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

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wge Working Group on Effects of the
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Abstract

The Integrated Monitoring Programme (ICP IM) is part of the effect-oriented activities under the 1979 Convention on Long-range Transboundary Air Pollution, which covers the region of the United Nations Economic Commission for Europe (UNECE). The main aim of ICP IM is to provide a framework to observe and understand the complex changes occurring in natural/semi natural ecosystems. This report summarizes the work carried out by the ICP IM Programme Centre and several collaborating institutes. The emphasis of the report is in the work done during the programme year 2022/2023 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
- Brief summary of forthcoming publication on heavy metal concentrations trend analysis
- Update on developing co-operation with eLTER
- Proposed workplan for the next period
- Report on IM participation in Canadian led mercury sampling
- National Reports on ICP IM activities

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

Abbreviations

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

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Preface

Welcome to the 32nd Annual Report, produced by the Programme Centre at the Swedish University of Agricultural Sciences. This year's Task Force meeting was held in Lunz, Austria, where our hosts were celebrating 30 years of the Zöbelboden site, which is an important part of the IM network. We thank them for hosting a successful meeting and wish them many more years of monitoring! The cover photo for this year's report is of one of the many steep slopes at Zöbelboden that house monitoring equipment. Minutes of the TF meeting may be downloaded from the IM website (link below).

As well as the usual updates, this edition of the annual report includes features on our suggestions for the next workplan, information on co-operation with the eLTER research infrastructure, an analysis of trends in heavy metal concentrations and an extended update from Sweden with an overview of the latest data.

James Kurén Weldon, on behalf of the Programme Centre

The Programme Centre team is as follows:

James Kurén Weldon – Head of Programme Centre

Karin Eklöf – Evaluation of heavy metals data

Martyn Futter – Senior researcher, with focus on modelling

Hampus Markensten – Validation of incoming data, updating the database, and handling data excerpts

Pernilla Rönnback – Database manager/administrator

The current co-chairs of IM, Ulf Grandin and Salar Valinia are also in Sweden, facilitating close co-operation with the Programme Centre.

The website for IM can be found at www.slu.se/en/icp-im and a PDF version of the monitoring manual can be downloaded from the link above, as well as minutes of meetings and previous editions of this report.

Comprehensive 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, <http://www.slu.se/en/icp-im>) 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 Swedish University of Agricultural Sciences, in Uppsala. 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 versions, which can be downloaded from the website).

Assessment activities within the ICP IM

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

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

Conclusions from international studies using ICP IM data

Input-output and proton budgets, C/N interactions

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

The most recent results from ICP IM studies are available from the study of Vuorenmaa et al. (2017). Site-specific annual input-output budgets were calculated for sulphate (SO₄) and total inorganic nitrogen (TIN = NO₃-N + NH₄-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 SO₄ and TIN were also analysed. Large spatial variability in the input and output fluxes of SO₄ 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 SO₄ deposition and bulk deposition of TIN was found at 90% and 65% of the sites, respectively. Output fluxes of non-marine SO₄ 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 SO₄ 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 SO₄ 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 SO₄ was explained by variations in runoff and SO₄ 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 SO₄ may lead to a slower recovery of surface waters than those predicted by the decrease in SO₄ 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.

Summary of earlier results from ICP IM studies

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 conjunction 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 & Kakebeke 2004, Working Group on Effects 2004).

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

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

Trend assessments

Empirical evidence on the development of environmental effects is of central importance for the assessment of success of international emission reduction policy. In order to assess the impacts of air pollution and climate change in the environment, a long-term integrated monitoring approach in remote

unmanaged areas including physical, chemical, and biological variables is needed. Vuorenmaa et al. (2018) evaluated long-term trends (1990–2015) for deposition and runoff water chemistry and fluxes, and climatic variables at 25 ICP IM sites in Europe that commonly belong also to the LTER Europe/ILTER networks. The trend assessment was published in a special issue in *Science of the Total Environment* with the title: “International Long-Term Ecological Research (ILTER) network”. The recent results from trend assessment at IM sites confirm that emission abatement actions are having their intended effects on precipitation and runoff water chemistry in the course of successful emission reductions in different regions in Europe. Concentrations and deposition fluxes of xSO_4 , and consequently acidity in precipitation, have substantially decreased in IM areas. Inorganic N (TIN) deposition has decreased in most of the IM areas, but to a lesser extent than that of xSO_4 . Substantially decreased xSO_4 deposition has resulted in decreased concentrations and output fluxes of xSO_4 in runoff and decreasing trends of TIN concentrations in runoff – particularly for NO_3 – are more prominent than increasing trends. In addition, decreasing trends appeared to strengthen over the course of emission reductions during the last 25 years. TIN concentrations in runoff were mainly decreasing, while trends in output fluxes were more variable, but trend slopes were decreasing rather than increasing. However, decreasing trends for S and N emissions and deposition and deposition reduction responses in runoff water chemistry tended to be more gradual since the early 2000s. Air temperature increased significantly at 61% of the sites, while trends for precipitation and runoff were rarely significant. The site-specific variation of xSO_4 concentrations in runoff was most strongly explained by deposition. Climatic variables and deposition explained the variation of TIN concentrations in runoff at single sites poorly, and as yet there are no clear signs of a consistent deposition-driven or climate-driven increase in TIN exports in the catchments.

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

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

Summary of earlier trend studies from IM

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 SO₄, NO₃ and NH₄ 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 SO₄ and base cation trends in runoff waters were commonly observed at the ICP IM sites. At some sites in the Nordic countries decreasing NO₃ 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 SO₄ 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 NO₃ 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.

A 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 SO₄ and TIN were evaluated for the period 1990–2012 (Vuorenmaa et al. 2017). These results clearly showed the regional-scale decreasing trends of SO₄ in deposition and runoff/soil water, and suggested that IM catchments have increasingly responded to the decreases in S emissions and depositions of SO₄ since the early 1990s. Decreased nitrogen emissions also resulted in decrease of inorganic N deposition, but to a lesser extent than that of SO₄, 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 SO₄ in forested catchments fuelled 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 SO₄ results in generally higher leaching fluxes of SO₄ than those of inorganic N in European forested ecosystems. SO₄ thus remains the dominant source of actual soil acidification despite the generally lower input of SO₄ 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. Recent studies using ICP IM data on dose-response relationships showed a weak recovery of epiphytic lichen communities in Sweden despite improvements in air quality (Weldon & Grandin, 2021), and found a relationship between increasing levels of nitrogen deposition and increased dominance of nitrophyllic bryophytes in forest understorey, but no effect on species richness (Weldon et al., 2022). Previous work was published by Dirnböck et al. (2014). This study utilised a new ICP IM database for biological data and focussed on effects on forest floor vegetation from elevated nitrogen deposition..

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 questions that the old database could not. As a direct consequence, Dirnböck et al. (2014) utilised the re-reported long-term monitoring data from 28 Integrated Monitoring sites to analyse the effect by nitrogen deposition on temporal trends in plant species cover and diversity. 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) found that 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.

Summary of earlier work on biological data from IM

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 SO₂ in air, NH₄⁺, NO₃⁻ and SO₄²⁻ 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.

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

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

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

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

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

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

Pools and fluxes of heavy metals

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

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

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

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

The objective of the aluminium (Al) contribution of Krám and Kleemola in the 28th Annual Report (2019) was to collect and present recently available information about Al fractions from the Integrated Monitoring (IM) database and stimulate the IM National Focal Points to checkout and add not yet reported Al fractions data to the IM database for a publication in peer-reviewed journal. Aluminium (Al) does not belong to the group of so-called heavy metals and is not transferred in large quantities by atmospheric deposition to forest catchments like most of the heavy metals. However, elevated inputs of strong acids from the anthropogenic atmospheric deposition to sensitive sites could mobilize Al from soils and stream sediments in a form of potentially toxic Al fractions to surface waters (Gensemer & Playle 1999). Different fractions of aqueous Al have very different toxicity levels for aquatic biota. Modified methods of the original Al fractionation procedure of Driscoll (1984) were applied and reported from fourteen IM catchments. Total monomeric Al (Al_m) and organic monomeric Al (Al_o , sometimes called non-labile Al) were measured in surface water by a colorimetry method. The Al_o was separated using a strong cation exchange resin, the method utilized charge exclusion by ion exchange. Potentially toxic inorganic monomeric Al (Al_i , sometimes called labile Al) was calculated as the difference between Al_m and Al_o . The ICP IM database contains relevant data about Al fractions in surface runoff from fourteen catchments so far. These catchments belong to seven countries: Finland (5), Norway (3), United Kingdom (2), Czech Republic (1), Estonia (1), Sweden (1) and Switzerland (1). Distinct patterns were evident in runoff waters of these catchments. The highest Al_i values were detected at CZ02 (median $340 \mu\text{g L}^{-1}$) and at SE04 (median $210 \mu\text{g L}^{-1}$). Very high Al_i concentrations were measured at NO01 and NO03 (median $170 \mu\text{g L}^{-1}$ and $130 \mu\text{g L}^{-1}$, 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).

Summary of earlier work on heavy metals

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 ($>1.4 \text{ mg m}^{-2} \text{ yr}^{-1}$) and Cd ($>0.15 \text{ mg m}^{-2} \text{ yr}^{-1}$) were observed in two mountainous Central European sites with high water discharge. All other sites had outputs below or

equal to 0.36 and 0.06 mg m⁻² yr⁻¹, respectively, for the two metals. Almost complete retention of Hg, 86–99% of input, was reported in the Swedish sites. These high levels of metal retention were maintained even in the face of recent dramatic reductions in pollutant loads. In the Progress report on heavy metal trends at ICP IM sites (Åkerblom & Lundin 2015) temporal trends were seen in forest floor with decreasing concentrations for Cd and Pb while Hg did not change. An increase in heavy metal concentrations was also seen in deeper mineral soil horizon indicating a translocation of heavy metals from upper to deeper soil horizons.

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

The critical load (CL) methodology has been a key science-based tool for assessing the environmental consequences of air pollution. Critical loads are deposition thresholds used to describe the sensitivity of ecosystems to atmospheric deposition. Critical loads for eutrophication and acidification were computed using a long-term dataset of intensively studied forested ecosystem ICP Integrated Monitoring sites (n = 17) in northern and central Europe (Forsius et al. 2021). The sites belong to the ICP Integrated Monitoring and eLTER networks. The link between the site-specific calculations and time-series of CL exceedances and measured site data was evaluated using long-term measurements (1990–2017) for bulk deposition, throughfall and runoff water chemistry. Novel techniques for presenting exceedances of CLs and their temporal development were also developed. Concentrations and fluxes of sulphate, total inorganic nitrogen (TIN) and acidity in deposition substantially decreased at the sites. Decreases in sulphur (S) deposition resulted in statistically significant decreased concentrations and fluxes of sulphate in runoff and decreasing trends of TIN in runoff were more common than increasing trends. The temporal developments of the exceedance of the CLs indicated the more effective reductions of S deposition compared to nitrogen (N) at the sites. There was a relation between calculated exceedance of the CLs and measured runoff water concentrations and fluxes, and most sites with higher CL exceedances showed larger decreases in both TIN and H⁺ concentrations and fluxes. Sites with higher cumulative exceedance of eutrophication CLs (averaged over 3 and 30 years) generally showed higher TIN concentrations in runoff. The results provided evidence on the link between CL exceedances and empirical impacts, increasing confidence in the methodology used for the European-scale CL calculations. The results also confirm that emission abatement actions are having their intended effects on CL exceedances and ecosystem impacts.

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

Planned activities

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

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ICP IM activities, monitoring sites and available data

Review of the ICP IM activities from June 2022 to June 2023

Meetings

- Salar Valinia, Ulf Grandin, and James Kurén Weldon represented ICP IM at the Eighth Joint Session of the Working Group on Effects and the Steering Body to EMEP, held in Geneva 12–15 September 2022.
- Ulf Grandin, Salar Valinia and James Kurén Weldon represented ICP IM at the videoconference of the joint meeting of EMEP Steering Body and Working Group on Effects Extended Bureau, held online 7-8 February 2023.
- James Kurén Weldon represented ICP IM and gave a presentation about current activities at the ICP Vegetation Task Force meeting, held on-line 13–15 Feb 2023.
- Salar Valinia represented ICP IM at the Saltsjöbaden VI workshop held in Gothenburg, Sweden 13-15 March 2023.
- James Kurén Weldon represented ICP IM and gave an online presentation about current activities, at the CDM/CCE/M&M meeting, held in Prague, Czechia 28-30 March 2023.
- Martyn Futter, and James Kurén Weldon took part in the eLTER PPP and PLUS project meeting, held in Frankfurt, Germany 17–21 April 2023. A workshop on co-operation between eLTER and the WGE was held.
- Ulf Grandin, Salar Valinia, James Kurén Weldon, Karin Eklöf and Martyn Futter represented ICP IM at the joint meeting of EMEP Steering Body and Working Group on Effects Extended Bureau, held in Uppsala, Sweden, 24–26 April 2023.
- The thirty-first meeting of the Programme Task Force on ICP Integrated Monitoring was held at Lunz, Austria (and online), 10–12 May 2023.
- James Kurén Weldon represented ICP IM and gave a presentation about current activities, at the ICP Forests Task Force meeting, held on-line 6–8 June 2023.

Data issues

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

Scientific work and activities in priority topics

- A scientific paper on Hg and heavy metal trends in concentrations across ICP Integrated Monitoring sites in Europe (Eklöf et al.) was submitted for publication (2023).
- New passive mercury samplers installed at participating sites as part of a large international collaboration led by Canadian researchers.
- Task Force Chairs together with the Programme Centre and a group of experts from the IM community developed the Extended IM concept.

Activities and tasks planned for 2023–2024

Carried over from the previous workplan, ICP IM will produce the following papers:

- Scientific paper on Hg and HM trends in concentrations across ICP Integrated Monitoring sites in Europe (WGE item 1.1.1.1.6), peer reviewed scientific publication, submitted for publication.

ICP IM activities on the WGE 2022–23 work plan:

- 2022: Operationalise and advertise Extended IM as an attractive monitoring protocol, aiming at adding more ecosystem types in the ICP IM monitoring.
- 2023: Scientific paper or report on modelling and assessment of biodiversity and ecosystem impacts, in co-operation with e.g., Centre for Dynamic Modelling (CDM).

Other activities

- Maintenance and development of central ICP IM database at Swedish Agricultural University (SLU).
- Arrangement of the 32nd Task Force meeting (2024).
- Preparation of the 33rd ICP IM Annual Report (2024).
- 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).

Published reports and articles 2022–2023

Evaluations of international ICP IM data and related publications

Weldon, J. (ed.), 2021. 31st Annual Report (2022). Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Annual Reports of the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems 31, Uppsala.

<https://res.slu.se/id/publ/118336>

Weldon, J., Merder, J., Ferretti, M., Grandin, U. (2022). Nitrogen deposition causes eutrophication in bryophyte communities in central and northern European forests. *Annals of Forest Science* 79(24). doi: 10.1186/s13595-022-01148-6

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

Monteith, D. T., Henrys, P. A., Hruška, J., de Wit, H. A., Krám, P., Moldan, F., . . . Evans, C. D. (2023). Long-term rise in riverine dissolved organic carbon concentration is predicted by electrolyte solubility theory. *Science Advances*, 9(3), eade3491. doi:10.1126/sciadv.ade3491

Liptzin, D., Boy, J., Campbell, J.L., Clarke, N., Laclauf, J.-P., Godoy, R., Johnson, S.L., Kaiser, K., Likens, G.E., Pihl Karlsson, G., Markewitz, D., Rogora, M., Sebestyen, S.D., Shanley, J.B., Vanguelova, E., Verstraeten, A., Wilcker, W., Worralls, F., McDowell, W.H. (2022). Spatial and Temporal Patterns in Atmospheric Deposition of Dissolved Organic Carbon. *Global Biogeochemical Cycles*, doi: 10.1029/2022GB007393.

Monteith D.T., Henrys P.A., Hruška J., de Wit H.A., Krám P., Moldan F., Posch M., Raike A., Stoddard J.L., Shilland E.M., Gloria Pereira M., Evans C.D. (2023) Long-term rise in riverine dissolved organic carbon concentration is predicted by electrolyte solubility theory. *Science Advances*, 9(3): eade3491, 1-11.

Petrash D.A., Krám P., Pérez-Rivera K.X., Buzek F., Čuřík J., Veselovský F., Novák M. (2023) Soil solution data from Bohemian headwater catchments record atmospheric metal deposition and legacy pollution. *Environmental Science and Pollution Research*, doi.org/10.1007/s11356-023-25673-7.

Yttri, K. E., Canonaco, F., Eckhardt, S., Evangeliou, N., Fiebig, M., Gundersen, H., Hjellbrekke, A.-G., Lund Myhre, C., Platt, S. M., Prévôt, A. S. H., Simpson, D., Solberg, S., Surratt, J., Tørseth, K., Uggerud, H., Vadset, M., Wan, X., and Aas, W.: Trends, composition, and sources of carbonaceous aerosol at the Birkenes Observatory, northern Europe, 2001–2018, *Atmos. Chem. Phys.*, 21, 7149–7170, <https://doi.org/10.5194/acp-21-7149-2021>, 2021.

Reports:

Austnes, K., Hjermann, D.O., Sample, J.E., Wright, R.F., Kaste, O., de Wit, H. 2022. Nitrogen in surface waters: time trends and geographical patterns explained by deposition levels and catchment characteristics. NIVA-rapport 7728-2022. ICP Waters report 149-2022.

Monitoring sites and data

The following countries have continued data submission to the ICP IM database during the period 2016–2022: Austria, Belarus, the Czech Republic, Estonia, Finland, Germany, Ireland, Italy, Lithuania, Norway, Poland, the Russian Federation, Spain, Sweden, and Switzerland.

The number of sites with on-going data submission for at least part of the data years 2015–2022 is 48 from fifteen countries. Sites from Canada, Latvia and United Kingdom only contain older data.

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

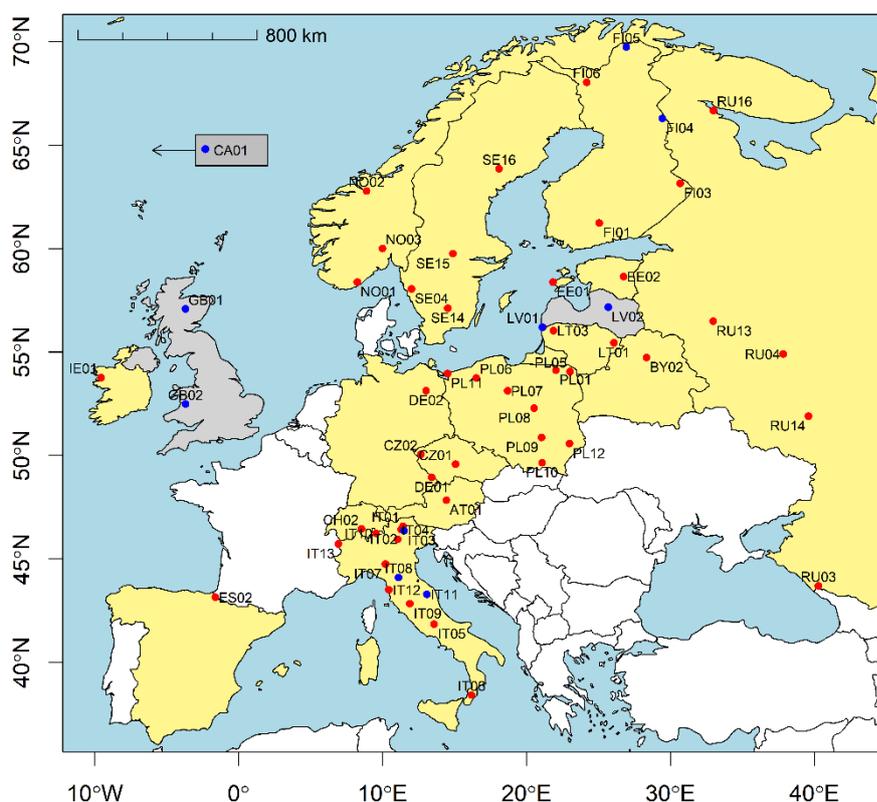


Figure 1: Location and status of ICP IM sites. For site names, see table 1.1. Red mark: active site. Blue mark: non-active site. Yellow: country with active IM sites. Grey: country with only non-active sites.

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

AREA	SUBPROGRAMME																*									
	AM meteorology	AC air chemistry	PC precipitation chemistry	MC moss chemistry	TF throughfall	SF stemflow	SC soil chemistry	SW soil water chemistry	GW groundwater chemistry	RW runoff water chemistry	LC lake water chemistry	FC foliage chemistry	LF litterfall	RB hydrobiology of streams	LB hydrobiology of lakes	FD forest damage	VG vegetation	BI bioelements	VS vegetation structure	EP trunk epiphytes	AL aerial green algae	MB microbial decomposition	BB bird inventory	BV vegetation inventory		
AT01 ZÖBELBODEN	95-21	95-21	93-21		93-21	99-04	04	93-21		95-21	-	92-21	93-21				93				93-98					
BY02 BEREZINA BR	89-15	89-15	89-15				95-98			95-15																
CH02 LAGO NERO	15-19	15-19	15-19				18	17-19		15-19	15-16						17									
CZ01 ANENSKÉ POVODI	89-21	89-21	89-21	89	89-21		02-20	07-21	08-21	89-21	-			07	-											
CZ02 LYSINA	67-20	93-96	90-21		91-21		93	90-21	89-21	89-21	91-21	94	08	07	11		15	94			14-15		10			
DE01 FORELLENBACH	90-21	90-21	90-21	90	90-21	90-05	90-11	90-21	88-21	90-21	-	90-21	90-21		-	90-14	90-08		00	92-95	94-21	91-02	90-95			
DE02 NEUGLOBSOW	67-21	98-21	98-21		98-21	04-21	04-16	98-21	98-21		98-21	06-21	04-21				04-17									
EE01 VILSANDI	95-21	94-21	94-21	94-20	94-21	94-21	94-20	94-21	95-96	-	-	94-21	94-21	-	-	94-17	94-21			94-21		94-21		94		
EE02 SAAREJÄRVE	94-21	98-21	94-21	94-21	94-21	94-21	94-20	95-21	95-21	94-21	94-21	96	94-21	94-21		96-17	96-12	12		94-21	94-17	96-21	98-14			
ES02 BERTIZ	08-17	08-21	07-21		07-21	08-19	10-15	07-18		07-21		08-21	08-21			07-12	07-21		07							
FI01 VALKEA-KOTINEN	88-21	94-20	88-21	88-96	89-17	89-99	88-89	89-17		88-21	87-21	88-17	90-16		90-93	88-91	88-09			88-97	90	87-89	87			
FI03 HIETAJÄRVI	88-21	93-00	88-21	89-96	89-17	89-99	88	89-17	93-21	88-21	87-21	88-17	90-16		90	88-91	90-09			90-97	90-91	87-89				
FI06 PALLASJÄRVI	13-21		14-21		16-17			02-17	18-21	04-21	04-21	95-17	07-16										88-89			
IE01 BRACKLOON WOOD			91-16		91-11	92-97		91-16		-	91-96	91-98	-	-												
IT01 RENON-RITTEN	90-19	93-19	93-14		93-13	93-13	93-11	93-13		00-13	-	93-10	00	-	-	92-13	09		05-09	92		93-11				
IT03 PASSO LAVAZE	92-21	93-21	92-13		94-13	94-00	93-95	95-07		01-13	-	93-05	94	-	-	93-19	95-09		99-09	92						
IT05 SELVA PIANA	97-21	97-21	97-19		97-19	97-19	95	02-08		-	-	97-19		-	-	97-19	09		99-09							
IT06 PIANO LIMINA	99-19	97-16	97-21		97-21	97-21	95	19-20		-	-	97-19		-	-	97-19	09		99-09							
IT07 CARRAGA	97-21	97-21	97-21		97-21	97-00	95	19-20		98-13	-	97-19		-	-	97-19	09		99-09							
IT09 MONTE RUFENO	97-21	97-21	97-21		97-21	97-00	95	02-08		97-14	-	97-19		-	-	97-19	09		99-09							
IT10 VAL MASINO	97-19	00-15	97-15		97-15		95	05-07		-	-	97-05		-	-	97-09	09		99-09							
IT12 COLOGNOLE	97-01	97-15	97-15		97-15	97-00	95			-	-	97-05		-	-	97-09	09		99-09							
IT13 LA THUILE	97-21	97-15	09-15		09-15		95			-	-	97-05		-	-	97-09			99-08							
LT01 AUKŠTAITIJA	93-13	93-19	93-19	93-20	93-19		93-20	94-20	93-20	93-20		06-19	99-20	12-20		00-20	93-20	20	02-20	93-20	93-20		93			
LT03 ZEMAITIJA	90-13	95-19	95-19	06-20	95-19		94-20	95-20	95-20	95-20		06-19	99-20	95-20		00-20	94-20	20	02-20	94-20	94-20		94			
NO01 BIRKENES	87-21	87-21	87-21	92	89-21		87-11	86-21	87-88	87-21	-	86-17	87-02		-	91-18	86-18		86							
NO02 KÄRVATN	87-91	87-21	87-21	88	89-11		89-13	89-10		87-21	-	89-09	89-02		-	92-10	89-09									
NO03 LANGTJERN		87-97	77-21		86-03		91-13	91-03		87-21		86-03	87-02													
PL01 PUSZCZA BORECKA	06-21	16-21	16-21		16-21	21	17	10-21		16-21	16-21	06-21				16										
PL05 WIGRY	06-21	16-21	16-21		16-21	21	19	06-21		16-21		05-21				16										
PL06 PARSENTA	10-21	16-21	94-21		96-21	21		10-21		94-21		10-21														
PL07 POJEZIERZE CHELMINSKI	16-21	16-21	16-21				18	20		16-21																
PL08 KAMPINOS	09-21	16-21	16-21		16-21	21	16	12-19		16-21		10-21				16										
PL09 LYSOGORY	05-21	16-21	16-21		16-21	21		05-21		16-21		05-21				16										
PL10 BESKIDY	94-21	16-21	94-21		02-21	21		11-21		94-21		09-21				16										
PL11 WOLIN	16-21	16-21	16-21		17-21	21	21	16-21		16-21		16-21														
PL12 ROZTOCZE	16-21	16-21	16-21		16-21	21		16-21		16-21		16-21				16										
RU03 CAUCASUS BR	89-94	89-21	89-98																							
RU04 OKA-TERRACE BR	89-06	89-21	89-98	90										93-99	93-21	93-02			93		94-96					
RU12 ASTRAKHAN BR	93-94	93-21	93-94																							
RU13 CENTRAL FOREST BR	93	93-94	93												09-21	18-20										
RU14 VORONEZH BR	94	94-21	94-98																							
RU16 VELIKIY ISLAND				89-90			89	89	89						93-99	93-21	91-94			89-94	93	94-95	91			
RU47 KURSK															18-21											
SE04 GÅRDSJÖN F1	87-21	88-21	87-21	95	87-21		95-10	87-21	79-21	87-21	-	99-21	96-20		-	97-01	95-19	91-20	91-20	96-21	92-21	95-21				
SE14 ANEBODA	96-21	96-21	96-21	95	96-21		96-11	95-21	96-21	96-21	-	99-21	95-20		-	97-01	82-19	96-21	06-16	97-17	97-21	95-21				
SE15 KINDLA	97-21	96-21	96-21		96-21		97-12	95-21	97-21	96-21	-	97-21	95-20		-	98-01	96-20	98-18	98-18	98-18	98-18	97-21	95-21			
SE16 GAMMTRATTEN	99-21	99-21	99-21		99-21		00-18	00-21	00-21	99-21		99-21	00-20			00-01	99-21	99-19	99-19	00-20	00-21	00-21				

Trend maps

Here we present a new feature, maps showing trends in the IM data series. The idea is to provide the reader with a quick visual overview of trends across the IM network over the last 10 years, with annual updates as new data become available. We are beginning with trends in throughfall concentrations of SO_4 , NH_4 and NO_2 but will expand the coverage in future editions of the annual report to encompass more subprogrammes. At each site trends were analysed using the non-parametric Seasonal Kendall test (Hirsch et al. 1982) applied to monthly data. The magnitude of trend was estimated by the slope estimation method (Sen 1968), which estimates the slope by calculating the median of all between-year differences in the variable of interest. For the analysed parameters, a calculated positive value of test statistics S indicates an increasing slope (increasing values with time), and a negative value indicates a decreasing slope. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis.

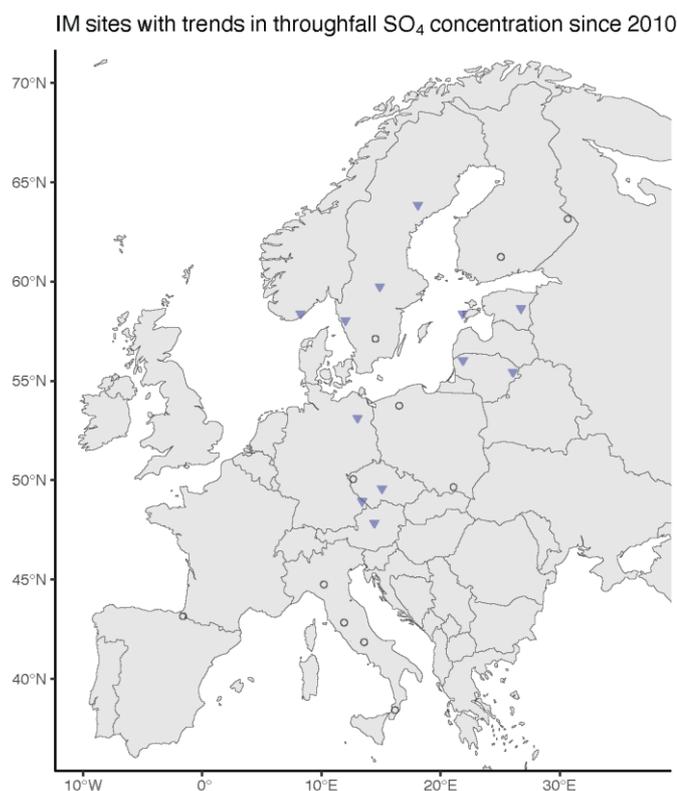


Figure 2: Trends in throughfall SO_4 concentrations since 2010, blue arrows indicate declining trend, red arrows increasing trend (Mann Kendall tests)

IM sites with trends in throughfall NO_2 concentration since 2010

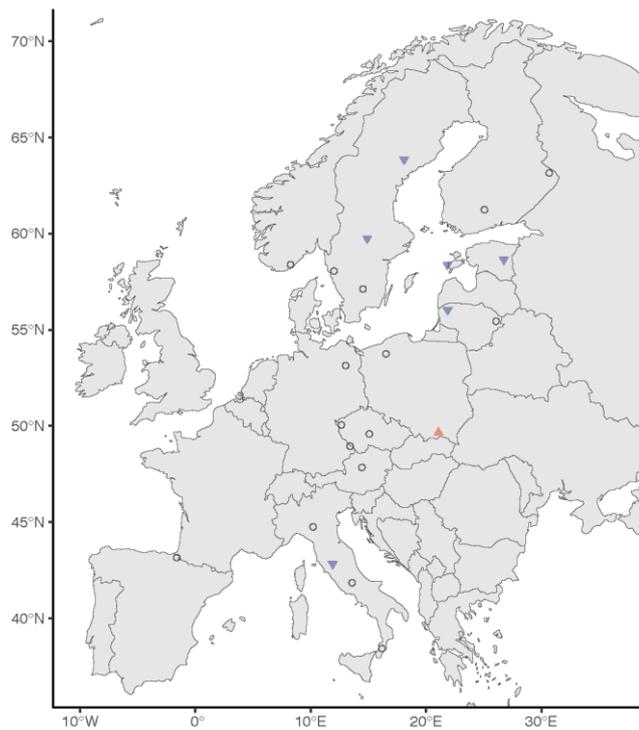


Figure 3: Trends in throughfall NO_2 concentrations since 2010, blue arrows indicate declining trend, red arrows increasing trend (Mann Kendall tests)

IM sites with trends in throughfall NH_4 concentration since 2010

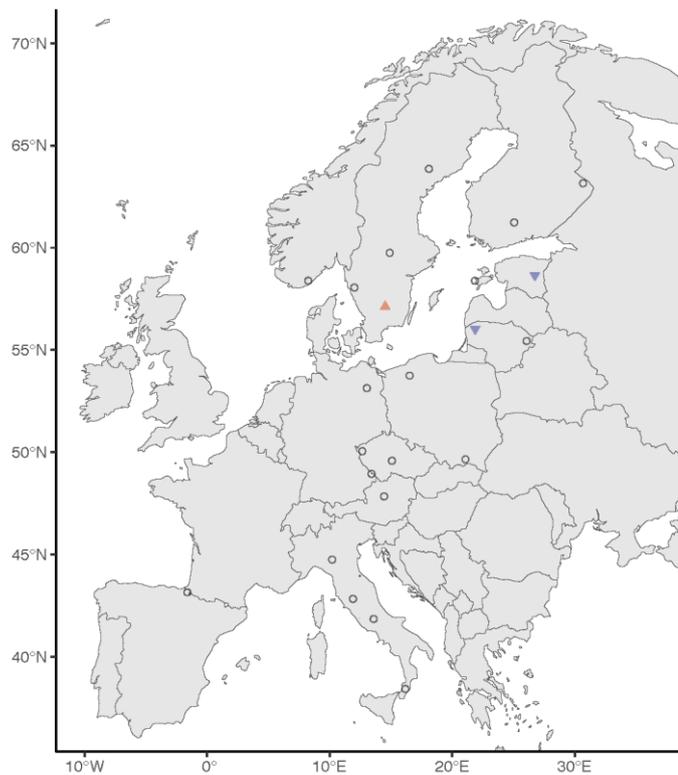


Figure 4: Trends in throughfall NH_4 concentrations since 2010, blue arrows indicate declining trend, red arrows increasing trend (Mann Kendall tests)

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International network of passive mercury samplers

In collaboration with Environment and Climate Change Canada, the University of Toronto Scarborough developed a passive air sampler for gaseous mercury (sold under the MerPAS[®] name). The Government of Canada committed to funding a pilot study to investigate applying this passive air sampler globally, the intent of which is to assess the feasibility of a globally implemented monitoring program using the samplers to establish a baseline concentration, especially in remote locations. Unusually for such a network, locations in Europe were lacking, and ICP IM were contacted to help. Participating IM sites have helped to fill important gaps in their global coverage (see map reflecting the network when we were first contacted). Their funding covers sending out the samplers on a quarterly basis, which are then returned to them for analysis, with the resulting data being freely available. As data from the network become available we look forward to incorporating them in analyses.



Figure 5: Canadian led mercury sampling network coverage before IM joined the study.

Passive air sampling of gaseous elemental mercury at Swedish IM stations

Pernilla Rönnback and Karin Eklöf

Passive air sampling of gaseous elemental mercury (GEM) is a relatively cheap and easy method to estimate the air concentrations of GEM, both for spatial mapping and temporal variations. Since January 2023 all four IM stations (Aneboda IM, Gårdsjön IM, Kindla IM and Gammtratten IM) in Sweden are participating in a study with passive air sampling of GEM using the MerPAS[®]. This effort is a 2-year pilot study led by the Environment and Climate Change Canada (ECCC) and the University of Toronto Scarborough, Canada.

The samplers at the four Swedish IM stations are installed in an open area at the same site where sampling of wet deposition occurs. Two samplers and one blank sampler are put out for three months and then replaced by new samplers. The collected samplers are sent back to the ECCC Mercury Research Laboratory for analysis. The MerPAS® is designed to absorb mercury from the air on a carbon trap. The amount of GEM taken up by the sampler generally increased linearly with time of sampling, and this is used to calculate the GEM concentration in air. Both wind speed and temperature could increase the Hg uptake by the passive air samplers. The samplers are thereby located in ICP IM stations where wind speed and temperature are also measured, to be used to adjust the GEM concentrations.

Due to the low cost of passive air samplers and no need of electricity (although electricity is needed for the climate station), it has been suggested that these samplers can greatly increase the global coverage of GEM measures. By ICP IM stations joining this large global network of GEM measures, we will contribute to the monitoring of the efforts within the Minamata Convention of Hg.



Figure 6: Mercury passive air sampling at Gammtratten IM, Sweden.

Trend analyses in heavy metal concentrations at IM sites

Karin Eklöf, the IM Programme Centre's heavy metals specialist and colleagues are submitting for publication a new study analysing trends in concentrations of mercury (Hg), lead (Pb), and cadmium (Cd) within the ICP Integrated Monitoring network and the Swedish environmental monitoring program. While the full results can be summarised in the next edition of the annual report, after publication in a scientific journal, some key findings are as follows.

Most of the water courses had no significant trends in Hg concentrations during 2000-2020. In the water courses with significant decreasing trends of Hg, trends were mainly observed during the period between 2000-2005.

Concentrations of Pb and Cd decreased in 35% and 70% of the water courses, respectively, and the concentrations of these elements have flattened out after 2005. However, it is important to note that towards the end of the evaluated time period, 2015-2020, more watercourses showed significant increasing, rather than decreasing trends in Hg, Pb and Cd concentrations and the reasons for this somewhat unexpected finding need further investigation.

The overall concentration trends could be driven by declining deposition of heavy metals over Europe, which is especially strong for Pb and Cd. Also, other changes related to metal transport and chemistry may contribute, such as processes related to the aqueous recovery from acidification in northern latitudes, and the ongoing browning of surface waters in the northern hemisphere. It is, however, noteworthy that most catchments have upward trends in dissolved organic matter, which would potentially counteract the decline in Hg and Pb since these are strongly complexed with organic matter.

Co-operation with eLTER

After many years of informal close co-operation between the eLTER (Integrated European Long-Term Ecosystem, critical zone and socio-ecological Research) network and the WGE, a formal letter of co-operation was drafted, circulated, revised, and officially adopted at the Geneva meeting in September 2022. Approximately half of the ca 200 sites that will form the core of the eLTER Research Infrastructure are also part of one or more ICP Networks, so in many ways this was an obvious step to take. At that Geneva meeting ICP IM was tasked with moving the process forward, and has raised the question of how to practically proceed at both the eLTER consortium meeting in Frankfurt (April 2023) and the Task Force meetings of several ICPs. The focus is on the following areas:

1. Standardisation and harmonisation
 - Measurement protocols and standards (eLTER standard observations)
 - Data and metadata standards
2. Methodological development for both cost efficiency and data accuracy (eDNA, remote sensing, drones)
3. Scientific co-operation in data evaluation and analyses
4. Network development
 - Exchange about the site networks, coverage and options for expansion
 - National level collaborations to secure cost-efficiency, sustainability and network alignment
5. Service Portfolio
 - Identification of services of joint interest (widen range of stakeholders)
 - Collaborate as potential user in service specification

One possibility that was raised at the Frankfurt meeting came out of a gap identification exercise done on the eLTER network. The results are shown in the figure below, and indicate four key geographic areas that are currently underrepresented. The participation of sites in the ICP networks located in these areas would be of great value and we would encourage them to consider joining the eLTER network.

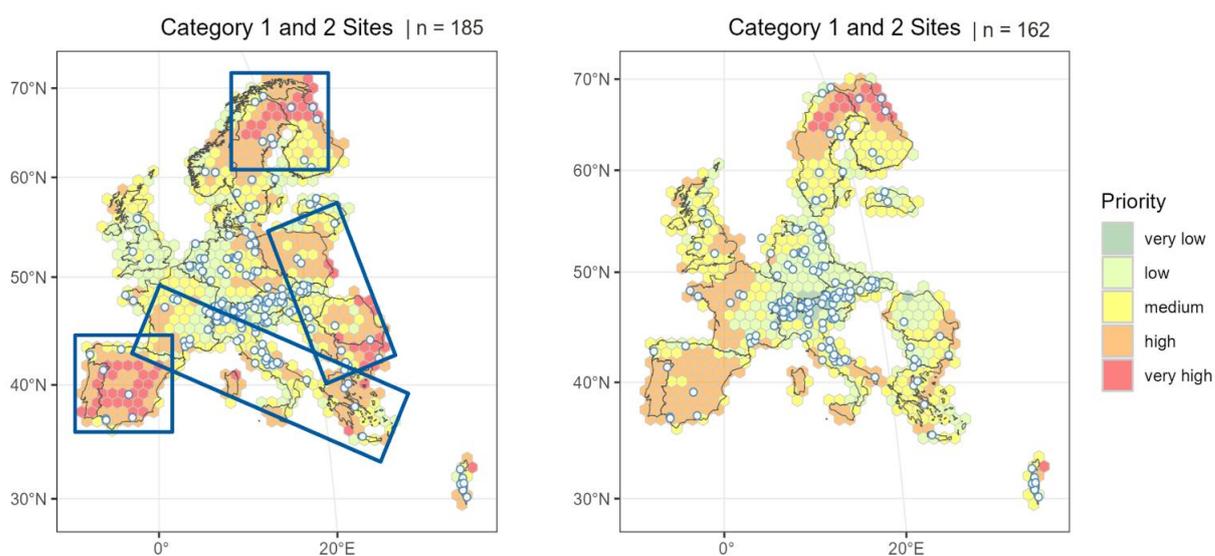


Figure 7 : Gaps identified in the eLTER RI network coverage (left panel all likely participating countries, right panel only those currently confirmed), boxes indicate the four broad areas identified as having high priority (a high proportion of orange and red coloured areas) for adding further sites.

Workplan for 2024-2025

The process for drawing up the next workplan is well underway. Suggestions from the Chairs, PC and others were discussed at the Task Force meeting in Lunz, and the following list was later submitted to the Secretariat for approval at the Geneva meeting in September 2023.

Suggested IM activities in the WGE 2024-25 Work Plan	Time frame
Scientific paper on the effects of N and S deposition on vegetation community stability over time.	2024
Scientific paper or report on trends in heavy metal fluxes across ICP Integrated Monitoring sites, and an assessment of the mercury data gathered by the newly installed passive samplers.	2024-25
Make the ICP IM database open access under a feasible licence and principles, and publish an associated data paper.	2024-25
Scientific paper or report - Update in long-term changes in the atmospheric deposition and runoff water chemistry of sulfate, inorganic nitrogen and acidity.	2024-25
Initiate a revision and update of the IM manual.	2024-25

One further item will be included in the work plan for this period, if the required external funding is obtained:

- Proof of concept for development of above ground vegetation monitoring in ICP IM sites using drone remote sensing.

National activity reports

The following reports of national activities were received - many thanks to all those who contributed, and we look forward to a wider coverage of the network in the next issue, especially from those countries who have not sent an update for a while.

Report on National ICP IM activities in the Czech Republic – CZ02 Lysina catchment

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List of publications (2022-2023) about the CZ02 Lysina for the ICP IM

Petrash D.A., Krám P., Pérez-Rivera K.X., Buzek F., Čuřík J., Veselovský F., Novák M. (2023) Soil solution data from Bohemian headwater catchments record atmospheric metal deposition and legacy pollution. *Environmental Science and Pollution Research*, doi.org/10.1007/s11356-023-25673-7.

Monteith D.T., Henrys P.A., Hruška J., de Wit H.A., Krám P., Moldan F., Posch M., Raike A., Stoddard J.L., Shilland E.M., Gloria Pereira M., Evans C.D. (2023) Long-term rise in riverine dissolved organic carbon concentration is predicted by electrolyte solubility theory. *Science Advances*, 9(3): eade3491, 1-11.

Report on National ICP IM activities in Ireland

Thomas Cummins

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Monitoring at Ireland's IM site Brackloon operated 1991–2017. New ecosystem monitoring under the EU National Emission Reduction Commitments Directive is underway by Ireland's Environmental Protection Agency, as a [National Ecosystems Monitoring Network](#). Initial developments will be on bulk precipitation collection and a network of passive and active samplers for ambient ammonia, use of fixed plots for monitoring of ground vegetation in open habitats (not yet forests), and development of further surveys across aquatic and terrestrial ecosystems in coming years, with a commitment to long-term monitoring now established at country level under the statutory requirements of NECD. Alignment with the IM-light initiative will allow data submission in future, and so the existing connection with ICP IM is intended to be maintained.

NFC Thomas Cummins

Report on National ICP IM activities in Norway

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Norway contributes data from three catchments (Level 3: catchment element budgets) to ICP IM: Birkenes, Langtjern and Kårvatn. Here, data on deposition chemistry (by NILU, Norwegian Institute for Air Research), hydrology, and stream water chemistry have been collected since the 1970s (by NIVA, Norwegian Institute for Water Research). Additionally, soil solution chemistry and throughfall is still monitored annually at Birkenes, in addition to regular surveys of vegetation (by NIBIO, Norwegian Institute for Bioeconomy).

At Birkenes, the Birkenes observatory is placed (<https://www.nilu.com/facility/nilus-observatories-and-monitoring-stations/birkenes-observatory/>), a key European site for long-term air quality monitoring. Langtjern is a highly instrumented site, with high-frequency monitoring taking place in inlet, lake and outlet and with an on-site weather station (www.aquamonitor.no/langtjern). The three sites are also included in ICP Waters, while Birkenes is included in ICP Forests.

Active research projects that use and collect data at Birkenes and Langtjern are the project 'CatchCan' (The fate and future of carbon in forests, 2020-2024; CZ RESEARCH Programme EEA and Norway Grants 2014-2021; NIVA is partner) and 'Biogov' (Biogeochemical processes governing boreal C cycling, 2022-2026, Norwegian Research Council; NIVA is partner).



Soil sampling at Langtjern (CatchCan project).
Photo: H. de Wit



Forest soil at Langtjern.
Photo: H. de Wit

Recent publications with data from Norwegian ICP IM stations

Peer-reviewed publications:

Clayer, F., Thrane, J. E., Brandt, U., Dorsch, P., & de Wit, H. A. (2021). Boreal Headwater Catchment as Hot Spot of Carbon Processing From Headwater to Fjord. *Journal of Geophysical Research-Biogeosciences*, 126(12). doi:10.1029/2021jg006359

de Wit, H. A., Stoddard, J. L., Monteith, D. T., Sample, J. E., Austnes, K., Couture, S., . . . Evans, C. D. (2021). Cleaner air reveals growing influence of climate on dissolved organic carbon trends in northern headwaters. *Environmental Research Letters*, 16(10). doi:10.1088/1748-9326/ac2526

Monteith, D. T., Henrys, P. A., Hruška, J., de Wit, H. A., Krám, P., Moldan, F., . . . Evans, C. D. (2023). Long-term rise in riverine dissolved organic carbon concentration is predicted by electrolyte solubility theory. *Science Advances*, 9(3), eade3491. doi:doi:10.1126/sciadv.ade3491

Liptzin, D., Boy, J., Campbell, J.L., Clarke, N., Laclauf, J.-P., Godoy, R., Johnson, S.L., Kaiser, K., Likens, G.E., Pihl Karlsson, G., Markewitz, D., Rogora, M., Sebestyen, S.D., Shanley, J.B., Vanguelova, E., Verstraeten, A., Wilcker, W., Worralls, F., McDowell, W.H. 2022. Spatial and Temporal Patterns in Atmospheric Deposition of Dissolved Organic Carbon. *Global Biogeochemical Cycles*, DOI: 10.1029/2022GB007393.

Yttri, K. E., Canonaco, F., Eckhardt, S., Evangeliou, N., Fiebig, M., Gundersen, H., Hjellbrekke, A.-G., Lund Myhre, C., Platt, S. M., Prévôt, A. S. H., Simpson, D., Solberg, S., Surratt, J., Tørseth, K., Uggerud, H., Vadset, M., Wan, X., and Aas, W.: Trends, composition, and sources of carbonaceous aerosol at the Birkenes Observatory, northern Europe, 2001–2018, *Atmos. Chem. Phys.*, 21, 7149–7170, <https://doi.org/10.5194/acp-21-7149-2021>, 2021.

Reports:

Austnes, K., Hjermann, D.O., Sample, J.E., Wright, R.F., Kaste, O., de Wit, H. 2022. Nitrogen in surface waters: time trends and geographical patterns explained by deposition levels and catchment characteristics. NIVA-rapport 7728-2022. ICP Waters report 149-2022.

Report on National ICP IM activities in Sweden

Lundin, L.¹, Löfgren, S.¹, Rönnback, P.¹, Kurén Weldon, J.¹, Bovin, K.², Evehorn, D.², Grandin, U.¹, Jutterström, S.³, Pihl Karlsson, G.³, Moldan, F.³ and Thunholm, B.².

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Introduction

The Swedish integrated monitoring programme is running 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 was influenced by long-term high deposition loads. The sites are well-defined catchments with mainly coniferous forest stands dominated by bilberry spruce forests on glacial till deposited above the highest coastline. Hence, there has been no water sorting of the soil material. Both climate and deposition gradients coincide with the distribution of the sites from south to north (Table 1).

Table 1 : Geographic location and long-term climate and hydrology at the Swedish IM sites (long-term average values, 1:1961–1990 and 2: 1991–2020)

	SE04	SE14	SE15	SE16
Site name	Gårdsjön F1	Aneboda	Kindla	Gammtratten
Latitude; Longitude	N 58° 03'; E 12° 01'	N 57° 05'; E 14° 32'	N 59° 45'; E 14° 54'	N 63° 51'; E 18° 06'
Altitude, m	114–140	210–240	312–415	410–545
Area, ha	3.7	18.9	20.4	45
Mean annual temperature (1), °C	+6.7	+5.8	+4.2	+1.2
Mean annual temperature (2), °C	+7.2	+6.7	+5.3	+2.4
Mean annual precipitation (1), mm	1000	750	900	750
Mean annual precipitation (2), mm	1080	730	900	660
Mean annual evapotranspiration, mm	480	470	450	370
Mean annual runoff, mm (1)	520	280	450	380

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

In the following, presented results mainly relate to 2021 and include climate, hydrology and water chemistry as well as some ongoing work at the four Swedish IM sites (Löfgren, 2023).

Climate and Hydrology in 2021

Based on long-term (1961-1990) mean values from the Swedish Meteorological and Hydrological Institute (SMHI), and measured data from climate monitoring at the IM sites, the 2021 annual mean temperatures were 0.1-1.1 °C higher for all four sites. Largest deviation occurred at the mid-southern site SE15 Kindla. A new climate period (1991-2020) now exists with higher values than 1961-1990 showing somewhat lower temperature deviations as noticed from the previous period (Table 1). Deviations from this period showed mainly lower annual temperature in 2021 with 0.2 – 1.0 °C with highest deviation at the northernmost site. Only SE04 had a higher value by 0.5 °C.

Compared with the on-site measured time series, 21 years at site SE16 Gammtratten and 25 years at the other sites, the temperatures in 2021 were lower at SE15 Kindla with 0.2 °C and SE16 Gammtratten with 0.5 °C. The other two sites were close to the long-term average. At SE04 Gårdsjön, the length of the vegetation period has increased by one month over the 25 years of monitoring. For 2021, the mean annual temperature was above zero degrees for all four sites but all sites had mean monthly temperatures below zero in Months Jan-Feb and Dec. Monthly average temperatures in June and July were higher compared to long-term means at all stations. SE15 Kindla and SE16 Gammtratten showed higher temperatures in beginning of the year (Jan-Mar) while SE04 Gårdsjön and SE14 Aneboda had slightly lower monthly mean the first five months (Jan-May). In the last months of the year Oct-Dec, SE16 Gammtratten showed higher temperatures compared with the long-term average while the three other sites showed only small deviations from the long-term average.

Annual precipitation amounts in 2021 were higher at the two sites SE04 Gårdsjön and SE14 Aneboda with about 7% compared to the average long-term average 1961-1990. SE16 Gammtratten had 114 mm, 16% lower precipitation and site SE15 Kindla was close to average. The previous years 2019 and 2020 showed similar patterns. This deviated somewhat from 2018 when all sites had lower amounts compared to the long-term mean.

The distribution between months in 2021 showed low values in spring months April and May, while both lower and higher amounts were recorded for three sites for the rest of the year. SE16 Gammtratten

had lower precipitation for all months except January and October. The month October showed higher amounts compared to average for all sites.

High groundwater levels during winter and lower levels in summer and early autumn characterize the annual hydrological patterns of the southern catchments. At the northern locations, the general picture is low groundwater levels in winter when precipitation is stored as snow, raising levels at spring snowmelt followed by lower levels in summer due to evapotranspiration and groundwater outflow. However, depending on rainfall events in summer and/or autumn, the groundwater levels could occasionally be elevated also during these periods. Common are elevated levels in autumn. In 2021, only the site SE16 Gammtratten started the year on comparably low levels (c. 2 m below ground surface) while all other sites had fairly high groundwater levels (0.2 m to c. 1 m). In SE16 Gammtratten, levels showed receding levels until mid-April (c. 2.8 m). Water storage in snow before snowmelt was 252 mm to be compared with 234 mm in 2020 and 210 mm in 2019. In end of May, the snowmelt peak occurred with the highest level during the year being 1.2 m below surface. Recession followed and the deepest level on 2.9 m was reached in mid-September. After this autumn rains elevated the level to 1.5 m before colder weather occurred and precipitation was stored as snow with lowered groundwater levels to 2.2 m in the end of the year.

At SE14 Aneboda, groundwater levels started high in January and turned lower until August-September when the deepest levels occurred. After this, autumn rains elevated the levels being comparably high in November and December.

At SE15 Kindla, the groundwater level mostly was close to ground surface being 0.2-0.5 m below the surface. Precipitation made the levels fluctuating all through the year providing rapid changes also in discharge. Deepest levels occurred in July on 0.7 m which was higher than the two previous years with levels below one metre. The year 2018 showed the deepest levels for the last five years with 1.5 m while the years 2016-2017 and 2019-2020 were more alike 2021.

The stream water discharge patterns (Fig. 8) reflect the groundwater levels.

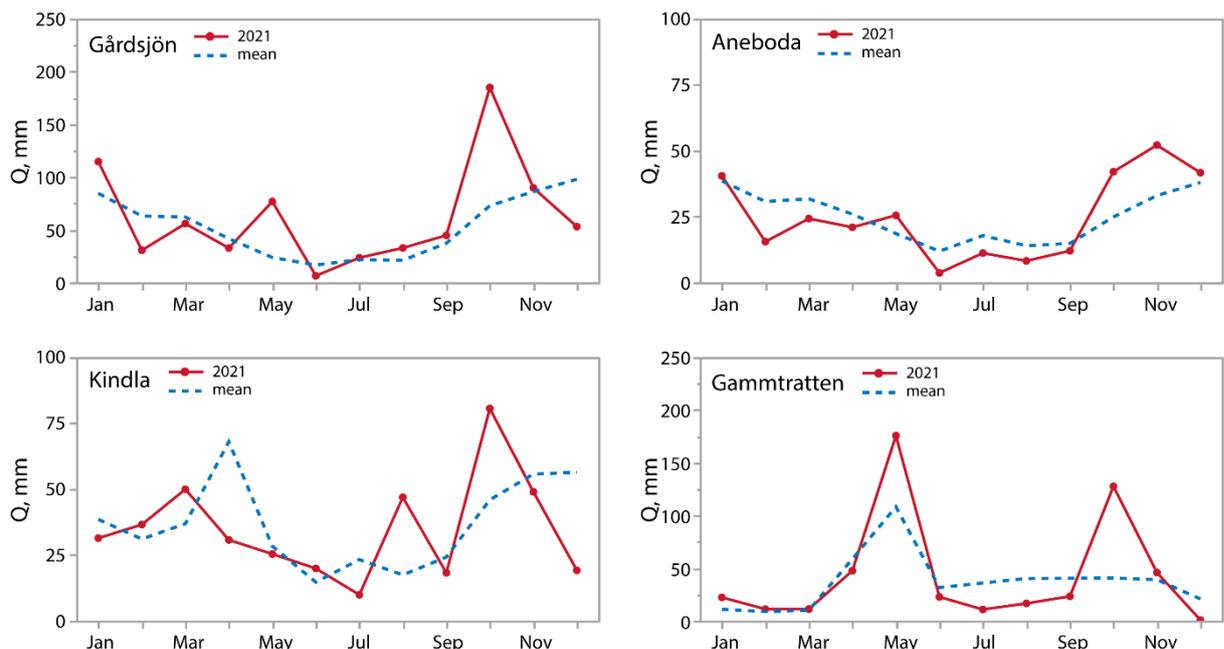


Figure 8 : Monthly discharges at the four Swedish IM sites in 2021 compared with the monthly averages for the period 1996–2020 (mean). Note the different scales at the Y-axis

Precipitation, evapotranspiration and groundwater levels affect the runoff patterns. At SE16 Gammtratten, this pattern was fairly typical with a snowmelt peak in April and May and lower discharges in summer and autumn but with, for later years, a peak in October-November not shown in the long-term mean. Later, low temperatures caused snowfall and snow accumulation with lower discharges. The snowmelt peak agreed with the long-term pattern deviating somewhat from 2019 when it occurred slightly earlier. At SE04 Gårdsjön, the pattern mainly agreed with the long-term mean apart from the high discharge peak in October. In 2020, there was high discharge in the period January-March which deviated from 2021. In the summer of 2020, the discharge was lower compared to the long-term mean but with a small peak in July. Autumn discharge resembled the long-term mean.

At SE15 Kindla, the snowmelt peak was early and occurred in March deviating from the long-term mean with the snowmelt peak in April. Summer runoff was mainly close to average but with higher runoff in August. This deviated from 2019 and 2020 when summer runoff was comparably low. Autumn rains caused high runoff in October and after that descending to runoff in December. This deviated from the high values in 2020 being usual at the end of the year. The runoff at SE14 Aneboda in 2021 was mainly lower compared to the long-term mean for February-August with slightly higher than average in May. In October-November there were higher runoff compared to the mean. Main difference to 2020 occurred in January-March with high runoff in 2020 not occurring in 2021 (Fig. 1).

Generally, snow accumulates during winter at the two northern IM sites, 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. However, only the most northern site SE16 Gammtratten had low flows throughout the 2021 first months. At SE15 Kindla, runoff was low in January, but the flow started to increase already in February which didn't occur at the other sites with fairly low runoff in beginning of the year (Fig. 1).

In 2021, the annual runoff made up 41–89% of the annual precipitation (Table 2), which is similar compared to the normal 40–60% apart from SE16 Gammtratten having a share of 89%. The high share at SE16 Gammtratten, partly reflects the colder climate in the North. However, the value perhaps is somewhat high caused by fairly low precipitation but high runoff. At SE04 Gårdsjön, it is quite normal that runoff constitutes almost 2/3 of the precipitation. The share was slightly lower in 2020 (59%), then due to the large precipitation. At SE14 Aneboda, storm felling, followed by bark beetle attacks, have reduced the forest canopy cover and thereby interception. For 2021, however, the total evapotranspiration was estimated to 438 mm, which was higher than 390 mm in 2020, which is within the range of previous years and partly agreeing with the ordinary ratio 50/50 E to R. However, runoff was low especially since influence from the low forest cover. Throughfall was very high even higher compared to precipitation which would be considered out of order and a negative interception is hardly reasonable. The low runoff would not reflect the high throughfall. The site SE15 Kindla showed almost the lowest runoff share with 43%, being in the low range to the 50/50 E to R. SE15 Kindla had the highest throughfall even though the stand canopy is slowly declining. However, 98% of precipitation would possibly be out of range. In 2016 and 2018, the annual runoff range was wider than usual and made up 31–83% of the annual precipitation at the four sites.

Table 2: Compilation of the 2021 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	1103	100	736	100	966	100	579	100
Throughfall, TF	925	84	838	114	948	98	566	98
Interception, P–TF	178	16	-102	-14	18	18	13	2
Runoff, R	748	68	298	41	418	43	518	89
P–R	355	32	438	59	548	57	61	11

At the northern site SE16 Gammtratten, throughfall and bulk precipitation used to be fairly similar also found in 2021, but in 2020 throughfall only reached 77%. The latter is probably a more reasonable estimate compared with earlier years and 2021. Presumably, snow deposition infers large uncertainties, which in earlier years could have caused erroneous estimates of especially bulk precipitation. In 2021, throughfall was high, interception and evapotranspiration being low. In 2020, however, both evapotranspiration and runoff gave reasonable amounts. In summary and based on the estimated evapotranspiration (P-R) 2021, low values could be concluded for SE04 Gårdsjön and SE16 Gammtratten. The years 2020 and 2019 showed more reasonable evapotranspiration. Even though, lowest values would be found at the northern SE16 Gammtratten site, the value 61 mm is too low (Table 2). In 2018, the very dry summer furnished low evapotranspiration at all four sites.

Water chemistry in 2021

Low ion concentrations in bulk deposition (electrolytical conductivity 1–2 mS m⁻¹) characterised all four Swedish IM sites. The concentrations of ions in throughfall that include dry deposition, were higher at the two southern sites. At the central and northern sites SE15 Kindla and SE16 Gammtratten, the conductivity in throughfall (0.8 mS m⁻¹) was almost the same as in bulk deposition indicating very low sea salt deposition and uptake of ions by the trees. At the two southern sites, sea salt deposition provides tangibly higher ion concentrations, especially at the west coast SE04 Gårdsjön site, 3 mS m⁻¹ in throughfall and c. 2 mS m⁻¹ in bulk deposition. 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 at three sites (0.0–0.6 units) in throughfall compared with bulk deposition. SE15 Kindla deviated from this with 0.1 pH-units higher in bulk deposition. In 2021, however, SE15 Kindla and SE16 Gammtratten had similar pH (5.1–5.3) in both bulk deposition and throughfall. The sites SE04 Gårdsjön and SE14 Aneboda had 0.4 and 0.6 pH units, respectively higher pH in throughfall compared with in bulk deposition (Table 3).

Table 3: Mean bulk deposition chemistry values 2021 at the four Swedish IM sites. S and N in kg ha⁻¹ yr⁻¹.

	SE04	SE14	SE15	SE16
pH, bulk deposition	5.1	5.0	5.4	5.1
pH, throughfall	5.5	5.6	5.3	5.1
S, bulk deposition	2.6	1.4	2.1	0.8
N, bulk deposition	5.5	5.0	5.7	1.8

During the soil solution passage through the catchment soils, organic acids were added and leached on its way to stream runoff. In the upslope recharge areas, pH in soil water in the upper soil layers (E-horizon) was mainly lower than in throughfall with ≤ 0.6 pH-units for the three southern sites. SE16 Gammtratten had the lowest pH-value in soil water with 4.1, being 1.0 units lower compared to throughfall. However, in the organic rich discharge areas at SE15 Kindla and SE16 Gammtratten, pH was higher in groundwater compared with throughfall while the opposite occurred at SE14 Aneboda and especially at SE04 Gårdsjön with 1.0 units lower pH in groundwater.

In the recharge areas, the buffering capacity (ANC) in soil water and groundwater varied between negative and positive values, but most frequently it was on the negative side, especially in the B-horizon at SE14 Aneboda with -0.07 mEq L⁻¹. This may be an effect of nitrification. In the discharge areas of this site, the buffering capacity in groundwater was considerable with 0.23 mEq L⁻¹. At SE15 Kindla, the groundwater ANC was 0.05 mEq L⁻¹, while SE04 Gårdsjön showed high ANC with 0.30 mEq L⁻¹ and SE16 Gammtratten showed lower values with 0.04 mEq L⁻¹. Bicarbonate (HCO₃⁻) occurred at SE15 Kindla, SE16 Gammtratten and SE14 Aneboda with 0.18 , 0.04 and 0.08 mEq L⁻¹, respectively, but possibly not at SE04 Gårdsjön. The latter is not measured but was indicated by the very low pH of 4.5 and high DOC concentration (22 mg L⁻¹).

The stream waters were acidic with pH values below 4.7 at all sites except SE16 Gammtratten having an annual average pH of 5.6. The stream water buffer capacity was positive at all sites (ANC ≥ 0.017 mEq L⁻¹). However, also SE15 Kindla had acidic streamwater with ANC close to zero. At SE16 Gammtratten anions of weak organic acids and bicarbonate alkalinity contributed to the positive ANC (0.1 mEq L⁻¹). Similar ANC was found at SE14 Aneboda dependent to comparably high DOC with 28 mg L⁻¹ and stream acidity had decreased. However, pH was still lower compared to SE16 Gammtratten. At SE15 Kindla and SE04 Gårdsjön, the stream waters were more acidic compared with the other two sites probably due to oxidation of organically bound sulphur related to natural sources and the legacy from earlier sulphur deposition. In SE04 Gårdsjön deposition of sea salt contributed to the acidic conditions as this provided ion exchange and release of protons and inorganic aluminium.

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. At SE14 Aneboda and SE15 Kindla concentrations of chloride and sulphate were on similar levels. In the recharge area of SE14 Aneboda,

nitrification have contributed to relatively high nitrate concentrations in the soil water. However, the nitrate concentrations turned lower in 2019 – 2021 compared to earlier.

At the northern site SE16 Gammtratten, the sulphate concentrations in soil water and stream water were considerably higher compared to throughfall, indicating release from the soil pool. Organic anions and HCO_3^- dominated the stream water anion flow with 2/3 of the anions to be compared with 1/3 at SE15 Kindla and SE14 Aneboda and 1/8 at SE04 Gårdsjön.

The cation and anion relations reflect different soil and soil water processes. In deposition, Na^+ dominated the base cations for all sites. This was valid also in stream water except for the northern site SE16 Gammtratten where Ca^{2+} showed similar values. At sites SE04 Gårdsjön and SE14 Aneboda, Cl^- concentrations were similar compared Na^+ while Na^+ dominated at the other two sites. A higher leakage of Na^+ compared to Cl^- in the streamwater indicate weathering of minerals and release of base cations from the soils. At SE04 Gårdsjön, Mg^{2+} was the second highest base cation in runoff water, also reflecting the marine influence, while Mg^{2+} and Ca^{2+} were quite equal at the other three sites. Hydrogen ions contributed considerably in the ion balance at SE15 Kindla. At SE04 Gårdsjön the contribution was moderate and for SE16 Gammtratten low.

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 29 mg L^{-1} while SE04 Gårdsjön showed 20 mg DOC L^{-1} . The other two sites had lower DOC concentrations with 10 and 12 mg L^{-1} at SE15 Kindla and SE16 Gammtratten, respectively. High DOC concentrations create prerequisites for metal complexation and transport as well as high fluxes of organic nitrogen. This was the dominating nitrogen fraction in all stream waters, ranging from 0.21 to $0.63 \text{ mg N}_{\text{org}} \text{ L}^{-1}$. Inorganic N concentrations were low ($\leq 0.061 \text{ mg N}_{\text{inorg}} \text{ L}^{-1}$) at the sites. The high inorganic contents at SE04 Gårdsjön and SE14 Aneboda were likely depended on deteriorated forest stands.

Total phosphorus deposition (P_{tot}) in bulk deposition varied between 0.04 and $0.25 \text{ kg ha}^{-1} \text{ year}^{-1}$ with the highest value at SE15 Kindla and lowest at SE16 Gammtratten. The stream water export rates reached 0.01 - $0.06 \text{ kg ha}^{-1} \text{ year}^{-1}$ with the lowest value at SE15 Kindla.

Inorganic aluminum (Al_i), toxic to fish and other gill-breathing organisms, is analyzed in soil solution, groundwater and surface waters at the IM sites. Relatively high total Al concentrations occurred in the soil solution (0.6 – 1.7 mg L^{-1}) at the three southern sites. The stream water Al_{tot} -concentrations were between 0.5 and 0.7 mg L^{-1} at the three sites with low pH (4.5–4.7). The Al_{tot} concentrations were lower, approximately 0.27 mg L^{-1} at the northern site SE16 Gammtratten with a pH of 5.6. Inorganic Al made up 17–54% of the total Al with the highest levels at low pH at SE15 Kindla and the lowest at SE16 Gammtratten, corresponding to 0.06 – $0.28 \text{ mg Al}_i \text{ L}^{-1}$. According to the SEPA classification system, the Al_i concentrations at SE14 Aneboda and SE15 Kindla are considered *extremely high*, but *moderate* at SE04 Gårdsjön and SE16 Gammtratten.

Concentrations of Fe and Mn were moderate with $\leq 1.5 \text{ mg L}^{-1}$ and $\leq 0.09 \text{ mg L}^{-1}$, respectively. The priority heavy metals Pb, Cd and Hg were still accumulating in the SE14 Aneboda catchment soils, while the stream concentrations were low compared with the levels causing biological effects. Only Pb had somewhat higher concentrations (1.1 mg L^{-1}) compared with the established limits for ecological effects. Methyl mercury (Hg_{Me}), only measured at SE14 Aneboda, was still relatively high creating prerequisites for bioaccumulation. In stream water, the mean Hg_{tot} and Hg_{Me} concentrations were 9.4 ng L^{-1} and 1.4 ng L^{-1} , respectively. About 20% of the Hg_{tot} deposition was accumulated in the catchment

soil. The four times higher runoff of Hg_{Me} in stream water $0.24 \mu\text{g m}^{-2}, \text{yr}^{-1}$ compared with in throughfall indicates ongoing methylation. SITES financed the heavy metal analyses.

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

References

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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 during the year since the previous annual report.

