaa et al. 2017), and as of yet there are no clear signs of a consistent increase in TIN concentrations or exports in unmanaged forested catchments in Europe (Vuorenmaa et al. 2018).

In order to assess the impacts of N deposition in the environment, a long-term integrated monitoring approach in remote unmanaged areas including physical, chemical and biological variables is needed. The multidisciplinary International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) is one of the activities set up under the UNECE CLTRAP to develop the necessary international co-operation in the assessment of the air pollutant effects and ecosystem impacts in forested and aquatic ecosystems. The recent trend assessments for TIN in runoff at ICP IM sites were focused on the long-term changes in annual input-output budgets and monthly concentrations and fluxes in runoff in the context of emission and deposition reduction responses and climatic variation (Vuorenmaa et al. 2017, 2018). The ICP IM sites are located in areas with very different N deposition gradients, and further analyses with specific catchment and soil data is needed to elucidate the variation of TIN concentrations in runoff and to better understand the regulating processes.

Critical loads (CLs) have been widely used to describe the limits of different ecosystems to sustain air pollutants (Amman et al. 2011, Posch et al. 2015a, Grennfelt et al. 2020). Ecosystems are considered at risk of becoming eutrophied if N deposition persistently exceeds the critical load of eutrophication ($CL_{eut}N$). The value of $CL_{eut}N$ is a static site-specific property of the ecosystem, based on empirical considerations of the vegetation response of the habitat (Bobbink & Hettelingh 2011), as well as on mass balance calculations involving observations and assumptions regarding the long-term site characteristics, such as hydrological regime, denitrification, N immobilisation, and nutrient uptake (Posch et al. 2015b). Critical loads are relevant tools for determining acceptable deposition levels if their exceedances are related to unwanted ecosystem effects the limits are designed to protect from. Higher TIN concentrations and fluxes in runoff have earlier been observed in ICP IM sites where the critical loads were exceeded (Holmberg et al. 2013). A decrease in TIN concentrations and fluxes occurred at some ICP IM sites that also experienced decreases in the exceedance of critical loads (Forsius et al. 2020).

The ICP IM Programme offers possibilities to analyse a wide range of internal catchment-related nitrogen parameters in a wide geographical range of sites within Europe. At the 26th meeting of the ICP IM Programme Task Force (7–9 May 2018 in Warsaw, Poland) it was agreed that the Programme Centre with NFPs will continue the analyses of TIN leaching in IM catchments involving collection and analysing available data on meteorology (air and soil temperature, subprogramme AM) and physical and chemical soil and foliar parameters from subprogrammes SW (soil water chemistry), SC (soil chemistry), LF (litterfall chemistry) and FC (foliage chemistry). In this interim report we 1) present the preliminary results if different internal N-related parameters can explain the variation of TIN trends in IM catchments, and 2) report year-to-year variation in runoff TIN concentration concurrently with the exceedance of critical loads of eutrophication for ICP IM sites.

3.2 The relationship between TIN leaching and internal catchment N-related parameters

3.2.1 Material and methods

Data between 1990 and 2017 was collected within the frame of ICP IM and reported by each country to the ICP IM Programme Centre's database at the Finnish Environment Institute (Table 3.2.1).

Table 3.2.1. Data used in the assessment of the impacts of internal N-related parameters.

Subprogramme	Parameters
Meteorology (AM)	air temperature, soil temperature
Precipitation chemistry (PC)	precipitation, NO_3-N, NH_4-N, Cl
Throughfall (TF)	precipitation, NO_3-N, NH_4-N, Cl
Runoff water chemistry (RW)	runoff volume, NO_3-N, NH_4-N
Soil water chemistry (SW)	tot N, NO_3-N, NH_4-N, TOC/DOC, pH
Soil chemistry (SC)	tot N, TOC, pH, bulk density
Litterfall chemistry (LF)	tot N, tot P, TOC, litterfall amount
Foliage chemistry (FC)	tot N, tot P, TOC

Temporal trends were evaluated for precipitation and runoff amount, and NO_3-N, NH_4-N and TIN (TIN = NO_3-N + NH_4-N) concentrations and fluxes in PC, TF and RW using monthly data in 1990–2017.

Trend slopes (i.e. annual change in 1990–2017) for concentrations and fluxes in bulk deposition (PC), throughfall (TF) and runoff water (RW), and annual means between 2010 and 2017 for concentrations in soil (SC), litterfall (LF) and foliage (FC), and for concentrations and fluxes in PC, TF, RW and soil water (SW) were used in multiple statistical analyses. Data was explored using multiple stepwise regression, correlation analysis and discriminant analysis. Based on available data (2010–2017) in the IM database, 17 sites from 10 countries with RW measurements (concentrations and/or fluxes) were included into the N assessment: AT01, CZ01, CZ02, DE01, EE02, ES02, FI01, FI03, LT01, LT03, NO01, NO02, PL06, PL10, SE04, SE15, SE16.

The mean values of concentrations and output fluxes in soil water (SW) were calculated using values measured from the depth of 30–40 cm. Data was best available from these depths and this soil layer was assumed to be in many soil water stations below the root zone. Due to the lack of hydrological measurements or modelled soil water flow estimates, the soil water recharge needed for output calculations was calculated using the chloride mass-balance method (e.g. Allison and Hughes 1983). The soil chemistry (SC) results from organic horizon were used in statistical analysis.

3.2.2 Results and discussion

Temporal trends of TIN in bulk deposition, throughfall and runoff

ICP IM network confirms the positive effects of the continuing emission reductions in Europe. IM sites showed dominantly negative trend slopes of TIN in concentrations and bulk/wet deposition between 1990 and 2017 (95% and 91% of the sites, respectively) (Fig. 3.2.1a). Decrease of NO_3 and NH_4 in concentrations was significant at 91% and 77% of the sites, and in fluxes 64% and 59% of the sites, respectively. Long-term trends in precipitation amounts in 1990–2017 showed dominantly increasing trend slopes (68% of the sites), but trends were rarely significant. The short and long-term variations in precipitation may mask long-term trends caused by N deposition (Wright et al. 2001).

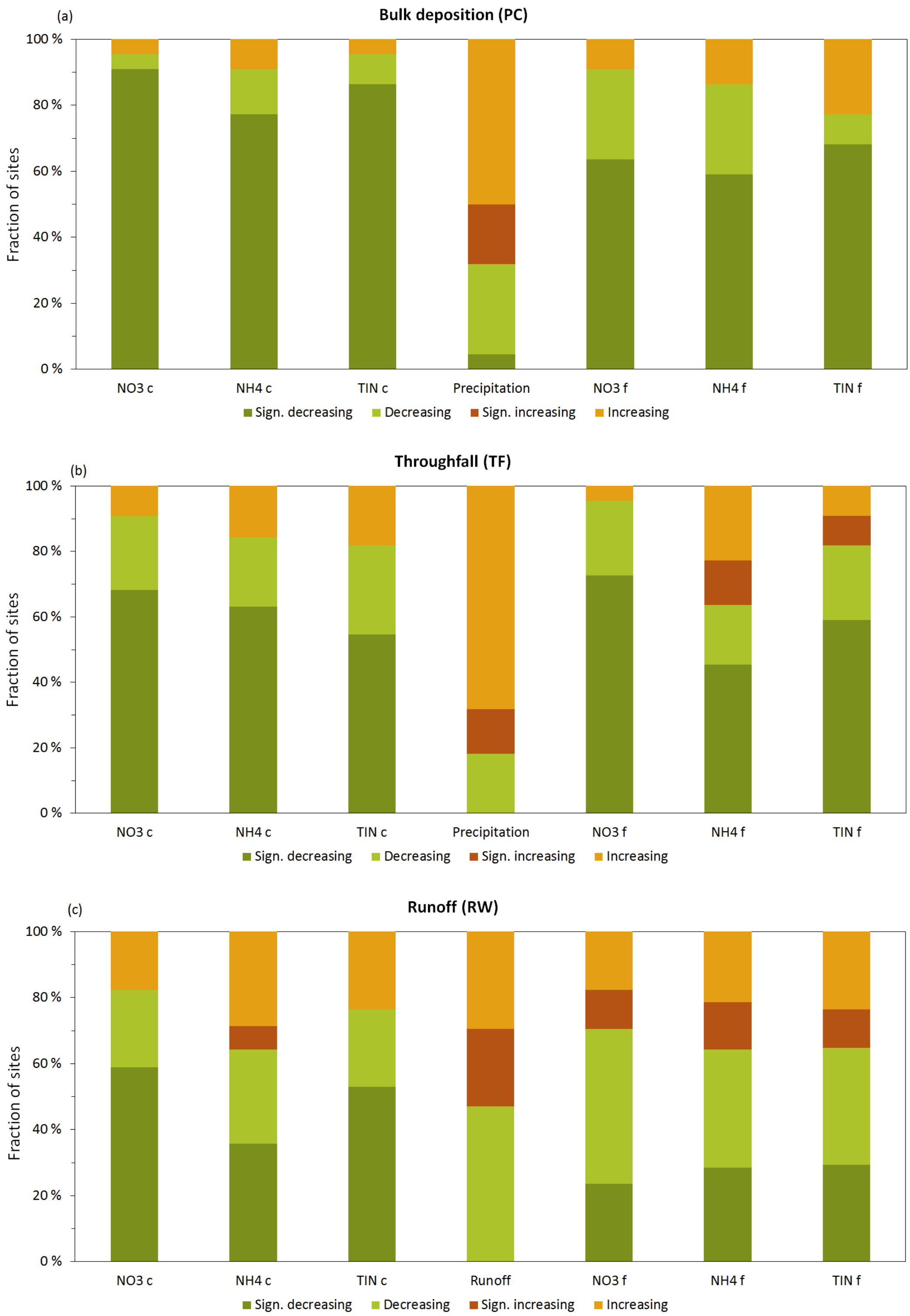

Figure 3.2.1. Percentage of Integrated Monitoring sites with a significant decreasing (green), insignificant decreasing (light green), significant increasing (dark orange) and insignificant increasing (yellow) trend in concentrations (denoted as c) and fluxes (denoted as f) for bulk deposition (a), throughfall (b) and runoff (c) in 1990–2017.

TIN concentrations in throughfall showed also predominantly decreasing trend slopes (81% of the sites) and decrease in NO_3 and NH_4 concentrations was significant at 62% and 54% of the sites, respectively (Fig. 3.2.1b). Deposition of TIN in throughfall decreased at 81% of the sites, and the decrease in NO_3 and NH_4 fluxes was significant at 69% and 46% of the sites, respectively. Only a few sites showed significant increases in inorganic N concentrations and fluxes in throughfall. Biological processes such as N uptake by plant tissue and through stomata and other complex canopy interactions control inorganic N fluxes in throughfall (Draaijers & Erisman 1995), and thus long-term trends can be largely controlled by factors other than direct deposition effect.

IM catchments have increasingly responded to the decreases in the emission and deposition of N in Europe. Concentrations and fluxes of TIN in runoff exhibited dominantly downward trend slopes (76% and 69% of the sites, respectively) (Fig. 3.2.1c). Decrease of NO_3 and NH_4 in concentrations was significant at 59% and 36% of the sites, but the decrease in fluxes was significant only at 25% and 31% of the sites, respectively.

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 (Table 3.2.2). 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 a 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 (Figs 3.2.2 and 3.2.3).

Status and future tasks

First results of multivariate statistical analysis if internal N-parameters in catchments can explain the variation of TIN trends in runoff, including N in deposition, and N, P and C in foliage, litterfall, soil water and soil organic horizon, were presented here. Next step will be inclusion of climatological parameters to the analysis e.g. changes in length of growing season and analysis of trends in runoff water volume, status and trends of TOC/TON-ratio in runoff water, fraction of atmospheric N deposition lost in stream water vs. TIN deposition at the study sites and N saturation stage and its changes. The work will continue on interpretation of the present results and further multivariate statistical analysis with additional parameters.

Table 3.2.2. Significant (p<0.05) Pearson correlations between annual change of TIN concentrations and fluxes in runoff (Δ TIN c, RW and Δ TIN f, RW, respectively) and mean TIN fluxes in TF (Mean TIN f, TF), annual change of TIN flux in bulk deposition (Δ TIN f, PC), annual change in runoff volume (Δ runoff volume), mean tot N concentration in foliage (Mean tot N, FC), N/P-ratio in foliage (FC), mean litterfall amount (Mean LF amount), mean tot N concentration in litterfall (Mean tot N, LF), N/P-ratio in litterfall (N/P, LF), mean TIN concentration in soil water (Mean TIN c, SW), mean tot N concentration in soil water (Mean tot N c, SW), mean TOC concentration in soil water (Mean TOC c, SW), mean TIN flux in soil water (Mean TIN f, SW), mean tot N flux in soil water (Mean tot N f, SW) and mean TOC concentration in soil (Mean TOC, SC).

	Mean TIN f, TF	Δ TIN f, PC	Δ run-off volume	Mean tot N, FC	N/P, FC	Mean LF amount	Mean tot N, LF	N/P, LF	Mean TIN c, SW	Mean tot N c, SW	Mean TOC c, SW	Mean TIN f, SW	Mean tot N f, SW	Mean TOC, SC
Δ TIN c, RW	−0.86 <.0001	0.58 0.02		−0.79 0.007		−0.85 0.001	−0.76 0.007	−0.89 <.001		−0.85 <.001	−0.63 0.03		−0.89 <.001	0.69 0.03
Δ TIN f, RW	−0.68 0.005	0.56 0.03	0.55 0.03	−0.77 0.01	−0.89 0.001	−0.68 0.02	−0.93 <.0001	−0.91 <.0001	−0.75 0.001			-0.67 0.006		
Mean TIN c, RW	0.66 0.005			0.71 0.01		0.90 <.0001	0.82 0.001	0.79 0.002	0.58 0.02					
Mean TIN f, RW	0.56 0.03			0.87 0.001	0.75 0.02	0.62 0.04	0.81 0.003	0.66 0.03						

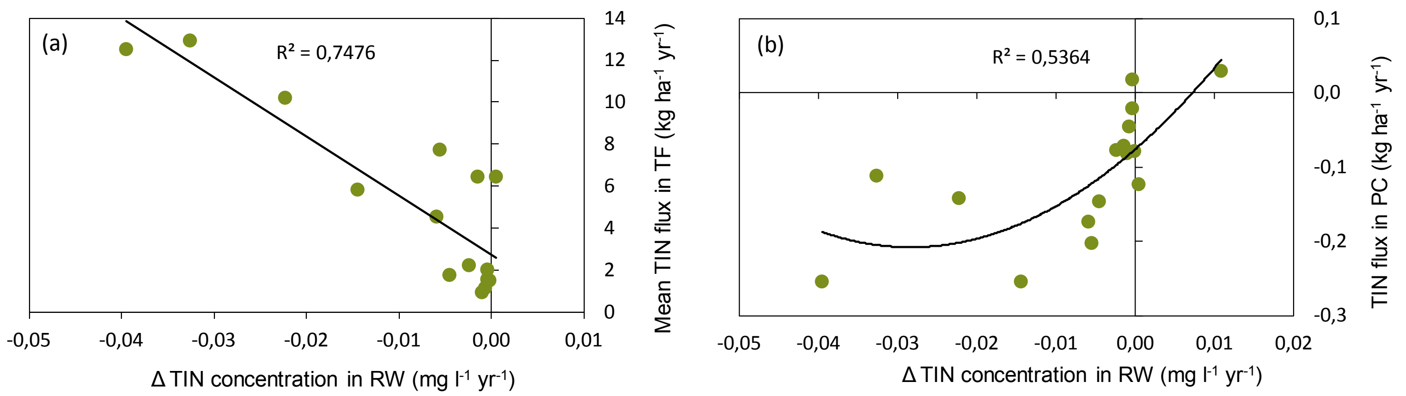

Fig. 3.2.2. Relationship between annual change of TIN concentrations (mg L^{-1} yr^{-1}) in runoff (RW) and mean TIN fluxes in throughfall (TF) (kg ha^{-1} yr^{-1}) (a) and annual change of TIN fluxes in bulk deposition (PC) (kg ha^{-1} yr^{-1}) (b).

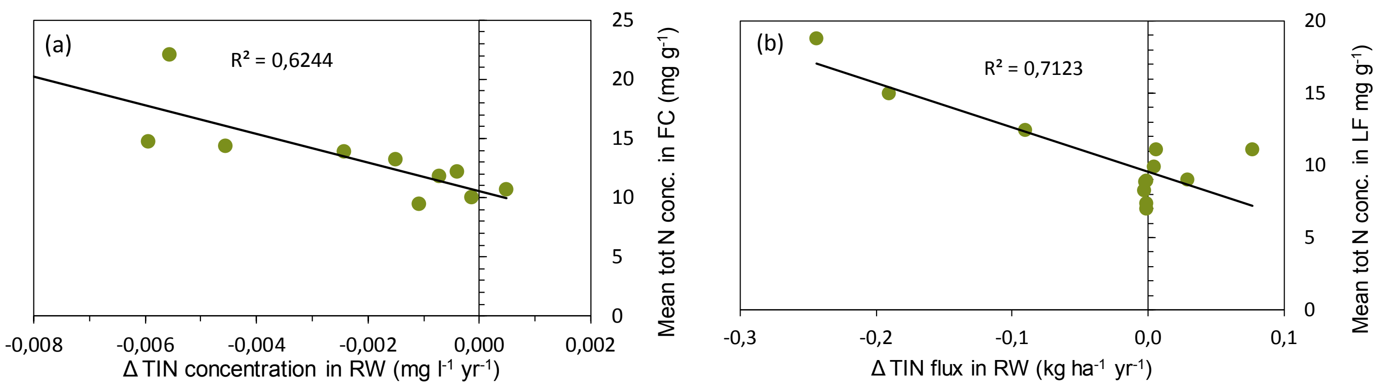

Fig. 3.2.3. Relationships between annual change of TIN concentrations in runoff (mg L^{-1} yr^{-1}) and mean tot N concentrations in foliage (FC) (mg g^{-1}) (a) and annual change of TIN fluxes in runoff (RW) (kg ha^{-1} yr^{-1}) and mean tot N concentrations in litterfall (LF) (b).

3.3 Assessment of critical load exceedances and ecosystem impacts of nitrogen

3.3.1 Material and methods

The eutrophication critical load, $CL_{eut}N$, is defined as the minimum of $CL_{emp}N$ and $CL_{nut}N$ (Mapping Manual, 2017). Empirical critical loads of nitrogen, $CL_{emp}N$, were set to the lower end of the range proposed by Bobbink and Hettelingh (2011) for the respective ecosystem types (EUNIS classes) present at each site (Holmberg et al. 2013).

The critical load of nutrient N, $CL_{nut}N$ (kg ha^{-1} yr^{-1}) was derived from the mass balance of N for an acceptable (or critical) value of N leaching, using the equation

$$CL_{nut}\, N = N_i + N_u + \frac{Q \cdot [N_{acc}]}{1 - f_{de}}.$$

In the equation above, N_i is the long-term immobilization of N in the soil, N_u is the net removal of N in harvested vegetation, f_{de} is the fraction of N input which is denitrified in the soil, and Q represents the annual runoff. In the absence of site-specific observations, a low value of N_i was used (0.5 kg ha^{-1} yr^{-1}) for all sites. Because these sites are not actively harvested, N_u was assumed zero for all sites. The average annual volume of runoff Q was calculated for a period of 10 years (2008 – 2017) for most sites, and for 8, 7 and 6 years for AT01, LT01, and LT03, respectively. The site-specific acceptable soil solution N concentrations $[N_{acc}]$ (mg L^{-1}) (Table 3.3.1) were chosen from the suggested values in the Mapping Manual of the ICP Modelling and Mapping (Mapping Manual, Table V.5, 2017), to minimize the risk of unwanted vegetation impacts, such as nutrient imbalances, or sensitivity to fungal disease or frost. The

Table 3.3.1. Site specific values of acceptable soil [N] N_{acc} (mg L^{-1}), long term average runoff Q (m yr^{-1}) and fraction of N denitrified fde at the 17 ICP IM sites. Critical loads of nutrient N ($CL_{nut}N$), empirical critical loads of N ($CL_{emp}N$), and critical loads of eutrophication ($CL_{eut}N$) (all in kg ha^{-1} yr^{-1}). The observed 2017 N deposition N_{dep} (kg ha^{-1} yr^{-1}).

Site code	$[N_{acc}]$ (mg L^{-1})	Q (m yr^{-1})	f_{de}	$CL_{nut}N$ (kg ha^{-1} yr^{-1})	$CL_{emp}N$ (kg ha^{-1} yr^{-1})	$CL_{eut}N$ (kg ha^{-1} yr^{-1})	N_{dep} (kg ha^{-1} yr^{-1})
AT01	2.6	0.39	0.10	11.5	10.0	10.0	16.5
CZ01	5.2	0.04	0.10	2.7	5.0	2.7	9.2
CZ02	0.8	0.37	0.14	3.9	10.0	3.9	7.6
DE01	0.5	0.91	0.31	7.1	10.0	7.1	6.8
EE02	2.0	0.27	0.17	6.9	10.0	6.9	2.1
FI01	1.3	0.17	0.23	3.3	5.0	3.3	2.3
FI03	1.0	0.40	0.35	6.7	5.0	5.0	1.8
LT01	3.9	0.11	0.17	5.6	5.0	5.0	4.8
LT03	2.0	0.19	0.24	5.5	5.0	5.0	5.0
NO01	0.5	1.22	0.15	7.7	5.0	5.0	13.2
NO02	0.3	1.71	0.11	6.3	5.0	5.0	2.1
PL06	0.8	0.25	0.11	2.7	10.0	2.7	6.7
PL10	0.8	0.40	0.10	4.1	10.0	4.1	11.7
SE04	0.8	0.71	0.17	7.3	10.0	7.3	7.1
SE14	0.8	0.28	0.22	3.3	10.0	3.3	3.2
SE15	0.8	0.47	0.27	5.7	5.0	5.0	4.1
SE16	0.8	0.42	0.21	4.8	5.0	4.8	1.3

denitrification fraction is related to the drainage properties of the catchment and was calculated from the fraction of peatlands (f_{peat}) in the terrestrial part of the catchment by $f_{de} = 0.1 + 0.7 \cdot f_{peat}$ (Table V.7 in the Manual, Posch et al. 1997).

For purposes of evaluating deposition reduction requirements, exceedances of critical loads are defined as the positive differences between deposition and critical loads (Mapping Manual, 2017), as a deposition lower than the critical load is not considered harmful for the ecosystem. Here we report, however, both negative and positive exceedance values ($Ex_{eut} = N_{dep} - CL_{eut}N$). This is done in order to illustrate the temporal trajectories of the pairs of calculated exceedances of eutrophication critical loads (Ex_{eut}) and observed TIN concentrations ([TIN]) as they vary from year to year in the (Ex_{eut}, [TIN]) plane.

3.3.2 Results and discussion

Being lower than the mass balance critical load, the empirical critical load of N was used for $CL_{eut}N$ at seven sites (AT01, FI03, LT01, LT03, NO01, NO02). At the other ten sites, the mass balance nutrient critical load was used (Table 3.3.1). The values of empirical critical loads ($CL_{emp}N$) depend on the allocation of the site vegetation to different EUNIS classes (Holmberg et al. 2013), as well as on the choice to use the lower end of the range of empirical CL for each EUNIS class (Bobbink & Hettelingh 2011). Site-specific considerations that determined the mass balance critical load values

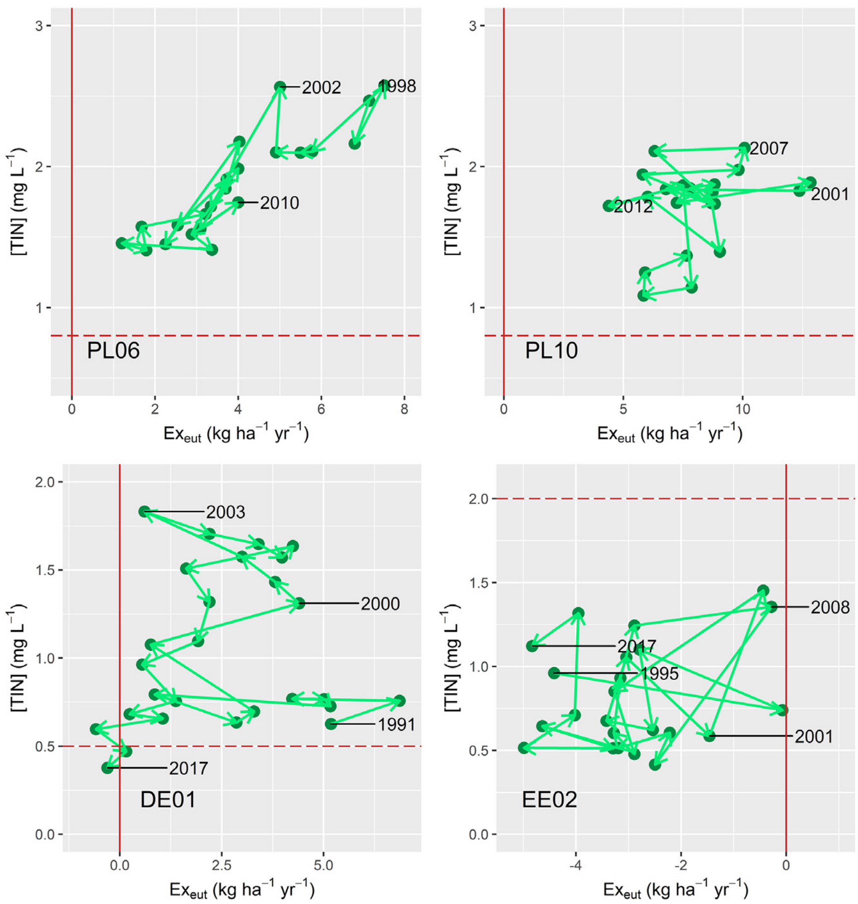

Figure 3.3.1. Response trajectories for sites PL06, PL10, DE01 and EE02. The horizontal dashed line indicates the acceptable nitrogen concentration, $[N_{acc}]$ (mg L^{-1}), and the vertical solid line represents the division between non-exceedance (Ex_{eut} <0) and exceedance (Ex_{eut} >0).

($CL_{nut}N$) include the choice of acceptable N concentrations [N_{acc}], the observed volume of runoff during the study period Q, and the fraction of peatland in the catchment f_{peat} (Table 3.3.1). For all sites, we assumed a low value of N immobilization (N_i = 0.5 kg ha^{-1} yr^{-1}), and no net removal of N in harvested vegetation (N_u = 0).

At the sites where $CL_{eut}N = CL_{nut}N$, the trajectory graphs (Figs. 3.3.1 – 3.3.2) show the acceptable nitrogen concentration [N_{acc}] as a horizontal dashed line. At all sites, the graphs (Figs. 3.3.1 – 3.3.5) show the division between non-exceedance and exceedance as a vertical solid line (Ex_{eut} = 0).

Some of the sites show a clear pattern of transition from earlier higher values of both Ex_{eut} and [TIN] towards currently lower values, especially PL06, PL10 and DE01. Of these, only DE01 reached non-exceedance and acceptable [TIN] during the observation period. At EE02, $CL_{eut}N$ was not exceeded, and [TIN] was acceptable during the observation period (Fig. 3.3.1, Table 3.3.1).

At the sites CZ01 and CZ02, [TIN] was acceptable during the observation period, despite $CL_{eut}N$ being clearly exceeded for these sites with relatively high deposition. At both SE04, which receives moderately high deposition, and at lower deposition SE14, [TIN] values occur despite positive Ex_{eut} values (Fig. 3.3.2, Table 3.3.1).

The high and the moderate deposition sites AT01 and LT03 remain exceeded with respect to $CL_{eut}N$ for the whole observation period. Low [TIN] values are combined

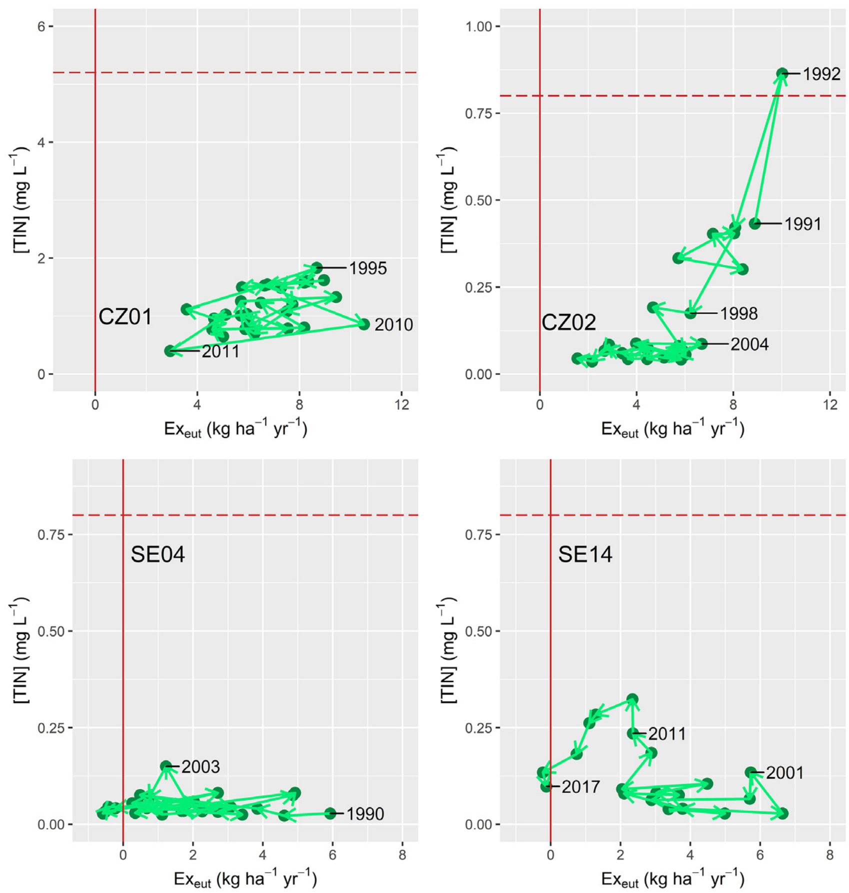

Figure 3.3.2. Response trajectories for sites CZ01, CZ02, SE04 and SE14. The horizontal dashed line indicates the acceptable nitrogen concentration, [N_{acc}] (mg L^{-1}), and the vertical solid line represents the division between non-exceedance (Ex_{eut} <0) and exceedance (Ex_{eut} >0).

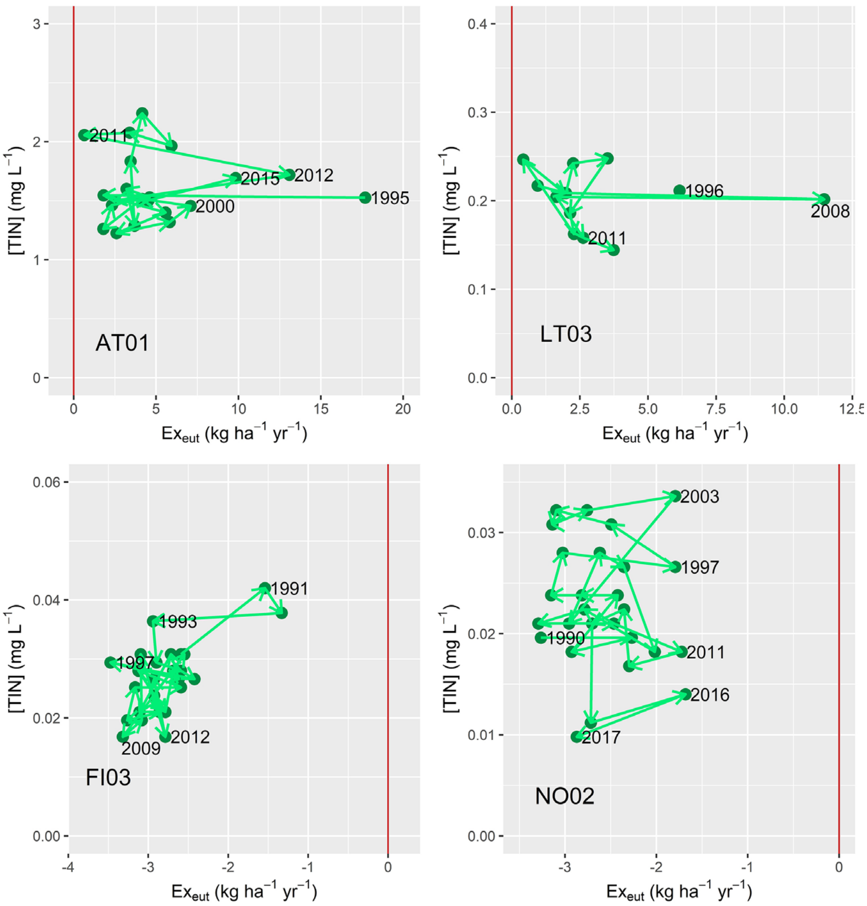

Figure 3.3.3. Response trajectories for sites AT01, LT03, FI03 and NO02. The vertical solid line represents the division between non-exceedance (Ex_{eut} <0) and exceedance (Ex_{eut} >0).

with lack of exceedance for the whole period at the low-deposition sites FI03 and NO02 (Fig. 3.3.3, Table 3.3.1).

A clear transition from exceedance to non-exceedance occurs at sites LT01 and SE15 receiving moderately high deposition. For LT01, this transition is accompanied by a clear decrease in [TIN] (Fig. 3.3.4, Table 3.3.1).

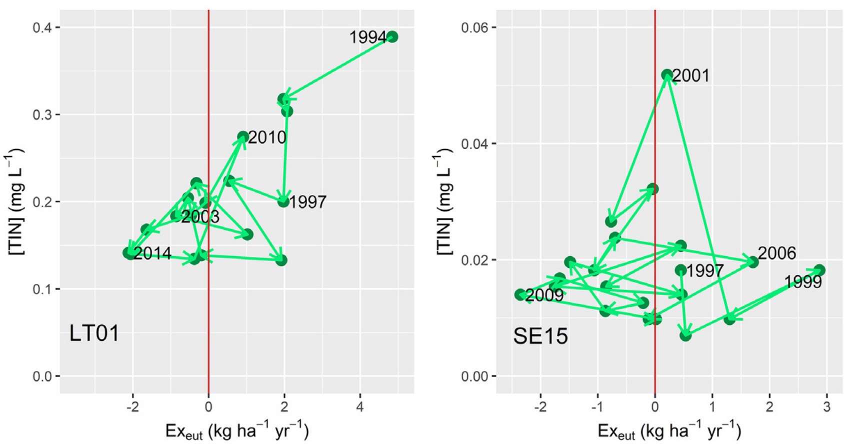

Figure 3.3.4. Response trajectories for sites LT01 and SE15. The vertical solid line represents the division between non-exceedance (Ex_{eut} <0) and exceedance (Ex_{eut} >0).

Critical loads are static quantities, designed to reflect long-term properties of the ecosystems. The trajectory graphs shown above (Figs. 3.3.1 to 3.3.4) combine the static nature of the critical loads with observations of actual ecosystem responses. Here we have assumed low nitrogen immobilization at all sites, and no net nitrogen uptake by vegetation. These are strong assumptions that may not be defendable at all sites. One has also to bear in mind that finite sized buffers in the soil – which are not part of steady-state CL models – can damped the response (output) of the system in reaction to changing inputs. The use of different approaches – static critical loads, dynamic modelling, empirical analysis - is useful to achieve a comprehensive view.

References

Allison, G.B. & Hughes, M.W. 1983. The use of natural tracers as indicators of soil water movement in a temperate semi-arid region. Journal of Hydrology 60: 157–173.

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F. & Winiwarter, W. 2011. Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications. Environmental Modelling & Software 26 (12): 1489–1501.

Bobbink, R. & Hettelingh, J.-P. 2011. Review and revision of empirical critical loads and dose-response relationships. Proceedings of an expert workshop, Noordwijkerhout 23-24 June 2010. Report 680359002, RIVM, Bilthoven, the Netherlands.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L. & de Vries, W. 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. Ecological Applications 20: 30–59.

Dirnböck, T., Grandin, U., Bernhardt-Römermann, M., Beudert, B., Canullo, R., Forsius, M., Grabner, M. T., Holmberg, M., Kleemola, S., Lundin, L., Mirtl, M., Neumann, M., Pompei, E., Salemaa, M., Starlinger, F., Staszewski, T. & Uzięblo, A.K. 2014. Forest floor vegetation response to nitrogen deposition in Europe. Global Change Biology 20: 429–440.

Dirnböck, T., Pröll, G., Austnes, K., Beloica, J., Beudert, B., Canullo, R., De Marco, A., Fornasier, M.A., Futter, M., Goergen, K., Grandin, U., Holmberg, M., Lindroos, A.J., Mirtl, M., Neirynck, J., Pecka, T., Nieminen, T.M., Nordbakken, J.F., Posch, M., Reinds, G.J., Rowe, E., Salemaa, M., Scheuschner, T., Starlinger, F., Uzięblo, A.K., Valinia, S., Weldon, J. Wamelink, W. & Forsius, M. 2018. Currently legislated decreases in nitrogen deposition will yield only limited plant species recovery in European forests. Environmental Research Letters 13 (2018) 125010.

Draaijers, G.P.J. & Erisman, J.W. 1995. A canopy budget model to assess atmospheric deposition from throughfall measurements. Water Air Soil Pollution 85: 2253–2258.

Grennfelt, P., Engleryd, A., Forsius, M., Hov, Ø., Rodhe, H. & Cowling. E. 2020. Acid rain and air pollution– 50 years of progress in environmental science and policy. Ambio 49: 849–864.

Holmberg, M., Vuorenmaa, J., Posch, M., Forsius, M., Lundin, L., Kleemola, S., Augustaitis, A., Beudert, B., de Wit, H.A., Dirnböck, T., Evans, C.D., Frey. J., Grandin, U., Indriksone. I., Krám, P., Pompei, E., Schulte-Bisping, H., Srybnyn, A. & Vána, M. 2013. Relationship between critical load exceedances and empirical impact indicators at Integrated Monitoring sites across Europe. Ecological Indicators 24: 256–265.

Holmberg, M., Aherne , J., Austnes, K., Beloica, J., DeMarco, A., Dirnböck, T., Fornasier, M.F, Goergen, K., Futter, M.J. , Lindroos, A-J.,, Krám, P., Neirynck, J., Nieminen, T.M., Pecka, T., Posch, M., Pröll, G., Rowe, E.C., Scheuschner, T. Schlutow, A., Valinia, S. & Forsius, M. 2018. Modelling study of soil C, N and pH response to air pollution and climate change using European LTER site observations. Science of the Total Environment 640–641: 387–399.

Mapping Manual 2017. German Environment Agency, Coordination Centre for Effects. www.icpmapping.org, Accessed date: 3 April 2020.

Posch, M., Kämäri, J., Forsius, M., Henriksen, A. & Wilander, A. 1997. Exceedance of critical loads for lakes in Finland, Norway and Sweden: Reduction requirements for acidifying nitrogen and sulfur deposition. Environmental Management. 21(2): 291–304

Posch, M., Duan, L., Reinds, G.J. & Zhao, Y. 2015a. Critical loads of nitrogen and sulphur to avert acidification and eutrophication in Europe and China. Landscape Ecology 30(3): 487–499.

Posch, M., De Vries, W. & Sverdrup, H.U. 2015b. Mass balance models to derive critical loads of nitrogen and acidity for terrestrial and aquatic ecosystems. Chapter 6 in: De Vries, W., Hettelingh, J.-P. & Posch, M. (eds), Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems. Environmental Pollution Series Vol. 25, Springer, Dordrecht, pp. 171–205.

Vuorenmaa, J., Augustaitis, A., Beudert, B., Clarke, N., de Wit, H.A., Dirnböck, T., Frey, J., Forsius, M., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Ruoho-Airola, T., Ukonmaanaho, L. & Váňa, M. 2017. Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990-2012). Ecological Indicators 76: 15–29.

Vuorenmaa, J. Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H., Dirnböck, T., Frey, J., Hakola, H., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Löfgren, S., Marchetto, A., Pecka, T., Schulte-Bisping, H., Skotak, K., Srybny, A., Szpikowski, J., Ukonmaanaho, L., Váňa, M., Åkerblom, S. & Forsius, M. 2018. Long-term changes (1990–2015) in the atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in Europe in relation to changes in emissions and hydrometeorological conditions. Science of the Total Environment 625:1129–1145.

Wright, R.F., Alewell, C., Cullen, J.M., Evans, C.D., Marchetto, A., Moldan, F., Prechtel, A. & Rogora, M. 2001. Trends in nitrogen deposition and leaching in acid-sensitive streams in Europe. Hydrology and Earth System Sciences 5: 299–310.

Annex I

Report on National ICP IM activities in Sweden in 2018

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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). 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, natural processes have considerably 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.

In the following, climate, hydrology and water chemistry related primarily to 2018 as well as some ongoing work at the four Swedish IM sites are presented (Löfgren 2019).

Climate and Hydrology in 2018

The measured data for 2018 from climate monitoring in the IM sites were compared to long-term (1961–1990) mean values from the Swedish Meteorological and Hydrological Institute (SMHI). The annual mean temperatures were higher (1.1–1.6 °C)

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

	SE04	SE14	SE15	SE16
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

compared to the long-term mean for all four sites. Largest deviation occurred at the northern site SE16 Gammtratten. Compared with the measured time series, 18 years at site SE16 Gammtratten and 22 years at the other sites, the temperatures in 2018 were somewhat higher at all four sites 1.1 and 1.2 °C at the two southern sites, and 1.5 and 1.6 °C at the two northern sites. 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 22 years average at three sites, while SE15 Kindla only deviated with 0.9 °C below this mean.

Compared to the SMHI long-term average values (1961–1990), the precipitation amounts in 2018 were considerably lower at all sites with only 64–74% of the long-term average at three sites. At SE04 Gårdsjön, the precipitation reached 86% of the long-term mean with monthly values varying between lower and higher values for seven and five months, respectively. The other sites had lower precipitation mainly from February to December with deviations at SE14 Aneboda for August (+31 mm) and SE16 Gammtratten also in February–March (+48 mm). Mainly summer and autumn showed low precipitation, especially for the two northern sites with low soil moisture content and extensive forest fires in those regions, however not hitting the IM sites.

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 groundwater levels often are low in winter when precipitation is stored as snow, with raising levels at snowmelt in spring and returning to lower levels in summer due to evapotranspiration. However, depending on rainfall amounts in summer and/or autumn, the groundwater levels could occasionally be elevated also during these periods. In 2018, the three sites SE14 Aneboda, SE15 Kindla and SE16 Gammtratten started the year on fairly high groundwater levels, receding to ordinary lower levels in March–April followed by elevated levels at snowmelt. Especially, groundwater levels at SE16 Gammtratten in the north reached relatively high levels. During the following months groundwater levels were lowered to unusually low levels in late summer and early autumn. Low evapotranspiration in autumn resulted in slightly elevated groundwater levels. However, only site SE15 Kindla reached ordinary high levels at the end of the year. At SE14 Aneboda and SE16 Gammtratten, the groundwater levels were lower than normal by c. 0.5 m. At site SE15

Kindla, a more varying pattern was observed with several peaks 0.2 m below the soil surface during snowmelt in March – April. However, the lowest levels in 2016–2017 c. 0.8 m below soil surface, were exceeded for five months in 2018 when the levels were at 1.5 m soil depth. The groundwater levels were reflected in the stream water discharge patterns (Fig. 1).

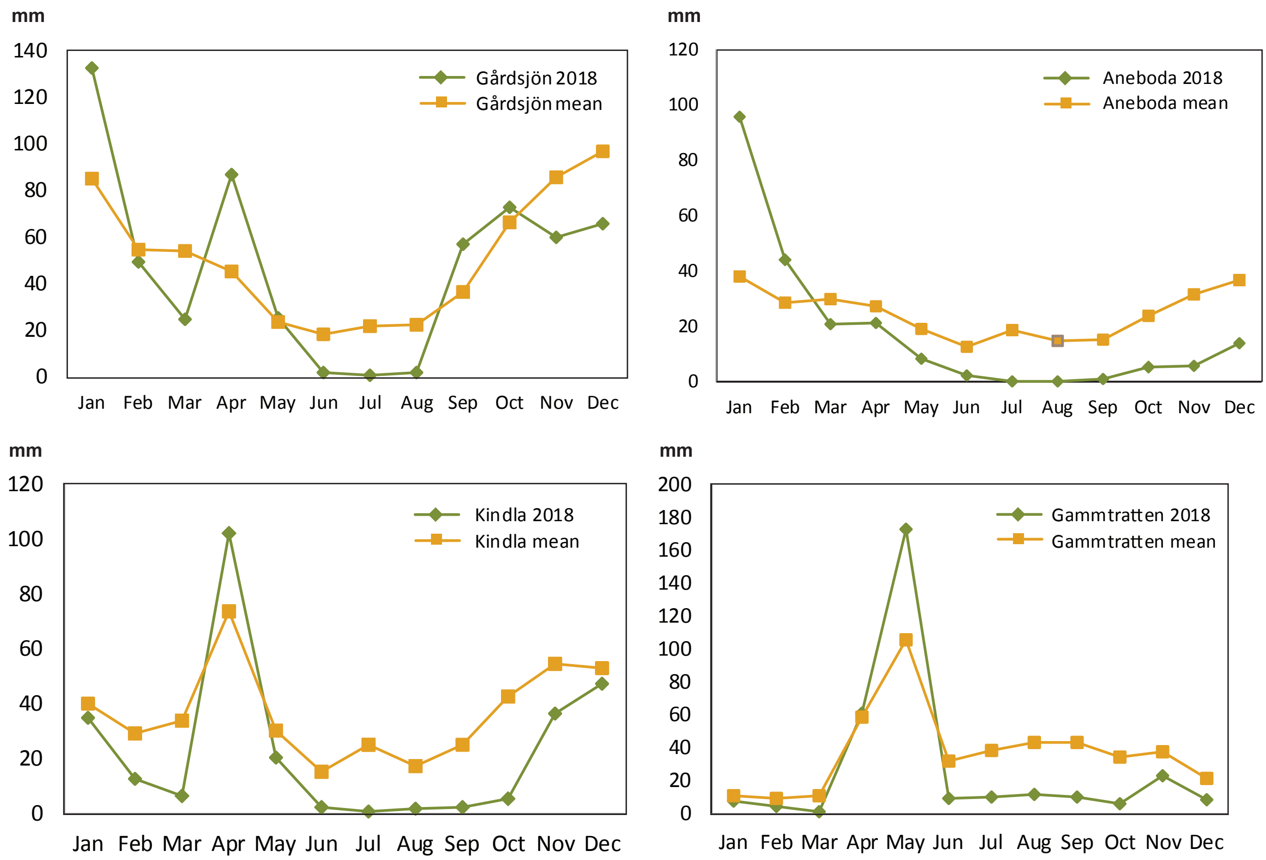

Figure 1. Discharge patterns at the four Swedish IM sites in 2018 compared to monthly averages for the period 1996–2018 (mean). Note the different scales at the Y-axis.

In addition to precipitation, evapotranspiration affects the runoff pattern. The runoff pattern for SE16 Gammtratten in 2018, was fairly typical with a snowmelt peak in May and lower discharges in summer and autumn, but with a small peak in November before low temperatures caused snowfall and snow accumulation. At SE04 Gårdsjön, the pattern was in accordance with the average except for high values in January and very low values in summer. Also comparably low flows occurred in November–December. Runoff at SE15 Kindla followed the ordinary pattern during 2018, but with considerably lower monthly runoff in June to November. Runoff at SE14 Aneboda showed high monthly values in the beginning of the year, turning to very low runoff by the end of the year (Fig. 1). In July surface water runoff actually ceased.

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. Runoff during 2018 exhibited a normal pattern with peaks in spring during snowmelt, but runoff was low throughout the year and especially during summer and autumn (Fig. 1). This pattern was not as evident at the two southern sites, where slightly higher runoff than normal was observed in January and at SE04 Gårdsjön also in April and September.

In 2018, the annual runoff made up 44–80% of the annual precipitation (Table 2), a wide range compared to the ordinary 40–60% but with mainly SE16 Gammtratten deviating with a large share. In 2016 the range was even larger (31–83%). At SE04 Gårdsjön, 2018 and 2017 were similar with shares of 64% and 63%, respectively, due to somewhat high runoff at the end of the year when evapotranspiration was low (Table 2). Runoff at this site, constituting almost 2/3 of the precipitation, is quite normal. At SE14 Aneboda, storm felling, followed by bark beetle attacks, have reduced the forest canopy cover, inducing low interception. The total evapotranspiration was estimated to 179 mm, which is considerably lower than in previous years with 477 mm in 2017 and 349 mm in 2016. Low precipitation and dry conditions seem to have contributed to this low evapotranspiration. At SE15 Kindla, the water balance was also influenced by low precipitation, resulting in low calculated evapotranspiration and runoff. However, the proportions related to precipitation were reasonably normal with 56% and 44%, respectively. At the northern site SE16 Gammtratten, throughfall and bulk precipitation were very similar (1% deviation), which indicates large uncertainties in these measurements. Similar patterns have been found for several years. Presumably, snow deposition infers the largest uncertainty, probably resulting in erroneous estimates of bulk precipitation. The precipitation observed at a nearby SMHI station showed slightly higher values, generating a higher and more realistic evapotranspiration. In summary and based on the estimated evapotranspiration (P-R), it must be concluded that the very dry summer 2018 furnished low evapotranspiration at all four sites (Table 2).

Table 2. Compilation of the 2018 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	906	100	397	100	619	100	409	100
Throughfall, TF	733	81	497	125	494	80	412	101
Interception, P–TF	173	19	-101	-25	125	20	-3	-1
Runoff, R	577	64	218	55	272	44	325	80
P–R	329	36	179	45	348	56	84	20

Water chemistry in 2018

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 (1.0 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 (5.6 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 (0–0.5 units) in throughfall compared with bulk deposition. In 2018, however, both SE04 Gårdsjön and SE16 Gammtratten had similar pH (ca 5.1) in both bulk deposition and throughfall. The two sites SE14 Aneboda and SE15 Kindla had 0.3–0.5 higher pH values in throughfall compared with bulk deposition (Table 3).

Table 3. Mean deposition chemistry values 2018 at the four Swedish IM sites. S and N in kg ha^{-1} yr^{-1}.

	SE04	SE14	SE15	SE16
pH, bulk deposition	5.1	4.9	5.0	5.1
pH, throughfall	5.1	5.4	5.3	5.0
S, bulk deposition	2.9	1.1	1.9	0.8
N, bulk deposition	7.9	3.8	6.5	1.4

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. Especially SE14 Aneboda and SE16 Gammtratten exhibited low pH-values in soil water, pH 4.4 and pH 3.6, respectively, compared with throughfall, pH 5.4 and pH 5.0, respectively. However, in the organic rich discharge areas at SE04 Gårdsjön and SE16 Gammtratten, pH was higher in soil solution compared with throughfall while the opposite was true at SE14 Aneboda and SE15 Kindla.

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 SE04 Gårdsjön with constantly negative values. In the discharge areas, the buffering capacity in groundwater varied between positive and negative values for the sites. Especially low values were found at Aneboda with ANC -0.11 mEq L^{-1} possibly dependent on nitrification in upslope locations. In groundwater in SE15 Kindla, ANC was comparably high with 0.18 mEq L^{-1} while SE04 Gårdsjön and SE16 Gammtratten showed values close to zero. Bicarbonate (HCO_3) occurred in 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 Gammtratten having a pH of 5.6. The stream water buffer capacity was positive at all sites (ANC ≥ 0.033 mEq L^{-1}), except for SE15 Kindla (ANC -0.005 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 SE14 Aneboda and SE04 Gårdsjön, the stream waters were more acidic compared with the other two sites, probably due to nitrification and the legacy of historically high acid deposition, respectively.

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. the sulphate concentrations were higher in stream water compared with deposition, indicating desorption or mineralization of previously accumulated sulphur in the soils. For Aneboda, nitrification contributed to fairly high nitrate values in the recharge area soil water (0.05 mEq L^{-1}), however, values being lower compared to previous year.

Considerably lower concentrations occurred in the discharge areas, probably due to nitrogen uptake and denitrification.

At site SE16 Gammtratten in the north, sulphate concentrations in soil water and stream water were considerably higher compared to throughfall, indicating release from the soil pool. Organic anions dominated anion flow in the stream with 2/3 of the content to be compared to 25% at SE15 Kindla, 15% at SE14 Aneboda and only 10% at SE04 Gårdsjön.

Soil and soil water processes could be evaluated through relationships between mainly sodium and chloride. In deposition, Na^+ dominated the base cations except for the northern site SE16 Gammtratten where Ca^{2+} showed the highest concentrations. At sites SE04 Gårdsjön and SE14 Aneboda, Cl^- was higher compared to Na^+, while the opposite occurred at the other two sites. A higher Cl^- outflow than Na^+ in stream water indicates ion exchange in the soil and release of other base cations, H^+ and/or Al^{3+} from the catchments. Mg^{2+} was the second highest base cation in runoff water at SE04 Gårdsjön, 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 with 18 mg L^{-1} while the other sites SE04 Gårdsjön, SE15 Kindla and SE16 Gammtratten showed lower values 16, 9, and 10 mg L^{-1}, respectively. High DOC concentrations create prerequisites for metal complexation and transport as well as high organic nitrogen fluxes. Organic nitrogen was the dominating nitrogen fraction in stream water, ranging from 0.19 to 0.44 mg N_{org} L^{-1}. The shares of N_{org}/N_{tot} were 81–97%, with SE14 Aneboda having the lowest share while SE16 Gammtratten and SE15 Kindla had the highest values. However, the nitrogen flux was higher at the two southern sites compared to the northern ones. Inorganic nitrogen (NO_3–N and NH_4–N) was low at the two sites SE15 Kindla and SE16 Gammtratten with 34 and 4 mg L^{-1}, respectively. At SE04 Gårdsjön, somewhat higher concentration (61 mg L^{-1}) was found, reflecting a somewhat higher deposition. The highest concentrations in stream water were found at SE14 Aneboda (102 mg L^{-1}), probably reflecting the poor forest stand condition. Compared to 2016, however, when the mean inorganic N concentration was 191 mg L^{-1}, a considerable decrease has occurred.

Total phosphorus (P_{tot}) in bulk deposition varied between 4 and 28 µg L^{-1} with the highest values at SE15 Kindla and lowest in site SE14 Aneboda. In stream water, SE14 Aneboda showed the highest P_{tot} (13 µg L^{-1}) as well as DOC concentrations. The other sites had average P_{tot} concentrations between 4 and 9 µg L^{-1} with the lowest value at SE15 Kindla.

Inorganic aluminum (Al_i), toxic to fish and other gill-breathing organisms, has been analyzed in soil solution, groundwater and surface waters at the IM sites. Relatively high total Al concentrations occurred in the soil solution (0.5–1.8 mg L^{-1}) in 2018, however, concentrations were only half of the concentrations observed in 2017. Sites SE04 Gårdsjön and SE14 Aneboda showed fairly high concentrations in groundwater in recharge areas with 1.2 mg L^{-1} and 1.6 mg L^{-1}, respectively. In stream water, Al_{tot}-concentrations were between 0.4 and 0.7 mg L^{-1} at three sites with low pH (4.4–4.8). Only at the northern site SE16 Gammtratten with a pH of 5.6, the total Al concentrations were lower, approximately 0.23 mg L^{-1}. Inorganic Al made up 13–51% of the total Al with the highest levels at low pH at SE15 Kindla, and the lowest at SE16 Gammtratten, corresponding to 0.04–0.27 mg Al_i L^{-1} and 0.03 mg Al_i L^{-1}, respectively. In 2018, both Al_{tot} and Al_i were somewhat higher in stream water compared with values in 2017. According to the SEPA classification system, the Al_i concentrations at SE04 Gårdsjön, SE14 Aneboda and SE15 Kindla are considered extremely high, but moderate at 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. However, methyl mercury, only measured at SE14 Aneboda and financed by SITES, was still relatively high creating prerequisites for bioaccumulation. In stream water, the mean Hg_{tot} and Hg-methyl concentrations were 5.3 ng L^{-1} and 0.4 ng L^{-1}, respectively, which is lower compared to values in 2017.

In summary, the four Swedish IM sites show low ion contents and permanently acidic conditions. 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 color, metal complexation, and phosphorus concentrations at all sites, but less at SE15 Kindla, where rapid soil water flow paths provide relatively low DOC and acidic waters. At SE14 Aneboda, the forest dieback provides a relatively high share of water runoff as well as high nitrate concentrations compared with the other three sites. At SE04 Gårdsjön, deposition is strongly influenced by the sea.

References

Löfgren, S., Grandin, U. & Stendera, S. 2014. Long-term effects on nitrogen and benthic fauna of extreme weather events: Examples from two Swedish headwater streams. Ambio 43, 58-76.

Löfgren, S. (ed.) 2019. Integrerad övervakning av miljötillståndet i svensk skogsmark – IM. Årsrapport 2018. Integrated monitoring of environmental status in Swedish forest ecosystems – IM. Annual Report for 2018. Rapport 2019:7. SLU. Uppsala. 30pp and 23 appendix. (In Swedish with English summary)

Annex II

Lago Nero observatory – Report of the five-years of activities as a contribution to ICP IM

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Background

We present an overview of the work conducted in 2015–2019 at Lago Nero, a high-alpine lake where the University of Applied Sciences and Arts of Southern Switzerland (SUPSI) established an ICP IM site supported by the Air Pollution Control and Chemicals Divisions of the Swiss Federal Office of the Environment (FOEN).

Mountain catchments are ideal indicators of large-scale pressures and, therefore, catchments in Europe and North America have been instrumented to monitor long-term effects of air pollution and climate change[1].

In Switzerland, since the 80s 20 mountain lakes on the southern side of the Alps, which is highly affected by long-range transport of the atmospheric pollutants originating from the highly industrialised Po plain in northern Italy, have been monitored extensively and included in ICP Waters program[2].

Owing to the predominance of base-poor rocks with very low buffer capacity, on the Southern Swiss Alps many high-altitude catchments and lakes are still sensitive to acidification despite the long term decreasing trends of N- and S- depositions.

Worldwide, extensive monitoring programs are increasingly complemented with intensive, integrated programs (e.g. ICP IM). We therefore proposed to the FOEN the integration of the extensive ICP Waters program with an intensive monitoring of one catchment on the Southern Swiss Alps and we identified the Lago Nero catchment as a potential site.

The catchment of Lago Nero was chosen based on the results of a pilot study[3], which indicated that it is well suited for the long-term monitoring of the ecological consequences of atmospheric pollutants and other environmental issues, considering that it is: (i) virtually unexposed to local anthropogenic impacts, (ii) sensitive to the impacts of interest including atmospheric pollutants and, at the same time, (iii) strongly exposed to these impacts. The other main criteria for the selection of the Lago Nero catchment were the nearby presence of an existing weather and deposition monitoring station (Lago Robièi, see Fig. 2) and the inclusion of the lake in the swiss ICP Waters program.

The monitoring project aimed at detecting trends in ecosystem responses (i.e. acidification recovery, N-nutrient enrichment effects, climate changes interactions) to atmospheric pollution and other environmental changes based on long-term monitoring of atmospheric pollutants (concentrations and types of pollutants) and

Figure 1. Lago Nero and its catchment in the Southern Swiss Alps.

climatic parameters (average and seasonal temperature, precipitation, freezing and thawing events, etc.). The various and complex effects of key atmospheric pollutants and environmental changes in general warrant an integrative monitoring of sensitive ecosystems.

The study site

The catchment of Lago Nero (Val Bavona, Ticino, Switzerland; Figs. 1 and 2). is south-west-facing, with altitude ranging from 2385 m to 2842 m a.s.l., an area of 77.5 ha and a mean slope of 84 %. The substrate is dominated by gneissic bedrock with patches of grassy vegetation and shallow soils. The catchment is snow-covered and the lake is ice-covered approximately from November to June. Lago Nero is an oligotrophic, soft-water lake with a surface of approximately 13 ha and a maximum depth of around 70 m. Monitoring of the site began in summer 2014, with an initial testing phase aimed at developing and testing methodologies and at evaluating the suitability of the catchment and the feasibility of the monitoring program[3].

Since the 80s, Lago Nero has been monitored by the cantonal administration and from 2000 it has been included in the Swiss network of ICP Waters sites, which focuses on a regional set of high-alpine lakes in Ticino. The double inclusion of Lago Nero in ICP W and ICP IM provides synergies in form of shared infrastructure and measurements and allows spatial and temporal comparison, i.e. with other similar lakes and – for some parameters – with long time series.

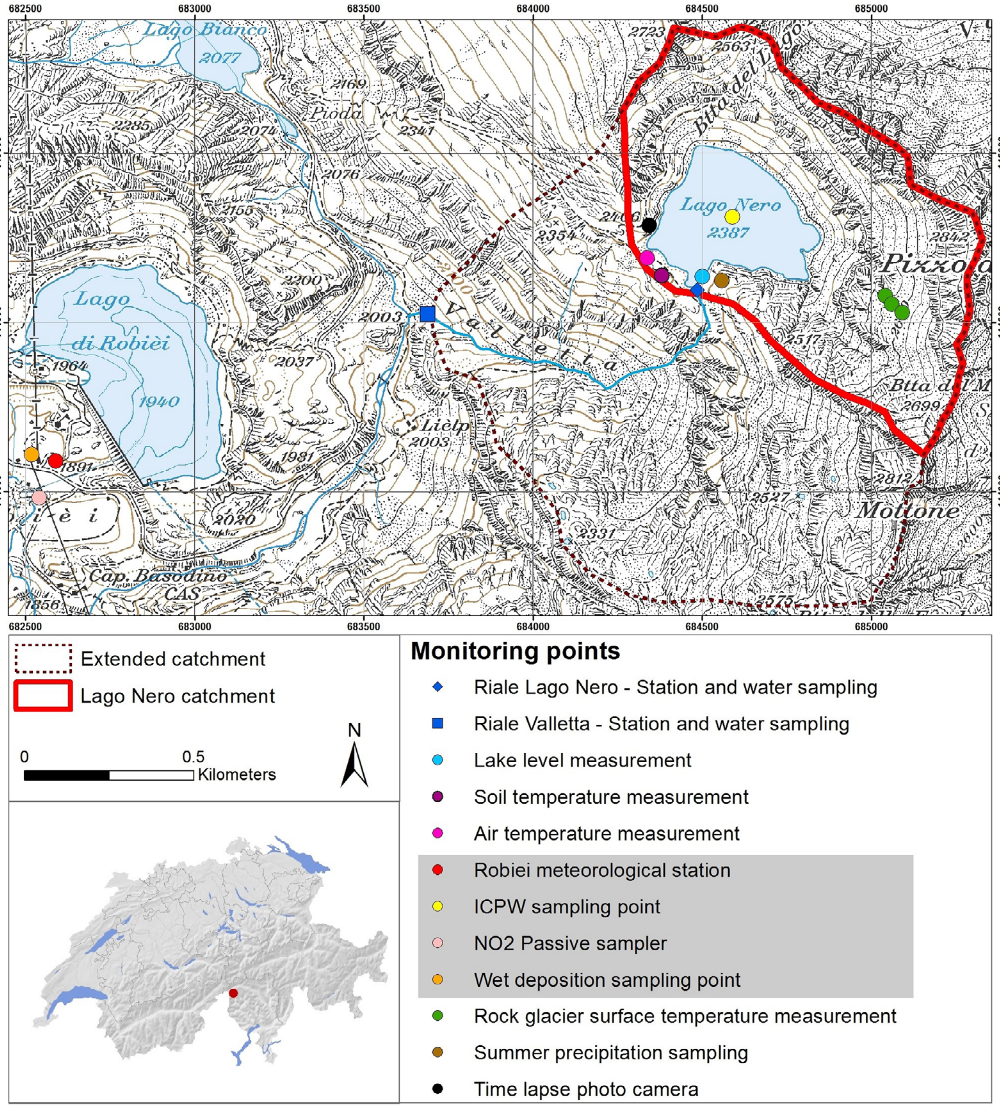

Figure 2. The Lago Nero catchment and its location in Switzerland. The monitoring program is based on various measurements made at the outflow of the lake and at the downstream confluence (extended catchment).

The monitoring program

Parameters and methodologies of the monitoring program generally followed ICP IM protocols and included the following: meteorology and air chemistry (based on the nearby station of Robièi managed by the Federal Office of Meteorology and Climatology MeteoSwiss, and on atmospheric models), precipitation chemistry (based on the nearby station Robièi, managed by the cantonal administration, Ufficio Aria, Clima ed Energie Rinnovabili, UACER, and the station in the catchment, managed by SUPSI), soil chemistry and soil-water chemistry, lake outflow discharge and water chemistry, vegetation composition. Because the Lago Nero catchment lies markedly above the treeline, the other ICP IM mandatory parameters (trunk epiphytes, throughfall, foliage and litterfall chemistry), were not surveyed.

In addition to parameters of the mandatory ICP IM program, data for parameters improving the understanding of the consequences of environmental changes on the

Table 1. Sampling frequency and data source of the mandatory (1–7), optional (8) ICP IM sub-programs and additional (9–12) monitoring run at the Lago Nero site, 2015–2019. Air and precipitation chemistry were sampled at the nearby Robièi Station.

	Sub-program	Sampling frequency	Data source
1	Meteorology	10 min 2 h for temperature	MeteoSwiss SUPSI
2	Air chemistry	monthly for NO_2-N yearly for AOT40 5-10 year for SO_2, NO_3^-, HNO_3, NH_3, NH_4^+	UACER MeteoTest (modelled)
3	Precipitation chemistry	weekly	UACER
4	Runoff water chemistry	monthly for chemical parameters 10 min for temperature and conductivity	SUPSI
5	Soil water chemistry	approx. monthly during summer	SUPSI
6	Vegetation survey	5-year intervals	SUPSI
7	Soil chemistry and structure	5-year intervals	SUPSI
8	Lake water chemistry	10 min for temperature and conductivity 3x/year for conductivity, pH, tot Alk, Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^-, SO_4^{2-}, NO_3^-, NO_2^--N, Cl^-, Ptot, N_{tot}, DOC, SiO2, heavy metals (tot and dissolved)	SUPSI UACER
9	Lake temperature profiles	1 h	SUPSI
10	Cryosphere (rock glacier and ice patches)	2x/year ice patches outflow	SUPSI
11	Snowpack and lake ice (including time lapse camera)	daily snow height in Robièi daily videos with camera	MeteoSwiss SUPSI
12	3D-model of the catchment (360° laser scan)	1x	SUPSI

ecosystems at this particular site were also collected. The mandatory ICP IM sub-programs were therefore integrated by measuring some additional physicochemical variables in order to provide further insight into the lake's and watershed's working and response to global changes. These parameters support interpretation of results from mandatory sub-programs or their extrapolation to the entire ecosystem and allowed to complete a detailed assessment of the chemical, physical and biological state of the Lago Nero catchment.

They included, for instance, measurements of the lake temperature profiles, ground surface temperature monitoring on an intact rock glacier and on perennial ice patches as well as of the water chemistry of a spring influenced by ground ice melting draining into the lake.

Table 1 reports the sub-programs ran at the Lago Nero monitoring site with indication of sampling frequency and their data source.

Main results and discussion

The results of the five-year monitoring program confirmed the preliminary outcomes of the pilot study. The runoff chemistry, the deposition data, and the input-output budgets calculated based on collected data[4] indicate that Lago Nero is affected by exceptionally high (for a remote high alpine environment) deposition of nitrogen and other key pollutants.

Temporal patterns of concentrations in nitrogen and other key chemicals in the outflow indicated that the snowmelt period contributes substantially (more than 50%, see Fig. 3) to the budgets and was therefore sampled with higher frequency.

For nitrogen, these inputs (wet deposition only and precipitation at Robièi) – ranging between 10.6–19.3 kg*ha^{-1}*a^{-1} during the observation period – are substantially higher than the critical load for eutrophication estimated at ~3 kg*ha^{-1}*a^{-1} for softwater alpine lakes[5,6,7] and 8 kg*ha^{-1}*a^{-1} for alpine grasslands[8]. These high levels of depositions can be partly explained by the very large precipitation rate at Robièi[9], which reaches a mean annual precipitation of 2400 mm*a^{-1} whereas the swiss mean annual precipitation is around 1500 mm*a^{-1}.

The N-load and the exceedance of critical loads for eutrophication are given in Table 2 and the latter, ranging between from 7.6 to 16.3 kg*ha^{-1}*a^{-1}, is likely to significantly alter aquatic and terrestrial biological communities[5,6,7].

Chemical budgets based on measurements of inputs and outputs (i.e. outflow concentrations and runoff) showed that much of the deposited nitrogen is retained in the catchment. The nitrogen percent net export (PNE), reported in Table 2, ranged from -73 to -86%, indicating a high retention (on average four fifths) of N-deposition in the catchment, whereas 20–30% of the deposited nitrogen still reaches the lake.

Another interesting result of the programme monitoring was that the catchment exports high amounts of sulphur relative to the atmospheric input (Table 2). Even though current deposition of sulphur, ranging between 3 to 6 kg*ha^{-1}*a^{-1}, is relatively low, the outputs of sulphur largely exceeded the inputs, ranging from +134% to a maximum of +253%. The release of a large amount of S from the catchment suggests the presence of other sources within the watershed, which might include the melting ground ice, which is hypothesized to release atmospheric sulphur stored in the past[10], or the weathering of S-bearing minerals (e.g., FeS_2 as pyrite or marcasite).

An assessment of the long term trends in input-output budgets of S and N in swiss high altitude lakes[11] indicates that high fluxes and high increasing rates of S release are limited to catchment with average altitudes higher than 2400 m a.s.l., showing that

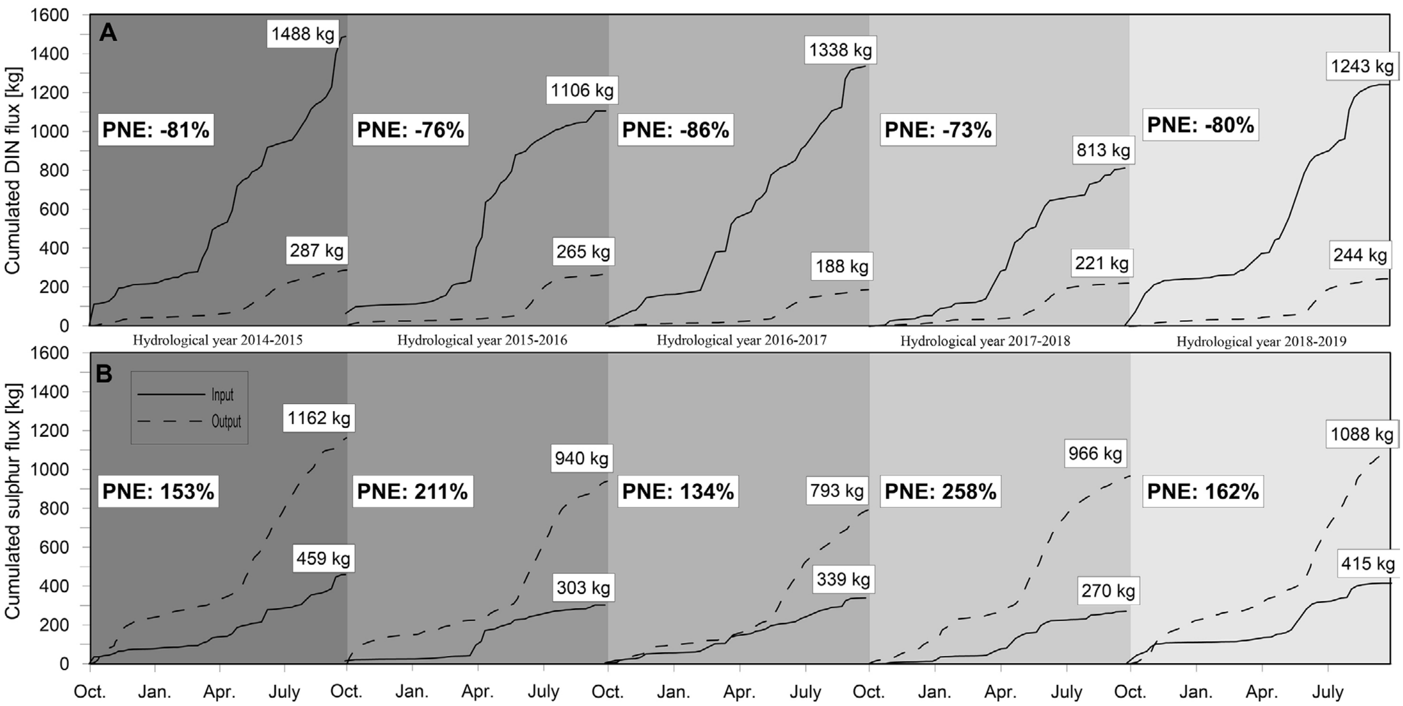

Figure 3. Multi-year comparison of the cumulated fluxes of nitrogen as DIN (dissolved inorganic nitrogen - upper panel) and sulphur (lower panel) at the Lago Nero catchment calculated during the hydrological years 2014–2015 to 2018–2019. Input and output fluxes in kg and percent net export (PNE) in %, defined as: PNE= (output – deposition) ∗ 100 / deposition.

the process of high amount release of S occurs only at high altitudes, thus strengthening the hypothesis of S release from the Alpine cryosphere.

Moreover the analyses of N and S budgets performed by the ICP IM program center[12] for several catchments across Europe showed that catchments retained SO_4 in the early and mid-90s but this shifted towards net release of S in the late-90s, whereas total inorganic N (TIN), in general, was strongly retained in the catchments not affected by natural disturbances, suggesting that the patterns observed for the catchment of Lago Nero, i.e. substantial retention of DIN and relevant release of sulphur, are relatively common.

Table 2. Interannual comparison of the chemical input/output balances for N and S, in kg and percent net export (PNE) in %, defined as: PNE= (output – deposition) * 100 / deposition. N- and S Load (calculated from their inputs), N Critical Load exceedance for eutrophication at Lago Nero in $kg{*}ha^{-1}{*}a^{-1}$.

Hydrological year		2014-2015	2015-2016	2016-2017	2017-2018	2018-2019
yearly precipitation (mm)		3082	2031	2313	1927	2696
Nitrogen (DIN)	Input ($kg.a^{-1}$)	1488	1106	1338	813	1243
	Output ($kg.a^{-1}$)	287	265	188	221	244
	PNE (%)	-81	-76	-86	-73	-80
	N Load, N $kg{*}ha^{-1}{*}a^{-1}$	19.3	14.4	17.4	10.6	16.1
	N Critical Load exceedance, CLexc , N $kg{*}ha^{-1}{*}a^{-1}$	16.3	11.4	14.4	7.6	13.1
Sulphur	Input ($kg.a^{-1}$)	459	303	339	270	415
	Output ($kg.a^{-1}$)	1162	940	973	966	1088
	PNE (%)	153	211	134	258	162
	S Load, S, $kg{*}ha^{-1}{*}a^{-1}$	6.0	3.9	4.4	3.5	5.4

Soil-water samples were collected during the snow free period 2017–2019. The total nitrogen concentration in soil-water samples showed a seasonal variation with generally increasing concentration with depth. In contrast, sulphate concentration did not show a seasonal or depth trend.

A vegetation survey was also performed at the beginning of the vegetative period, i.e. in mid-July 2017, consisting of a quantitative and a qualitative analysis of the plant species composition. The quantitative survey indicated that species richness was relatively poor on the vegetation plot (average species richness per subplot: 7.8) and that the community is dominated by species adapted to nutrient-poor and acid soils (e.g. including species like *Carex curvula*, *Leodonton helveticus*, *Salix herbacea* and *Homogyne alpina*). Communities in other areas of the catchment differed due to different orientation, slope, and humidity of the soils.

A permanent homogeneous soil chemistry plot was established in September 2018. In 2019, the sub-programme 'Soil chemistry and structure' was completed by the soil profile description, the soil base map and the pedological base map (see Fig. 4). The soil samples were analysed by measuring pH and quantifying concentrations of total sulphur, total phosphorus, total nitrogen, Ca^{2+}, Mg^{2+}, K^+, Na^+, Al^{3+}, and TOC. All the analysed profiles (soil profiles 1-7, see Fig. 4), showed common characteristics: an extremely low pH (around ~4.6), high content of organic carbon and strong mineral alteration, mostly due to the high amount of rainfall.

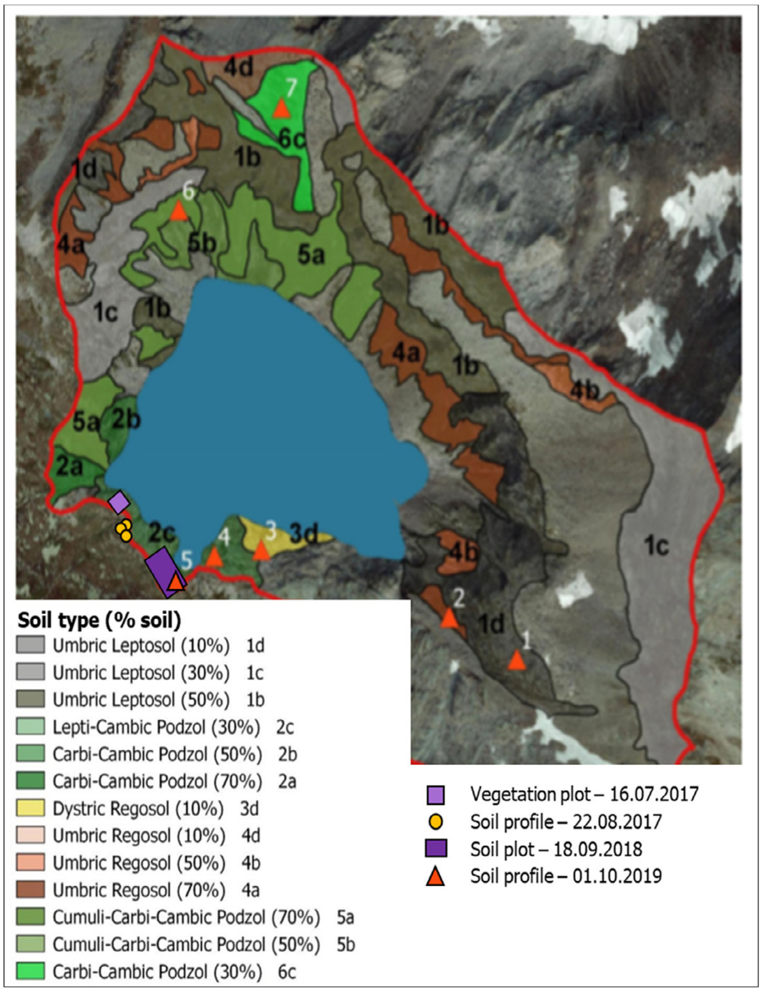

Figure 4. Soil map of the Lago Nero catchment. The cartographic units of the soil type and their percentage are defined according the FAO-UNESCO 1990 system[13].

At the beginning of summer 2017, a thermistor chain was installed until a depth of 40 m. The collected data during two years indicated that the waters of Lago Nero follow a typical dimictic regime for a small mountain lake, mixing from the surface to at least 40 m of depth twice a year, the first time in July and the second time in autumn (October-November). These turnovers are separated by periods of winter/spring (November through June) and summer (July through October) stratification. The timing of the turnovers and duration of the summer stratification period identified by the temperature profiles are particularly interesting because they are potentially sensitive to climate change. i.e., warmer climates are expected to lead to earlier summer turnovers and later autumn turnovers, and therefore longer summer stratification.

Cryosphere monitoring at Lago Nero site began in autumn 2015, to characterise the catchment geomorphology, assess the permafrost distribution, and monitor the intact rock glacier and the two perennial ice patches present in the catchment. Catchment geomorphology was assessed by digital mapping on swissimage orthophoto and on swissALTI3D 2m hillshaded Digital Elevation Model, focusing on landforms, ground texture and the presence of vegetation. The geomorphological and geological characteristics of the area as well as the potential local permafrost distribution were already presented in the ICP IM annual report 2017[14] and were coupled with measurements of ground surface temperatures, chemistry of Lago Nero outflow and meltwater chemistry from several perennial ice patches.

The comparison of elemental concentrations between the periglacial terrains and the Lago Nero outflow (see Fig. 5) unveiled the presence of high concentration of nitrogen and sulphur coming from the cryosphere[10,14]. These results suggest that Lago Nero is particularly sensitive to changes in the cryosphere, particularly concerning thickness of snow cover, snowmelt date and duration, and length of ice-free period of the lake surface.

Probable storage of ground ice during the 1966–1985 period and its significant melting in the last decades may explain the high amounts of sulphur measured at the ice patch outflow. The ground ice-fed headwaters with high levels of sulphur are likely to have ecological effects on the sensitive biota of the Lago Nero catchment, for instance by retarding the recovery from past acidification and representing peculiar ecosystems from both a chemical and biological point of view[15].

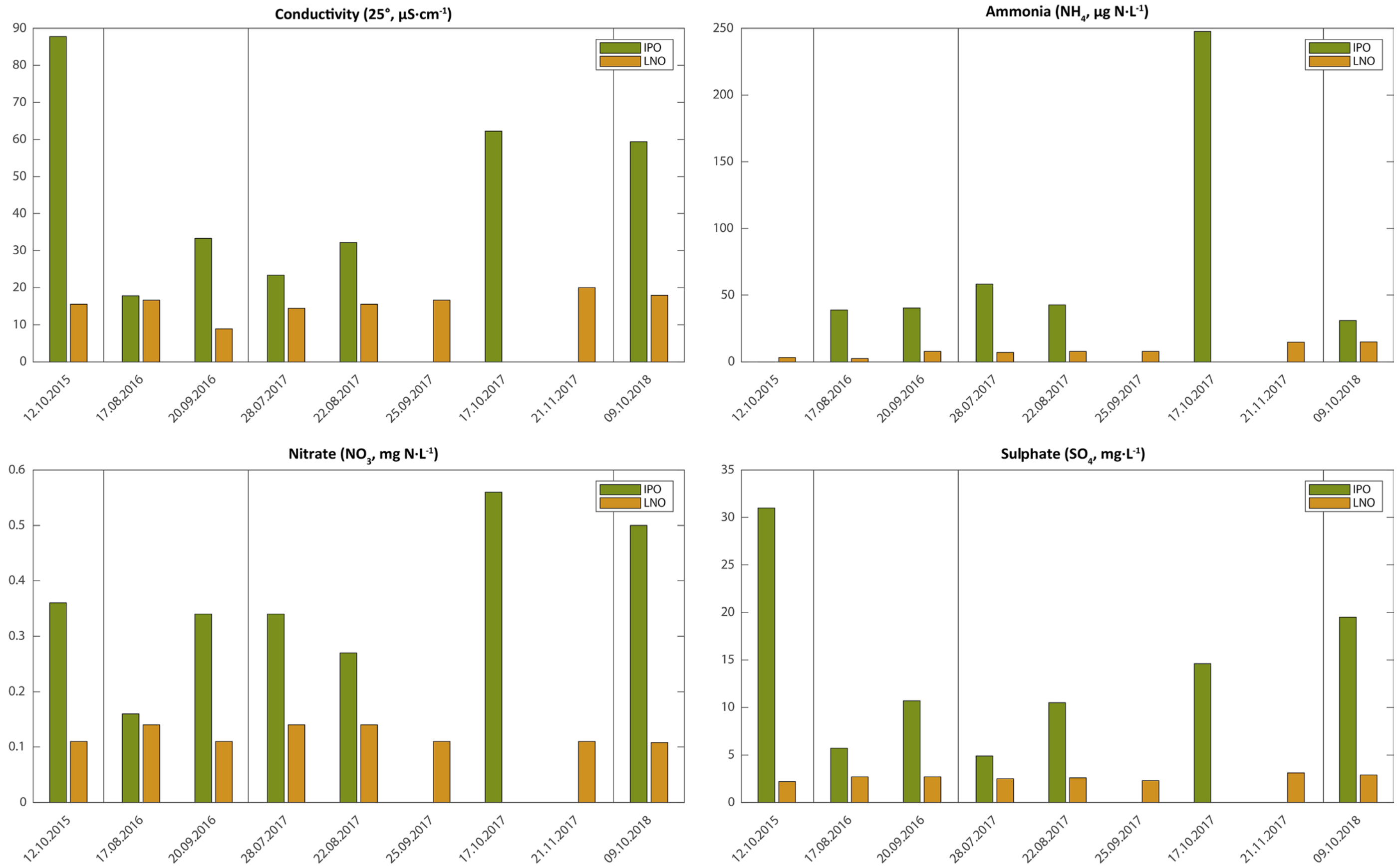

Figure 5. Comparison between the ice patch outflow (IPO) and the Lago Nero outflow (LNO), measured values for the 2015/2016, 2016/2017 and 2017/2018 hydrological years. Source: Scapozza et al. 2020[10].

Conclusions

The results of the environmental parameters studied during the period 2014–2019 improved our understanding of the Lago Nero catchment and the methods required to assess the impacts of atmospheric pollutants on the different ecosystem compartments (air, soil, water and terrestrial vegetation) of the catchment.

From a methodological point of view the Lago Nero site has proven to be suitable for long term monitoring of air pollution impacts on alpine ecosystems due to his peculiar location in the middle of the Alps, which is relevant and characteristic for ICP IM program monitoring at European level. The site is exposed to long-range transport of air pollutants from the south; very few sites with these characteristics exist at global scale (high altitude, high N-depositions, not forested).

The Lago Nero monitoring site is instrumented as agreed with ICP IM program center, considering the peculiar characteristics of this not forested high alpine site.

Regarding the scientific outcomes and considering that input-output budgets of eutrophying/acidifying N and S compounds derived from atmospheric deposition play a central role in ICP IM activities, three major conclusions can be proposed:

Assessment of acidification: the findings on element budgets at the catchment scale have shown how sulphur retention may delay the acidification recovery and contribute to maintain high sulphate concentrations in surface waters despite declining atmospheric deposition. Deposition and runoff fluxes of nitrogen are still high. The S positive net release could still affect the acidification recovery. The monitoring of the permafrost terrains revealed the presence of high concentration of S and partially N in water coming from a perennial ice patch, which may contribute to maintain high release of acidifying elements in the catchment and is likely to have ecological effects on the sensitive biota of the watershed.

Assessment of N-enrichment: retention and uptake of N is a relevant indicator of the nutrient enrichment. The runoff and deposition chemistry integrated by the input-output budgets point out a very high deposition of N, confirming that the nitrogen nutrient enrichment critical load for high-alpine lakes and grassland are still largely exceeded. The effects of the imbalance of N/P ratio due to the high deposition and retention rate of N should also be addressed and the phytoplankton would be an important indicator to estimate the impact on the biological recovery.

Climate changes effects: Global warming has been so rapid and intense over the last decades and the Alps are experiencing one of the highest increase in average annual temperature (up to ~2°C)[9,16,] that reconstructing long-term trends in surface water and following the temperature profile of the water column of the lake are key elements to characterise the environmental ecosystem. Ground ice in the rock glaciers and in the ice patches has been affected by the increasing temperature and therefore evaluations of permafrost distribution and monitoring of periglacial landforms can contribute to understand the water chemistry in the whole catchment and would be relevant to assess N- and S-fluxes which are partly dependent on deposition amount.

The five-years intensive monitoring was very valuable and allowed deeper insights into the characterisation of the Lago Nero catchment. Due to the long-term funding reduction and based on the main findings of the monitoring project, the future research at the Lago Nero will be redirected towards shorter-term projects aimed at addressing unanswered questions raised by the five-year programme and will focus on: i) the impact of N deposition on contemporary biological communities, ii) the reconstruction of the effects of atmospheric deposition on the biological communities of the lake from pre-industrial conditions to the present based on fossil diatoms and other proxies, iii) the relative contributions of atmospheric deposition, natural weathering and melting ground ice to the S budget of the catchment and the effect of legacy effects on the recovery from past S deposition, iv) the assessment of the microbial communities of the lake and his tributaries.

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References

[1] Lepori, F. & Keck, F. 2012. Effects of atmospheric nitrogen deposition on remote freshwater ecosystems, Ambio, 41, 235–246.

[2] Steingruber, S. & Colombo, L. 2006. Impacts of Air Pollution on Alpine Lakes and Rivers. Chemistry and biology in Alpine lakes and rivers in Southern Switzerland related to acidification from long-range transboundary air pollution: Monitoring results from 1980–2004. Environmental studies no. UW–0619. Federal Office for the Environment, Bern. 74 p.

[3] Bruder, A., Lepori, F., Pozzoni, M., Pera, S., Scapozza, C., Rioggi, S., Domenici, M. & Colombo, L. 2016. Lago Nero – a new site to assess the effects of environmental change on high-alpine lakes and their catchments, Report on National ICP IM activities in Switzerland. In: Kleemola, S. & Forsius, M. (Eds.) 25th Annual Report 2016. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 29/2016, pp. 52–56.

[4] Pozzoni, M., Bruder, A., Bulgheroni, M., Pera, S., Scapozza, C., Rioggi, S., Domenici, M. & Colombo, L. 2018. Lago Nero: monitoraggio della risposta di un ecosistema all'inquinamento atmosferico e ai cambiamenti ambientali, Bollettino della Società ticinese di scienze naturali, 106, pp. 23–33.

[5] De Wit, H. A. & Lindholm, M. 2010. Nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters – a review. ICP Water Report 101/2010. NIVA, Oslo, Norway, 41 p.

[6] Baron, J., Driscoll, C., Stoddard, J. & Richer, E. 2011. Empirical Critical Loads of Atmospheric Nitrogen Deposition for Nutrient Enrichment and Acidification of Sensitive US Lakes, BioScience, Vol. 61 No. 8, pp. 602–613.

[7] Posch, M., Eggenberger, U., Kurz, D. & Rihm, B. 2007. Critical Loads of Acidity for Alpine Lakes. A weathering rate calculation model and the generalized First-order Acidity Balance (FAB) model applied to Alpine lake catchments. Environmental Studies no. 0709. Federal Office for the Environment, Berne, 69 p.

[8] Achermann, B., Rihm, B. & Kurz, D. 2011, National Focal Centre Report – Switzerland. In: Posch, M. Slootweg, J. & Hettelingh, J.-P. (eds), CCE Status Report 2011, Coordination Center for Effects, RIVM, Bilthoven, pp. 155–165.

[9] MeteoSvizzera, 2012, Rapporto sul clima – Cantone Ticino 2012, rapporto di lavoro MeteoSvizzera, p. 63.

[10] Scapozza, C., Deluigi, N., Bulgheroni, M., Pera, S., Pozzoni, M., Colombo, L. & Lepori, F. 2020. Assessing the impact of ground ice degradation on high mountain lake environments (Lago Nero catchment, Swiss Alps), Aquatic Sciences, 82; 5, 2020.

[11] Steingruber, S. 2017. Trends in input-output budgets of S and N in swiss high-altitude lakes, ICP Waters report 131/2017, Proceedings of the 33rd Task Force meeting of the ICP Waters Programme in Uppsala, May 9-11, 2017, pp. 14–22.

[12] Vuorenmaaa, J., Augustaitis, A., Beudert, B., Clarke, N., De Wit, H., Dirnböck, T., Frey, J., Forsius, M., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A-J., Lundin, L., Ruoho-Airola, T., Ukonmaanaho, L. & Vána, M. 2017. Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990–2012), Ecological Indicators 76, pp. 15–29.

[13] FAO UNESCO 1990. Soil map of the World. Revised legend, World Soil Resources Report 60. Rome 1990.

[14] Scapozza, C., Colombo, L., Domenici, M., Lepori, F., Pera, S., Pozzoni, M., Rioggi, S. & Bruder, A. 2017. Lago Nero – an example of mountain lake monitoring in a changing Alpine cryosphere. Report on National ICP IM activities in Switzerland. In: Kleemola, S. & Forsius, M. (Eds.) 26th Annual Report 2017. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of the Finnish Environment Institute 24/2017, pp. 43–49.

[15] Tolotti, M., Cerasino, L., Donati, C., Pindo, M., Rogora, M., Seppi, R. & Albanese, D. 2020. Alpine headwaters emerging from glaciers and rock glaciers host different bacterial communities: Ecological implications for the future. Science of the Total Environment 717, 2020, 137101.

[16] Dokulil, M.T., Teubner, K., Jagsch, A., Nickus, U., Adrian, R., Straile, D., Jankowski, T., Herzig, A. & Padisak, J. 2010. The impact of climate change on lakes in Central Europe. In: George, G. (Ed.), The Impact of Climate Change on European Lakes. Springer, Dordrecht, The Netherlands, pp. 387–409.

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 summarises 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 2019/2020 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 temporal trends and input - output budgets of heavy metals in ICP IM catchments
- An interim assessment of the impact of internal nitrogen-related parameters and exceedances of critical loads of eutrophication on long-term changes in the inorganic nitrogen output in European ICP Integrated Monitoring catchments
- National Reports on ICP IM activities are presented as annexes.

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