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 2012/2013 including:

• A short summary of previous data assessments
• A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
• A final report on relations between vegetation changes and nitrogen Critical Load exceedance
• A progress report on baseline heavy metal approach, estimation of the extent of metal turnover in European forest catchments over the last decades
• A final report on sulphur and nitrogen input-output budgets at ICP IM sites in Europe
• National Reports on ICP IM activities are presented as annexes.
22nd Annual Report 2013

Convention on Long-range
Transboundary Air Pollution

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

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FINNISH ENVIRONMENT INSTITUTE
ABBREVIATIONS

AMAP  Arctic Monitoring and Assessment Programme
ANC  Acid neutralising capacity
ALTER-Net  A Long-Term Biodiversity, Ecosystem and Awareness Research Network
CCE  Coordination Center for Effects
CL  Critical Load
CNTER  Carbon-nitrogen interactions in forest ecosystems
ECEconomic  Commission for Europe
EMEP  Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
EMEP50  EMEP grid system with a resolution of 50 km by 50 km
EnvEurope  EU LIFE project “Environmental quality and pressures assessment across Europe; the LTER network as an integrated and shared system for ecosystem monitoring”
EU  European Union
EU LIFE  EU’s financial instrument supporting environmental and nature conservation projects throughout the EU
ExpeER  Experimentation in Ecosystem Research
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 Wers  International Cooperative Programme on Assessment and Monitoring of 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
LifeWatch  EU-infrastructure project, e-science and infrastructure for biodiversity data and observatories
LRTAP Convention  Convention on Long-range Transboundary Air Pollution
LTER-Europe  European Long-Term Ecosystem Research Network
LTER-Network  Long Term Ecological Research Network
MFR  Maximum feasible reductions scenario
NFP  National Focal Point
UNECE  United Nations Economic Commission for Europe
WGE  Working Group on Effects
Summary

Background and objectives of ICP IM

Integrated monitoring of ecosystems means physical, chemical and biological measurements over time of different ecosystem compartments simultaneously at the same location. In practice, monitoring is divided into a number of compartmental sub-programmes which are linked by the use of the same parameters (cross-media flux approach) and/or same or close stations (cause-effect approach).

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

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

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

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

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

Assessment of data collected in the ICP IM framework is carried out at both national and international levels. Key recent 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 biological data using multivariate gradient analysis
- 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
- Empirical thresholds for N deposition (soil C/N ratios, input-output budgets)
- Compilation of available information on cause-effect relationships of forest ecosystems
- Calculation of critical loads for sulphur and nitrogen compounds, and assessment of critical load exceedance, as well as links between critical load exceedance and empirical impact indicators.

Conclusions from international studies using ICP IM data

Input-output and proton budgets, C/N interactions

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

The 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). Soil water fluxes for budget calculations have been estimated using a water balance model and were presented in Starr 1999. More recent 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\(^{-1}\) a\(^{-1}\), 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\(^{-1}\) a\(^{-1}\). When stratifying data based on C/N ratios less than or equal to 25 and greater than 25, highly significant relationships were observed between N input and nitrate leached (Dise et al. 1998, MacDonald et al. 2002, Gundersen et al. 2006). Such statistical relationships from intensively studied sites can be efficiently used in conjugation with regional monitoring data (e.g. ICP Forests and ICP Waters data) in order to link process level data with regional-scale questions.

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 recent European plot and catchment studies.

The reduction in deposition of S and N compounds at the ICP IM sites, caused by 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. 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 and Kakebeeke 2004, Working Group on Effects 2004).

ICP IM has contributed to an assessment report on reactive nitrogen (N\(_r\)) of the WGE. This report has been 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, aiming to abate the emission of air pollutants contributing to acidification, eutrophication and ground-level ozone.

It should also be recognized 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 analysis**

Empirical evidence on the development of environmental effects is of central importance for the assessment of success of international emission reduction policy. First results from a trend analysis of monthly ICP IM data on bulk and throughfall deposition as well as runoff water chemistry were presented in Vuorenmaa (1997). ICP IM data on water chemistry were also used for a trend analysis carried out by the ICP Waters and results were presented in the Nine Year Report of that programme (Lükewille et al. 1997).

Calculations on the trends of N and S compounds, base cations and hydrogen ions were made for 22 ICP IM sites with available data across Europe (Forsius et al. 2001). The site-specific trends were calculated for deposition and runoff water fluxes using monthly data and non-parametric methods. Statistically significant downward trends of SO\(_4^{2-}\), NO\(_3^-\) and NH\(_4^+\) bulk deposition (fluxes or concentrations) were observed at 50% of the ICP IM sites. Sites with higher N deposition and lower C/N-ratios clearly showed higher N output fluxes, and the results were consistent with previous observations from European forested ecosystems. Decreasing SO\(_4^{2-}\) and base cation trends
in runoff waters were commonly observed at the ICP IM sites. At some sites in the Nordic countries decreasing NO$_3$ and H$^+$ trends (increasing pH) were also observed. The results partly confirm 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 et al. 2006). Statistically significant decreases in SO$_4$ concentrations were observed at a majority of sites in both deposition and runoff/soil water quality. Increases in ANC (acid neutralising capacity) were also commonly observed. For NO$_3$, the situation was more complex, with fewer decreasing trends in deposition and even some increasing trends in runoff/soil water.

Results from several ICPs and EMEP were used in an assessment report on acidifying pollutants, arctic haze and acidification in the arctic region prepared for the Arctic Monitoring and Assessment Programme (AMAP, Forsius and Nyman 2006, www.amap.no). Sulphate concentrations in air generally showed 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.

Vuorenmaa et al. (2009) made a more recent trend evaluation using ICP IM data. These new results of the ICP IM sites confirmed the previously observed regional-scale decreasing trends of S in deposition and runoff/soil water. Acid-sensitive ICP IM sites in northern Europe also indicated recovery from acidification. The situation regarding N was quite different with few decreasing trends in deposition and both decreasing and increasing trends in runoff/soil water. Critical load calculations for Europe also indicate exceedances of the N critical loads over large areas. It was concluded that the N problem thus clearly requires continued attention as a European air pollution issue.

A new assessment on changes in the retention of S and N compounds at the ICP IM sites was prepared for 21st Annual Report (Vuorenmaa et al. 2012). Updated and revised data were included in the continuation of the work presented in this report. In addition, 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).
**Assessment of biological data using multivariate gradient analysis**

The effect of pollutant deposition on natural vegetation, including both trees and understorey vegetation, is one of the central concerns in the impact assessment and prediction. The 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 multivariate statistical gradient analysis of possible causes underlying the aspect of forest damage at ICP IM sites. These results suggested that coniferous defoliation, discoloration 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.

From the previous ordination exercises it was concluded that the applied statistical techniques are capable of revealing underlying structure and possible cause-effect relationships in complex ecological data, provided that analysed gradients have an adequate range to be interpolated. Since the data obtained were unexpectedly poor in the span of environmental gradients, the results of the presented statistical ordination only indicated correlative cause-effect relationships with a limited validity. The poor span of gradients could be attributed to the relative scarcity of biological effect data and the occurrence of missing observations both in the chemical and biological data sets. It was concluded, that the power of the vegetation monitoring in impact assessment would increase considerably with improvements in the ICP IM data reporting and inclusion of additional sites.

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.

Concepts for biodiversity monitoring and research have been developed in the ALTER-Net project (www.alter-net.info).

All available information on biodiversity changes and indicators at ICP IM sites were collected and the progress and preliminary results were published in the 20th and 21st Annual Reports (Bergander et al. 2011, Dirnböck et al. 2012). The final report on relations between vegetation changes and CL-exceedances was prepared for the present Annual Report (Dirnböck et al. 2013a). It is based on the key finding of the scientific manuscript (submitted) ‘Forest floor vegetation response to nitrogen deposition in Europe’ (Dirnböck et al. 2013b) which indicates that oligotrophic plant species are on the downgrade in European forest ecosystems that are sensitive to eutrophication effects.
Dynamic modelling and assessment of the effects of emission/deposition scenarios

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

Dynamic models have 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, Futter et al. 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 reductions 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.

A summary on the use of dynamic modelling forecasts to derive target loads for sulphur and nitrogen in atmospheric deposition, including climate change impacts, was included in the 16th Annual Report (Hutchins 2007).

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 concentrations, stores and fluxes of heavy metals at ICP IM sites is led by Sweden. Preliminary results on concentrations, fluxes and catchment retention were reported to the Working Group on Effects (document EB.AIR/WG.1/2001/10). Considerable retention of Cd, Cu, Ni, Pb and Zn (80−95 % of total input) was observed at some sites with available detailed information. The main findings on heavy metals budgets and critical loads at ICP IM sites presented in the 15th, 16th and 20th Annual Reports (Bringmark et al. 2006, Bringmark and Lundin 2007, Bringmark 2011). Input/output budgets and catchment retention for Cd, Pb and Hg in the years 1997−2011 were recently determined for 14 ICP IM catchments across Europe (Bringmark et al. 2013). Litterfall plus throughfall was taken as a measure of the total deposition of Pb and Hg (wet + dry) on the basis of evidence suggesting that, for these metals, internal circulation is negligible. The same is not true for Cd. Excluding a few sites with high discharge, between 74 and 94 % of the input Pb was retained within the catchments; significant Cd retention was also observed. High losses of Pb (>1.4 mg m⁻² year⁻¹) and Cd (>0.15 mg m⁻² year⁻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 year⁻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 many national studies on ICP IM sites, detailed site-specific budget calculations of heavy metals (including mercury) 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.

Compilation of available information on cause-effect relationships of forest ecosystems

A report summarising available information from the ICP Forests and ICP IM programmes on cause-effect relationships of forest ecosystems was prepared by de Vries et al. 2002. The results were also officially reported to the Working Group on Effects in 2002 (EB.AIR/WG.1/2002/15).

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

Empirical impact indicators of acidification and eutrophication were determined from stream water chemistry and runoff observations at ICP IM catchments. The indicators were compared with exceedances of critical loads of acidification and eutrophication obtained with deposition estimates for the year 2000. Critical loads for acidification were calculated for the stream water with the Steady-State Water Chemistry model feeding into the First-order Acidity Balance model. Critical loads for eutrophication were established as vegetation-specific empirical values for sites with only vegetation plots without runoff observations. Empirical impact indicators agreed well with the calculated exceedances. Annual mean fluxes and concentrations of acid neutralizing 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. With deposition estimates for 2020, although only one more catchment (7 cf. 6 of 18) would be protected from acidification, the average exceedance would decrease from 1 000 eq ha⁻¹ yr⁻¹ in 2000 to 300 eq ha⁻¹ yr⁻¹ in 2020. For the vegetation plots, the protection from eutrophication would rise from 15 (of 83) protected in 2000 to 54 protected in 2020. Progress reports of these
activities were presented in Holmberg et al. (2009) and Vuorenmaa and Holmberg (2010). A scientific paper on the key findings from these studies was published in 2013 (Holmberg et al. 2013), concluding that data from the ICP IM provide evidence of a connection between modelled critical loads and empirical monitoring results for acidification parameters and nutrient nitrogen.

**Planned activities**

- Maintenance and development of a central ICP IM database at the Programme Centre.
- 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 Ecological Research network, www.lter-europe.net), and the related EU-infrastructure project LifeWatch (www.lifewatch.eu).
- Cooperation with other external organisations and programmes, particularly the International Long Term Ecological Research network (ILTER, www.ilter-net.edu).
- Participation in projects with a global change perspective.
References


ICP IM activities, monitoring sites and available data

1.1 Review of the ICP IM activities in 2012–2013

Meetings

- The Chairman Lars Lundin represented the ICP IM programme at the 28th ICP Forests Task Force meeting in Bialowieża, Poland, 30 May–1 June 2012.
- Lars Lundin attended the EU FP7 project Soil Transformations in European Catchments (SoilTrEC) meeting and 4th International Congress of the European Soil Science Societies (Eurosoil) 2012 in Bari, Italy, 2–6 July 2012.
- The Programme Manager Martin Forsius and Maria Holmberg participated in the BIOGEMON conference in Maine, the United States, 15–20 July 2012.
- Martin Forsius and Maria Holmberg took part in the Second Nordic International Conference on Climate Change in Helsinki, Finland, 29–31 August 2012.
- Lars Lundin and Martin Forsius represented ICP IM in the Working Group on Effects (WGE) 31st meeting in Geneva, Switzerland, 20–21 September 2012 and Lars Lundin also in the WGE (Extended) Bureau meeting on 23–24 September.
- Martin Forsius attended a coordination meeting for ICP funding on 2 October 2012 in Berlin, Germany.
- Lars Lundin took part in the EU FP7 project SoilTrEC meeting in Chania, Greece, 3–5 October 2012.
- Martin Forsius and Jussi Vuorenmaa represented the ICP IM programme at the ICP Waters 28th Task Force meeting in Pallanza, Italy, 8–10 October 2012.
- Martin Forsius participated in the LifeWatch Nordic meeting in Uppsala, Sweden, 8–9 November 2012.
- Martin Forsius participated in the 10th LTER Europe Conference and Joint EnvEurope and ExpeER (Experimentation in Ecosystem Research) Meeting in Sofia, Bulgaria, 4–7 December 2012.
- Lars Lundin represented ICP IM in the 26th ICP Vegetation Task Force meeting in Halmstad, Sweden, 28–31 January 2013.
- Martin Forsius took part in the second annual ExpeER meeting in Florenze, Italy, 4–7 February 2013.
• Lars Lundin represented ICP IM in the WGE (Extended) Bureau meeting and Workshop with EMEP. Workshop was held on 20 February and the Bureau meeting on 21–22 February 2013.
• Lars Lundin represented the ICP IM programme at the 23rd CCE workshop and the 29th Task Force Meeting of the ICP M&M in Copenhagen, Denmark, 8–11 April 2013.
• Maria Holmberg also took part in the 23rd CCE workshop and 29th Task Force Meeting of the ICP M&M in Copenhagen, Denmark, 8–11 April 2013.
• Lars Lundin participated in the ALTER-Net and EC Conference “Science underpinning the EU 2020 Biodiversity Strategy” in Ghent, Belgium, 15–18 April 2013.
• The twenty-first meeting of the Programme Task Force on ICP Integrated Monitoring was held in Obninsk, Russia 22 May 2013. A one-day workshop on the assessment of ICP IM data was held prior to the Task Force meeting on 21 May.

Projects, data issues

After December 1st 2012 the National Focal Points (NFPs) reported their 2011 results to the IM Programme Centre. The Programme Centre carried out standard check up of the results and incorporated them into the IM database.

Scientific work in priority topics

• The Programme Centre prepared the ICP IM contribution to the Joint Report 2012 of the ICPs, TF health and Joint Expert group on Dynamic Modelling for the WGE.
• Progress report on base line heavy metal approach is included in the present Annual Report (Bringmark et al.). This report summarises the main findings of the scientific article “Trace metal budgets for forested catchments in Europe – Pb, Cd, Hg, Cu and Zn” (Bringmark et al. 2013).
• Report on sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe 1990-2010 is presented in the present Annual Report (Vuorenmaa et al.). A scientific paper on mass balances for sulphur and nitrogen at ICP IM sites is planned for 2014.
• Final report on relations between vegetation changes and CL-exceedance is included as a chapter in the present Annual Report 2013 (Dirnböck and Grandin). A scientific article on this topic has been submitted in April 2013.
• ICP IM participates in a joint coordinated exercise on dynamic modelling together with other ICPs (Joint Expert Group on Dynamic Modelling, JEG DM). Priority in the ICP IM work is given to site-specific modelling activities and development/testing of new methodologies for assessing the connections between air pollution and climate change.
• The ICP IM Programme Centre has contributed to the preparation of a special issue on “Ecosystem services: climate change and policy impacts” (Fu et al. 2013), and a review article “Using long-term ecosystem service and biodiversity data to study the impacts and adaptation options in response to climate change: insights from the global ILTER sites network” (Vihervaara et al. 2013), Ecosystem services assessment is one of the new priorities of the WGE.
1.2 Activities and tasks planned for 2013–2014

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

According to the workplan of the Working Group on Effects, ICP IM will produce the following reports:

2013: Report and scientific paper on relations between biodiversity and CL-exceedances
2013: Progress report and scientific paper on base line heavy metal approach
2013: ICP IM contribution to the WGE report on ecosystem services
2014: Report and scientific paper on mass balances for sulphur and nitrogen
2014: Report on dynamic modelling on vegetation changes in relation to N deposition
2015: Report and scientific paper on long-term trends in ecosystem effects

Other activities

• Maintenance and development of central ICP IM database at the Programme Centre
• Arrangement of the 22nd Task Force meeting (2014)
• Preparation of the 23rd ICP IM Annual Report (2014)
• Preparation of the ICP IM contribution to assessment reports of the WGE
• Participation in meetings of the WGE, other ICPs and the JEG DM

Activities/tasks aimed at further development of the programme

• Participation in the development of the European LTER-network (Long Term Ecological Research network, www.lter-europe.net), and the related EU-infrastructure project LifeWatch (www.lifewatch.eu)
• Participation in the activities of other external organisations, particularly the International Long Term Ecological Research Network (ILTER, www.ilternet.edu)

1.3 Published reports and articles 2012–2013

Evaluations of international ICP IM data and related publications


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


1.4 Monitoring sites and data

The following fifteen countries have continued data submission to the ICP IM data base during the period 2008 - 2012: Austria, Belarus, Canada, Czech Republic, Estonia, Finland, Germany, Italy, Latvia, Lithuania, Norway, Russian Federation, Spain, Sweden, and United Kingdom. Ireland has re-established the site IE01 in 2012 and will provide data in 2013. Ukrainian site UA01 was also re-established in 2013.

Presently the number of ICP IM sites with on-going data submission, data for at least part of the period 2007-2011, is forty-four. Most of the sites are European. 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 Figure 1.1. These sites have either been suspended or taken out of the IM network and used for regional monitoring. Locations of the ICP IM monitoring sites with data from recent years are shown in Figure 1.1.
| AREA | SUBPROGRAMME | AC | MC | PC | SF | TF | SC | SW | GW | RW | LC | LC | FC | PC | PF | RB | LB | LD | BB | BV |
|------|--------------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
|      | vegetation inventory |   |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | bird inventory |   |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | microbial decomposition |     |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | aerial green algae |         |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | trunk epiphytes |             |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | vegetation structure |               |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | bioelements |                 |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | vegetation |                       |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | forest damage |                        |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | hydrobiology of lakes |                      |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | hydrobiology of streams |                    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | litterfall |                        |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | foliage chemistry |                      |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | lake water chemistry |                    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | run-off water chemistry |                |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | groundwater chemistry |                   |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | soil water chemistry |                   |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | soil chemistry |                      |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | stemflow |                          |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | throughfall |                        |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | mass chemistry |                      |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | precipitation chemistry |                  |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | air chemistry |                        |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
|      | meteorology |                      |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |

Table 1.1 Internationally reported data from ICP-IM sites (subprogramme possible to carry out, * or forest health parameters in former Forest stand Trees).
Figure 1.1 Geographical locations of ICP IM sites with data from recent years.
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2 Final report on relations between vegetation changes and CL-exceedance

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2.1 Extended summary

Chronic nitrogen deposition poses a threat to biodiversity as a result of eutrophication of sensitive ecosystems. We studied long-term monitoring data from 28 forest sites from the ICP Integrated Monitoring and ICP Forests Programme to analyse temporal trends in species cover and diversity (Figure 2.1). We found that the cover of plant species that prefer nutrient-poor soils (oligotrophic species) significantly decreased at sites where the measured N deposition exceeded the empirical critical load for eutrophication effects (Figure 2.2). Forest plant species that prefer nutrient-rich soils (eutrophic species) showed an opposite trend but only with a marginal significance, p = 0.1. Contrary to changes in species cover, species diversity did not correlate with nitrogen critical load exceedance. We conclude that while the cover of oligotrophic plant species has decreased in European forest ecosystems, diversity is still not affected by airborne N deposition.

With regard to indicators we prove that existing critical loads are very useful to describe the sensitivity of forest floor vegetation to N deposition. We also show that the use of critical load exceedances is particularly suitable for revealing the eutrophication signal of N deposition. It is superior to N deposition alone, which is ignoring the differences in sensitivity among ecosystems. Further details of the study will soon be published in a peer review journal.
Figure 2.1 Location of study sites in Europe.
Figure 2.2 Forest plant species that prefer low soil nutrient levels have decreased during the last 10–50 years in 28 sites across Europe owing to the exceedance of the nitrogen Critical Loads. The Y-axis indicates the strength of the cover change of all oligotrophic species in a study site (negative values indicate a decrease, positive values an increase). The Critical Load exceedances are shown as the difference between the N deposition and the empirical Critical Load (negative values indicate no exceedance, positive values an exceedance) (Dirnböck et al., submitted to Global Change Biology).
3 Progress report on base line heavy metal approach

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3.1 Introduction

Metals and especially heavy metals are part of the air pollution. Deposition has been ongoing for a long-time period and metals are stored in the soil, primarily in the organic top layer but metals have also partly been translocated to deeper soil layers, though still accumulated in the terrestrial ecosystem (Ukonmaanaho et al. 2001, Eriksson 2002, Kobler et al. 2010). High concentration of heavy metals may deteriorate the soil microbiological activities in soils with hampered decomposition as one effect (Rademacher 2001). Further effects concern the discharge to surface water where heavy metals exert influences on the aquatic biota (Johanssson et al. 2001).

The base line understanding regarding heavy metal turnover for forest ecosystem based on catchment budgets shows ongoing accumulation and there is seldom net release to receiving systems. The approach addressed here is to estimate the recent and current extent of metal turnover in European forest catchments over the last decades. The conditions refer to calculations for 15 integrated monitoring sites in eight European countries (Bringmark et al. 2013).

Such evaluations would preferably be based on the catchment concept that provides well-defined boundaries of the systems considering inputs and outputs. Such investigations are comprehensive, but the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) for the ECE region made it possible to perform Europe-wide comparisons of sites in various climates and exposure to pollution loads. The program is one of six environmental monitoring
and modelling programs initiated to support the work of the UNECE Convention on Long-Range Transboundary Air Pollution, CLRTAP (Sliggers and Kakebeeke 2004).

The investigated current situation reveals still ongoing accumulation of heavy metals in most of the forest land and it continues despite considerable decrease in emission and deposition.

3.2 Methods

The study reflects conditions in 15 ICP IM sites distributed over north-south and coastal-continental gradients in Europe (Figure 3.1). The monitored sites are generally located in protected areas with unmanaged forests that have been allowed to develop naturally. The site altitudes range from near sea level to mountain locations of up to 1292 meters above sea level. The annual precipitation ranged from 590 to 1650 mm, being higher in coastal and high altitude locations. Runoff water amounts correlate to precipitation and the size of discharging water correlate to element flow with high discharge and high flow.

Figure 3.1 Sites of the ICP Integrated Monitoring (ICP IM) network included for the assessment of trace metal budgets.

Determinations of heavy metal variables include bulk deposition (BD), throughfall (TF), litter fall (LF) and runoff (RW) and have in different extent been carried out at the sites but available data in the ICP IM database have been compiled for the assessments. Measurements have followed the ICP IM manual with analyses mainly by AAS and ICP-OES or ICP-MS methods (DIN 38406-6, ISO 5961, ISO 11885) in laboratories adherent to quality control using certified reference waters and materials included in sample series as routine and with participation in laboratory inter-comparisons.
3.3 Results

The variation in precipitation and hydrology would reflect the deposition, runoff and turnover of the heavy metal elements, and therefore the sites were grouped in three groups based on the precipitation and discharge values. This resulted in one high water turnover group, one average and one with low water flows (Table 3.1).

Table 3.1 Regionalisation of the sites based on precipitation (P) and discharge (R). In the table mean values of P and R are presented. Site No. refers to country code and site number (Figure 3.1). Average catchment size and altitude range are also included.

<table>
<thead>
<tr>
<th>Regionalisation</th>
<th>P, R level</th>
<th>Site No.</th>
<th>Area (ha)</th>
<th>Altitude (m)</th>
<th>P (mm)</th>
<th>R (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low P, R</td>
<td>FI01, LV01, LV02, LT01, LT03, CZ01</td>
<td>167</td>
<td>189 - 218</td>
<td>718</td>
<td>145</td>
<td></td>
</tr>
<tr>
<td>Mean P, R</td>
<td>FI03, SE04, SE14, SE15, SE16</td>
<td>111</td>
<td>242 - 311</td>
<td>864</td>
<td>429</td>
<td></td>
</tr>
<tr>
<td>High P, R</td>
<td>CZ02, DE01, AT01, GB01</td>
<td>296</td>
<td>598 - 1075</td>
<td>1348</td>
<td>897</td>
<td></td>
</tr>
</tbody>
</table>

Estimations of atmospheric input would be of crucial importance for calculations of turnover and element budgets. The use of only bulk deposition samplers was considered to underestimate the total deposition (TD) and instead both throughfall (TF) and litterfall (LF) should be considered. Budgets for metals involve important processes and could be illustrated in a flux balance model (Figure 3.2). For Pb and Hg the addition of TF+LF would be the most appropriate estimation while for Cd, Cu and Zn there is known vegetation uptake meaning an internal circulation and the sums of TF and LF somewhat overestimate the total input, but is probably the best approximation of input. The resulting estimates for the deposition of Pb, Cd and Hg seems to exceed those obtained by calculations of EMEP, the UN program for monitoring and evaluation of the long range transmission of air pollutants in Europe (Bringmark et al 2013).

Figure 3.2 Processes involving metals in forest catchment. Uptake from the soil to the vegetation via the roots is not shown, and is not significant for Pb and Hg.
An update of earlier studies was recently presented (Bringmark et al. 2013) and results are reflected here. After this update of metal flux data, this study covers the entire period 1997−2011 and the general picture of metal retention in catchments reported before (Bringmark 2011) still remains. Disregarding sites with exceptionally high outflow, the average outflow of Pb from catchments with coniferous forest was 16% of that in throughfall, for Cd the figure was 26%. As noted in the previous report, this means that metal accumulation is still going on in catchments in spite of considerably lowered deposition. According to moss surveys in Sweden and a few long term deposition measurements in Denmark, Pb deposition has been reduced by about 85% since 1970, Cd and Hg by 75% and 70%, respectively (Rühling and Tyler 2001). Also in later moss surveys decreased contents have been observed (Harmens et al. 2008).

The geographical patterns of relative Pb release from catchments were analysed by comparing annual RW/TF ratios in the three groups of ICP IM sites in which this variable was available, i.e. group A; DE01, CZ02, group B; SE14, SE15, SE16 and AT01, and group C; CZ01, LV01 and LV02. Based on logarithmically transformed data, the differences between groups were evident (ANOVA and Tukey-Kramer test, p < 0.001). The mean RW/TF values were 1.0, 0.26 and 0.06 for the groups A, B and C, respectively (Bringmark et al. 2013).

The retention of Cd was tested on logarithmically transformed RW/TF data for the same groups of ICP IM sites as for Pb. The Tukey-Kramer test showed that group B and C did not differ (p=0.35), while group A differed significantly from groups B and C (p < 0.001). Mean RW/TF values were 2.3, 0.15 and 0.08 in respective groups.

The Cu exports from sites varied significantly, ranging from 0.04 mg m⁻² yr⁻¹ to 0.95 mg m⁻² yr⁻¹; the British moorland site GB01 exhibited a particularly large Cu outflow on 0.95 mg m⁻² yr⁻¹. Strong retention on the catchment scale was commonplace, reaching 80−97% of TF. Large quantities of Zn were transported in the runoff at CZ02 and GB01; at the other sites, the amount of Zn exported was between 0.3 mg m⁻² yr⁻¹ and 3.5 mg m⁻² yr⁻¹. Zn was heavily retained in the catchments to a degree of 38–96% of deposited Zn in TF.

Hg levels were monitored at Swedish sites during special one year campaigns that moved from site to site in repetition. Additionally, in some years, Hg levels in the litter fall and stream water were measured. The Hg flows in TF and LF followed this pattern (Figure 3.3). The four sites had average annual Hg deposition by TF of 17, 14, 12 and 4 µg m⁻² yr⁻¹, respectively, corresponding values by LF at 39, 23, 12 and 8 µg m⁻² yr⁻¹.

There were north-south gradients for deposition and litter fall but no such gradient for runoff amounts, which were remarkably similar from site to site with annual values on average 1.7–2.7 µg m⁻² yr⁻¹. Measurements of Hg in RW were performed from 2 to 14 years at the sites. Correlation analysis of annual values showed that Hg in RW had no significant relation to the Hg deposition using Hg in LF as proxy (p = 0.52, n = 23). The degree of retention in the catchments was 86–99% of the amount deposited by TF+LF.

3.4 Conclusions

Summarizing results would conclude the budget studies showing on-going accumulation of heavy metals in the organic soil top layer, i.e. the forest floor. A mobility of Cd and Pb would relocate these metals within the soil profile, thus at lowered pollution loads reducing the strain on biota that fulfill important ecosystem functions in the upper soil layers. A lowering of Pb and Cd contents in the organic top soils of two Swedish IM sites in the period from 1995/1996 to 2003/2005 was earlier reported (Bringmark and Lundin 2007).
For Cd, the reduction in soil storage was only evident in the upper F-layer and not in the humus layer as a whole and Pb as well was most sharply reduced in the uppermost F-layer. Earlier determinations of metal contents in the humus layer of the same ICP IM sites show that Cd was substantially reduced already in an earlier decade (Table 3.2). However, the most recent determinations now available tell that decrease of Pb and Cd has stopped. A striking observation is that reduction of Hg levels in humus layers did not occur during the entire period, this metal being tightly bound to organic matter.

Table 3.2 Metal contents in the humus layer of plots in the Swedish ICP IM sites Gårdsjön (SE04) and Aneboda (SE14) for the period 1983−2011. Site Svartedalen is located close to site Gårdsjön and was monitored in 1983 and 1988.

<table>
<thead>
<tr>
<th>Year/Site</th>
<th>Pb µg g⁻¹</th>
<th>Cd µg g⁻¹</th>
<th>Hg µg g⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Gårdsjön/Svartedalen</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1983, Svartedalen</td>
<td>83</td>
<td>0.94</td>
<td>0.41</td>
</tr>
<tr>
<td>1988, Svartedalen</td>
<td>113</td>
<td>0.74</td>
<td>0.29</td>
</tr>
<tr>
<td>1995, Gårdsjön</td>
<td>102</td>
<td>0.35</td>
<td>0.36</td>
</tr>
<tr>
<td>2003, Gårdsjön</td>
<td>87</td>
<td>0.35</td>
<td>0.41</td>
</tr>
<tr>
<td>2010, Gårdsjön</td>
<td>75</td>
<td>0.38</td>
<td>0.40</td>
</tr>
<tr>
<td><strong>Aneboda</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1983</td>
<td>98</td>
<td>0.81</td>
<td>0.31</td>
</tr>
<tr>
<td>1988</td>
<td>110</td>
<td>0.69</td>
<td>0.34</td>
</tr>
<tr>
<td>1996</td>
<td>81</td>
<td>0.45</td>
<td>0.27</td>
</tr>
<tr>
<td>2005</td>
<td>42</td>
<td>0.42</td>
<td>0.33</td>
</tr>
<tr>
<td>2011</td>
<td>62</td>
<td>0.42</td>
<td>0.36</td>
</tr>
</tbody>
</table>

Figure 3.3 Mercury balances for the four Swedish catchments SE04, SE14, SE15 and SE16 showing throughfall (TF), litterfall (LF), throughfall+ litterfall (TF+LF) and runoff (RW).
References


Rademacher, P. 2001. Atmospheric heavy metals in forest ecosystems. Federal Research Centre for Forestry and Forest Products.


4 Sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe

Final report

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4.1 Introduction

Due to the implementation of successful emission reduction measures, the emissions of sulphur and nitrogen in Europe have substantially decreased during the past 30 years. The protocols of the Convention on Long-range Transboundary Air Pollution (UNECE LRTAP Convention) and legislation of the European Union have been key international instruments causing this positive development. The emission control programmes have been most successful for SO₂. Following a slight decrease during the 1980s, the emission reduction agreements resulted in accelerated decrease of SO₂ emissions during the 1990s. Similar to sulphur, total European emissions of N compounds exhibited a gradual decrease during the 1990s, but reductions were smaller. In Europe, overall emissions of sulphur dioxide (SO₂) have declined by 73 % between the years 1990 and 2009 and those of nitrogen (N) compounds by 36 % (NOₓ) and 31 % (NH₃) (Fagerli et al. 2011). Following the drastic decrease in S emissions and deposition, a widespread recovery from acidification of sensitive ecosystems has taken place in Europe, and has been extensively evaluated and documented (e.g. Vuorenmaa et al. 2009, Skjelkvåle and de Wit 2011).
In order to assess the ecosystem benefits of costly emission reduction policies, the importance of long-term integrated environmental monitoring approach including physical, chemical and biological variables is clearly indicated. Mass balance budgets integrate information about the complex chemical and biological recovery processes that govern the retention or release of sulphur and nitrogen compounds and regulate acid production and buffering in both the terrestrial and aquatic portions of catchments in the ecosystem. Long-term assessment of mass balances in hydrologically and geologically well-defined ICP IM catchments gives important information for the identification of ecological effects of different anthropogenically derived pollutants, and for documenting the effects of emission reduction measures. An earlier report (Vuorenmaa et al. 2012) presented first estimates of annual mass balance budgets for sulphur and nitrogen at Integrated Monitoring sites for the period 1990–2010. The same assessment is described in the present report, but with updated and revised data. In addition, the role of organic nitrogen in nitrogen fluxes is derived and trends of S and N in fluxes are analysed.

4.2 Materials and methods

Annual input-output budgets for sulphate (SO$_4$) and total inorganic nitrogen (TIN=NO$_3$+NH$_4$) for the period 1990–2010 were calculated for a selection of 17 IM sites (CZ01, CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16). Annual output fluxes for organic nitrogen were calculated for 16 sites (CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16) during the period 1995–2010, according to data availability. For the location of the sites see Fig.1.1 in Chapter 1. The selection of catchments was guided by the availability of deposition (bulk and throughfall) data and surface water chemistry and runoff volume data in the ICP IM database. Total deposition (meq m$^{-2}$ yr$^{-1}$) i.e. the input of wet and dry deposition of sulphate to the catchment was estimated from bulk deposition (open area) and throughfall (forest stands) measurements. Because of the strong impact of canopy processes, bulk deposition measurements were used for nitrogen (NH$_4$ + NO$_3$) as total deposition estimates. Annual total deposition fluxes to the basins were calculated as the sum of monthly values. Output fluxes for TIN (meq m$^{-2}$ yr$^{-1}$) from the catchments were calculated as the product of measured catchment discharge and ion concentrations. Output fluxes of TIN were calculated as a sum of total inorganic nitrogen (TIN = NO$_3$ + NH$_4$) (meq m$^{-2}$ yr$^{-1}$). Loss of organic nitrogen was calculated as total nitrogen loss minus TIN loss. Annual runoff water element fluxes were calculated by summing mean monthly fluxes, obtained from monthly mean water flux and monthly mean solute concentration.

At each site trends in fluxes outlined above were analysed using the non-parametric Seasonal Kendall test (Hirsch et al. 1982) applied to annual data. The magnitude of trend was estimated by the Theil-Sen slope estimation method, which is equivalent to Sen slope estimator (Sen 1968). This method estimates the slope by calculating the median of all between-year differences in the variable of interest. The unit of the slope estimate for yearly based data is meq m$^{-2}$ yr$^{-1}$ for fluxes. For the analysed parameters, a calculated positive value of slope estimate indicates an increasing trend (increasing values with time), and a negative value indicates a decreasing trend. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis i.e providing at least 95 % confidence that the detected trend was significantly different from a zero trend.

In order to quantify retention/release of sulphur and nitrogen in the catchment, a percent net export (pne) was calculated. The percent net export is defined as: pne = (output–deposition)100 / deposition. Positive pne values indicate release and negative pne values indicate retention in the catchment.
4.3 Results and discussion

Large differences in the deposition of sulphur and nitrogen can be observed between the different sites, with the highest values in southern Scandinavia and central Europe and lowest values at sites in northern regions (Fig. 4.1). This reflects well-known gradients in European emissions and deposition of air pollutants. Bulk deposition of TIN (NO$_3$ + NH$_4$) generally exceeded SO$_4$ deposition on an equivalent basis at most of the sites. The relative importance of the N deposition generally has increased in more southern regions (Forsius et al. 2005). At most sites, throughfall deposition of N has been lower than that of bulk deposition, indicating canopy uptake (Vuorenmaa et al. 2012).

Figure 4.1 Mean annual total deposition of sulphur (SO$_4$, meq m$^{-2}$ yr$^{-1}$) and total inorganic nitrogen (TIN = NO$_3$ + NH$_4$, meq m$^{-2}$ yr$^{-1}$) at ICP IM sites in 1995−2010.

A statistically significant downward trend (p < 0.05) of total S deposition from 1990 to 2010 was observed at all studied IM sites (Table 4.1). As a response to decreased S deposition, sulphate fluxes in runoff have decreased at 16 out of 17 sites, being significant at 60% of the sites. Thus, the regional-scale decreases of sulphate deposition and runoff water fluxes observed in the earlier trend assessments (Forsius et al. 2001, Kleemola 2005, Kleemola and Forsius 2006, Vuorenmaa et al. 2009) have continued to decline. Nitrogen deposition has also decreased at almost all sites (15 out of 17), being significant at 53% of the sites. In contrast to sulphate, inorganic nitrogen (TIN) fluxes in runoff showed mixed response with both decreasing and increasing trends. Statistically significant decreasing trends were observed at four sites and increasing trends at two sites (DE01, Forellenbach, Germany and SE14, Aneboda, Sweden). The significant increasing trends for these two sites are probably due to excess N mineralization and increased NO$_3$ leaching, resulted from forest damage and dieback in the areas due to storm logging and bark beetle infestation (Beudert et al. 2007, Löfgren et al. 2011).
Calculated sulphate budgets showed increasing percent net exports for SO\textsubscript{4} at majority of the sites, indicating a net release of previously stored SO\textsubscript{4}, particularly during the 2000s (Fig. 4.2, Table 4.1). This process is clearly seen e.g. at the site CZ02 (Lysina, Czech Republic), which has been exposed to high sulphur deposition (> 200 meq m\textsuperscript{-2} a\textsuperscript{-1}) in the early 1990s, but has also been subjected to drastic decrease of S deposition during the 1990s (Fig. 4.3, Table 4.1). In low deposition area at the site FI03 (Hietajärvi, Finland) sulphate restrains to the catchment, but net retention rate is decreasing over the past 20 years (Fig. 4.3). These results are consistent with budget calculations for a number of other studies from European forested catchments (de Vries et al. 2001, Prechtel et al. 2001) indicating that forest soils are now releasing stored airborne S that had accumulated in the past. A net release of stored SO\textsubscript{4} is considered to act as a H\textsuperscript{+} source at many IM sites (Forsius et al. 2005), and SO\textsubscript{4} remains the dominant source of actual soil acidification despite the generally lower input of S than N in European forested ecosystems (deVries et al. 2003). Several processes, including desorption and excess mineralisation, regulate the long-term response of soil S, and a differentiation is necessary for assessing the effects of emission reductions on acidification recovery and for predictions of future responses (Markewitz et al. 1998, Prechtel et al. 2001). Many of these S retention processes are also sensitive to changes in climatic variables, and would therefore be affected by climate change (e.g. Wright 1998).

Nitrogen is generally the growth-limiting nutrient in forest ecosystems, and the uptake of available N compounds is efficient. In contrast to sulphur, nitrogen deposition is usually retained in boreal terrestrial ecosystems; typically < 10 % is leached in runoff, mostly as NO\textsubscript{3}. Nitrate is a strong acid anion and so can acidify soil and

![Figure 4.2. Percentiles (25 %, median 50 %, 75 %) of percent net export (pne, %) of sulphate (SO\textsubscript{4}) and total inorganic nitrogen (TIN) for the IM sites CZ01, CZ02, FI01, FI03, NO01, NO02, SE04 in 1990–2010 (a and b, respectively) and for the sites CZ01, CZ02, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16 in 2000–2010 (c and d, respectively).](image-url)
water like $\text{SO}_4$ (e.g. Wright et al. 2005). The percent net export (pne) of nitrogen has generally ranged between −97 % and −90 % at the studied IM sites during the 2000s (Fig. 4.2), indicating a strong retention of N in the catchment. Although nitrogen has played a minor role in the acidification in the past, its relative importance is increasing because N emissions have decreased much less than sulphur emissions. The role of nitrate as an acidifying agent may increase, when continued high nitrogen deposition may result in N-saturation of terrestrial ecosystems, and excess NO$_3$ leach to surface waters (e.g. Aber et al. 1989, Dise and Wright 1995, Macdonald et al. 2002, Oulehle et al. 2012). During the past 20−30 years, there are no signs of widespread regional increases in nitrate concentrations in sensitive freshwaters in Europe. However, nitrogen continues to accumulate in catchment soils and vegetation. N-saturation may thus require many decades to occur, at least at levels of N deposition typical for Europe (Wright et al. 2001).

Although the effects of anthropogenic nitrogen inputs on the dynamics of inorganic N in watersheds have been studied extensively, the influence of N enrichment on organic N loss is not as well understood (Pellerin et al. 2006). Studies comparing dissolved organic nitrogen (DON) losses from old-growth forests have reported that DON may account for 60−95 % of total dissolved nitrogen (TDN) losses from minimally disturbed watersheds (Perakis and Hedin 2002, Van Breeman 2002). Similar results were observed in the IM catchments (Fig. 4.4). Part of this DON may decompose in the downstream surface waters or sea areas (releasing inorganic N compounds) and may thus contribute to detrimental N effects.

In forested watersheds, there is evidence of the effects of N enrichment on DON production in soils. Plot-scale inorganic N fertilization studies have reported increased DON concentrations in both the forest floor (McDowell et al. 2004) and mineral soils (Pregitzer et al. 2004). There is a link between N deposition and organic nitrogen loss at IM sites, suggesting that the sites with higher N deposition exhibit also higher organic N loss in runoff (Fig. 4.5). However, other studies have shown that N enrichment has only a little impact on DON dynamics (Raastad and Mulder 1998, Gundersen et al. 1998, Hagedorn et al. 2001, Pilkington et al. 2005), as detected also at some IM sites. The effects of elevated N loading on watershed DON loss likely vary with the type and magnitude of human disturbance, as well as inherent ecosystem characteristics, and abiotic and biotic factors ultimately limit the loss of DON from watersheds (Pellerin et al. 2006). Anyway, DON leaching is receiving increasing attention because climate change impacts on mineralization of organic nitrogen and leaching of organic matter, and potential risk for elevated N loss from watersheds to surface waters may be anticipated in the future.
Environmental factors other than air pollutants – so-called ‘confounding factors’ – may largely affect the ecosystem behavior (e.g. de Wit et al. 2007). Spruce forest stands at German site DE01 were exposed to bark beetle infestation in 1996–1997, and
resulted in forest dieback in the area: by 2003, the total area share of spruce stands fell about 30% (Beudert et al. 2007). The rapid change in the vegetation cover caused dramatic biogeochemical changes on catchment scale, e.g. resulted in increased leaching of nitrogen due to excess mineralization (Fig. 4.6) (Beudert et al. 2007). A similar episode took place at Swedish site SE14 during 2005−2009; in 2009 approximately 50% of the trees were dead or seriously affected (Löfgren et al. 2011), and resulted in increased N leaching (Table 4.1). Storminess and insect pests are anticipated to increase in the future due to climate change, and may have a large impact on terrestrial and aquatic ecosystems. In general, many of S and N retention/release processes are sensitive to changes in climatic variables, and would therefore be affected by future climate changes (e.g. Wright et al. 1998, Benčoková et al. 2011).

### Table 4.1. Trends of annual input (deposition) and output (runoff water) fluxes and percent net export (pne) for sulphate (SO$_4^-$) and TIN (NO$_3^-$ + NH$_4^+$) at studied IM sites in 1990–2010. Statistically significant trend ($p < 0.05$) of annual change (meq m$^{-2}$ yr$^{-1}$ for fluxes, % yr$^{-1}$ for pne) is in bold.

<table>
<thead>
<tr>
<th></th>
<th>SO$_4$ dep (meq m$^{-2}$ yr$^{-1}$)</th>
<th>SO$_4$ out (meq m$^{-2}$ yr$^{-1}$)</th>
<th>SO$_4$ pne (% yr$^{-1}$)</th>
<th>TIN dep (meq m$^{-2}$ yr$^{-1}$)</th>
<th>TIN out (meq m$^{-2}$ yr$^{-1}$)</th>
<th>TIN pne (% yr$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CZ02, Lysina</td>
<td>-10.7</td>
<td>-7.30</td>
<td>7.6</td>
<td>-1.70</td>
<td>-0.90</td>
<td>-0.94</td>
</tr>
<tr>
<td>LT03, Zemaitija</td>
<td>-2.53</td>
<td>-5.37</td>
<td>-5.45</td>
<td>-1.38</td>
<td>0.01</td>
<td>0.07</td>
</tr>
<tr>
<td>CZ01, Anenske Povodi</td>
<td>-5.58</td>
<td>2.38</td>
<td>8.3</td>
<td>-0.87</td>
<td>-0.07</td>
<td>-0.08</td>
</tr>
<tr>
<td>NO01, Birkenes</td>
<td>-3.36</td>
<td>-3.81</td>
<td>1.57</td>
<td>-1.11</td>
<td>-0.18</td>
<td>-0.04</td>
</tr>
<tr>
<td>SE04, Gårdsjön</td>
<td>-3.40</td>
<td>-5.02</td>
<td>1.52</td>
<td>-1.51</td>
<td>0.06</td>
<td>0.15</td>
</tr>
<tr>
<td>DE01, Forellenbach</td>
<td>-2.88</td>
<td>-0.94</td>
<td>9.37</td>
<td>-1.00</td>
<td>4.82</td>
<td>7.45</td>
</tr>
<tr>
<td>EE02, Saarejärve</td>
<td>-1.83</td>
<td>-0.45</td>
<td>7.53</td>
<td>0.27</td>
<td>0.26</td>
<td>0.77</td>
</tr>
<tr>
<td>LV01, Rucava</td>
<td>-1.33</td>
<td>-4.43</td>
<td>-3.57</td>
<td>-0.93</td>
<td>0.1</td>
<td>0.19</td>
</tr>
<tr>
<td>LV02, Zoseni</td>
<td>-2.10</td>
<td>-9.21</td>
<td>-7.98</td>
<td>-4.15</td>
<td>-0.26</td>
<td>-0.10</td>
</tr>
<tr>
<td>SE15, Kindla</td>
<td>-1.51</td>
<td>-4.06</td>
<td>-6.87</td>
<td>-1.04</td>
<td>-0.03</td>
<td>-0.03</td>
</tr>
<tr>
<td>LT01, Aukstaitija</td>
<td>-1.40</td>
<td>-10.9</td>
<td>-3.30</td>
<td>-1.54</td>
<td>-0.26</td>
<td>-0.32</td>
</tr>
<tr>
<td>SE14, Aneboda</td>
<td>-1.59</td>
<td>-3.15</td>
<td>3.39</td>
<td>-0.67</td>
<td>0.15</td>
<td>0.37</td>
</tr>
<tr>
<td>IT01, Renon-Ritten</td>
<td>-2.28</td>
<td>-0.16</td>
<td>1.13</td>
<td>-1.08</td>
<td>-0.05</td>
<td>-0.10</td>
</tr>
<tr>
<td>FI01, Valkea-Kotinen</td>
<td>-1.11</td>
<td>-0.46</td>
<td>2.57</td>
<td>-0.41</td>
<td>-0.02</td>
<td>0.04</td>
</tr>
<tr>
<td>NO02, Kärvatni</td>
<td>-0.46</td>
<td>-0.52</td>
<td>0.35</td>
<td>0.09</td>
<td>0.01</td>
<td>0.05</td>
</tr>
<tr>
<td>FI03, Hietsjärvi</td>
<td>-0.62</td>
<td>-0.39</td>
<td>0.96</td>
<td>-0.19</td>
<td>-0.02</td>
<td>-0.04</td>
</tr>
<tr>
<td>SE16, Gammaratten</td>
<td>-0.67</td>
<td>-1.18</td>
<td>-4.60</td>
<td>-1.20</td>
<td>-0.04</td>
<td>-0.14</td>
</tr>
</tbody>
</table>

### 4.4 Conclusions

Forest soils are now releasing S that had accumulated in the past. The more efficient retention of N than S results in generally higher leaching fluxes of SO$_4^-$ than those of NO$_3^-$ in European forested ecosystems. Sulphate thus remains the dominant source of actual soil acidification despite the generally lower input of S than N. Organic nitrogen may account for significant fraction of total nitrogen, and N deposition may increase organic N loss in forested catchments. Continued work on processes regulating both N and S retention and release in terrestrial ecosystems is therefore needed. This would be important for assessing the effects of emission reductions on acidification recovery as well as other N pollution problems in semi-natural ecosystems. Many of these S and N retention processes are also sensitive to changes in climatic variables, and would therefore be affected by future climate changes.

The next phase of the work on mass balances of S and N for IM sites will be preparation of a scientific paper, involving further updating and revision of data, and more detailed assessment of the role of organic nitrogen in mass balance budget. The national focal points and the representatives for the sites will be invited to assist with these activities.
References


Annex 1

Report on National ICP IM Activities in Estonia

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ICP IM Programme has been carried out at two monitoring sites in Estonia since 1995. Vilsandi (EE01) area is located on Estonia’s westernmost island (58°23’ N, 21°50’ E) and Saarejärve (EE02) is located in the forested sub-catchment area (109 ha) of Lake Saare in eastern Estonia (58°39’ N, 26°45’ E). Stand characteristics of permanent plots of the monitoring sites are presented in Table 1.

Reporting year 2012 was a year of higher precipitation and, hence, it was characterized by higher bulk precipitation, throughfall, soilwater and runoff water fluxes compared to the average annual water fluxes for the period 1995-2011 (Table 1). Precipitation distribution between months was rather homogeneous, and the usual summer drought period in July and August was not observed.

In 2012 sampling and measurements were run under the sub-programmes: AM, AC, PC, TF, SF, SC, SW, GW, LF, FD, EP, MB at both areas and, additionally, under RW and AL at the Saarejärve (EE02) area.

Table 1 Stand characteristics and 2012 water fluxes (mm) compared with average annual water fluxes during 1995−2011 (in brackets) at permanent plots of Estonian integrated monitoring areas.

<table>
<thead>
<tr>
<th>Permanent plot</th>
<th>Vilsandi (EE01) pine stand</th>
<th>Saarejärve (EE02) pine stand</th>
<th>Saarejärve (EE02) spruce stand</th>
</tr>
</thead>
<tbody>
<tr>
<td>Site type</td>
<td>Fragario-Pinetum</td>
<td>Rhodococco-vitis-idae-Pinetum</td>
<td>Vaccinio-myrtilli-Piceetum</td>
</tr>
<tr>
<td>Soil type</td>
<td>Calcar-Gleyic Leptosol</td>
<td>Haplic Podzol</td>
<td>Haplic Podzol</td>
</tr>
<tr>
<td>Age of dominant trees</td>
<td>107</td>
<td>127</td>
<td>100</td>
</tr>
<tr>
<td>Bulk precipitation (mm)</td>
<td>891 (538)</td>
<td>693 (667)</td>
<td></td>
</tr>
<tr>
<td>Throughfall (mm)</td>
<td>507 (304)</td>
<td>636 (542)</td>
<td>639 (464)</td>
</tr>
<tr>
<td>Soil water amount from depth of 10 cm (mm)</td>
<td>152 (112)</td>
<td>64 (59)</td>
<td>140 (116)</td>
</tr>
<tr>
<td>Soil water amount from depth of 40 cm (mm)</td>
<td>116 (95)</td>
<td>40 (29)</td>
<td>101 (80)</td>
</tr>
<tr>
<td>Runoff water (mm)</td>
<td>180 (117)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Anthropogenic anions in deposition at Vilsandi (EE001) and Saarejärve (EE02) IM sites

The shares of cations and anions in bulk precipitation at IM areas EE01 and EE02 are quite different from each other. Precipitation at Vilsandi station is dominated by chloride (49 %) from marine impacts, followed by two of the most of anthropogenic origin ions: sulphate (34 %) and nitrate (15 %). Precipitation at Saarejärve station is mainly dominated by HCO₃ (43 %) of natural origin, followed by anthropogenic sulphate (32 %), while the share of nitrate is low(7 %).
Anthropogenic sulphate reduction has lasted throughout the monitoring period in bulk precipitation and throughfall of the pine and spruce stands at Saarejärve station. Annual open area deposition of sulphur at Saarejärve was only 2.2 kg S ha\(^{-1}\) in 2012, which is the lowest load measured during the 18-year monitoring period. The annual load of sulphur has declined from 12 kg S ha\(^{-1}\) in 1996 to 3–5 kg S ha\(^{-1}\) during 1997–2001, to 3–4 kg S ha\(^{-1}\) during 2002–2008, to 2.3 in 2010, 2011 and finally to 2.2 kg S ha\(^{-1}\) in 2012. The continuous reduction of S load by deposition at Saarejärve station indicates effectiveness of new cleaner technologies for power generation, as well as flue gas cleaning of local oil shale industry and oil shale power plants in the industrial northeastern region of Estonia.

In 2012 the annual S load by bulk precipitation at Vilsandi was 2.8 kg S ha\(^{-1}\) while in 1996 the S deposition was 4.9 kg ha\(^{-1}\). The higher deposited load of S at the western island of Vilsandi station indicates more stable long-range transboundary S pollution over the past five years.

Higher shares of anionic and cationic inorganic nitrogen (\(\text{NO}_3^-\) and \(\text{NH}_4^+\)) in bulk precipitation at Vilsandi (Figure 1) show a higher proportion of the impact of air pollution compared to Saarejärve station. At Saarejärve station inorganic nitrogen (\(\text{NO}_3^-,\text{N} + \text{NH}_4^-,\text{N}\)) load by bulk precipitation was only 2.3 (1.2+1.1) kg N ha\(^{-1}\) in 2012, which is the lowest value ever recorded over the observation period. Also, throughfall deposition of inorganic nitrogen is characterized by low quantities: 2.7 (1.3+1.4) kg N ha\(^{-1}\) in the pine stand and 2.6 (1.0+1.6) kg N ha\(^{-1}\) in the spruce stand – which should not have a direct effect on the growth of green algae communities on needles, or species composition changes of field layer. Meanwhile, at Vilsandi area, inorganic N deposition by bulk precipitation was 5.5 (2.7+2.8) kg N ha\(^{-1}\) and by throughfall 6.7 (3.8+2.9) kg N ha\(^{-1}\) in 2012, which is double the concentration recorded at
Saarejärve IM area. The relatively high inorganic N loads at Vilsandi are of long-range transboundary origin as on this small island there is no industrial action, not much traffic or livestock husbandry (Figure 2).

Figure 2 Summed flow of NO$_2$ at Vilsandi and Saarejärve IM area in 2012.
Annex 2

Report on National ICP IM Activities in Spain

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  e-mail: chusmi@unav.es

The ICP IM activities in ES02 - Bertiz catchment have continued uninterrupted since 2007. Additionally, the following complementary activities were completed or started during 2012:

1 Ecological processes that determine the spatial structure of Carex remota and other species in forest riparian environments

The spatial structure of plant populations results from the combined effect of abiotic and biotic interactions that their individuals and their ancestors have been exposed to in the past. Therefore, the study of the spatial structure of populations and environment becomes crucial in the understanding and management of the ecological processes that define it.

This study focused in three different forest stream areas (P1, P2, and P3) within ES02 Bertiz catchment, which bear a number of populations of sedge (Carex remota), hard fern (Blechnum spicant) and wood sorrel (Oxalis acetosella). In order to understand the dependence of the three species on abiotic factors and biotic processes, their spatial patterns were modelled by means of hierarchical spatial point process models of growing complexity. The bivariate Ripley’s function was used to analyse the spatial relationships between the different species present in the studied areas.

Our results showed that the importance of the different factors that determine the spatial distribution of the studied plant species varies according to the scale and heterogeneity of the environment. Thus, abiotic factors play a major role in the spatial distribution of individuals as long as they remain heterogeneous within a given area (Figure 1c and d). When abiotic factors are homogeneous, the biotic interspecific and intraspecific interactions between individuals that become decisive (Figure 1e and f). Even so, both types of factors act together to a greater or lesser extent.

Soil moisture was identified as the key abiotic factor conditioning the spatial distribution of Carex remota and Blechnum spicant, which favour high (Figure 1a, c and e) and low (Figure 1d) soil moisture respectively. Intraspecific competition was identified as the major biotic factor influencing the distribution of Carex remota individuals. Finally, the distribution of Oxalis acetosella was chiefly mediated by facilitation processes from Carex remota (Figure 2).
Figure 1 Soil moisture map (mV) and soil moisture variation coefficient, respectively, in plots P1 (a and b), P2 (c and d), and P3 (e and f). Point patterns are also represented for the three studied species: *Carex remota* in P1 (a), P2 (c) P3 (e) (left column); *Blechnum spicant* (green triangles) and *Oxalis acetosella* (blue points) in P2 (d); and *O. acetosella* en P3 (f).

Figure 2 Bivariate Ripley $L_{ij}(r)$ function between *C. remota* and *O. acetosella*, in P2 (left) and P3 (right).
2 Fish and amphibian populations monitoring and ecological status assessment in ES02 stream waters

Since 2007 fish and amphibian populations are being monitored in the Suspiro stream, which drains the ES02 catchment and constitutes a typical example of a headwaters brook within the Atlantic valleys of the Pyrenees. Its ecological status is also being assessed, based on the composition of its macroinvertebrate community. What follows is the summary of 5 years of activity in the two control areas (upper/lower sections) that was presented in 2012.

All the species found in the stream are native of the region. The upper, lower-flow section is characterised by the absence of fish species and the presence of stable populations of the amphibian species Salamandra salamandra (fire salamander) and endemic Calotriton asper (Pyrenean brook salamander/newt), which are listed as vulnerable and near threatened species in Spain, respectively (Pleguezuelos et al. 2002). Both are very sensitive to disturbances and their presence is considered as a good indicative of habitat quality. The lower section presents a stable and well established population of Salmo trutta (brown trout), plus presence of the IUCN critically endangered Anguilla anguilla (European eel) and Cottus aturi, a bullhead species described as endemic of the Pyrenean Adour and Nivelle river catchments (Freyjofh et al. 2005), which is listed as endangered in Spain and whose presence constitutes a good indicator of undisturbed mountain brooks.

The ecological status of the stream was assessed using the Iberian Biological Monitoring Work Party (IBMWP; Alba-Tercedor & Sánchez-Ortega 1988) and the Iberian Average Score Per Taxon (IASPT) indexes; their recorded values during the period reported are shown in figure 3. The IBMWP values of both sections have consistently ranked as Category I ‘very good’, which corresponds to unpolluted/unimpacted sites. The observed oscillations correspond to lower runoff levels, lower availability of suitable habitats, and sampling issues related with those factors -specially true in the case of the upper stream section. Despite these minor differences, the IASPT values indicate a consistent, major presence of taxa sensitive to pollution in both cases, fact which again remarks the optimal quality of the stream.
3 Ozone uptake assessment in a Pyrenean beech (*Fagus sylvatica* L.) stand

Widespread evidence on the deleterious effects of *O*$_3$ on vegetation and crops in Europe has been extensively documented during the last decades. The DO$_3$SE model constitutes a feasible tool for the estimation of the dose absorbed by plants and is thus extensively used for the assessment of *O*$_3$ risk to vegetation and crops within the framework of the LRTAP Convention (Mills et al. 2011). The study aims at inferring the *O*$_3$ dose absorbed by the trees and providing data from old-growth trees growing in field conditions to contrast with DO$_3$SE estimations.

Sap flow was estimated at ES02 Bertiz using the THB approach (Čermák et al. 2004, Figure 4) from the leaf sprout in April to the leaf drop in mid November during 2012. Soil water content, meteorological parameters and *O*$_3$ concentrations are also registered at the site. The AOT40 critical level for the protection of forests was exceeded as early as in May; the *O*$_3$ levels scored an hourly maximum of 158 μg m$^{-3}$ (Figure 5). Mean temperature was 17.5°C and ranged from 0°C to 41°C. There were 89 days of rainfall that totalled 828 mm of precipitation. Maximum solar radiation values were over 1200 W m$^{-2}$. This field experience is being expanded into a second season during 2013.

References


Čermák, J., Kučera, J., Nadezhina, N. 2004. Sap flow measurements with some thermodynamic methods, flow integration within trees and scaling up from sample trees to entire forest stands. Trees 18, 529-546.


Annex 3

Report on National ICP IM activities in Sweden 2011−2013

Lundin, L.¹, Löfgren, S.¹, Bovin, K.², Bringmark, L.¹, Grandin, U.¹, Pihl Karlsson, G.³, Moldan, F.³ and Thunholm, B.³.

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The programme is funded by the Swedish Environmental Protection Agency.

Introduction

Swedish integrated monitoring programme is run on four sites distributed from south central Sweden (SE14 Aneboda) over the middle part (SE15 Kindla), to a northerly site (SE16 Gammtratten) representing north Sweden. The long-term monitoring site SE04 Gårdsjön F1 is complementary on the inland of the West Coast and has been influenced by long-term high deposition loads. The Swedish group now compiled results from the four Swedish IM sites for the year 2011. The sites are well-defined catchments with mainly coniferous forest stands dominated by bilberry spruce forests on glacial till deposited above the highest coastline, meaning no water sorting of the soil material. Both climate and deposition gradients coincide with site distribution from south towards north (Table 1). Forest stands are mainly over 100 years and at least three of them have several hundred years of natural continuity but were up to c. 50 years ago partly lightly grazed woodlands. In early 2005, a heavy storm struck the Swedish IM site Aneboda, SE14. However, in comparison to the surrounding forests this site managed rather well and a rough estimate considered 20−30% of the area affected. Results from the 2006 inventory show increased number of logs. In 1996 the total number of logs in all plots was 317. In the inventory of 2001, this number had decreased to 257. In 2006, after the storm, the number of logs increased to 433 and with a calculation for the whole IM site SE14, this means an increase with 2711 logs. In later years, 2007−2010, bark beetle, Ips typographus, attacks have almost totally erased the old spruce trees.

Table 1 Geographic location and long-term climate at the Swedish IM sites.

<table>
<thead>
<tr>
<th></th>
<th>SE04</th>
<th>SE14</th>
<th>SE15</th>
<th>SE16</th>
</tr>
</thead>
<tbody>
<tr>
<td>Latitude; Longitude</td>
<td>N 58° 03’; E 12° 01’</td>
<td>N 57° 05’; E 14° 32’</td>
<td>N 59° 45’; E 14° 54’</td>
<td>N 63° 51’; E 18° 06’</td>
</tr>
<tr>
<td>Altitude, m</td>
<td>114-140</td>
<td>210-240</td>
<td>312-415</td>
<td>410-545</td>
</tr>
<tr>
<td>Area, ha</td>
<td>3.7</td>
<td>18.9</td>
<td>20.4</td>
<td>45</td>
</tr>
<tr>
<td>Mean annual temperature, °C</td>
<td>+6.7</td>
<td>+5.8</td>
<td>+4.2</td>
<td>+1.2</td>
</tr>
<tr>
<td>Mean annual precipitation, mm</td>
<td>1000</td>
<td>750</td>
<td>900</td>
<td>750</td>
</tr>
<tr>
<td>Mean annual evapotranspiration, mm</td>
<td>480</td>
<td>470</td>
<td>450</td>
<td>370</td>
</tr>
<tr>
<td>Mean annual runoff, mm</td>
<td>520</td>
<td>280</td>
<td>450</td>
<td>380</td>
</tr>
</tbody>
</table>

In the following, some special conditions and ongoing work for the four Swedish IM sites during 2011 to 2013 are presented. A special issue of the Journal Ambio was finalised at the end of 2011 (Starr 2011).
Climate and Hydrology in 2011

Temperature climate for the year 2011 showed higher annual averages compared to the long-term mean 1961–1990. This was a shift from 2010 when temperatures mainly were lower. However, temperatures have during several years been higher compared to long-term averages with values between +0.5 °C and 2.3 °C. In 2011, annual average temperatures in the southern sites were c. 0.7 °C higher than long-term averages and further north deviations were even greater with +2.3 °C at site SE16.

Precipitation amounts in 2011 were higher than the long-term average for the two southern sites (SE14: 7 % and SE04: 25 %) while the two sites further north had only c. 70 % of the long-term averages. Fairly low precipitation occurred in November while some summer months had exceedances, being rather similar to the year before.

The characteristic annual hydrological patterns of the catchments are high groundwater levels during winter and lower levels in summer and early autumn. This pattern should also be reflected in runoff. However, warm periods in winter have during a number of years furnished snowmelt and runoff in winter resulting in lower spring discharges in the snowmelt period. For 2009–2011, this pattern was somewhat changed with rather ordinary discharges over the year with low discharge in the winter period at the sites. In 2011 SE04 and SE16 deviated somewhat from this and in SE16 snowmelt occurred in January furnishing a fairly low and early snowmelt peak already in April. Late summer and early autumn peaks occurred and especially at the two southern sites runoff was somewhat high (Figure 1).

Runoff in 2011 made up 39–62 % of annual precipitation and could be compared with an average for previous three years on c. 40–60 %, i.e. ordinary. Higher rates occur especially on the west coast Gårdsjön (62 %) while the two northern sites showed similar values of 48–50 % and the southern site SE14 only had 39 % of precipitation as runoff. The runoff could be considered normal at the central site Kindla (SE15). Gårdsjön (SE04) had high flow and partly also Gammtratten (SE16), while Aneboda in the south had fairly low flows (Table 2).

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Table 2 Compilation of the 2011 water balances for the four Swedish IM sites. P – Precipitation, TF – Throughfall, I – Interception, R – Water runoff.

<table>
<thead>
<tr>
<th>Gårdsjön SE04</th>
<th>Aneboda SE14</th>
<th>Kindla SE15</th>
<th>Gammtratten SE16</th>
</tr>
</thead>
<tbody>
<tr>
<td>mm</td>
<td>% of P</td>
<td>mm</td>
<td>% of P</td>
</tr>
<tr>
<td>Bulk precipitation, P</td>
<td>1321</td>
<td>100</td>
<td>885</td>
</tr>
<tr>
<td>Throughfall, TF</td>
<td>921</td>
<td>70</td>
<td>685</td>
</tr>
<tr>
<td>Interception, P-TF</td>
<td>400</td>
<td>30</td>
<td>200</td>
</tr>
<tr>
<td>Runoff, R</td>
<td>820</td>
<td>62</td>
<td>349</td>
</tr>
<tr>
<td>P-R</td>
<td>501</td>
<td>38</td>
<td>535</td>
</tr>
</tbody>
</table>

Water chemistry

Low ion content characterises the deposition and throughfall for the three inland sites (c. 1–3 mS m⁻¹) while sea salt provides higher ion content in the west coast SE04 site (8.0 mS m⁻¹ in bulk deposition). Water pathways through the soils of the catchments are fairly short and shallow, and most of the surface water formation depends on short connections between infiltration and surface water. Acidity in the deposition was mainly the same at all sites with partly higher pH (0–0.1 units) in throughfall (TF) compared to bulk deposition (BD), though SE14 Aneboda site deviated from this, pH in TF was 0.5 units lower compared to BD. At this site there was a considerable die-back of the spruce forest. For all areas, pH was close to 5.0 in BD (Table 3). This was similar to the previous years.
Chemical reactions during water flow through the catchments buffered the acid water fairly little and pH values in the stream water were between 4.4 and 4.8 in the three southern sites, while in the northern site SE16, pH in stream water was c. 5.6 with an ANC of c. 0.1 meq L\(^{-1}\). At SE14, the pH in stream water was 4.7 compared to bulk deposition 5.1 and ANC increased as a consequence of increased DOC (27 mg L\(^{-1}\)) and reached about 0.09 meq L\(^{-1}\). Organic anions contribute to positive ANC. In stream water from SE15, ANC was 0.01 meq L\(^{-1}\) partly influenced by low DOC (c. 10 mg L\(^{-1}\)).

In relation to the decrease in sulphur deposition (2−7 kg ha\(^{-1}\) yr\(^{-1}\) during the period 1996 to 2010), the increase in pH has been 0.3 to 0.5 pH-units. In stream water pH has increased by c. 0.3 units while sulphur decreased by c. 30%, but the northernmost site Gamtratten shows small changes in pH, however, this site has higher pH in stream water c. 5.6. Sulphur in deposition equals stream water flux at low levels on c. 2 kg ha\(^{-1}\) yr\(^{-1}\).

Surface water chemistry is to a considerable extent influenced by the content of organic matter. In the Aneboda site (SE14), the DOC concentration was comparably high 27 mg L\(^{-1}\), while the other sites Gårdsjön (SE04), Kindla (SE15) and Gammtatten (SE16) showed lower values on 18, 11 and 9 mg L\(^{-1}\), respectively. The fairly high DOC concentrations at sites Aneboda and Gårdsjön furnished high potential for metal complexion and transport. The content of organic nitrogen also correlates positively to organic matter and fluxes turn high where DOC is high. The Norg values ranged between 0.19−0.59 mg L\(^{-1}\) and have importance for the inorganic nitrogen and transformations from the mainly inorganic nitrogen deposition to the organic nitrogen output flux. Organic nitrogen needs further consideration in nitrogen balance estimations.

Heavy metal deposition and runoff were studied in the IM catchments and studies are still showing accumulation in the sites. However, the storage of lead and cadmium showed decreased concentrations in the organic humus layer from where metals were translocated to deeper soil layers. Mercury concentrations, however, continued to increase and all sites show much lower runoff compared to deposition.

### Table 3
Deposition chemistry 2011 for the four Swedish IM sites. S and N in kg ha\(^{-1}\) yr\(^{-1}\).

<table>
<thead>
<tr>
<th></th>
<th>SE04</th>
<th>SE14</th>
<th>SE15</th>
<th>SE16</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH, bulk deposition</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.1</td>
</tr>
<tr>
<td>pH, throughfall</td>
<td>5.9</td>
<td>5.5</td>
<td>5.1</td>
<td>5.0</td>
</tr>
<tr>
<td>SO(_4)-S, bulk deposition</td>
<td>6.8</td>
<td>6.8</td>
<td>5.7</td>
<td>4.1</td>
</tr>
<tr>
<td>N-tot, bulk deposition</td>
<td>8.6</td>
<td>6.8</td>
<td>5.7</td>
<td>4.1</td>
</tr>
</tbody>
</table>
Swedish Integrated Monitoring data used in scientific projects

Besides the environmental assessment and applied research directed towards policy and decision making within the CLRTAP framework, Integrated monitoring data furnish great opportunities to perform basic research on different biogeochemical processes in the semi-natural forests and this offers information of reference character. Basic forest hillslope processes modifying the N, Al and DOC chemistry in soil solution and groundwater along transects from recharge areas to discharge areas and ultimately the streams have been studied (Fölster 2000, Löfgren and Cory 2010, Löfgren et al. 2010). Additionally, the IM data have been used as references to results from manipulation experiments and man-made activities (Löfgren et al. 2009b). Trends in acidity related variables in the IM and PMK5 (an antecedent monitoring program of semi-natural forests) streams were e.g. compared with trends in streams subjected to experimental low dose forest soil liming in southwest Sweden (SKOKAL, Swedish Forest Agency Forest Soil Liming program) (Löfgren et al. 2009a).

Related to the semi-natural forest status, the long-term monitoring data from the IM and PMK5 streams are often used as references and for estimating background concentrations and fluxes of different elements under undisturbed conditions (Löfgren 2007, Löfgren et al. 2009b). This applied research is of great value for the Swedish Environmental Protection Agency (SEPA), the Swedish Agency for Marine and Freshwater Management and the Swedish Water Authorities using the data for national assessments of acidification status, nutrient source apportionments, Pollution Load Compilations (PLC’s, HELCOM and OSPARCOM) and classifications of ecological status according to the EU Water Framework Directive.

Recently, some of the major Swedish forest and forestry research groups have decided to use the Swedish IM data time series and other data to study environmental impacts. Here could be mentioned:

- Climate change and Environmental Objectives [CLEO], a SEPA research program, http://www.cleoresearch.se/; for modelling potential environmental impacts of forestry and climate change,
- ForWater, The Swedish Research Council Formas' strong research environments, https://sites.google.com/site/sluforwater/, for studying forest hillslope processes,
- Future Forest, MISTRA program, http://www.futureforests.se/, for studying reference conditions compared with managed forests
- Acidification of surface waters due to forest biofuel extraction, Swedish Energy Agency Fuel System research program, http://www.energimyndigheten.se/en/Innovations-R--D/Fuel-systems/, for modelling potential effects of whole tree harvesting,

Additionally, research conducted at the Swedish IM sites were in December 2011 published in a special issue of Ambio (http://www.springerlink.com/content/0044-7447/40/8/). The 7 scientific articles deal with issues such as different rates of recovery from acidification in different compartments of the catchments (Löfgren et al. 2011) the impact of N and S deposition on epiphytic algae and lichens and understory vegetation (Grandin 2011a, b), geographical litter decomposition rate
gradients (Bringmark et al. 2011), uncertainties in acidification modelling (Köhler et al. 2011) and simulations of DOC in runoff, based on catchment and riparian zone models, respectively (Futter et al. 2011, Winterdahl et al. 2011). The results of the seven scientific Ambio articles published in 2011 were summarized in the 21st Annual Report 2012 and are not further presented here.

The following section presents summaries of scientific articles based primarily on IM data or where the IM data had a significant impact on the study’s conclusions.

The long-term effects of catchment liming and reduced sulphur deposition on forest soils and runoff chemistry in southwest Sweden (Löfgren et al. 2009)

Whole catchment liming or forest liming has been proposed and implemented as a countermeasure to the effects of elevated sulphur deposition. Since the end of the 1980’s, the Swedish Forest Agency has undertaken experimental forest liming experiments in selected catchments in southern Sweden. These studies were with low doses (3 tonnes ha$^{-1}$) of lime (CaCO$_3$) and dolomite (CaMg(CO$_3$)$_2$). Data from both soil samples and stream water samples have been collected for the 16 years following treatment. The stream data has been complemented with data from untreated catchments, from the Swedish monitoring stream network. Significant differences due to treatment were seen for Ca, cation exchange capacity (CEC) and base saturation (BS) in the humus layer; none of these variables showed a statistically significant change in the mineral soil due to treatment alone. Soil samples from both the treated and untreated sites showed temporal changes in both the humus layer and the mineral soils with increases in pH, Ca and CEC and decreases in BS and Al which were independent of treatment. A combination of treatment and time, gave significant changes in BS and TA (total exchangeable acidity) down to 10 cm in the mineral soil. In the stream water samples, no statistically significant differences were observed between treated and untreated sites. Regardless of treatment, the streams exhibited a general pattern of declining concentrations of SO$_4^{2-}$, Ca, sum of base cations (BC) and increasing acid neutralizing capacity (ANC). In summary, the application of a low dose of lime (3 tonnes ha$^{-1}$) did not result in significant changes in surface water chemistry in the study catchments and changes in soil chemistry were mainly restricted to the humus layer during the 16 years following treatment. The natural recovery, as a result of reductions in sulphur deposition, dominated the effects and was clearly seen in both the treated and untreated study sites. MAGIC simulations indicate that this recovery will continue in the coming decades.

Groundwater Al dynamics in boreal hillslopes at three integrated monitoring sites along a sulphur deposition gradient in Sweden (Löfgren & Cory 2010)

Data from four soil water and groundwater transects from three small, boreal catchments (IM) situated along a south–north sulphur deposition gradient in Sweden were studied to assess whether the soils in the near stream zone can significantly modify the groundwater aluminium (Al) chemistry just before it enters the stream and to what extent different levels of acid deposition influences this. The results show that the groundwater aluminium species composition (Alt = total, Alo = organic, Ali = inorganic) and concentrations reflected the variations in groundwater pH ($r^2 = 0.74$) or TOC ($r^2 = 0.93$) or a combination of both ($r^2 = 0.89$). The highest Al concentrations were recorded in shallow groundwater, creating the prerequisites for large lateral Al-fluxes along the hillslopes during episodes of high flow when superficial flow paths are active. A downhill gradient was also seen, with increasing Alo and TOC concentrations towards the stream. Reduced Ali, in absolute as well as relative terms, but increased Alt concentrations in the discharge areas, indicate complex reactions favouring Alo formation and a local input of Alo from the soils. Results from the transect with the most detailed riparian sampling showed that in the last few meters...
before lateral flow reaches the stream, the mixing of superficial acid soil/groundwater and well-buffered groundwater that had moved along deeper flow paths increased pH and reduced the Ali and Alt concentrations, tangibly. The Alo concentrations were little affected by this pH increase, but at the soil and stream water interface Ali formation was favoured due to the low pH and DOC concentrations in the surface water. Hydrological and soil forming processes within the catchments were more important than acid deposition for the Al dynamics along the hillslopes.

**Decreasing DOC trends in soil solution along the hillslopes at two IM sites in southern Sweden – Geochemical modeling of organic matter solubility during acidification recovery (Löfgren et al. 2010)**

Numerous studies report increased concentrations of dissolved organic carbon (DOC) during the last two decades in boreal lakes and streams in Europe and North America. Recently, a hypothesis was presented on how various spatial and temporal factors affect the DOC dynamics. It was concluded that declining sulphur deposition and thereby increased DOC solubility, is the most important driver for the long-term DOC concentration trends in surface waters. If this recovery hypothesis is correct, the DOC levels should increase both in the soil solution as well as in the surrounding surface waters as soil pH rises and the ionic strength declines due to the reduced input of SO$_4^{2-}$ ions. In this project a geochemical model was set up to calculate the net humic charge and DOC solubility trends in soils during the period 1996–2007 at two integrated monitoring sites in southern Sweden, showing clear signs of acidification recovery. The Stockholm Humic Model was used to investigate whether the observed DOC solubility is related to the humic charge and to examine how pH and ionic strength influence it. Soil water data from recharge and discharge areas, covering both Podzol and riparian soils, were used. The model exercise showed that the increased net charge following the pH increase was in many cases counteracted by a decreased ionic strength, which acted to decrease the net charge and hence the DOC solubility. Thus, the recovery from acidification does not necessarily have to generate increasing DOC trends in soil solution. Depending on changes in pH, ionic strength and soil Al pools, the trends might be positive, negative or indifferent. Due to the high hydraulic connectivity with the streams, the explanations to the DOC trends in surface waters should be searched for in discharge areas and peatlands.
References


**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 2012/2013 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
- A final report on relations between vegetation changes and nitrogen Critical Load exceedance
- A progress report on base line heavy metal approach, estimation of the extent of metal turnover in European forest catchments over the last decades
- A final report on sulphur and nitrogen input-output budgets at ICP IM sites in Europe
- National Reports on ICP IM activities are presented as annexes.

**Keywords**

Integrated Monitoring, ecosystems, small catchments, air pollution, critical loads
Yhdennetyn ympäristön seuranta (ICP IM) kuuluu kansainvälisen ilman epäpuhtauksien kaukokulkeutumista koskevan yleissopimuksen ”Convention on Long-range Transboundary Air Pollution” (1979) alaisiin seurantaohjelmiin. Yhdennetyn seurannan ohjelmassa selvitetään kaukokulkeutuvien saasteiden ja muiden ympäristömurtoisten vaikutuksia elinympäristööme. Muutosten seurantaa ja ennusteita muutosten laajuudesta ja nopeudesta tehdään yleensä pienillä metsäisillä valuma-alueilla, mutta verkostoon kuuluu myös muita alueita. Tämä julkaisu on kooste ohjelmakeskuksen ja yhteistyölaitosten toiminnasta kaudella 2012/2013, joka sisältää:

- Lyhyen yhteenvedon ohjelmassa aiemmin tehdyistä arvioinnista
- Kuvauksen ICP IM ohjelman toiminnasta ja ohjelman seurantaverkosta
- Loppuraportin aluskäsitteissä ja yhteisyydessä typen kriittisen kuormituksen ylityksestä
- Katsauksen raskasmetallien luonnon ekosysteemeissä ja raskasmetallien ainetaseista metsäisillä valuma-alueilla
- Päivityksiä rikitsi- ja tyyppisissä ICP IM alueille
- Kuvauksia kansallisesta ICP IM toiminnasta eri maissa liitteenä.
**Sammandrag**

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

Rapporten sammanfattar de utvärderingar som gjorts av ICP IM Programme Centre och de samarbetande instituten under programåret 2012/2013. Rapporten innehåller:

- En sammanfattning av programmets nuvarande omfattning och databasens innehåll
- En syntes av tidigare utvärderingar av data från programmet
- En slutrapport av sambandet mellan förändringar i markflora och kritisk belastning av kväve
- En lägesrapport beträffande tungmetaller i naturliga ekosystem, inklusive beräkningar av processer och massbalanser på skogliga avrinningsområden i Europa under de senaste årtiondena
- En slutrapport om massbalanser av svavel (S) och kväve (N) på ICP IM områden
- Beskrivning av nationella ICP IM aktiviteter

**Nyckelord**

Integrerad miljöövervakning, ekosystem, små avrinningsområden, luftföroreningar, kritisk belastning

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**Publikationens titel**

22nd Annual Report 2013
Convention on Long-range Transboundary Air Pollution
International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems

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**Publikationens tema**

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