

# 25<sup>th</sup> Annual Report 2016

**Convention on Long-range  
Transboundary Air Pollution**

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

**Sirpa Kleemola and Martin Forsius (eds.)**







REPORTS OF THE FINNISH ENVIRONMENT  
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**wge** Working Group on Effects of the  
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Helsinki 2016

FINNISH ENVIRONMENT INSTITUTE



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Finnish Environment Institute

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Subject Editor: Tapio Lindholm  
Financier: Swedish Environmental Protection Agency, Ministry of the Environment, Finland,  
Working Group on Effects of the LRTAP Convention  
Publisher of publication: Finnish Environment Institute (SYKE)  
P.O. Box 140, FI-00251 Helsinki, Finland, Phone +358 295 251 000, syke.fi

Layout: Pirjo Lehtovaara  
Cover photo: Stefano Rioggi, A view from the new Swiss monitoring site Lago Nero

The publication is available in the internet (pdf): [syke.fi/publications](http://syke.fi/publications) | [helda.helsinki.fi/syke](http://helda.helsinki.fi/syke) and  
in print: [syke.juvenesprint.fi](http://syke.juvenesprint.fi)

Juvenes Print, Tampere 2016

ISBN 978-952-11-4588-9 (pbk.)  
ISBN 978-952-11-4589-6 (PDF)  
ISSN 1796-1718 (print)  
ISSN 1796-1726 (online)

Year of issue: 2016



## 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 2015/2016 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
- A report on dynamic vegetation modelling at ecosystem monitoring and research sites
- An interim report on trend assessment for deposition and runoff water chemistry and climatic variables at ICP IM sites in 1990–2013
- National Reports on ICP IM activities are presented as annexes.

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

## TIIVISTELMÄ

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

Tämä julkaisu on kooste ohjelmakeskuksen ja yhteistyölaitosten toiminnasta kaudella 2015/2016, joka sisältää:

- Lyhyen yhteenvedon ohjelmassa aiemmin tehdyistä arvioinneista
- Kuvauksen ICP IM ohjelman toiminnasta ja ohjelman seurantaverkosta
- Tiivistelmät toiminnasta ohjelman prioriteetti aihealueilla:
  - dynaaminen kasvillisuus- ja maaperämallinnus ICP IM alueilla
  - trenditarkastelut ICP IM alueiden laskeuma- ja valuntadatoille
- Kuvauksia kansallisesta ICP IM toiminnasta eri maissa liitteenä.

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

## SAMMANDRAG

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

Rapporten sammanfattar de utvärderingar som gjorts av ICP IM Programme Centre och de samarbetande instituten under programåret 2015/2016. Rapporten innehåller:

- En sammanfattning av programmets nuvarande omfattning och databasens innehåll
- En syntes av tidigare utvärderingar av data från programmet
- Sammanfattning beträffande utvärderingar inom följande sektorer:
  - modellering av markkemi och markflora
  - trendanalys av ICP IM depositions- och avrinningsdata
- Beskrivning av nationella ICP IM aktiviteter

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



## CONTENTS

Abstract .....	3
Tiivistelmä.....	4
Sammandrag .....	5
Contents .....	6
Abbreviations .....	7
<b>Summary .....</b>	<b>9</b>
<b>I ICP IM activities, monitoring sites and available data .....</b>	<b>19</b>
1.1 Review of the ICP IM activities in 2015–2016 .....	19
1.2 Activities and tasks planned for 2017–2018 .....	20
1.3 Published reports and articles 2015–2016 .....	21
1.4 Monitoring sites and data.....	22
1.5 National Focal Points (NFPs) and contact persons for ICP IM sites .....	25
<b>2 Dynamic vegetation modelling at ecosystem monitoring and research sites .....</b>	<b>27</b>
2.1 Introduction .....	27
2.2 Sites .....	27
2.3 Methods and models .....	29
2.4 Modelled and observed soil chemistry .....	30
2.5 Vegetation modelling examples.....	32
2.6 Status and future tasks .....	32
<b>3 Trend assessments for deposition and runoff water chemistry concentrations and fluxes and climatic variables at ICP Integrated Monitoring sites in 1990–2013 .....</b>	<b>34</b>
3.1 Introduction.....	35
3.2 Material and methods .....	36
3.3 Results and discussion.....	38
<b>Annex 1. Switzerland: Lago Nero – a new site to assess the effects of environmental change on high-alpine lakes and their catchments ...</b>	<b>52</b>
<b>Annex 2. Report on National ICP IM activities in Austria .....</b>	<b>57</b>
<b>Annex 3. Report on National ICP IM activities in Lithuania in 2015 .....</b>	<b>60</b>
<b>Annex 4. Russia: Effect of temperature and precipitation on the annual height increment of Scots pine on the Kandalaksha Gulf Coast and ICP IM site RUI6 .....</b>	<b>62</b>
<b>Annex 5. Report on National ICP IM activities in Sweden 2014–2015 ..</b>	<b>65</b>

## ABBREVIATIONS

<b>AMAP</b>	Arctic Monitoring and Assessment Programme
<b>ANC</b>	Acid neutralising capacity
<b>ALTER-Net</b>	A Long-Term Biodiversity, Ecosystem and Awareness Research Network
<b>CCE</b>	Coordination Center for Effects
<b>CL</b>	Critical Load
<b>CNTER</b>	Carbon-nitrogen interactions in forest ecosystems
<b>ECE</b>	Economic Commission for Europe
<b>EMEP</b>	Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
<b>EnvEurope</b>	EU LIFE project “Environmental quality and pressures assessment across Europe; the LTER network as an integrated and shared system for ecosystem monitoring”
<b>EU</b>	European Union
<b>EU LIFE</b>	EU’s financial instrument supporting environmental and nature conservation projects throughout the EU
<b>ExpeER</b>	Experimentation in Ecosystem Research
<b>Horizon 2020</b>	H2020, EU Research and Innovation programme
<b>ICP</b>	International Cooperative Programme
<b>ICP Forests</b>	International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests
<b>ICP IM</b>	International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems
<b>ICP Materials</b>	International Cooperative Programme on Effects on Materials
<b>ICP M&amp;M</b>	ICP Modelling and Mapping, International Cooperative Programme on Modelling and Mapping of Critical Loads and Levels and Air Pollution Effects, Risks and Trends
<b>ICP Waters</b>	International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes
<b>ICP Vegetation</b>	International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops
<b>ILTER</b>	International Long Term Ecological Research Network
<b>IM</b>	Integrated Monitoring
<b>JEG</b>	JEG DM, Joint Expert Group on Dynamic Modelling
<b>LRTAP Convention</b>	Convention on Long-range Transboundary Air Pollution
<b>LTER-Europe</b>	European Long-Term Ecosystem Research Network
<b>LTER-Network</b>	Long Term Ecological Research Network
<b>NFP</b>	National Focal Point
<b>TF</b>	Task Force
<b>Task Force on Health</b>	The Joint Task Force on the Health Aspects of Air Pollution
<b>UNECE</b>	United Nations Economic Commission for Europe
<b>WGE</b>	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](http://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-one sites from fourteen countries, two additional countries are re-joining the network. 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 tasks regarding international ICP IM data have been:

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

## Conclusions from international studies using ICP IM data

### Input-output and proton budgets, C/N interactions

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

The most recent results from ICP IM studies are available from the study of Vuorenmaa et al. (2016). Site-specific annual input-output budgets were calculated for sulphate ( $\text{SO}_4$ ) and total inorganic nitrogen ( $\text{TIN} = \text{NO}_3\text{-N} + \text{NH}_4\text{-N}$ ) and temporal trends were analysed for input (deposition) and output (runoff water) fluxes and net retention/net release of  $\text{SO}_4$  and TIN at 17 European ICP IM sites in 1990 to 2012. Large spatial variability in input and output fluxes of  $\text{SO}_4$  and TIN were observed between the sites, with the highest deposition and runoff water fluxes in South Scandinavia, Central and Eastern Europe and lowest fluxes at more remote sites in northern European regions. A significant decrease in total  $\text{SO}_4$  (wet + dry) deposition and bulk deposition of TIN was found at 90% and 65% of the sites, respectively. Output fluxes of  $\text{SO}_4$  decreased significantly at 60% of the sites, while TIN output fluxes showed mixed response with both decreasing (9 sites) and increasing (8 sites) trend slopes, but trends were rarely significant. Catchments retained  $\text{SO}_4$  in the early 1990s, but they shifted towards net loss in the late 1990s. This indicates that forest soils are now releasing former accumulated  $\text{SO}_4$ . TIN retention also showed a mixed response with increasing or declining retention rates, but generally TIN was strongly retained in the catchments not affected by natural disturbances. The long-term variation of net losses for  $\text{SO}_4$  was explained by changes in runoff and  $\text{SO}_4$  concentrations in deposition, while variation of TIN retention was dominantly explained by changes in TIN concentrations in runoff. Net losses of  $\text{SO}_4$  may lead to a slower recovery of surface waters than those predicted by the decrease in  $\text{SO}_4$  deposition. Continued enrichment of N in catchment soils poses a threat to terrestrial biodiversity and may ultimately lead to higher TIN runoff through N saturation or climate change. The results confirm the effects of emission reduction measures, but large uncertainties still remain regarding many regulating ecosystem processes.

Earlier results from ICP IM studies are summarized below.

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

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

of N deposition. When the deposition increases also N processes become increasingly important as net sources of acidity.

A critical deposition threshold of about 8–10 kg N ha<sup>-1</sup> yr<sup>-1</sup>, 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<sup>-1</sup> yr<sup>-1</sup>. When stratifying data based on C/N ratios less than or equal to 25 and greater than 25, highly significant relationships were observed between N input and nitrate leached (Dise et al. 1998, MacDonald et al. 2002, Gundersen et al. 2006). Such statistical relationships from intensively studied sites can be efficiently used in conjugation with regional monitoring data (e.g. ICP Forests and ICP Waters data) in order to link process level data with regional-scale questions.

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

Sulphur budgets calculations indicated a net release of S from many ICP IM sites, indicating that the soils are releasing previously accumulated S. Similar results have been obtained in other 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 & Kakebeeke 2004, Working Group on Effects 2004).

ICP IM contributed to an assessment report on reactive nitrogen (N<sub>r</sub>) of the WGE. This report was prepared for submission to the TF on Reactive Nitrogen and other bodies of the LRTAP Convention to show what relevant information has been collected by the ICP programmes under the aegis of the WGE to allow a better understanding of N<sub>r</sub> effects in the ECE region. The report contributed relevant information for the revision of the Gothenburg Protocol. A revised Gothenburg Protocol was successfully finalised in 2012.

It should also be 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. The



study of Vuorenmaa et al. (2016) referred to above, contained results also regarding temporal trends. The latest results are included in the present Annual Report (see Chapter 3).

Earlier work is summarized below.

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

Calculations on the trends of N and S compounds, base cations and hydrogen ions were made for 22 ICP IM sites with available data across Europe (Forsius et al. 2001). The site-specific trends were calculated for deposition and runoff water fluxes using monthly data and non-parametric methods. Statistically significant downward trends of  $\text{SO}_4$ ,  $\text{NO}_3$  and  $\text{NH}_4$  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  $\text{SO}_4$  and base cation trends in runoff waters were commonly observed at the ICP IM sites. At some sites in the Nordic countries decreasing  $\text{NO}_3$  and  $\text{H}^+$  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 15<sup>th</sup> Annual Report (Kleemola et al. 2006). Statistically significant decreases in  $\text{SO}_4$  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  $\text{NO}_3$  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](http://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 results 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 only a 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.

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

## Detected responses in biological data

The effect of pollutant deposition on natural vegetation, including both trees and understorey vegetation, is one of the central concerns in the impact assessment and prediction. The most recent ICP IM study on dose-response relationships was published by Dirnböck et al. (2014). This study utilized a new ICP IM database for biological data (see below) and focussed on effects on forest floor vegetation from elevated nitrogen deposition.

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

Previous work on biological data is summarized below.

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

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

As a separate exercise, the epiphytic lichen flora of 25 European ICP IM monitoring sites, all situated in areas remote from local air pollution sources, was statistically related to measured levels of SO<sub>2</sub> in air, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> in precipitation, annual bulk precipitation, and annual average temperature (van Herk et al. 2003, de Zwart et al. 2003). It was concluded that long distance transport of nitrogen air pollution is

important in determining the occurrence of acidophytic lichen species, and constitutes a threat to natural populations that is strongly underestimated so far.

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

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

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

Dynamic vegetation modelling at ICP IM sites has been initiated with contributions from ICP M&M and ICP Forests. First results have been reported by Holmberg and Dirnböck (2015). The VSD+ model was applied to simulate soil chemistry at ten sites in four countries (Austria, Italy, Poland and Finland). The next steps include application and calibration at further sites and, after the soil chemistry simulations are satisfactory at all sites, the vegetation responses will be estimated with the PROPS model. The impact on biodiversity is evaluated using the habitat suitability index HS (Posch et al. 2014). The latest results are included in the present Annual Report (see Chapter 2).

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

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). The main findings on heavy metals budgets and critical loads at ICP IM sites were presented in Bringmark (2011). Input/output budgets and catchment retention for Cd, Pb and Hg in the years 1997–2011 were determined for 14 ICP IM catchments across Europe (Bringmark et al. 2013). Litterfall plus throughfall was taken as a measure of the total deposition of Pb and Hg (wet + dry) on the basis of evidence suggesting that, for these metals, internal circulation is negligible. The same is not true for Cd. Excluding a few sites with high discharge, between 74 and 94 % of the input, Pb was retained within the catchments; significant Cd retention was also observed. High losses of Pb ( $>1.4 \text{ mg m}^{-2} \text{ yr}^{-1}$ ) and Cd ( $>0.15 \text{ mg m}^{-2} \text{ yr}^{-1}$ ) were observed in two mountainous Central European sites with high water discharge. All other sites had outputs below or equal to  $0.36$  and  $0.06 \text{ mg m}^{-2} \text{ yr}^{-1}$ , respectively, for the two metals. Almost complete retention of Hg, 86–99 % of input, was reported in the Swedish sites. These high levels of metal retention were maintained even in the face of recent dramatic reductions in pollutant loads.

In 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.

## 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. 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. 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](http://www.lter-europe.net)), and the related EU/H2020-infrastructure project eLTER.
- Cooperation with other external organisations and programmes, particularly the International Long Term Ecological Research network (ILTER, [www.ilternet.edu](http://www.ilternet.edu)).
- Participation in projects with a global change perspective.

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# 1 ICP IM activities, monitoring sites and available data

## 1.1

### Review of the ICP IM activities in 2015–2016

#### Meetings

- The Chairman Lars Lundin represented the ICP IM programme at the 31<sup>st</sup> ICP Forests Task Force meeting in Ljubljana, Slovenia, 20–22 May 2015.
- ICP IM Programme Manager Martin Forsius and Maria Holmberg participated in the eLTER H2020 kick-off meeting in Chania, Greece, 22–24 June 2015.
- Lars Lundin and Martin Forsius represented ICP IM in the First Joint session of the Steering Body to the EMEP and the Working Group on Effects in Geneva, Switzerland, 14–18 September 2015.
- Martin Forsius participated in the 23<sup>rd</sup> International Long-Term Ecological Research (ILTER) 2015 Annual Meeting in Rome and Science days in Central Italy, 23–30 September 2015.
- Jussi Vuorenmaa represented the ICP IM programme at the 31<sup>st</sup> Task Force meeting of ICP Waters in Switzerland, 5–9 October 2015.
- Jussi Vuorenmaa took part in the Acid Rain 2015 Conference in Rochester, NY, USA, 19–25 October 2015.
- Martin Forsius participated in the eLTER H2020 project Core Team Meeting in Paris, France, 1–2 December 2015.
- Martin Forsius took part in the Joint Workshop between AMAP and LRTAP bodies in Potsdam, Germany, 15–17 February 2016.
- Lars Lundin represented ICP IM in the Joint meeting of the Extended Bureaux of the EMEP Steering Body and the Working Group on Effects in Geneva, Switzerland, 14–17 March 2016.
- Filip Moldan represented ICP IM in the 26<sup>th</sup> CCE Workshop and 32<sup>nd</sup> Meeting of the ICP M&M Programme Task Force in Dessau, Germany, 19–22 April 2016.
- The twenty-fourth meeting of the Programme Task Force on ICP Integrated Monitoring was organized as joint 2016 Task Force Meeting of ICP Waters and ICP Integrated Monitoring in Asker, Norway from May 24 to May 26, 2016.

#### Projects, data issues

After December 1<sup>st</sup> 2015 the National Focal Points (NFPs) reported their 2014 results to the ICP IM Programme Centre. The Programme Centre carried out standard check-up of the results and incorporated them into the IM database.

#### Scientific work in priority topics

- The Programme Centre prepared the ICP IM contribution to the Joint Report 2015 of the ICPs, TF health and Joint Expert group on Dynamic Modelling for the WGE (ECE/EB.AIR/GE.1/2015/3–ECE/EB.AIR/WG.1/2015/3).
- Programme Centre finalized the contribution to the joint WGE trend report.
- Scientific paper: Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990–2012)

(Jussi Vuorenmaa et al.), in review (to be finalized according to review comments received)

- Report on dynamic responses of vegetation changes in relation to nitrogen; Included as a chapter in the present Annual Report: Dynamic vegetation modelling at ecosystem monitoring and research sites (Maria Holmberg, Thomas Dimböck, et al.) A scientific paper on this topic is planned for 2017.
- Report on long-term trends of S and N effects; Presented in this report as: Trend assessment for deposition and runoff water chemistry and climatic variables at ICP IM sites in 1990–2013 – Interim report (Jussi Vuorenmaa et al.).
- 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.

## 1.2

### Activities and tasks planned for 2017–2018

#### 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:

2016–2017: Report on assessing long-term trends in ecosystem effects of sulphur and nitrogen

2017: Report on mercury in the aquatic environment; Joint Report together with ICP Waters

2017: Scientific paper on dynamic response of vegetation changes in relation to nitrogen deposition

#### Other activities

- Maintenance and development of central ICP IM database at the Programme Centre
- Arrangement of the 25<sup>th</sup> Task Force meeting (2017)
- Preparation of the 26<sup>th</sup> ICP IM Annual Report (2017)
- 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](http://www.lter-europe.net)), and the EU/H2020 eLTER-project.
- Participation in the activities of other external organisations, particularly the International Long Term Ecological Research Network (ILTER, [www.ilternet.edu](http://www.ilternet.edu))



## Published reports and articles 2015–2016

### Evaluations of international ICP IM data and related publications

Kleemola, S. & Forsius, M. (Eds.) 2015. 24<sup>th</sup> Annual Report 2015. Convention on Long-range Transboundary Air Pollution, ICP Integrated Monitoring. Reports of Finnish Environment Institute 31/2015, Finnish Environment Institute, Helsinki. 58 p. <http://hdl.handle.net/10138/156295>

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

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#### 1.4

### Monitoring sites and data

The following countries have continued data submission to the ICP IM data base during the period 2011–2015: Austria, Belarus, the Czech Republic, Estonia, Finland, Germany, Ireland, Italy, Lithuania, Norway, the Russian Federation, Spain, Sweden, and Ukraine. Two additional countries are re-joining: Switzerland included a new site in 2016, description of the site included as an Annex to this report; Poland is preparing data and will soon include one or more sites to the network.

The number of sites with on-going data submission for at least part of the data years 2010–2014 is 41 from fourteen countries. Sites from Canada, Latvia and United Kingdom only contain older data.

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



Figure I.I. Geographical location of ICP IM sites.



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## 2 Dynamic vegetation modelling at ecosystem monitoring and research sites

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### 2.1

#### Introduction

One severe negative effect of elevated N deposition is the threat to biodiversity that results from the eutrophication of sensitive ecosystems. Accumulation of N from anthropogenic activity into natural ecosystems contributes also to the stress caused by climate warming. Observed effects include changes in species composition (Bobink et al. 2010) and changes in primary production (Stevens et al. 2015). Changes in biodiversity caused by drivers of environmental change, including anthropogenic nitrogen deposition, may in turn be a major factor determining how global environmental changes affect ecosystem stability (Hautier et al. 2015).

The monitoring and research activities at the sites of the ICP IM, ICP Forests programmes and LTER-Europe network produce high quality data that is valuable for identifying ecosystem responses. Our aim is to utilize the monitoring data from selected sites of these networks in a dynamic modelling study to evaluate future vegetation responses to continuing nitrogen deposition. This report summarizes the approach and the progress. The VSD+ model has been applied to simulate soil properties: soil solution pH, soil base saturation (BS) and soil organic carbon to nitrogen ratio (C:N). Draft VSD+ calibrations have been compiled at nearly 20 sites in eight countries (Austria, Germany, Italy, Norway, Poland, Serbia, Sweden and Finland). Successful calibration results are presented for 13 of the sites. The PROPS model has been tested in evaluating habitat suitability (HS) at Austrian sites. The next steps include improving the calibrations, applications at further sites and estimation of vegetation responses with PROPS. The final results of the project will be used for air pollution impact assessment work of the CLRTAP Working Group on Effects, and the work is also related to the EU/H2020 project eLTER.

### 2.2

#### Sites

A set of sites with data on soil properties and vegetation has been examined for inclusion in the dynamic modelling study. The sites hold comprehensive infrastructure, operated by national institutes that report to ICP IM or ICP Forests, providing long term intensive data and research on vegetation, element mass balances as well as studies on regulating processes. A number of the sites form part of the LTER-Europe network. The dynamic modelling of soil and vegetation can be carried out at sites

with available appropriate information on vegetation as well as on soil physical and chemical properties. Even if site-specific vegetation data is missing, some results can be obtained concerning future vegetation. Currently, 27 sites are intended for the modelling study (Table 2.1). The sites are located from south N41° to north N63° and from W09° to east E30°, including Mediterranean, Atlantic, temperate and boreal sites (Holmberg & Dirnböck 2015).

Table 2.1. List of sites intended for dynamic modelling study of soil chemistry and vegetation response.

Country	Site Code	Site	Latitude	Longitude	Data available for modelling S: soil V: vegetation	Model (draft) calibrations S: soil V: vegetation	Model (draft) simulations S: soil V: vegetation
Austria	AT01_I	Zöbelboden	N47°50'	E14°26'	S/V	S/V	S/V
	AT01_2	Zöbelboden	N47°50'	E14°26'	S/V	S/V	S/V
Belgium	BE001	Brasschaat	N51°18'	E04°31'	S		
Czech Republic	CZ01	Anenské Povodi	N49°35'	E15°05'	S		
	CZ02	Lysina	N50°03'	E12°40'	S		
Germany	DE01	Forellenbach	N48°56'	E13°25'	S/V	S	S
	DE02	Neuglobsow	N53°08'	E13°02'	S/V	S	S
	DE301	Lüss	N52°50'	E10°16'	S	S	S
	DE507	Monschau	N50°24'	E06°09'	S	S	S
Finland	FI01	Valkea-Kotinen	N61°14'	E25°03'	S/V	S	
	FI03	Hietajärvi	N63°09'	E30°40'	S/V	S	
Ireland	IE01	Brackloon Wood	N53°46'	W09°33'	S/V		
Italy	IT05	Selvapiana	N41°50'	E13°35'	S/V	S	
	IT07	Carrega	N44°43'	E10°12'	S/V	S	
	IT08	Brasimone	N44°06'	E11°07'	S/V	S	
	IT09	Monte Rufeno	N42°49'	E11°54'	S/V	S	
	IT10	Val Masino	N46°14'	E09°33'	S/V	S	
Norway	NO01	Birkenes	N58°23'	E08°15'	S/V	S	S
Poland	PL05	Puszcza Borecka	N54°07'	E23°02'	S/V	S	S
	SNP	Słowiński National Park	N54°07'	E23°02'	S/V		
	TNP	Tatrzański National Park	N49°10'	E19°55'	S/V		
Serbia	RS1	Kopanoik	N43°17'	E20°48'	S	S	S
	RS2	Crni Vrh	N44°07'	E21°58'	S	S	S
Sweden	SE04	Gårdsjön	N58°03'	E12°01'	S/V	S	
	SE14	Aneboda	N57°07'	E14°32'	S/V	S	
	SE15	Kindla	N59°45'	E14°54'	S/V	S	
	SE16	Gammtratten	N63°51'	E18°06'	S/V	S	

## Methods and models

An elaborate chain of models has been applied to study the impact of air pollution on habitat suitability (Fig. 2.1). An extension of the Very Simple Dynamic (VSD) model (Reinds et al. 2009), called VSD+ (CCE 2015, Bonten et al. 2016) was used to calculate the impact of historic and future air-borne deposition of air pollutants (S, N) and base cations (Ca, Mg, K). VSD+ includes the formulations of the RothC-26.3 model (Coleman & Jenkins 2014) to calculate changes in soil organic C. The mineralisation and immobilisation of N is calculated from the turnover of the C pools. Nitrification and denitrification depend on the total amounts of  $\text{NH}_4$  and  $\text{NO}_3$  available after deposition, uptake and mineralisation, and their rates are further modified by site-specific climate conditions (Bonten et al. 2016).

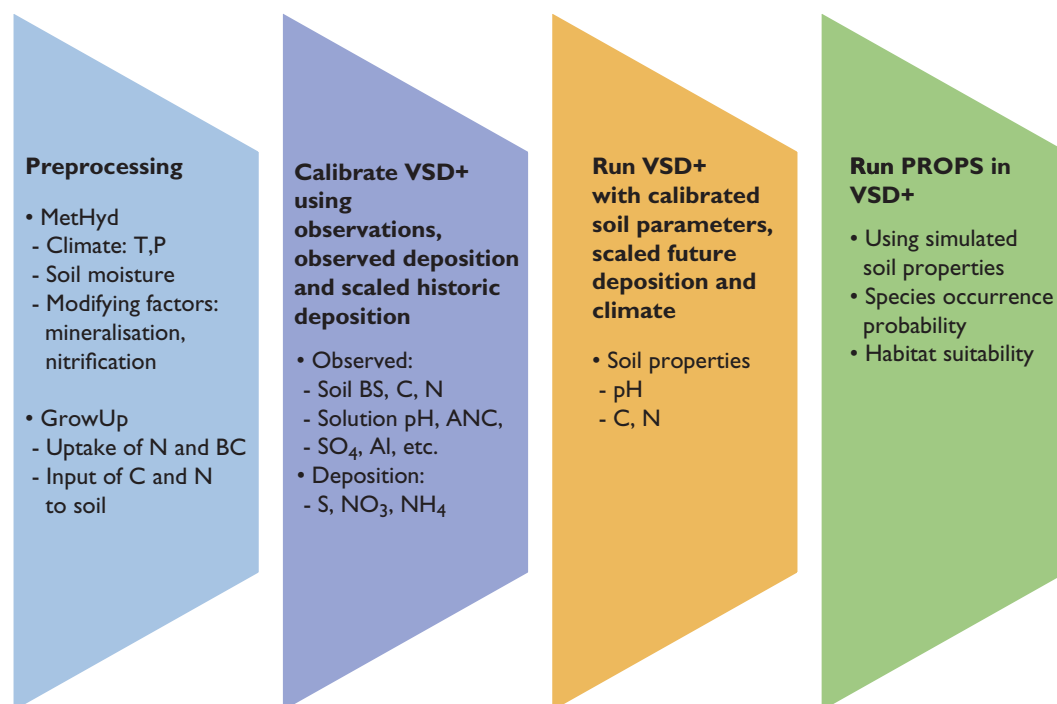


Figure 2.1 Schematic view of work flows in the application of VSD+ model chain including PROPS plant response functions to study impact of air pollution on habitat suitability.

VSD+ is integrally combined with the PROPS model (Reinds et al. 2015), which enables the assessment of changes in plant species diversity in response to changes in air pollution and climate. The PROPS model estimates the occurrence probability of plant species as a function of soil chemistry and climate, using a logistic regression technique. In PROPS, response curves have been derived for about 4 000 European plant species, based on a large data set with observed plant species occurrences and abiotic variables. A comparison for about 40 species with results from Finland (Heikkinen & Mäkipää 2010) showed similar responses (Reinds et al. 2015). PROPS also computes an aggregate indicator for species occurrence in a habitat. The habitat suitability index (HSI) is defined as the arithmetic mean of the normalised probabilities of occurrence of the species of interest (Posch et al. 2015, Reinds et al. 2015).

$$HS = \frac{1}{K} \sum_{k=1}^K \frac{p_k}{p_{k, \max}}$$

$K$	number of species
$p_k$	occurrence probability of species $k$
$p_{k, \max}$	the maximum occurrence probability of species $k$

In this study, the application of VSD+ was set up by first applying the preprocessing models MetHyd and GrowUp (Bonten et al. 2016). MetHyd is used for calculating daily evapotranspiration, soil moisture and percolation, as well as parameters related to (de)nitrification and mineralization. GrowUp was used for producing time series of nutrient uptake and litterfall as input to VSD+ (Bonten et al. 2016).

Observations of soil BS, C:N and soil solution pH were the key variables used for calibration of the VSD+ soil model. Additional observations are available at some sites, e.g., observations of soil solution concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Al}^{3+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  that can be used to further guide the application. The soil model calibration was considered successful and applicable as a starting point for the vegetation simulations if the model performed acceptably for the key variables BS, C:N and pH. The chosen performance criterion was related to the normalized mean error ( $-0.1 < \text{NME} < 0.1$ ).

The PROPS model was not yet applied for all the sites, only tested for Austrian sites, to calculate the HS index for sites AT01\_1 and AT01\_2.

Estimated time series of deposition of N and S are used as input to the VSD+ model. The historic deposition is based on older EMEP-model versions (Schöpp et al. 2003), while the deposition values for 2005, 2010, 2020 and 2030 are based on the latest EMEP model version (Simpson et al. 2012), using the current legislation scenario (CLE) with revised Gothenburg Protocol emissions.

## 2.4

### Modelled and observed soil chemistry

The VSD+ model was calibrated individually for each of the sites. The calibration results are presented for 13 sites. The modelled values slightly underestimated the observed present day conditions, with largest errors for pH ( $\text{NME} = -0.04$ , Fig. 2.2), while smaller errors were obtained for BS ( $\text{NME} = -0.01$ , Fig. 2.3) and C:N ( $\text{NME} = -0.02$ , Fig. 2.4). The errors were acceptable for a successful calibration and the soil model can be applied to project future soil conditions, as a basis for a later application of the vegetation model PROPS.

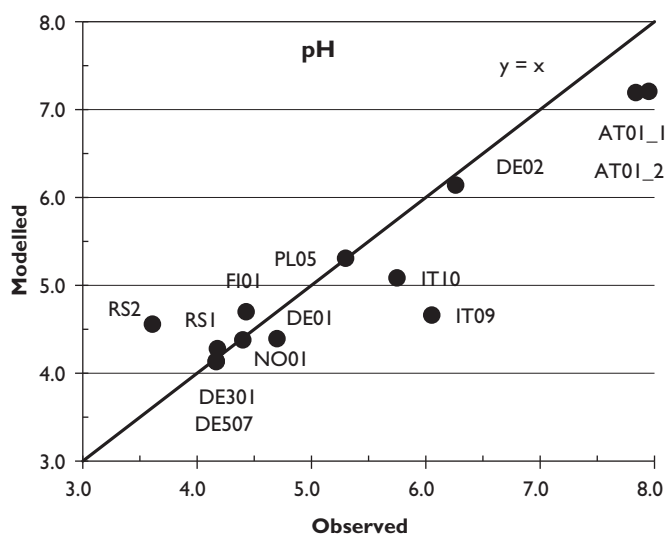


Figure 2.2. Comparison of modelled to observed values of soil pH at 13 sites. NME = -0.04.

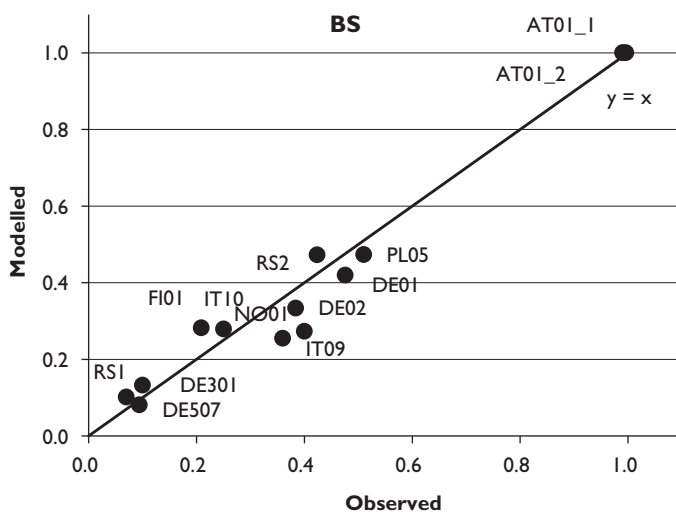


Figure 2.3. Comparison of modelled to observed values of soil base saturation at 13 sites. NME = -0.01.

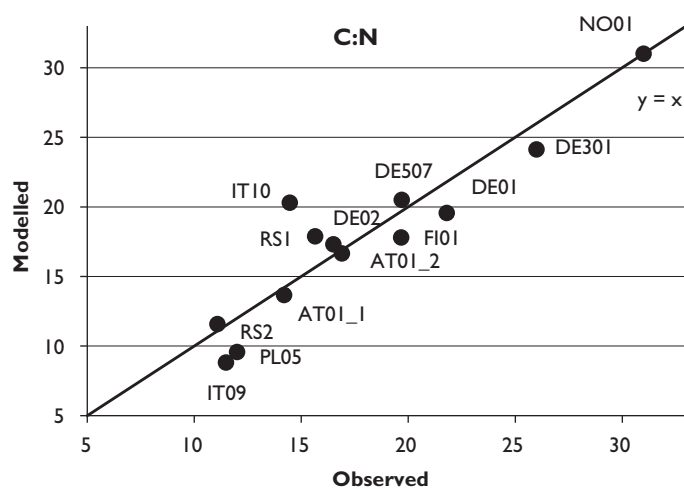


Figure 2.4. Comparison of modelled to observed values of soil C to N ratio at 13 sites. NME = -0.02.

## Vegetation modelling examples

Since additional data has been introduced into PROPS (Reinds et al. 2015), we were able to test its performance at 21 Austrian sites. Two of these sites will be used in this project. Vegetation observations were used to calculate HSI from the characteristic plant species of these forest types and use it as a comparison with modelled values. These measured HSI values were relatively low ( $<0.5$ ) because some characteristic plant species did occur with a low frequency owing to variations in site conditions but also forest management. Regression of modelled and observed values had an  $R^2=0.27$  ( $p=0.021$ ). Hence, we could predict HSI with PROPS but the achieved accuracy was not overwhelming. We did also model the sites with BERN (Shlütow et al. 2010), another plant response model mostly applied in Germany, and found that the results are very much in line with those from PROPS ( $R^2=0.65$ ). The low predictive accuracy is not specific to either PROPS or BERN but rather lies in the impact of management, which is not implemented in these types of models. We conclude from these results that by applying PROPS (or BERN), we are able to find effects of N deposition on the suitability of forest habitats to host their characteristic plant species. We also think that predictive accuracy will improve by using sites with low management impact, which is the case in most IM sites.

## Status and future tasks

Draft VSD+ calibrations are available for nearly 20 sites and the results for 13 sites were presented here. Future tasks are to improve the VSD+ calibrations, apply the model at the rest of the sites and test the PROPS model at further sites. The extension of PROPS with new data was a significant step forward because it allows us to model HSI for most of our sites in a reliable way. There may still be limitations with the northern- and southernmost sites because not many vegetation records from these regions were used in PROPS. It will be essential for the success of the project to include a wide variation of sites with different N deposition and N critical loads.

## Acknowledgements

We acknowledge the input of the model developers Luc Bonten, Janet Mol, Maximilian Posch, Gert Jan Reinds, Wieger Wamelink, as well as the collaboration in model application and data provision by several colleagues representing ICP IM and ICP Forests sites including Julian Aherne, Kari Austnes, Jelena Beloica, Burkhard Beudert, Nicholas Clarke, Natalie Cools, Alessandra de Marco, Francesca Fornasier, Martin Forsius, Martyn Futter, Ulf Grandin, Juha Heikkinen, Sirpa Kleemola, Pavel Krám, Antti-Jussi Lindroos, Lars Lundin, Tiina Nieminen, Jørn-Frode Nordbakken, Tomasz Pecka, Maija Salemaa, Thomas Scheuschner, Hubert Schulte-Bisping, Volkmar Timmermann, Salar Valinia, Milán Váña, Jussi Vuorenmaa.

We acknowledge the support of ICP IM and our institutes and those of our collaborators as well as support of the eLTER project (EU/H2020 grant agreement No. 654359).



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### 3 Trend assessments for deposition and runoff water chemistry concentrations and fluxes and climatic variables at ICP Integrated Monitoring sites in 1990–2013

#### Interim report

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## Introduction

Detrimental effects of transboundary air pollution led to international agreements to reduce emissions of sulphur and nitrogen in Europe and North America. The protocols of the United Nations Economic Commission for Europe's Convention on Long-Range Transboundary Air Pollution (UNECE CLRTAP) and legislation of the European Union have been key international instruments causing this positive development. It is essential that empirical scientific evidence based on environmental monitoring programmes is available for assessing and documenting the success and ecosystem benefits of costly international emission reduction policy. Acidification of sensitive lakes and rivers is still an environmental concern despite reduced emissions of sulphur and nitrogen (e.g. Garmo et al. 2014), and changing climate may have large impacts on water chemistry and freshwater biology (e.g. Wright & Jenkins 2001, Skjelkvåle et al. 2003, Wright et al. 2006, Adrian et al. 2009, Shimoda et al. 2011). Moreover, several studies have demonstrated the complexity of recovery processes from acidification and interactions within and between the aquatic and terrestrial ecosystems and atmosphere (e.g. Rask et al. 2014). Sustained accumulation of deposited inorganic N poses also a threat to ecosystems through nutrient enrichment and nutrient imbalance (Bergström et al. 2005, Bergström & Jansson 2006, Stevens et al. 2011, Lepori & Keck 2012). It poses also a threat to biodiversity as a consequence of the eutrophication of sensitive ecosystems, shown by results from both ICP Integrated Monitoring and ICP Forests sites (Dirnböck et al. 2014) and other studies (Sala et al. 2000, MEA 2005, Bobbink et al. 2010, Bleeker et al. 2011). Therefore, the importance of integrated long-term monitoring approach including physical, chemical and biological variables for detecting a variety of impacts of changing environmental conditions on ecosystems and long-term changes is clearly needed.

The multidisciplinary International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) under the LRTAP Convention quantifies air pollution effects on the environment through monitoring, modelling and scientific review, using data from catchments/plots located in predominantly unmanaged or semi-natural forested areas across Europe with different deposition, climate and acidification and eutrophication potential (Forsius et al. 2001, Forsius et al. 2005, Bringmark et al. 2013, Holmberg et al. 2013). ICP IM programme has a cause-effect approach and it provides a valuable means to study the effects of air pollution and climate change in catchments.

As monitoring and evaluation of long-term changes of air pollution effects on ecosystems are one of the main objectives of the ICP IM programme, it was agreed at the ICP IM Task Force meeting in 2007 that trend analysis of air pollution compartments should be carried out regularly. The most recent extensive trend assessment for ICP IM sites was presented for the period 1993–2006 in the 18<sup>th</sup> Annual Report, including 34 sites from 13 countries (Vuorenmaa et al. 2009). In addition, trends for S and N fluxes were presented for selected 17 IM sites from nine countries in 1990–2012 (Vuorenmaa et al. 2014, 2016). The 23<sup>rd</sup> ICP IM Task Force discussed the collaboration with ICP Waters, with interest to harmonize IM trend assessments with ICP Waters and possibilities of aiming at common trend reporting in the future. With respect to trends in surface water chemistry, the recent ICP Waters trend reports were focused on changes i) in 1990–2008 with a comparison of trend periods 1990–1999 and 1999–2008 (Garmo et al. 2011) and ii) in 2000–2011 with a prognosis for water chemical status and expected recovery in 2020 (Garmo et al. 2015). For method validation for the prognosis of future water chemistry, data from ICP IM were used.

Changes in emission reductions and emission reduction responses on deposition and surface water chemistry were more pronounced in the 1990s compared to the

2000s. Reductions for  $\text{SO}_x$ ,  $\text{NO}_x$  and  $\text{NH}_3$  emissions in Europe were larger in the 1990s (1990–2000) than in the 2000s (2000–2013) (Fig. 3.1), and a similar pattern with more gradual change during the 2000s was seen in S and N deposition in Europe (Aas & Vet 2011). Correspondingly, the decrease of  $\text{SO}_4$  concentrations in acid-sensitive surface waters was clearly stronger in the 1990s than in the 2000s, and also trends in concentrations for other indicators of chemical recovery from acidification tended to be less pronounced during the 2000s, suggesting that the rate of improvement of water quality has slowed (Garmo et al. 2014). Studies of input-out budgets for S and N at ICP IM sites have shown a net loss (output > input) of  $\text{SO}_4$  from internal S soil sources during the 2000s, indicating that forest soils are now releasing stored airborne  $\text{SO}_4$  that had accumulated in the past, whereas deposited inorganic N is still strongly retained in unmanaged catchments (Vuorenmaa et al. 2014, 2016). Many of these S and N retention processes are sensitive to changes in climatic variables, and would therefore be affected by climate change. Evidently, long-term assessment of air pollution effects on ecosystems including both trends in input and output fluxes, and changes in climatic variables, gives important information for the identification of emission reduction responses, changing climate and interactions between different drivers in European forested catchments in the course of recent two decades. This interim report presents the first results of the new trend assessment for deposition and runoff water chemistry concentrations and fluxes and climatic variables at ICP Integrated Monitoring sites from 1990 through 2013.

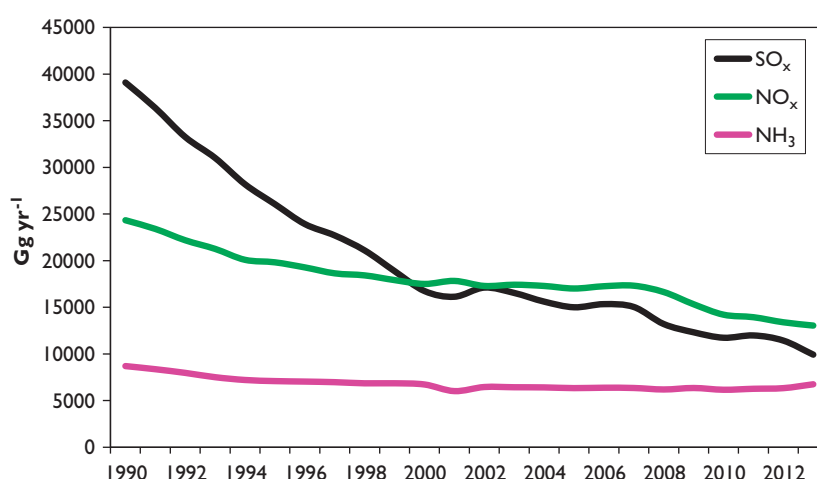


Figure 3.1. Anthropogenic S and N emissions ( $\text{Gg yr}^{-1}$ ) in Europe in 1990–2013 (www.emep.int).

### 3.2

## Material and methods

### Study sites

Long-term trends for compartments of air pollution effects and climatic variables were evaluated for a selection of 25 IM sites (AT01, BY02, CZ01, CZ02, DE01, DE02, EE01, EE02, FI01, FI03, IT01, IT03, IT07, IT09, LT01, LT03, LV01, LV02, NO01, NO02, NO02, SE04, SE14, SE15 and SE16) from 11 countries in 1990–2013. The selection of the sites was guided by the availability of deposition (bulk/wet and throughfall) data

and surface water chemistry and runoff volume data in the ICP IM database. For the location of the studied IM sites see Fig.1.1 in Chapter 1.

## Concentrations and fluxes in deposition and runoff water chemistry

Monthly concentrations and fluxes for bulk/wet deposition (PC), throughfall deposition (TF) and runoff water (RW) or soil water (SW, if no RW data were available) were used in the trend assessment for the individual ICP IM sites. Trends were evaluated for monthly based concentrations ( $\mu\text{eq l}^{-1}$ ) and fluxes ( $\text{meq m}^{-2}$ ) of non-marine (x denotes non-marine fraction) sulphate ( $\text{xSO}_4$ ) and base cations ( $\text{xBC}=\text{xCa} + \text{xMg}$ ), hydrogen ion ( $\text{H}^+$ ), nitrate ( $\text{NO}_3\text{-N}$ ), ammonium ( $\text{NH}_4\text{-N}$ ) and ANC (Acid Neutralising Capacity). ANC was calculated as  $\Sigma$  (base cations) –  $\Sigma$  (strong acid ions) equal to  $\Sigma(\text{Ca} + \text{Mg} + \text{Na} + \text{K}) - \Sigma(\text{SO}_4 + \text{NO}_3 + \text{Cl})$ .

Bulk deposition samples were collected in an open area, and represent largely wet deposition. As IM sites are almost totally forested catchments, dry deposition (gases and particles filtered by the canopy) highly contributes to the total deposition to the forest floor. Because precipitation under the forest canopy (wash-off of dry deposition and leachates produced by the canopy to the forest floor) differs in quality and quantity from that of precipitation collected in an open area, deposition trends under the canopy (throughfall) were thus analysed. Monthly deposition and runoff/soil water fluxes were calculated as the product of the respective ion concentrations with monthly precipitation sums and mean monthly runoff/soil water flux values, respectively.

## Climatic variables

Precipitation amount, runoff water volume and air temperature are regularly monitored at ICP IM sites, and monthly-based long-term trends for climatic variables were evaluated using monthly sum of precipitation ( $\text{mm month}^{-1}$ ), mean monthly runoff ( $\text{mm month}^{-1}$ ) and mean monthly air temperature ( $^{\circ}\text{C}$ ).

## Statistics

For detecting long-term monotonic trends in chemical concentrations and fluxes and climatic variables for each of the study sites, the non-parametric Mann-Kendall test (MKT) (Hirsch et al. 1982) was applied to monthly data. MKT is not particularly sensitive to missing data and outliers, and requires no assumption of normality. The magnitude of trend slope was estimated by the Theil-Sen slope estimation method (Sen 1968), and was expressed as  $\mu\text{eq l}^{-1} \text{ yr}^{-1}$  for chemical concentrations,  $\text{meq m}^{-2} \text{ yr}^{-1}$  for fluxes,  $\text{mm yr}^{-1}$  for precipitation and runoff and  $^{\circ}\text{C yr}^{-1}$  for air temperature. A statistical significance threshold of  $p < 0.05$  was applied to the trend analysis. For climatic parameters, a p value of  $< 0.1$  was used to indicate weak and not statistically significant trend, which can, however, be considered to show an increasing/decreasing tendency in the record.

## Results and discussion

### Deposition

According to the previous assessments at IM sites across Europe, the deposition of  $\text{SO}_4$  and inorganic N showed large spatial variability, with the highest values at IM sites in Central and Eastern Europe and lowest values at more remote IM sites in northern regions (e.g. Vuorenmaa et al. 2014, 2016). These numbers reflect well-known emission and deposition gradients of air pollutants (Lövgren et al. 2004, Waldner et al. 2014). Central and Eastern Europe were historically large sources of emission, and thus sites in the region (e.g. CZ01, CZ02, LT03, DE01, LV01, AT01) received the highest anthropogenic  $\text{SO}_4$  and/or inorganic N deposition. The sites in southern Scandinavia (NO01, SE04) were also exposed to high  $\text{SO}_4$  and inorganic N deposition due to the elevated long-range transport of these air pollutants (e.g. Vuorenmaa et al. 2014, 2016).

Successful emission reduction measures in Europe over the past 30 years have led to declining deposition of air pollutants in countries as shown at IM sites. The emission control programmes have been most successful for sulphur, and both bulk deposition (i.e. largely wet) and throughfall deposition (surrogate for dry deposition) of  $\text{SO}_4$  decreased significantly in all studied catchments between 1990 and 2013. The study sites that have been exposed to the highest  $\text{SO}_4$  deposition in the 1990s showed strongest reductions in deposition (Table 3.1.). Dry deposition of  $\text{SO}_4$  decreased more than bulk (wet) deposition ( $\Delta\text{Throughfall} > \Delta\text{Bulk deposition}$ ) (Tables 3.1. and 3.2.), which is in agreement with previous studies for a number of European forested catchments (Prechtel et al. 2001, Waldner et al. 2014).  $\text{SO}_4$  concentrations in throughfall are influenced by interception deposition where the relative decrease has been even more pronounced, because improved emission control techniques and fuel-switching away from high S-containing solid and liquid fuels to low S fuels have markedly reduced S-containing gases and particles in emissions and ambient air concentrations in Europe (Amann et al. 2013).

Along with decreased S emissions and  $\text{SO}_4$  deposition, base cation deposition ( $\text{Ca} + \text{Mg}$ ) has also decreased at majority of the sites, but less than  $\text{SO}_4$  in general. This has resulted in increase in acid neutralizing capacity (ANC) in precipitation, being significant at more than 60% of the sites both in bulk and throughfall deposition. Along with decreased  $\text{SO}_4$  concentrations and increased ANC in precipitation, hydrogen ion ( $\text{H}^+$ ) concentration i.e. acidity of precipitation decreased (increase of pH) in bulk and throughfall deposition as well, being significant in ca 80% of the sites (Tables 3.1. and 3.2.).

European nitrogen emissions have also decreased which have resulted in decrease in inorganic N deposition at majority of IM sites between 1990 and 2013. The IM sites showed dominantly negative trend slopes in  $\text{NO}_3$  and  $\text{NH}_4$  concentrations in bulk/wet deposition (at more than 90% of the sites), and decrease in  $\text{NO}_3$  and  $\text{NH}_4$  concentrations was significant at 76% and 68% of the sites, respectively. The corresponding records for deposition fluxes showed significant decrease in  $\text{NO}_3$  and  $\text{NH}_4$  at ca 40% of the sites. Only one site (NO02) showed significant increase in concentration and bulk deposition of  $\text{NH}_4$ . Long-term trends in precipitation amounts in 1990–2013 showed dominantly increasing trend slopes (18 out of the 22 sites) (Table 3.3.), but annual trends were rarely significant. The short and long-term variations in precipitation may mask long-term trends caused by N deposition (Wright et al. 2001).

Inorganic N concentrations in throughfall also showed predominantly decreasing trend slopes (ca 80–90% of the sites) and decrease in  $\text{NO}_3$  and  $\text{NH}_4$  concentrations was significant at ca 50% of the sites.  $\text{NO}_3$  and  $\text{NH}_4$  deposition in throughfall decreased



Table 3.1. Long-term trends (1990–2013) for monthly bulk/wet deposition chemistry concentrations and fluxes in studied IM catchments. For the annual change, a significant trend ( $p < 0.05$ , Mann-Kendall test, Sen's slope) is in bold. Site-specific annual changes and their mean and median (Md.) values for deposition concentrations and fluxes are given in  $\mu\text{eq l}^{-1} \text{yr}^{-1}$  and  $\text{meq m}^{-2} \text{yr}^{-1}$ , respectively. Data not available (n.d.). The IM sites are grouped into three different geographical regions reflecting gradients in long-range transport and deposition of pollutants and climate (NoC=Nordic Countries, NEE=North Eastern Europe, CE=Central Europe).

Site	Region	Data	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>	ANC	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>
			Concentration, $\mu\text{eq l}^{-1} \text{yr}^{-1}$						Flux, $\text{meq m}^{-2} \text{yr}^{-1}$				
FI01	NoC	1990–2013	<b>-1.05</b>	<b>-0.11</b>	<b>-0.19</b>	<b>-0.23</b>	<b>-0.77</b>	<b>1.00</b>	<b>-0.04</b>	<b>-0.01</b>	<b>-0.01</b>	<b>-0.01</b>	<b>-0.04</b>
FI03	NoC	1990–2013	<b>-0.82</b>	-0.06	<b>-0.19</b>	<b>-0.15</b>	<b>-0.77</b>	<b>0.92</b>	<b>-0.04</b>	-0.00	-0.01	-0.00	<b>-0.03</b>
SE04	NoC	1990–2013	<b>-1.47</b>	0.04	<b>-0.59</b>	<b>-0.60</b>	<b>-1.24</b>	<b>2.18</b>	<b>-0.12</b>	0.00	<b>-0.04</b>	<b>-0.05</b>	<b>-0.10</b>
SE14	NoC	1996–2013	<b>-0.97</b>	0.01	<b>-0.31</b>	<b>-0.50</b>	<b>-0.95</b>	<b>1.64</b>	<b>-0.07</b>	-0.00	-0.03	<b>-0.04</b>	<b>-0.06</b>
SE15	NoC	1996–2013	<b>-0.97</b>	-0.03	<b>-0.49</b>	<b>-0.57</b>	<b>-0.73</b>	<b>1.43</b>	<b>-0.06</b>	0.00	<b>-0.03</b>	<b>-0.04</b>	<b>-0.05</b>
SE16	NoC	1999–2013	<b>-0.49</b>	0.13	-0.18	-0.24	<b>-0.55</b>	<b>1.07</b>	<b>-0.03</b>	0.00	-0.02	-0.01	<b>-0.04</b>
NO01	NoC	1990–2013	<b>-1.27</b>	<b>0.09</b>	<b>-0.36</b>	<b>-0.36</b>	<b>-1.29</b>	<b>1.06</b>	<b>-0.11</b>	<b>0.01</b>	-0.02	-0.02	<b>-0.12</b>
NO02	NoC	1990–2013	<b>-0.15</b>	0.00	-0.00	<b>0.11</b>	<b>-0.19</b>	0.18	<b>-0.01</b>	0.00	-0.00	<b>0.01</b>	<b>-0.02</b>
NO03	NoC	1998–2013	<b>-0.70</b>	0.01	0.24	0.16	<b>-0.61</b>	0.59	<b>-0.03</b>	0.00	0.01	0.01	<b>-0.03</b>
EE01	NEE	1994–2013	<b>-1.87</b>	0.80	<b>-0.48</b>	<b>-0.46</b>	0.04	<b>4.78</b>	<b>-0.03</b>	<b>0.07</b>	0.02	0.01	0.01
EE02	NEE	1994–2013	<b>-2.11</b>	<b>-2.31</b>	-0.14	-0.24	-0.01	-0.35	<b>-0.08</b>	-0.06	0.01	0.00	0.00
LV01	NEE	1994–2009	<b>-2.30</b>	-0.01	-0.97	<b>-3.03</b>	<b>-0.27</b>	<b>3.29</b>	<b>-0.08</b>	0.02	-0.04	<b>-0.14</b>	<b>-0.01</b>
LV02	NEE	1994–2009	<b>-2.09</b>	-0.15	<b>-0.89</b>	<b>-2.14</b>	<b>-0.15</b>	<b>4.45</b>	<b>-0.10</b>	0.00	<b>-0.04</b>	-0.08	<b>-0.01</b>
LT01	NEE	1993–2013	<b>-2.70</b>	n.d.	<b>-0.65</b>	<b>-1.29</b>	-0.13	n.d.	<b>-0.13</b>	n.d.	<b>-0.04</b>	<b>-0.07</b>	-0.01
LT03	NEE	1995–2013	<b>-2.64</b>	n.d.	<b>-1.10</b>	-1.00	<b>0.31</b>	n.d.	<b>-0.15</b>	n.d.	<b>-0.04</b>	-0.04	<b>0.02</b>
BY02	NEE	1990–2013	<b>-1.87</b>	0.15	-0.07	<b>-0.82</b>	<b>-0.52</b>	1.37	<b>-0.11</b>	0.00	-0.01	-0.05	<b>-0.02</b>
DE01	CE	1991–2013	<b>-1.28</b>	<b>-0.57</b>	<b>-0.43</b>	<b>-0.50</b>	<b>-0.82</b>	<b>1.59</b>	<b>-0.10</b>	<b>-0.05</b>	<b>-0.03</b>	<b>-0.04</b>	<b>-0.07</b>
DE02	CE	1998–2013	<b>-1.59</b>	-0.50	<b>-0.62</b>	-0.68	<b>-0.78</b>	<b>1.19</b>	<b>-0.04</b>	-0.01	-0.00	-0.02	<b>-0.02</b>
CZ01	CE	1990–2013	<b>-2.56</b>	0.05	<b>-0.92</b>	<b>-1.30</b>	<b>-1.39</b>	<b>3.83</b>	<b>-0.11</b>	0.00	<b>-0.04</b>	<b>-0.05</b>	<b>-0.06</b>
CZ02	CE	1990–2013	<b>-2.22</b>	<b>-0.44</b>	<b>-0.90</b>	<b>-1.04</b>	<b>-1.22</b>	<b>2.77</b>	<b>-0.14</b>	<b>-0.03</b>	<b>-0.05</b>	<b>-0.07</b>	<b>-0.08</b>
AT01	CE	1993–2013	<b>-0.88</b>	<b>-0.70</b>	<b>-0.23</b>	-0.06	<b>-0.54</b>	0.39	<b>-0.09</b>	<b>-0.08</b>	-0.01	-0.00	<b>-0.05</b>
IT01	CE	1993–2013	<b>-0.86</b>	-0.16	<b>-0.57</b>	<b>-0.29</b>	<b>-0.31</b>	<b>1.78</b>	<b>-0.06</b>	-0.00	<b>-0.03</b>	<b>-0.01</b>	<b>-0.02</b>
IT03	CE	1997–2013	<b>-1.09</b>	<b>-2.75</b>	<b>-0.51</b>	<b>-0.53</b>	<b>0.11</b>	<b>-1.42</b>	<b>-0.05</b>	<b>-0.15</b>	<b>-0.02</b>	-0.01	<b>0.01</b>
IT07	CE	1997–2013	<b>-2.52</b>	<b>-1.79</b>	<b>-1.10</b>	<b>-1.48</b>	<b>-0.04</b>	1.22	<b>-0.13</b>	<b>-0.09</b>	-0.04	-0.05	<b>-0.00</b>
IT09	CE	1997–2013	<b>-1.46</b>	<b>-1.63</b>	<b>-0.48</b>	-0.30	0.01	-0.68	<b>-0.11</b>	<b>-0.13</b>	-0.02	-0.01	0.00
Mean			-1.52	-0.43	-0.49	-0.70	-0.51	1.49	-0.08	-0.02	-0.02	-0.03	-0.03
Md.			-1.46	-0.06	-0.48	-0.50	-0.54	1.22	-0.08	0.00	-0.02	-0.02	-0.02

at 76% and 52% of the sites, but the decrease was significant only at 36% and 24% of the sites, respectively. Three sites (DE01, EE01, NO02) showed significant increases in inorganic N concentrations and fluxes in throughfall (Table 3.2.). Biological processes such as N uptake by plant tissue and through stomata and other complex canopy interactions control inorganic N fluxes in throughfall (Draaijers and Erisman 1995), and thus long-term trends can be largely controlled by factors other than direct deposition effect.

Emissions of nitrogen have decreased less than those of sulphur, and decrease in inorganic N deposition has generally been smaller than that of SO<sub>4</sub> deposition, and

Table 3.2. Long-term trends (1990–2013) for monthly throughfall deposition chemistry concentrations and fluxes in studied IM catchments. For the annual change, a significant trend ( $p < 0.05$ , Mann-Kendall test, Sen's slope) is in bold. Site-specific annual changes and their mean and median (Md.) values for deposition concentrations and fluxes are given in  $\mu\text{eq l}^{-1} \text{yr}^{-1}$  and  $\text{meq m}^{-2} \text{yr}^{-1}$ , respectively (Forest plot: PA=*Picea abies*, PS=*Pinus sylvestris*, FS=*Fagus sylvatica*, QC=*Quercus cerris*). Data not available (n.d.). The IM sites are grouped into three different geographical regions reflecting gradients in long-range transport and deposition of pollutants and climate (NoC=Nordic Countries, NEE=North Eastern Europe, CE=Central Europe).

Site	Region	Forest plot	Data	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>	ANC	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>
				Concentration, $\mu\text{eq l}^{-1} \text{yr}^{-1}$						Flux, $\text{meq m}^{-2} \text{yr}^{-1}$				
FI01	NoC	PA	1990–2013	<b>-2.56</b>	0.32	-0.17	-0.08	<b>-0.92</b>	<b>4.96</b>	<b>-0.10</b>	-0.01	-0.01	-0.00	<b>-0.03</b>
FI03	NoC	PS	1990–2013	<b>-1.27</b>	0.08	<b>-0.20</b>	<b>-0.18</b>	<b>-0.92</b>	<b>2.20</b>	<b>-0.05</b>	0.01	-0.00	-0.00	<b>-0.04</b>
SE04	NoC	PA	1990–2013	<b>-4.65</b>	<b>-1.64</b>	<b>-1.76</b>	<b>-0.92</b>	<b>-2.22</b>	<b>5.02</b>	<b>-0.25</b>	<b>-0.06</b>	<b>-0.07</b>	<b>-0.03</b>	<b>-0.11</b>
SE14	NoC	PA	1996–2013	<b>-2.35</b>	<b>-1.23</b>	0.00	0.14	<b>-0.67</b>	0.07	<b>-0.08</b>	<b>-0.04</b>	0.00	0.01	<b>-0.03</b>
SE15	NoC	PA	1996–2013	<b>-1.77</b>	0.08	<b>-0.37</b>	-0.15	<b>-0.53</b>	<b>2.51</b>	<b>-0.08</b>	0.00	<b>-0.02</b>	-0.01	<b>-0.02</b>
SE16	NoC	PA	1999–2013	<b>-0.46</b>	0.19	-0.19	-0.03	<b>-0.33</b>	<b>0.88</b>	<b>-0.03</b>	0.00	<b>-0.01</b>	-0.00	<b>-0.02</b>
NO01	NoC	PA	1990–2013	<b>-2.54</b>	<b>-0.61</b>	<b>-0.71</b>	<b>-0.36</b>	<b>-1.88</b>	<b>3.46</b>	<b>-0.19</b>	<b>-0.04</b>	<b>-0.05</b>	<b>-0.03</b>	<b>-0.15</b>
NO02	NoC	PS	1990–2011	<b>-0.29</b>	<b>0.00</b>	<b>-0.10</b>	<b>0.15</b>	<b>-0.47</b>	0.21	<b>-0.02</b>	0.00	<b>-0.01</b>	<b>0.01</b>	<b>-0.03</b>
NO03	NoC	n.d.												
EE01	NEE	PS	1994–2013	<b>-3.82</b>	<b>6.74</b>	-0.29	<b>1.75</b>	-0.05	<b>17.3</b>	<b>-0.06</b>	<b>0.19</b>	0.01	<b>0.05</b>	0.00
EE02	NEE	PS	1994–2013	<b>-3.66</b>	<b>-4.54</b>	<b>-0.52</b>	-0.20	<b>-0.11</b>	0.99	<b>-0.14</b>	<b>-0.14</b>	-0.02	-0.00	<b>-0.00</b>
		PA	1994–2013	<b>-5.42</b>	<b>-6.06</b>	<b>-0.48</b>	-0.19	<b>-0.14</b>	2.15	<b>-0.14</b>	-0.11	-0.01	0.00	<b>-0.01</b>
LV01	NEE	PS	1994–2009	<b>-3.03</b>	1.96	-0.04	<b>-2.50</b>	-0.47	<b>5.02</b>	<b>-0.10</b>	<b>0.12</b>	0.02	<b>-0.12</b>	<b>-0.01</b>
LV02	NEE	PS	1994–2009	<b>-2.25</b>	2.05	<b>-1.76</b>	<b>-1.70</b>	<b>-0.67</b>	<b>12.3</b>	<b>-0.08</b>	<b>0.16</b>	<b>-0.08</b>	<b>-0.06</b>	<b>-0.02</b>
LT01	NEE	PA	1993–2013	<b>-4.25</b>	n.d.	<b>-1.30</b>	<b>-2.10</b>	-0.33	n.d.	<b>-0.12</b>	n.d.	<b>-0.06</b>	<b>-0.06</b>	0.00
LT03	NEE	PA	1995–2013	<b>-5.99</b>	n.d.	-1.23	<b>-1.30</b>	-0.04	n.d.	<b>-0.27</b>	n.d.	-0.03	-0.03	0.00
BY02	NEE	n.d.												
DE01	CE	PA	1993–2013	<b>-2.74</b>	0.67	<b>1.92</b>	<b>0.93</b>	<b>-1.49</b>	<b>3.93</b>	<b>-0.21</b>	0.01	<b>0.09</b>	<b>0.04</b>	<b>-0.11</b>
		FS	1990–2013	<b>-1.56</b>	<b>-0.54</b>	0.02	-0.05	<b>-0.54</b>	<b>1.46</b>	<b>-0.12</b>	<b>-0.05</b>	-0.00	-0.00	<b>-0.05</b>
DE02	CE	FS	1998–2013	<b>-3.41</b>	<b>-2.06</b>	-1.92	-1.06	<b>-0.57</b>	2.38	<b>-0.09</b>	-0.03	<b>-0.05</b>	-0.02	<b>-0.02</b>
CZ01	CE	PA	1990–2013	<b>-15</b>	<b>-3.62</b>	-0.44	0.57	<b>-4.40</b>	<b>12</b>	<b>-0.39</b>	<b>-0.09</b>	-0.00	0.02	<b>-0.08</b>
CZ02	CE	PA	1991–2013	<b>-11.4</b>	<b>-3.25</b>	<b>-1.67</b>	<b>-0.86</b>	<b>-4.17</b>	<b>8.85</b>	<b>-0.54</b>	<b>-0.15</b>	<b>-0.06</b>	<b>-0.03</b>	<b>-0.21</b>
AT01	CE	PA	1993–2013	<b>-1.81</b>	<b>-1.43</b>	-0.63	0.04	<b>-0.51</b>	<b>1.28</b>	<b>-0.16</b>	<b>-0.11</b>	-0.05	0.02	<b>-0.04</b>
		FS	1996–2013	<b>-1.01</b>	<b>-1.44</b>	-0.61	-0.37	<b>-0.17</b>	0.19	<b>-0.06</b>	<b>-0.08</b>	-0.02	-0.01	<b>-0.01</b>
IT01	CE	PA	1994–2013	<b>-2.64</b>	<b>-4.80</b>	<b>-0.93</b>	<b>-0.54</b>	<b>-0.25</b>	<b>-3.59</b>	<b>-0.11</b>	<b>-0.18</b>	-0.02	-0.01	<b>-0.01</b>
IT03	CE	PA	1997–2013	<b>-1.25</b>	<b>-2.01</b>	-0.51	-0.28	<b>0.15</b>	-0.28	<b>-0.05</b>	<b>-0.08</b>	-0.01	0.00	<b>0.01</b>
IT07	CE	QC	1997–2013	<b>-5.04</b>	<b>-3.15</b>	<b>-2.24</b>	<b>-2.38</b>	-0.00	<b>4.52</b>	<b>-0.22</b>	<b>-0.15</b>	-0.09	-0.09	-0.00
IT09	CE	QC	1997–2013	<b>-1.89</b>	-1.12	-0.10	-0.30	0.01	0.14	<b>-0.11</b>	-0.03	-0.01	-0.01	0.00
Mean				-3.54	-1.06	-0.62	-0.46	-0.83	3.67	-0.15	-0.04	-0.02	-0.01	-0.04
Md.				-3.22	-1.72	-0.57	-0.33	-0.29	2.26	-0.12	-0.08	-0.02	-0.01	-0.01

Table 3.3. Significant long-term trends (bold,  $p < 0.05$ ) and tendencies (italics,  $p < 0.1$ ) for monthly air temperature (T, °C yr<sup>-1</sup>), precipitation (P, mm yr<sup>-1</sup>) and runoff (R, mm yr<sup>-1</sup>) at studied IM sites in 1990–2013. Significant trends and tendencies are shaded in grey. Data not available (n.d.).

Site	Data	Period	Period												
			Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan-Dec
AT01	T	1995–2013	0.03	-0.23	0.08	<b>0.18</b>	0.03	0.03	0.06	0.07	0.13	-0.09	0.11	0.11	<i>0.06</i>
	P	1993–2013	4.54	-0.42	-5.45	-1.67	-3.97	3.52	1.87	<b>4.88</b>	-0.61	1.74	-1.18	-0.32	0.30
	R	1995–2013	<b>1.16</b>	0.10	-0.61	-0.83	-0.63	1.09	0.58	0.35	-0.45	-0.10	-0.21	-0.27	0.04
BY02	T	1992–2013	-0.15	-0.27	-0.06	0.03	0.11	0.05	0.06	0.02	0.05	0.06	<b>0.22</b>	0.22	<b>0.04</b>
	P	1990–2013	-0.01	0.49	-0.21	0.00	0.36	-0.06	-1.18	1.82	<b>-2.37</b>	0.57	-0.27	-0.54	-0.13
	R	n.d.													
CZ01	T	1990–2013	-0.07	-0.10	-0.05	<b>0.11</b>	0.01	0.07	0.05	0.00	0.04	0.02	0.09	0.06	0.03
	P	1990–2013	<b>2.21</b>	0.34	-0.34	-0.58	<i>1.21</i>	0.84	0.03	1.64	-0.22	-0.03	-0.97	-0.51	0.22
	R	1990–2013	0.02	0.06	0.08	0.00	-0.02	0.03	-0.01	0.01	0.00	0.02	0.00	-0.03	0.01
CZ02	T	1990–2013	-0.05	-0.08	-0.09	<i>0.09</i>	0.00	0.07	0.01	-0.01	0.04	0.02	0.05	0.03	0.01
	P	1990–2013	<b>3.37</b>	0.08	-0.81	0.79	<b>2.75</b>	-2.50	-0.86	0.41	-0.51	0.96	-0.54	1.00	0.52
	R	1990–2013	0.67	-0.26	0.38	0.28	0.27	-0.39	<b>-0.90</b>	-0.05	-0.15	-0.20	-0.61	0.48	-0.11
DE01	T	1990–2013	-0.08	<i>-0.16</i>	0.00	<b>0.13</b>	0.04	<b>0.07</b>	0.02	0.01	0.05	0.00	<i>0.11</i>	0.01	0.03
	P	1991–2013	1.10	1.58	-1.94	-0.50	2.96	-0.67	1.53	1.54	-0.58	-0.43	-0.84	-0.89	0.25
	R	1991–2013	0.33	0.18	1.03	2.32	0.97	0.82	0.10	0.51	0.31	-0.02	-0.52	-1.07	0.30
DE02	T	1990–2013	-0.12	-0.10	-0.03	<i>0.06</i>	0.05	<b>0.11</b>	0.07	0.02	0.05	0.05	<i>0.08</i>	0.06	<i>0.03</i>
	P	1998–2013	<i>1.58</i>	0.21	-0.64	-0.25	1.41	1.35	2.33	2.89	0.56	-1.07	0.96	1.38	<b>0.91</b>
	R	n.d.													
EE01	T	1995–2013	-0.11	<b>-0.37</b>	-0.10	-0.10	<b>0.10</b>	-0.03	0.00	0.04	0.03	-0.02	0.08	0.02	-0.01
	P	1994–2013	1.67	0.30	<b>1.11</b>	0.38	0.60	<i>1.90</i>	2.36	<b>3.80</b>	1.95	0.74	1.59	<b>2.96</b>	<b>1.45</b>
	R	n.d.													
EE02	T	1994–2013	-0.18	-0.25	-0.11	<b>-0.19</b>	<b>0.22</b>	0.06	-0.04	0.00	0.03	0.00	0.12	0.08	0.02
	P	1994–2013	-0.57	-1.07	0.19	-0.44	0.37	0.14	0.06	<b>4.00</b>	1.08	0.73	<b>2.32</b>	<b>2.39</b>	0.76
	R	1995–2013	0.12	0.60	<i>0.90</i>	<b>1.40</b>	<b>0.71</b>	0.27	0.16	0.06	0.07	<i>0.54</i>	<b>0.91</b>	0.77	<b>0.60</b>
FI01	T	1990–2013	-0.09	-0.20	<i>-0.10</i>	0.05	<b>0.13</b>	0.03	<b>0.09</b>	<i>0.06</i>	<b>0.13</b>	0.06	<b>0.19</b>	0.07	<b>0.06</b>
	P	1990–2013	-0.71	-0.33	-0.81	0.10	0.28	0.86	-0.28	-0.13	0.69	0.41	-0.17	0.43	0.05
	R	1990–2013	0.02	-0.14	-0.52	-0.28	-0.72	-0.03	0.04	0.05	0.07	0.28	0.69	0.54	0.02
FI03	T	1990–2013	0.00	-0.19	-0.12	0.02	<b>0.14</b>	0.04	0.09	0.06	<b>0.12</b>	0.06	<b>0.27</b>	0.03	<b>0.06</b>
	P	1990–2013	0.12	-0.68	-0.51	0.21	1.23	1.11	-0.01	0.81	0.79	0.34	0.24	0.33	0.23
	R	1990–2013	0.37	0.09	-0.06	0.10	-0.50	<i>-0.61</i>	-0.26	-0.03	-0.05	-0.13	0.33	0.64	-0.02
IT01	T	1990–2013	-0.07	-0.02	-0.06	<b>0.16</b>	0.10	<b>0.15</b>	0.07	0.07	<i>0.12</i>	0.06	0.10	0.00	<b>0.06</b>
	P	1993–2013	0.48	0.85	0.71	0.57	-1.28	-1.37	-1.29	-0.39	-1.28	0.18	2.44	<b>2.01</b>	0.26
	R	n.d.													
LT01	T	1993–2013	-0.18	-0.10	0.01	0.01	<b>0.18</b>	0.06	0.06	0.05	0.06	0.05	<b>0.26</b>	0.14	<b>0.06</b>
	P	1993–2013	-0.93	<b>-1.81</b>	-0.89	<b>-2.33</b>	<b>3.47</b>	-0.01	0.90	3.71	-2.03	-0.52	0.45	<b>-1.98</b>	-0.48
	R	1994–2012	-0.28	-0.33	-0.46	-0.58	-0.25	-0.14	-0.19	-0.19	-0.29	-0.30	-0.24	-0.24	-0.29
LT03	T	1990–2013	<b>-0.15</b>	<i>-0.28</i>	-0.07	0.00	<i>0.07</i>	0.07	0.09	0.02	<i>0.08</i>	0.05	<b>0.12</b>	0.07	<i>0.03</i>
	P	1993–2013	0.75	-2.59	-0.89	0.28	0.20	-0.61	2.14	<b>8.20</b>	1.66	-1.00	0.22	1.49	0.40
	R	1996–2012	<i>0.78</i>	0.48	<i>1.16</i>	0.28	<b>0.81</b>	<b>0.96</b>	<b>0.88</b>	<b>0.95</b>	<b>1.02</b>	0.26	0.54	0.51	<b>0.78</b>
LV01	T	1993–2008	0.09	-0.04	0.03	-0.01	0.00	0.10	0.06	0.11	<i>0.16</i>	0.10	0.20	<b>0.39</b>	<b>0.10</b>
	P	1994–2009	0.86	-1.48	1.83	-1.10	-0.29	-0.06	2.32	1.76	0.76	2.68	2.46	1.47	0.65
	R	1995–2009	-0.05	-3.42	0.40	-0.43	<b>-0.68</b>	<b>-0.38</b>	-0.09	-0.17	-0.08	-0.18	0.09	1.42	-0.23
LV02	T	1993–2008	0.04	-0.10	0.09	-0.03	-0.05	0.02	0.03	0.10	0.12	0.14	0.18	0.30	<b>0.07</b>
	P	1994–2009	0.91	-3.36	0.62	1.33	-2.38	0.24	2.40	<b>4.56</b>	-0.58	<b>3.85</b>	2.19	1.58	<b>1.05</b>
	R	1994–2009	-0.59	<i>-1.65</i>	<i>-1.72</i>	-1.18	<b>-1.66</b>	<b>-1.05</b>	-0.81	<b>-0.94</b>	<i>-0.84</i>	-0.68	-0.74	-0.23	<b>-0.86</b>
NO01	T	1990–2013	-0.09	-0.22	-0.08	-0.01	-0.04	-0.03	-0.03	-0.05	0.01	0.05	0.06	-0.01	-0.03
	P	1990–2013	-1.65	-1.80	-1.73	0.01	<i>3.08</i>	1.88	2.15	2.29	-0.38	3.93	0.29	3.54	1.00
	R	1990–2013	-2.43	-1.70	-0.80	2.87	0.43	0.24	0.58	0.78	-0.50	1.50	1.40	1.26	0.23
NO02	T														
	P	1990–2013	-1.12	0.89	-1.13	1.19	1.21	1.45	-0.51	-2.29	1.78	-0.15	1.49	-3.54	0.01
	R	1990–2013	0.84	0.79	<b>1.41</b>	<b>2.97</b>	4.77	-4.44	<b>-7.43</b>	-2.53	3.26	0.33	2.14	<b>2.08</b>	0.93
NO03	T														
	P	1998–2013	-1.95	-0.01	-1.55	-3.26	<b>8.04</b>	-0.10	4.33	<b>8.09</b>	-1.55	-0.64	0.38	2.68	0.50
	R	1990–2013	0.10	-0.23	-0.27	0.17	0.67	1.24	<b>2.26</b>	<b>2.84</b>	0.55	0.01	0.71	-0.12	<b>0.32</b>

Site	Data	Period	Period												
			Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan-Dec
SE04	T	1990–2013	-0.09	-0.06	0.07	-0.11	0.03	0.07	-0.01	0.00	0.09	0.09	0.12	-0.01	0.03
	P	1990–2013	-1.07	-1.85	-1.16	0.15	1.29	-1.11	2.28	1.49	1.66	1.32	2.40	0.80	0.56
	R	1990–2013	-1.13	-1.98	0.20	-0.16	0.14	0.10	0.25	1.36	1.93	1.96	2.59	2.14	0.40
SE14	T	1996–2013	-0.01	-0.14	0.01	0.00	0.04	0.03	0.11	-0.03	-0.02	0.06	0.14	0.13	0.03
	P	1996–2013	-0.50	-2.48	-1.90	-1.77	-1.38	0.10	-0.30	0.84	1.52	-0.63	-0.50	-0.21	-0.45
	R	1996–2013	-1.07	-1.26	-1.61	-0.55	-0.07	0.28	0.38	0.79	0.83	0.69	0.89	-1.19	-0.08
SE15	T	1997–2013	-0.12	-0.23	-0.10	0.09	0.03	0.07	0.06	0.02	0.00	-0.02	0.11	0.06	0.01
	P	1996–2013	-0.92	-0.82	-0.57	-0.92	-0.90	-1.64	3.66	2.74	-0.28	0.40	-1.84	1.55	0.00
	R	1996–2013	-1.74	-2.96	-1.00	-0.36	-0.54	-0.55	0.12	0.92	1.71	0.11	1.73	0.45	-0.11
SE16	T	1999–2013	-0.12	-0.23	-0.01	-0.05	0.02	-0.06	0.00	0.05	-0.03	-0.03	-0.05	0.04	-0.01
	P	1999–2013	-0.64	-0.95	-0.75	-2.11	1.26	-4.44	-2.86	1.74	2.21	-2.25	-2.06	1.13	-0.68
	R	1999–2013	0.37	0.09	0.20	-2.72	-1.71	-1.37	-1.27	-0.35	2.91	1.17	-0.11	0.13	0.10

bulk/wet deposition of inorganic N has generally exceeded SO<sub>4</sub> deposition on an equivalent basis since the late 1990s (Forsius et al. 2005, Vuorenmaa et al. 2014, 2016). Like for SO<sub>4</sub>, decreases of inorganic N in throughfall deposition at majority of the IM sites may indicate the pronounced effect of declining dry deposition as well.

Sulphur emissions in Europe decreased substantially from 1990 until 2000, and after that emissions have slowed down (Fig. 3.1.). Followed by a steeper decrease from 1990, emissions of NO<sub>x</sub> also experienced a more gradual decrease since 2000. These emission patterns are reflected by a steeper decrease in concentrations and deposition fluxes for both SO<sub>4</sub> and inorganic nitrogen and in acidity (H<sup>+</sup>) in precipitation chemistry and deposition fluxes at IM sites in the 1990s compared to the 2000s (Figs. 3.2 and 3.3).

Figure 3.2. Comparison of trend slopes for bulk/wet deposition concentrations ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) and fluxes ( $\text{meq m}^{-2} \text{yr}^{-1}$ ) between the periods 1990–2000 and 2000–2013 for  $\text{SO}_4$ ,  $\text{NO}_3$ -N,  $\text{NH}_4$ -N and  $\text{H}^+$ .

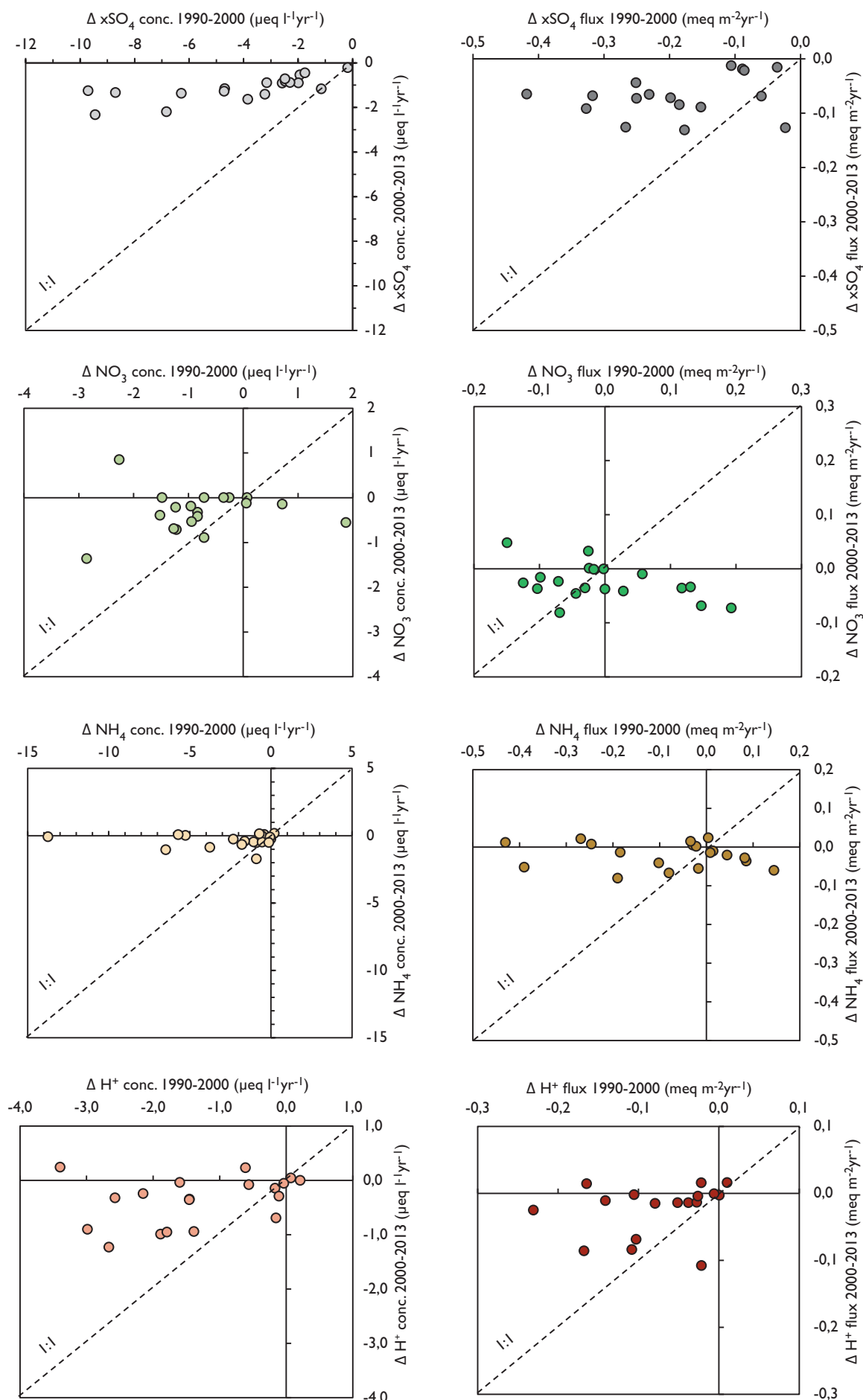
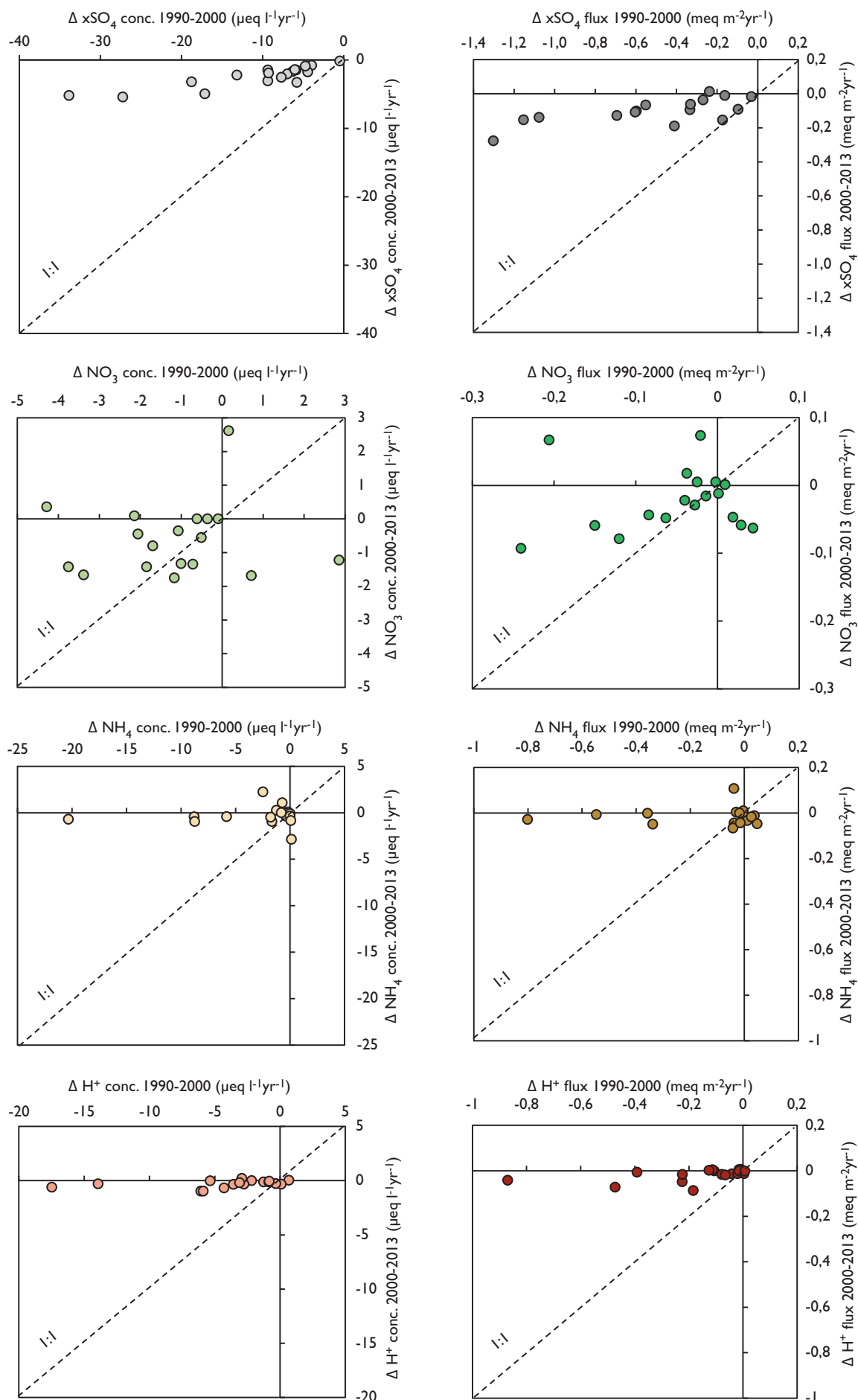


Figure 3.3. Comparison of trend slopes for throughfall deposition concentrations ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) and fluxes ( $\text{meq m}^{-2} \text{yr}^{-1}$ ) between the periods 1990–2000 and 2000–2013 for  $\text{SO}_4$ ,  $\text{NO}_3$ -N,  $\text{NH}_4$ -N and  $\text{H}^+$ .





## Runoff

The substantial decrease of SO<sub>4</sub> deposition has evidently resulted in decreased output fluxes of SO<sub>4</sub> in runoff in IM catchments. Although runoff water volume increased rather than decreased (11 out of the 18 sites, Table 3.3.), ca 60% of the sites exhibited a significant decrease in output fluxes in 1990–2013. Decreasing trend in SO<sub>4</sub> concentrations was more evident than in the fluxes, and a significant decrease was detected at 86% of the sites (Table 3.4.). SO<sub>4</sub> concentrations and acidity (H<sup>+</sup> concentration) in soil water mostly decreased (Table 3.5.).

The previous trend assessment for the IM sites in 1993–2006 showed that SO<sub>4</sub> output fluxes in catchments used in the present study decreased significantly at only 40% of the sites (Vuorenmaa et al. 2009). This suggests that IM catchments have increasingly responded to the decreases in emissions and deposition of SO<sub>4</sub> in Europe. The recent trend analysis of surface water chemistry in 173 acid-sensitive sites from 12 regions in Europe and North America as part of the UNECE ICP Waters programme (Garmo et al. 2014) and another recent European assessment of surface water SO<sub>4</sub> concentrations (Helliwell et al. 2014) have also demonstrated clear decreases of SO<sub>4</sub> concentrations in surface waters that eventually resulted from decreased SO<sub>4</sub> fluxes into the water courses.

Weaker trends in runoff water fluxes compared to trends in concentrations at IM catchments may be due to the net release of stored airborne sulphate in forest soils that had accumulated in the past (Vuorenmaa et al. 2014, 2016). Short-term inter-annual fluctuations in runoff water volume, that largely modify the output fluxes of SO<sub>4</sub>, can mask long-term changes in matter dynamics in ecosystems (e.g. Prechtel et al. 2001). Long-term mass balance budgets in IM catchments have shown that variation in annual retention and net release of SO<sub>4</sub> can be partly explained by variation in annual runoff (Vuorenmaa et al. 2014, 2016).

The IM catchments vary in their sensitivity to acidification, and based on the buffering capacity (Acid Neutralizing Capacity, ANC) of surface water, the sites in Sweden and Norway and the Finnish site FI01 and Czech site CZ02 are considered to be susceptible to acidification (ANC < 100 µeq l<sup>-1</sup>) (e.g. Vuorenmaa et al. 2009, Holmberg et al. 2013). The most acid-sensitive IM catchments in the present study with negligible/low buffering capacity are experiencing a recovery from sulphate-driven acidification, as indicated by decreases in H<sup>+</sup> concentrations (increases in pH) and increases in ANC in the soil-water ecosystem that resulted from decreased SO<sub>4</sub> loss (Table 3.4.). Proceeding recovery from acidification in acid-sensitive ICP monitoring sites has been one of the most significant responses to the decreasing sulphur emissions (de Wit et al. 2015)

The present trend of inorganic N deposition at IM sites is decreasing, which should generally lead to decreased NO<sub>3</sub> concentrations in runoff (Wright et al. 2001, Forsius et al. 2005, Holmberg et al. 2013). Concentrations and fluxes of inorganic N in runoff, however, showed a mixed response with both decreasing and increasing trend slopes. Trends for NO<sub>3</sub> concentrations were decreasing (14 out of the 21 sites) rather than increasing, while NH<sub>4</sub> concentrations decreased at seven out of the 18 sites and increased at 11 sites. NO<sub>3</sub> concentrations decreased significantly at eight sites, but increased significantly at four sites (AT01, BY02, LV01, SE14) and NH<sub>4</sub> concentrations increased significantly at two sites (BY02, SE14). Correspondingly, trend slopes for fluxes of NO<sub>3</sub> in runoff were decreasing at nine out of the 18 sites, being significant at five sites (Table 3.4.).

A significant increase in output fluxes of inorganic N was detected in six catchments (AT01, DE01, EE02, LT03, SE04, SE14), but trends were likely not linked to direct N deposition effects. Increasing trends at sites DE01 and SE14 were due to substantially altered

Table 3.4. Long-term trends (1990–2013) for monthly runoff water chemistry concentrations and fluxes in studied IM catchments. For the annual change, a significant trend ( $p < 0.05$ , Mann-Kendall test, Sen's slope) is in bold. Site-specific annual changes and their mean and median (Md.) values for runoff water concentrations and fluxes are given in  $\mu\text{eq l}^{-1} \text{yr}^{-1}$  and  $\text{meq m}^{-2} \text{yr}^{-1}$ , respectively. Data not available (n.d.). The IM sites are grouped into three different geographical regions reflecting gradients in long-range transport and deposition of pollutants and climate (NoC=Nordic Countries, NEE=North Eastern Europe, CE=Central Europe).

Site	Region	Data	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>	ANC	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>
			Concentration, $\mu\text{eq l}^{-1} \text{yr}^{-1}$						Flux, $\text{meq m}^{-2} \text{yr}^{-1}$				
FI01	NoC	1990–2013	<b>-3.02</b>	<b>-1.79</b>	<b>-0.02</b>	0.03	<b>-0.35</b>	<b>1.30</b>	-0.02	-0.01	0.00	0.00	-0.00
FI03	NoC	1990–2013	<b>-0.87</b>	0.08	<b>-0.01</b>	<b>-0.01</b>	0.00	<b>1.12</b>	<b>-0.03</b>	0.00	<b>-0.00</b>	<b>-0.00</b>	0.00
SE04	NoC	1990–2013	<b>-8.32</b>	<b>-2.86</b>	0.02	0.00	<b>-1.70</b>	<b>6.27</b>	<b>-0.17</b>	<b>-0.06</b>	<b>0.00</b>	0.00	-0.02
SE14	NoC	1996–2013	<b>-5.46</b>	<b>-2.54</b>	<b>0.27</b>	<b>0.04</b>	<b>-1.14</b>	<b>1.88</b>	<b>-0.12</b>	-0.06	<b>0.00</b>	<b>0.00</b>	<b>-0.03</b>
SE15	NoC	1996–2013	<b>-5.64</b>	<b>-1.72</b>	-0.01	0.00	<b>-0.94</b>	<b>3.59</b>	<b>-0.14</b>	<b>-0.04</b>	-0.00	0.00	<b>-0.03</b>
SE16	NoC	1999–2013	<b>-1.16</b>	-0.06	-0.02	<b>-0.02</b>	-0.01	1.10	-0.02	0.00	-0.00	-0.00	0.00
NO01	NoC	1990–2013	<b>-3.01</b>	<b>-1.23</b>	-0.04	n.d.	<b>-0.44</b>	<b>2.30</b>	<b>-0.18</b>	<b>-0.08</b>	-0.00	n.d.	-0.01
NO02	NoC	1990–2013	<b>-0.20</b>	<b>0.38</b>	-0.01	n.d.	<b>-0.01</b>	<b>0.92</b>	-0.01	<b>0.07</b>	0.00	n.d.	-0.00
NO03	NoC	1990–2013	<b>-2.00</b>	<b>-1.06</b>	<b>-0.03</b>	n.d.	<b>-0.24</b>	<b>1.08</b>	<b>-0.03</b>	<b>-0.00</b>	<b>-0.00</b>	n.d.	<b>-0.00</b>
EE01	NEE	n.d.											
EE02	NEE	1994–2013	<b>-10.1</b>	13.6	0.29	0.00	<b>-0.00</b>	22.7	0.01	<b>2.16</b>	<b>0.01</b>	<b>0.00</b>	<b>-0.00</b>
LV01	NEE	1993–2009	<b>-21.0</b>	4.34	<b>0.46</b>	0.00	<b>0.00</b>	20.3	<b>-0.35</b>	-0.56	0.00	-0.00	<b>-0.00</b>
LV02	NEE	1993–2009	-2.01	<b>-28.7</b>	<b>-0.14</b>	<b>-0.54</b>	<b>0.01</b>	<b>-23.6</b>	<b>-0.39</b>	<b>-2.50</b>	<b>-0.01</b>	<b>-0.01</b>	<b>0.00</b>
LT01	NEE	1994–2013	<b>-41.4</b>	<b>-26.7</b>	-0.33	-0.01	<b>-0.00</b>	11.4	-0.72	-1.21	-0.01	-0.00	0.00
LT03	NEE	1996–2012	<b>-28.8</b>	<b>-49.7</b>	<b>-0.45</b>	0.00	-0.00	-18.2	0.34	<b>1.35</b>	<b>0.01</b>	0.00	<b>-0.00</b>
BY02	NEE	1995–2013	-7.10	1.95	<b>2.33</b>	<b>1.67</b>	-0.00	9.96	n.d.	n.d.	n.d.	n.d.	n.d.
DE01	CE	1991–2013	<b>-1.23</b>	<b>1.52</b>	1.88	-0.11	<b>-0.01</b>	<b>2.77</b>	-0.05	<b>0.16</b>	<b>0.13</b>	-0.01	<b>-0.00</b>
DE02	CE	n.d.											
CZ01	CE	1990–2013	6.35	6.87	<b>-2.68</b>	<b>-0.12</b>	-0.00	1.50	0.02	0.02	<b>-0.01</b>	<b>-0.00</b>	0.00
CZ02	CE	1990–2013	<b>-16.4</b>	<b>-8.26</b>	<b>-0.08</b>	0.00	<b>-1.85</b>	<b>8.14</b>	<b>-0.36</b>	<b>-0.20</b>	<b>-0.01</b>	0.00	<b>-0.03</b>
AT01	CE	1995–2013	<b>-2.35</b>	<b>18.4</b>	<b>2.14</b>	0.00	-0.00	<b>17.8</b>	<b>-0.07</b>	0.78	<b>0.07</b>	0.00	0.00
IT01	CE	2000–2013	<b>-3.51</b>	<b>13.1</b>	<b>-0.47</b>	-0.04	<b>-0.00</b>	<b>18.5</b>	n.d.	n.d.	n.d.	n.d.	n.d.
IT03	CE	2001–2013	<b>-0.75</b>	-0.32	-0.05	0.02	<b>-0.00</b>	0.65	n.d.	n.d.	n.d.	n.d.	n.d.
IT07	CE	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
IT09	CE	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Mean			-7.52	-3.08	0.15	0.05	-0.32	4.36	-0.14	-0.01	0.01	0.00	-0.01
Md.			-3.02	-0.32	-0.02	0.00	0.00	2.30	-0.06	-0.01	0.00	0.00	0.00

Table 3.5. Long-term trends for monthly soil water chemistry concentrations and fluxes at different measuring depths at IM sites DE02 and EE01. For the annual change, a significant trend ( $p < 0.05$ , Mann-Kendall test, Sen's slope) is in bold. Site-specific annual changes and their mean and median (Md.) values for soil water concentrations and fluxes are given in  $\mu\text{eq l}^{-1} \text{yr}^{-1}$  and  $\text{meq m}^{-2} \text{yr}^{-1}$ , respectively. Data not available (n.d.).

Site, Region	Plot	Depth (cm)	Data	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>	ANC	xSO <sub>4</sub>	xBC	NO <sub>3</sub> -N	NH <sub>4</sub> -N	H <sup>+</sup>
				Concentration, $\mu\text{eq l}^{-1} \text{yr}^{-1}$						Flux, $\text{meq m}^{-2} \text{yr}^{-1}$				
EE01, NEE	8	17	1994–2013	<b>-3.49</b>	<b>53.3</b>	<b>4.56</b>	0.29	<b>-0.09</b>	<b>54.5</b>	-0.01	<b>0.80</b>	<b>0.07</b>	<b>0.01</b>	<b>-0.00</b>
		35	1994–2013	-2.23	<b>50.1</b>	<b>5.45</b>	<b>0.54</b>	-0.00	<b>47.2</b>	-0.00	0.22	0.03	0.00	-0.00
DE02, CE	10	30	1998–2013	<b>-3.48</b>	-21.9	0.04	<b>0.38</b>	-0.00	-21.7	n.d.	n.d.	n.d.	n.d.	n.d.
		50	1998–2013	<b>-2.82</b>	11.2	<b>-1.35</b>	<b>0.34</b>	<b>-0.00</b>	<b>17.7</b>	n.d.	n.d.	n.d.	n.d.	n.d.
		70	1998–2013	<b>-4.24</b>	-13	-1.09	<b>0.41</b>	<b>-0.00</b>	-3.92	n.d.	n.d.	n.d.	n.d.	n.d.
	20	30	1998–2013	-6.24	<b>-27.5</b>	-13.6	0.17	-0.08	-3.03	n.d.	n.d.	n.d.	n.d.	n.d.
		50	1998–2013	<b>-7.82</b>	<b>-44.9</b>	-7.94	<b>0.36</b>	<b>-0.06</b>	<b>-18.1</b>	n.d.	n.d.	n.d.	n.d.	n.d.
		120	1998–2013	<b>-11.6</b>	15.4	-10.7	<b>0.44</b>	<b>-0.01</b>	<b>40.6</b>	n.d.	n.d.	n.d.	n.d.	n.d.
Mean				-5.24	2.84	-3.08	0.37	-0.03	14.2	-0.01	0.51	0.05	0.00	0.00
Md.				-3.86	-0.90	-1.22	0.37	0.00	7.36					

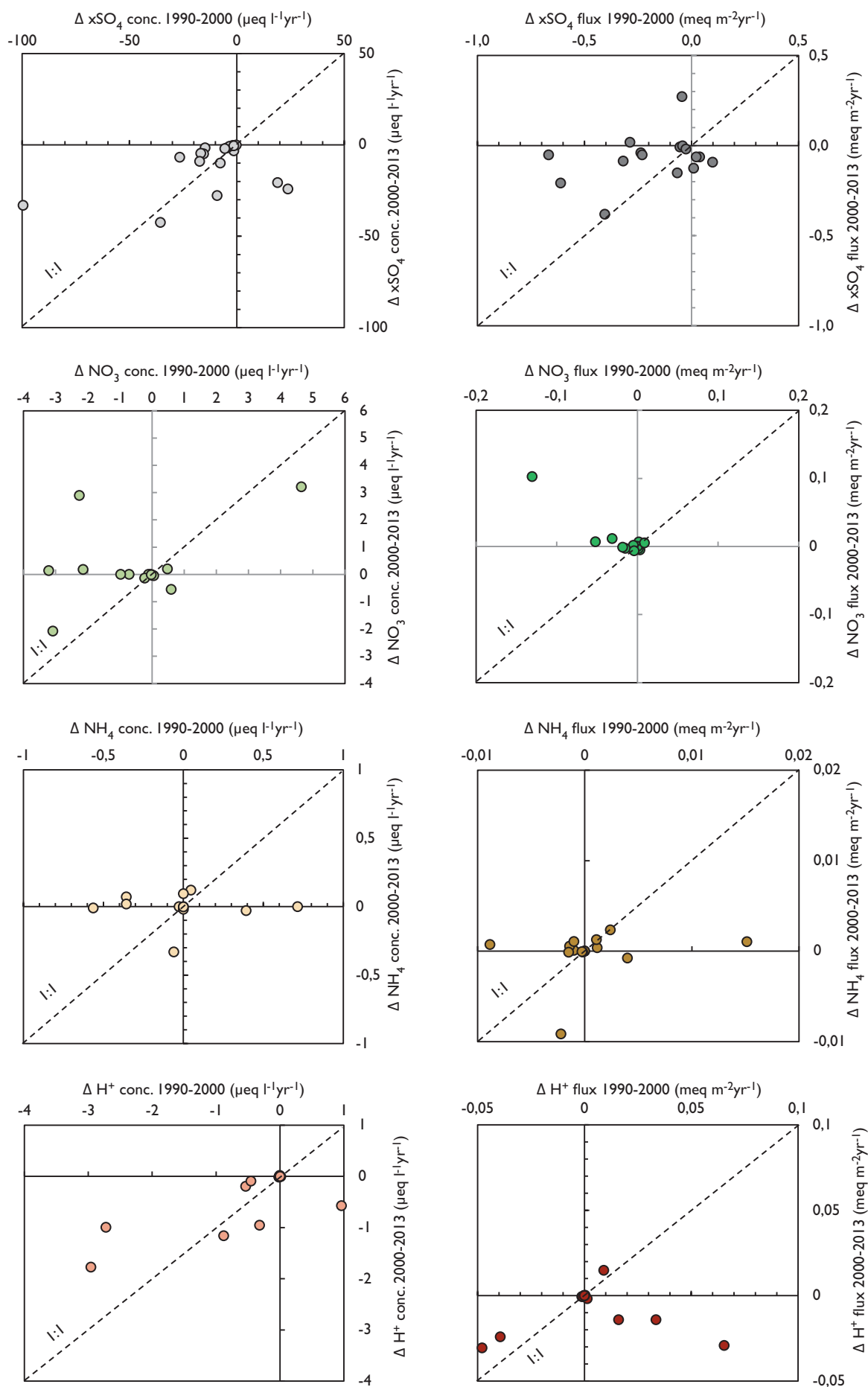
biogeochemical N cycles within the catchments by well-known forest disturbance regimes (i.e. wind-throw, bark beetle infestation) (Löfgren et al. 2011, Beudert et al. 2014, Vuorenmaa et al. 2016). The sites EE02 and SE04 did not show any significant increases in inorganic N concentrations in 1990–2013, and therefore the increase in N fluxes may be partly related to increased runoff (Table 3.3). The Austrian site AT01 is a leaky karst catchment, where high inorganic N deposition causes high nitrate loss even if the forests are not N-saturated. The catchment has a fast runoff dynamic, and snowmelt periods and heavy rain events cause strong throughflow, dictating not only annual but also the long-term N budgets (Jost et al. 2011).

The previous trend assessment (1993–2006) for the IM sites used in the present study (Vuorenmaa et al. 2009) also showed a mixed response with both decreasing and increasing trend slopes for  $\text{NO}_3$  concentrations and fluxes but trends for concentrations and fluxes were increasing (12 out of the 19 sites and nine out of the 16 sites, respectively) rather than decreasing. Significant decreases of  $\text{NO}_3$  fluxes in runoff were detected at four sites (CZ02, FI03, LT01 and LV02), while  $\text{NO}_3$  flux increased significantly at five sites (DE01, FI01, LT03, NO02 and SE04). Thus, the trends for output fluxes of  $\text{NO}_3$  are still highly variable, indicating that surface water-watershed nitrogen dynamics are inherently complex as nitrogen is strongly affected by biological processes, and nitrate concentrations in surface waters may highly fluctuate with season and spatially across ecosystems (e.g. Aber et al. 2003). Moreover, the short-term and long-term variations in climate may mask long-term trends caused by N deposition (Wright et al. 2001). Nevertheless, the present trends in concentrations and output fluxes of  $\text{NO}_3$  are now decreasing at a majority of the sites, and significant increasing trends have disappeared on some sites. Correspondingly, other surveys from Europe have not shown signs of consistent and widespread increase in nitrate concentrations or exports in sensitive undisturbed freshwaters (Wright et al. 2001, Watmough et al. 2005, Garmo et al. 2014, Helliwell et al. 2014, de Wit et al. 2015).

The emission and deposition patterns were also reflected to trends in runoff water chemistry and fluxes at IM sites. Decrease in concentrations and output fluxes for both  $\text{SO}_4$  and inorganic nitrogen and in acidity ( $\text{H}^+$ ) was steeper in the 1990s than in the 2000s (Fig. 3.4). Besides, the more gradual decrease in concentrations and fluxes of  $\text{SO}_4$  in the 2000s compared to that in the 1990s may also be due to increased net release of  $\text{SO}_4$  (Vuorenmaa et al. 2014, 2016).

Many of the retention and release processes for  $\text{SO}_4$  and inorganic nitrogen are sensitive to climatic variables, such as temperature, precipitation, discharge and drought/re-wetting events, and leaching of S and N compounds would therefore be affected by climate change (e.g. Wright & Jenkins 2001, Moore et al. 2010, Templer et al. 2012, Mitchell et al. 2013, Dirnböck et al. 2016). Many IM sites exhibit seasonal trends in climatic variables with a significant increase ( $p < 0.05$ ) or increasing tendency ( $p < 0.1$ ) in air temperature and precipitation particularly in spring and autumn (Table 3.3.). Climate-driven changes in hydrological conditions together with internal  $\text{SO}_4$  sources are becoming increasingly important as atmospheric  $\text{SO}_4$  input has declined (Dillon et al. 1997, Wright 1998, Wright & Jenkins 2001, Benčoková et al. 2011, Mitchell et al. 2013). Effects of climate variability and change are expected to offset and delay recovery of acid-sensitive waters (de Wit et al. 2015). While continued decrease in N deposition is anticipated at the ICP IM sites in the future (Forsius et al. 2005, Holmberg et al. 2013), nitrogen continues to accumulate in catchment soils and vegetation, and concern remains that chronic N deposition may ultimately lead to decreased soil capacity to retain N and to increased leaching of inorganic N. Many of the processes regulating N mineralization and inorganic N leaching are driven by climatic variables, and potential risk for climate-induced elevated N loss from watersheds to surface waters may be anticipated in the future.

Figure 3.4. Comparison of trend slopes for runoff water chemistry concentrations ( $\mu\text{eq l}^{-1} \text{yr}^{-1}$ ) and fluxes ( $\text{meq m}^{-2} \text{yr}^{-1}$ ) between the periods 1990–2000 and 2000–2013 for  $\text{SO}_4$ ,  $\text{NO}_3\text{-N}$ ,  $\text{NH}_4\text{-N}$  and  $\text{H}^+$ .



## Conclusions

The results from the ICP IM network document the positive effects of the international emission abatement actions that have led to decreased deposition and output fluxes of  $\text{SO}_4$  in forested catchments across Europe from 1990 through 2013. Acid-sensitive ICP IM sites also exhibit continuing recovery from acidification. Decreased nitrogen emissions have also resulted in decrease of inorganic N deposition, but to a lesser extent than that of  $\text{SO}_4$ . Inorganic nitrogen fluxes in runoff were decreasing rather than increasing, but trends were highly variable due complex processes in terrestrial catchment that are not yet fully understood. Besides, the net release of  $\text{SO}_4$  in forested catchments fueled by the mobilization of legacy S pools accumulated during times of high atmospheric sulphur deposition may delay the recovery from acidification. The more efficient retention of inorganic N than  $\text{SO}_4$  results in generally higher leaching fluxes of  $\text{SO}_4$  than those of inorganic N in European forested ecosystems.  $\text{SO}_4$  thus remains the dominant source of actual soil acidification despite the generally lower input of  $\text{SO}_4$  than inorganic N. The sulphur and nitrogen problem thus clearly requires continued attention as a European air pollution issue, and further long-term monitoring and trend assessments of different ecosystem compartments and climatic variables are needed to evaluate of the effects not only of emission reduction policies but also of changing climate.

The next phase of the work on trend assessment will be preparation of a scientific paper, involving inclusion of additional sites. The national focal points and the representatives for the sites will be invited to assist with these activities.

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## Annex I

### **Lago Nero – a new site to assess the effects of environmental change on high-alpine lakes and their catchments**

#### **Report on National ICP IM activities in Switzerland**

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#### **Background**

The consequences of deposited atmospheric pollutants on ecosystems is a key environmental issue, especially in the southern slopes of the Alps which receive substantial inputs of pollutants from the Po valley, northern Italy<sup>1</sup>. High-alpine catchments are particularly sensitive to atmospheric pollutants but also to other environmental issues including climate change, mainly as a consequence of their low chemical and physical buffer capacity and their often highly specialised biological communities<sup>2</sup>. In particular, lakes in these catchments can be seen as sentinels of environmental change<sup>3</sup>. The various and complex effects of atmospheric pollutants and environmental change in general warrant an integrative monitoring of ecosystems. This perspective reflects the goals of the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM).

#### **The study site**

We established a monitoring program in a high-alpine catchment located at the head of Val Bavona, Canton Ticino, southern Switzerland, which includes a small alpine lake, Lago Nero (Figs 1, 2). The catchment is southwest-facing, with altitude ranging from 2385 m to 2842 m.a.s.l., an area of 77.5 ha and a mean slope of 84 %. The substrate is dominated by gneissic bedrock with patches of grassy vegetation and shallow soils. The catchment is snow-covered approximately from November to May. For a similar period, the lake is ice-covered. Lago Nero is an oligotrophic, soft-water lake with a surface of approx. 13 ha and a maximal depth of 73 m. Monitoring of the site began in summer 2014, with an initial phase aimed at developing and testing methodologies and at evaluating the suitability of the catchment and the feasibility of the monitoring program.

Since the year 2000, Lago Nero has been monitored by an ICP Waters program, which focuses on summer surface water chemistry of a regional set of high-alpine lakes in Ticino (Fig. 2). This ICP Waters program thus provides synergies to our monitoring program in the form of shared infrastructure and measurements but it also allows spatial and temporal comparison, i.e. with other similar lakes and – for some parameters – with long time series.



Figure 1. Lago Nero and its catchment in the Alps of southern Switzerland.

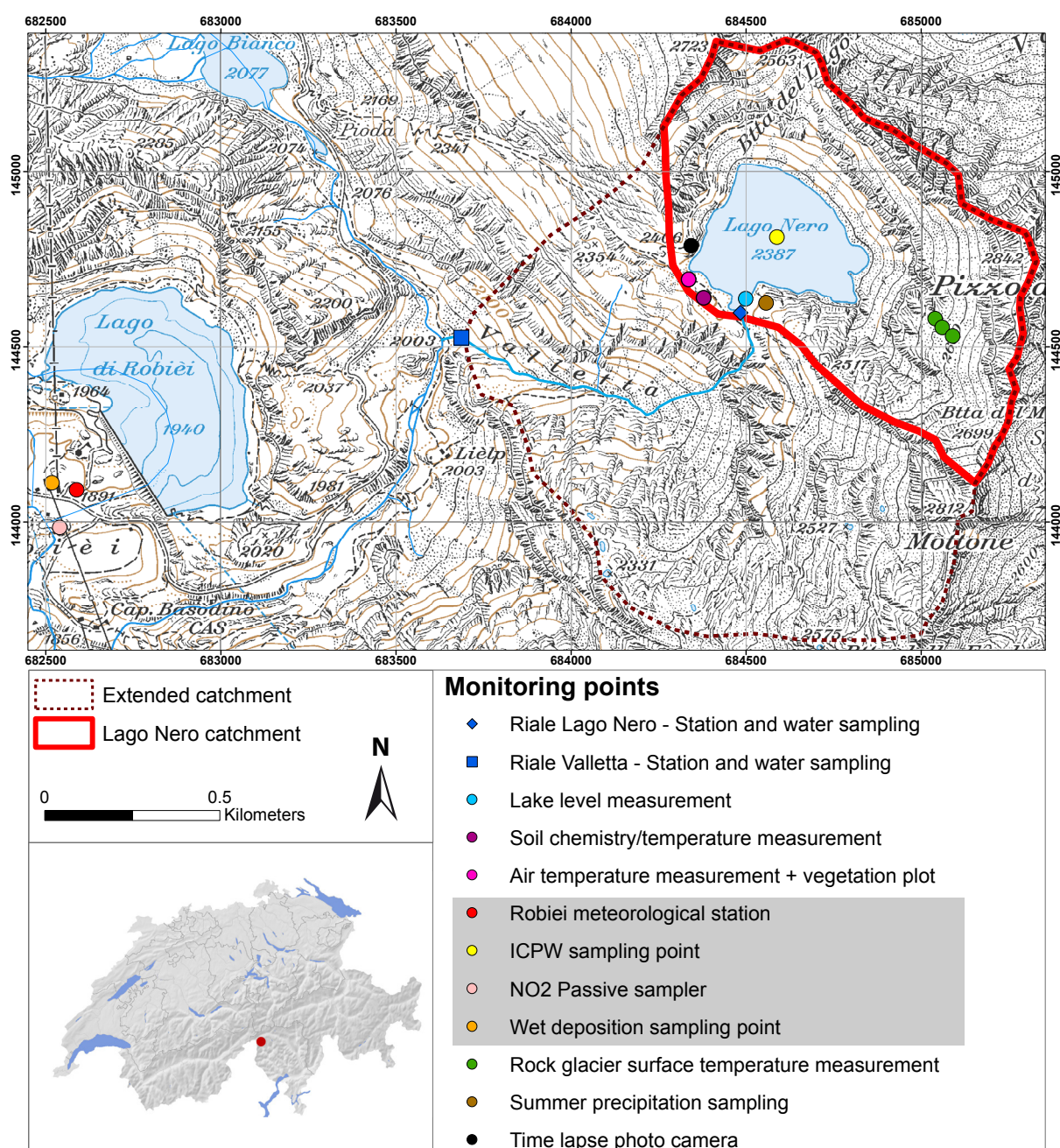


Figure 2. Lago Nero and its catchment (outlined in red) and location in Switzerland (bottom-left panel). The monitoring program is based on various measurements made at the outflow of the lake and at the downstream confluence (extended catchment). The grey-shaded box in the legend indicates monitoring stations from existing cantonal or federal monitoring programs and from ICP Waters. Coordinates are based on the Swiss grid (CHI903).



## The monitoring program

Parameters and methodologies of the monitoring program were chosen according to ICP IM protocols and include the following: meteorology and air chemistry (based on a nearby station by the federal meteorological office and on atmospheric models), precipitation chemistry (based on a nearby station by the cantonal administration plus a station in the catchment), soil chemistry and soil water chemistry, discharge and water chemistry of the outflow, vegetation composition. Due to the absence of trees in the catchment, ICP IM parameters like throughfall, foliage and litterfall chemistry, and trunk epiphytes are not assessed. However, additional parameters that seemed particularly interesting at this site are monitored. They include for instance measurements of runoff from the extended, terrestrially dominated catchment, as well as of the water chemistry of a rock glacier outflow draining into the lake.

## First results

The first full hydrological year covered by our monitoring program ran from October 2014 to September 2015. During this period, measurements revealed high inputs of key atmospheric pollutants including nitrogen and sulphur, which can cause acidification and eutrophication of sensitive ecosystems. These high inputs are probably a consequence of very high precipitation during the study period compared to long-term averages (i.e. 27 % higher than the long-term average based on the previous 24 years). For nitrogen, these inputs are substantially higher than the critical load estimated at 3–5 kg/ha·year<sup>-1</sup> for softwater alpine lakes<sup>4</sup> and 8 kg/ha·year<sup>-1</sup> for alpine grasslands<sup>5</sup>. Exceedance of critical loads for nitrogen is likely to alter aquatic and terrestrial biological communities<sup>2,4,5</sup>. Sulphur exceeded the critical load estimated for the Lago Nero catchment by a factor of 4.2<sup>ref 6</sup>. Critical loads for sulphur indicate thresholds above which acidification of ecosystems is expected<sup>6</sup>.

Chemical budgets calculated based on measurements of inputs (wet deposition only) and outputs (i.e. outflow concentrations) suggest that much of the deposited nitrogen was retained in the catchment or lost due to yet unmonitored processes, such as output of particulate organic nitrogen and denitrification (Fig. 3). For similar high-alpine lakes, the contribution of particulate organic nitrogen to total nitrogen had been estimated between 6 and 26 %<sup>7,8</sup>, which might explain a small fraction of the difference between input and output of nitrogen. Output of sulphur on the other hand matched inputs, suggesting that fluxes of this element in the catchment were in balance. Analyses of nitrogen and sulphur budgets for 17 catchments across Europe suggest that the patterns observed for the catchment of Lago Nero, i.e. substantial retention of nitrogen and balanced fluxes for sulphur, are common<sup>9</sup>. Integrative ecosystem responses will in the future provide mechanistic understanding of the processes controlling budgets of key chemicals in the catchment. Temporal patterns of concentrations in nitrogen and other key chemicals in the outflow indicate that the snowmelt period contributes substantially to the budgets and should be covered with high frequency sampling (e.g. with an autosampler). Additionally, certain physical characteristics of the catchment and the lake have strong influence on biological processes. For instance, estimates of the heat budget of the lake suggest a very low heat content building up during summer, suggesting that Lago Nero is particularly sensitive to consequences of climate change, including a prolonged ice-free period<sup>10</sup>. These changes can also increase the heat content of the lake and its stratification in the future, which would have important consequences on lake biota and interlinked ecosystem compartments.

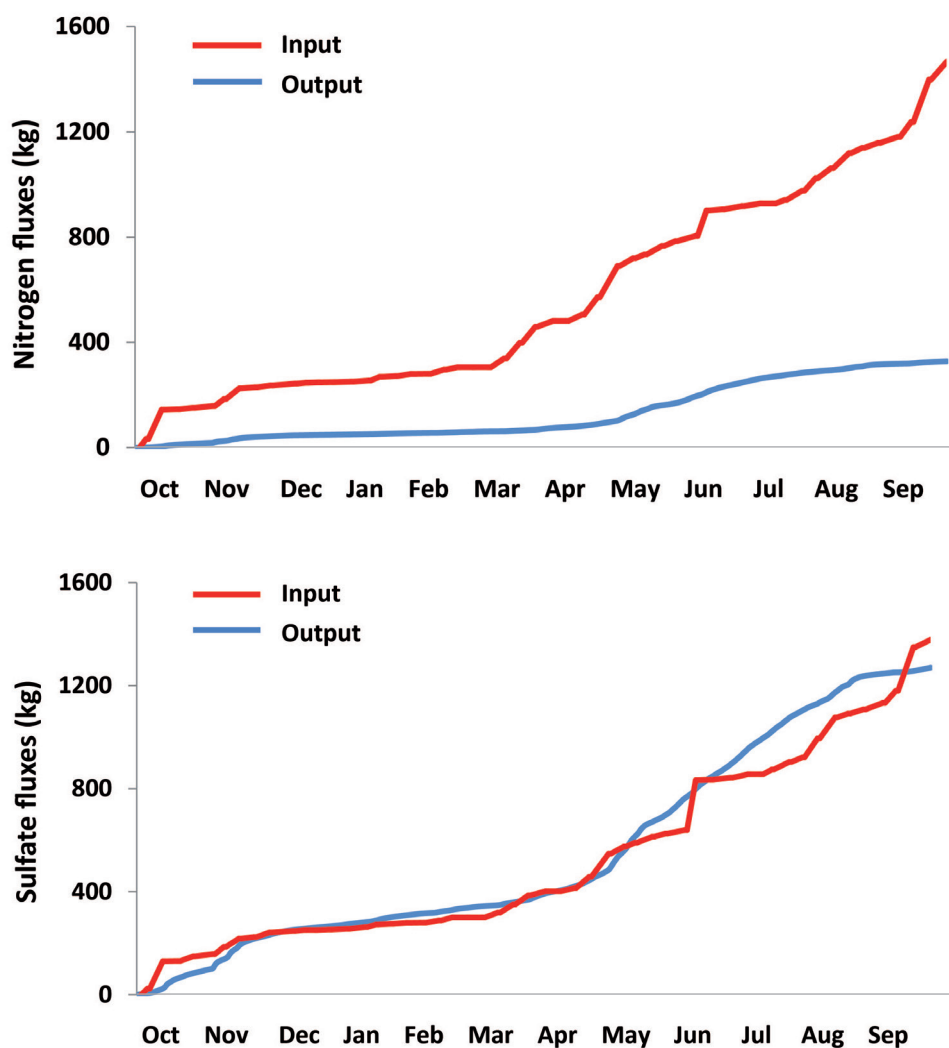


Figure 3. Budgets of nitrogen (upper panel) and sulphur (lower panel) estimated for the catchment of Lago Nero during the hydrological year from October 2014 to September 2015. For nitrogen, only dissolved fractions are considered.

## Conclusions

The experiences and findings made during this first phase of the monitoring program suggest that the Lago Nero catchment is a well-suited site for long-term monitoring of the ecological consequences of atmospheric pollutants and other environmental issues. This conclusion is based on the following characteristics of the Lago Nero catchment: it is (i) virtually unexposed to local anthropogenic impacts (e.g. hydro-power, grazing, tourism), (ii) sensitive to the impacts of interest including atmospheric pollutants, and at the same time (iii) strongly exposed to these impacts of interest. Based on these findings and the experiences made, the Swiss Federal Office for the Environment (FOEN) provided funding for the continuation of the program.

## Acknowledgements

We acknowledge funding by the Air Quality Management Section of the Swiss Federal Office for the Environment (FOEN). Furthermore, we greatly appreciate the support and contribution of Dr. Sandra Steingruber of the Ufficio dell'aria, del clima e delle energie rinnovabili of Canton Ticino and of ICP Waters as well as data provided by MeteoSwiss, Meteotest and the Swiss Federal Institute for Snow and Avalanche Research. We also appreciated the fruitful discussions with Gaston Theis-Goldener, Reto Meier and Beat Achermann (all FOEN) and with colleagues from ICP IM, ICP Waters, and our institute.

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## Annex 2

### Report on National ICP IM activities in Austria

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

Integrated Monitoring is carried out since 1992. The site also hosts air pollution monitoring in the framework of EMEP. Since 2006 Zöbelboden is part of Long-term Ecological Research (LTER Austria) and serves as a research station for several universities and research institutes. This development led to additional instrumentation, most prominently a CO<sub>2</sub> flux tower and soil respiration measurements. By the year 2008 an additional intensive plot was established due to severe wind and bark beetle disturbances at the plateau.

Monitoring of all obligatory and many mandatory parameters of the ICP Integrated Monitoring have continued and the respective data has been reported. During the last couple of years emphasis was on nitrogen deposition effects on nitrate leaching and biodiversity. Several research institutes have carried out in-depth studies and cross-site comparison analyses have further increased knowledge. This data and knowledge was used to inform policy, both at the national scale (e.g. in the form of critical load exceedance assessment) and at the international level (e.g. CLRTAP Assessment Report).

#### Chronic N deposition worsened ecosystem functioning

The IM site Zöbelboden is exposed to elevated N deposition (> 30 kg N ha<sup>-1</sup> yr<sup>-1</sup>, taking all deposition pathways into account) above the empirical critical load for coniferous or deciduous forests (10–20 kg N ha<sup>-1</sup> yr<sup>-1</sup>). High deposition together with high precipitation, snow melt and porous soils causes relatively high nitrate leaching (Jost et

al. 2011). By using measurements and modelling (LandscapeDNDC), annual N loss via leaching and outgassing sum up to 68% of the N deposition (54% leaching, 14% gaseous) at deeper soils (Chromic Cambisols with spruce forests) in the catchment. Although shallow slope soils (i.e. Lithic and Rendzic Leptosols with semi-natural mixed mountain forest) seem to be more efficient in N retention, we can conclude that chronic N deposition has rendered the site more susceptible to elevated nitrate loss. As a consequence, nitrate loss after clear-cut management or after disturbances such as climate change driven bark beetle attacks, will most likely be more severe (Dirnböck et al. 2016). This is important as major parts of the Northern Limestone Alps are used for water resources. Hartmann et al. (2016), by using the Zöbelboden as a model catchment, showed that karst water memorises elevated nitrate runoff following wind throws (e.g. during the storms Kyrill, Paula and Emma between 2007 and 2008) because of some slow water pathways (see also Hartmann et al. 2011). This might become an issue when forest disturbances are increasing with climate change. The knowledge gained from these analyses is used to improve Critical Loads, both at the national and at the UNECE level.

### **N deposition effects differ between organism groups**

Long-term decreasing trends of epiphytic lichen diversity at Zöbelboden could be related to elevated N deposition (Mayer et al. 2013). This temporal trajectory is in line with broader scale trends and proves that lichens, though they were recovering from former acidifying S deposition for some time, are now strongly affected by airborne N pollution. Vascular plants in the forest understorey are another effect related indicator for biodiversity. A trend analysis of data between 1993 and 2014 showed that N deposition, which had an effect until the year 2005, became less important relative to disturbance and climate change (Helm et al. in prep). Data from Zöbelboden were also part of a European cross-site analysis corroborating these results and showing that N deposition rendered oligotrophic plant species less abundant across Europe, mostly so in sensitive sites with acid soils (Dirnböck et al. 2014). Data and results from Zöbelboden are currently used in a collaborative project of ICP Integrated Monitoring and Modelling and Mapping to improve Critical Loads for biodiversity through the use of dynamic soil-plant models.

### **Reduced carbon sequestration in disturbed forests**

Not directly related to the CLRTAP air pollution issues, but very relevant for other policies (e.g. UNFCCC) are the studies carried out regarding forest carbon sequestration. The forests at Zöbelboden are sinks for CO<sub>2</sub> at a rate of approximately 1.5 t C ha<sup>-1</sup> yr<sup>-1</sup> (Kobler et al. 2015), which is typical for European temperate forests. However, forest gap disturbance, which might become more frequent with climate change, substantially reduce this sink. The major effect attributes to reduced tree C sequestration, because release of C from the soils through respiration is not strongly affected in small to medium size gaps and may be further buffered by regrowth of understorey vegetation. Carbon data from Zöbelboden is used in the assessment of ecosystem carbon sinks for the national emission inventory under the UNFCCC.

### **Contributions to cross site analyses**

Data from the Austrian site Zöbelboden has been used for a number of cross-site analyses carried out by the IM Programme centre and national contributions. Bringmark et al. (2013) showed that the retention of airborne lead and cadmium in forested catchments across Europe is high, even after significant reduction in deposition. Holmberg

et al. (2013) found that nitrate leaching is elevated in sites with an exceedance of the N critical load, which is one of the most important indicators for air pollution in the UNECE region. Dirnböck et al. (2014), for the first time, could find a European-wide eutrophication signal in forest floor vegetation which was due to N deposition. Vuorenmaa et al. (in prep) showed, that while catchments exposed to transboundary N and S deposition are characterised by relatively high N retention, sulphate is currently released in elevated amounts due to historic accumulation of S.

## Outreach

Integrated Monitoring Zöbelboden celebrated its 20 year anniversary in 2012 with an event targeted at the state and federal government, scientists and the wider public [www.umweltbundesamt.at/ueberuns/netzwerke/oekosystem\\_monitoring/20jahre\\_zoebelboden/](http://www.umweltbundesamt.at/ueberuns/netzwerke/oekosystem_monitoring/20jahre_zoebelboden/). As part of this event, a short movie was produced which is publicly available at [www.youtube.com/watch?v=i6KIU0POKkQ](http://www.youtube.com/watch?v=i6KIU0POKkQ) showing the monitoring program, its aims and results. The Austrian Focal Centre also contributed to the CLRTAP Assessment Report most prominently through its biodiversity expertise.

## Integration into EU research

Zöbelboden is also part of LTER Austria and LTER Europe. Owing to its excellent infrastructure and data, it was and is included in a number of national and EU research and research infrastructure projects (FWF DICE, ÖAW C-Alps, ACRP CCN-Adapt, ACRP CentForCSink, ACRP WoodNClimate, EU Life+ EnvEurope, EU SEE Orientgate, EU ExpeER, EU eLTER, EU Horizon2020 EcoPotential).

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## Annex 3

### Report on National ICP IM activities in Lithuania in 2015

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In 2015, tree crown defoliation at Lithuanian IM site Aukštaitija (LT01) was assessed 22 times, and at Žemaitija IM site (LT03) 21 times. After a period of increasing defoliation, the condition of monitored trees improved at both IM sites. This process continued during 2012-2015 period. At Aukštaitija the decrease in crown defoliation of birch trees was most significant and decreased from 22.7% to 14.5%. This improvement of crown condition was statistically significant ( $p < 0.05$ ). Decrease in mean defoliation of spruce and pine crowns were close to the level of significance and changed from 24.5% to 22.3% and from 16.8% to 15.8%, respectively.

Comparison of the last data on mean defoliation of the monitored trees in Žemaitija did not show significant change. Only during the last few years the defoliation of monitored trees started to improve. The reduction in mean defoliation of birch trees was the most significant from 19.2% to 12.4%. Improvement of spruce and pine crown condition was also statistically significant from 21.9% to 19.3% and 21.1% to 18.7%, respectively.

In 2015 crown condition of monitored trees at Žemaitija was one of the best over the whole period of investigation and reached 18.9%, meanwhile at Aukštaitija mean defoliation of all monitored trees was 19.7%.

Comparison of the long term research data on crown defoliation in the sites revealed that from the beginning of investigation up to 2004, the condition of trees in Aukštaitija in most cases was worse than that in Žemaitija. Afterwards, the situation changed and condition of trees in Žemaitija became worse than that in Aukštaitija. This period lasted until 2008. From 2009 to 2015 the condition of monitored trees in Aukštaitija deteriorated and was recorded to be worse than that in Žemaitija. Key factors contributing to such a variation in crown condition in the sites were first of all unfavourable climatic conditions (wind and snow breaks, and stem falls) resulting in outbreaks of *Ips typographus*. The effect of these factors was most pronounced in Žemaitija. Effect of 2015 drought on crown condition was not detected. Based on state of knowledge in this field, negative effect of this meteorological event can be expected to be detected only in the next year 2016.

The effect of air concentrations of the acidifying species on tree crown defoliation was found to be significant as well. In Aukštaitija key factors contributing to mean defoliation variation of Scots pine were: air concentrations of  $\text{SO}_2$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  ions. The correlation coefficients between these contaminants and pine defoliation exceeded 0.6 and were significant ( $p < 0.05$ ). Key factors contributing to mean defoliation variation of Norway spruce were: first of all air concentration of  $\text{NH}_4^+$  ions, followed by concentration of  $\text{SO}_4^{2-}$ .

Norway spruce trees were more sensitive to the effect of acidifying species at Žemaitija. It was the first time when the effect of air concentrations of ammonium and  $\text{SO}_2$ ,  $\text{SO}_4^{2-}$  ions on Norway spruce condition was found to be significant. Correlation coefficient exceeded 0.6. No significant relationships were established between the

defoliation of birch trees and air concentrations of acidifying species in the IM sites considered, which allowed to conclude that key factors contributing to tree crown condition of birch trees were climatic conditions. The effect of acid deposition had a remarkably lower significance on the tree species considered than that of air concentrations in both IM sites.

Chemical content of the needles and leaves are often presented as indicators of tree health. At Aukštaitija the 11 year long data set revealed that N concentrations in birch leaves had a tendency to decrease by 0.17g/kg per year, meanwhile in spruce and pine needles N concentrations fluctuated around 12 mg/g. Concentrations of P in monitored tree leaves and needles did not demonstrate significant trends. Tendencies towards increasing were detected in P concentrations in needles of pine and spruce – approximately by 0.03 mg/g per year, and the tendencies towards decreasing – in birch leaves by 0.06 mg/g per year. Most significant changes were detected in K concentrations in leaves and needles of the monitored trees in Aukštaitija. K concentration in spruce and pine needles increased significantly by 0.18 and 0.20 g/kg per year, respectively, and least significantly in birch leaves – by 0.14 g/kg per year. Concentrations of Ca started to decrease significantly in pine needles of the second year, by 0.18 g/kg per year. In remaining foliage samplers Ca concentration remains stable as well as Mg concentration.

In Žemaitija N concentrations in birch leaves and spruce and pine needles remained quite stable. P concentration remained stable also in needles and only in birch leaves demonstrated a decreasing tendency by approximately 0.05 mg/g per year. K concentrations increased in all foliage samplers, but statistically significantly only in pine needles by 0.22g/kg per year. Ca concentrations in needles demonstrated a decreasing tendency, meanwhile concentrations in birch leaves increased by 0.33 mg/g per year. Changes in meteorology and air concentrations of acidifying species together with their concentrations in precipitation were found to be responsible for the detected changes in leaves and needles at the considered IM sites.

Data on abundance of green algae on spruce needles indicated a more intensive pollution level by nitrogen species in Žemaitija than in Aukštaitija. Data on air concentrations of these species confirmed this bio indication.

From 2008 up to 2015 the abundance of epiphytic lichens increased indicating the improved ecological situation in Aukštaitija. The total coverage of monitored tree stems by epiphytic lichens exceeded 4%, 2% by *Hypogymnia physodes* (L.) Nyl. The specific composition remained stable during the entire period considered, i.e. 3 species were under investigation: *Hypogymnia physodes* (L.) Nyl., *Parmeliopsis ambigua* (Wulfen) Nyl., and *Cladonia* spp. (Hill.) Vain.

In Žemaitija the specific composition also remained stable over the entire period considered, i.e. 3 species are under investigation: *Hypogymnia physodes* (L.) Nyl., *Platismatia glauca* (L.) W.L. Culb. & C.F. Culb., and *Cladonia* spp. (Hill.) Vain. Key parameters resulting in these changes were hard to determine because data on air concentrations of sulphur species, as well as their wet deposition demonstrated a decreasing trend or were stable.



## Annex 4

### Effect of temperature and precipitation on the annual height increment of Scots pine on the Kandalaksha Gulf Coast and ICP IM site RU16

#### Report on National ICP IM activities in Russia

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The process of plant growth strongly depends on climatic factors. There are numerous dendrochronological investigations and issues concerning height and radial tree increment response to meteorological factors. It has been shown that there is a tight connection between time course of growth characteristics and total precipitation<sup>1,2,3,4,5</sup>.

Most dendrochronological studies consider mature trees; and the undergrowth aspect is usually not in the focus of interest of dendrologists. Our work deals with pine undergrowth, it does not require cutting of sample trees for tree height and internodes length measuring. Resulting dendrochronological data permit to reconstruct life conditions of former forest stands and response of trees to climatic impacts.

The main objective of this study is to analyze the dependence of Scots pine undergrowth height increment on temperature and precipitation variability in the current and previous years, and identification of climatic factors that are most important for the development of Scots pine undergrowth on Kandalaksha Gulf Coast, the White Sea (Murmansk region).

The studied area is located in the vicinity of Gorodetsky Ledge on Veliky Island (the Kandalaksha State Nature Reserve) of the White Sea Biological Station, Moscow State University (WSBS), on Cape Kindo (the Polarny Krug Local Nature Reserve) (ICP IM site RU16). The dependence of tree height increment on series of climatic variables during the growing season has been analyzed in Scots pine (*Pinus sylvestris* L.) growing in dry, mesic, and moist biotopes. Dry biotopes were presented by pine forests on the rocks. It is the most typical forest type on rock outcrops, where the soil cover is almost absent, as well as on the tops of well-drained glacial hills and ridges and on ancient alluvial marine terraces. Mesic biotopes were represented by smooth plateaus and the upper parts of slopes are usually covered by cowberry pine forests, with bilberry pine forests prevailing in the middle and basal parts of the slopes. And the moist biotopes hold areas with excessive (but not stagnant) moisture supply (in depressions, around lakes and bogs, etc.). They are characterized by dominance of hygrophytic mosses (mainly *Sphagnum* spp.).

The pine undergrowth was the object of investigation. The object trees were not older than 7 years and not above 2.5 m. The route survey method was used to establish sample plots. In total 45 sample plots (225 trees, 5 trees on each plot) were chosen. Standard dendrochronological methods were used.

For the statistical analysis the Openoffice Calc was used. The relationships between increment ranges and meteorological variables were to estimate the degree of dependence of inter annual fluctuations in tree height increment on climatic factors. The data



bases of the Federal Service for Hydrometeorology and Environmental Monitoring of Russia (Roshydromet)<sup>6</sup> and hydrometeorological station “Kandalaksha” were used for the study. The Pearson correlation coefficient was calculated; the confidence level was identified according to the manual<sup>7</sup>.

Ecosystem parameters, including soil, hydrological and vegetation conditions are homotypic for these areas. It was shown that all areas could be regarded as structural components of the same biogeocenosis ( $R = 0.496$  at a 90% confidence level, Fig. 1). Because of that dry, mesic, and moist biotopes on the both two coasts could be regarded as structural components of the same biogeocenosis.

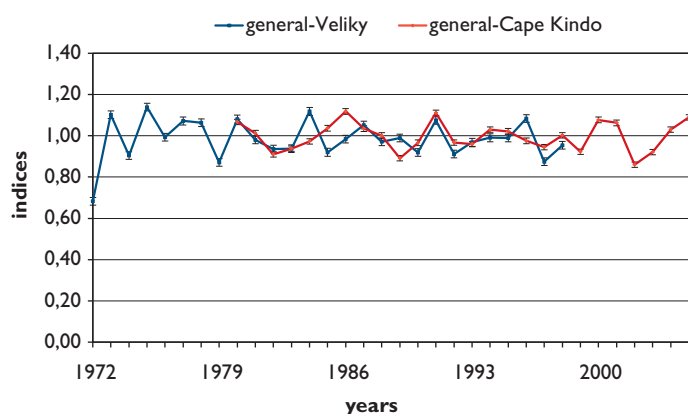


Figure 1. General averaged increment series on Veliky Island and Cape Kindo (two standard errors are shown).

Studies on Scots pine undergrowth in the Veliky Island (the Kandalaksha State Nature Reserve) and Cape Kindo (the Polarny Krug Local Nature Reserve) have shown that there was no common response of height increment to temperature among all tree stands studied (Table 1). The analysis results showed that the pattern of temperature dependence of tree height increment was determined primarily by various local factors (in particular, microclimate and soil cover).

Table 1. Significant correlations between series of height increment indices and meteorological variables in biotopes on Veliky Island and Cape Kindo in various moisture conditions.

Biotope type	Veliky Island				Cape Kindo			
	Temperature		Precipitation		Temperature		Precipitation	
	Current year	Previous year	Current year	Previous year	Current year	Previous year	Current year	Previous year
Moist	–	–	–	–	March (0.474)		June (–0.686) July (–0.415)	May (–0.432)
Mesic	April (0.333)		August (–0.325)				June (–0.390) July (–0.414)	
Dry	June (–0.485)	August (–0.341)	March (–0.491) May (–0.390) June (–0.350)	May (0.417)	March (–0.555)			April (0.367)

The relationship between fluctuations of height increment and meteorological variables in mesic biotopes of the studied region was characterized by a spectrum of parameters differing in sign and value. There was a high level of noise from individual variability of undergrowth and from local factors, which interferes with analysis of relevant general trends. Apparently, soil moisture was not the factor limiting tree growth in mesic biotopes. In contrast, this relationship in dry and moist biotopes was fairly distinct: tree height increments showed statistically significant negative responses to water stress (developing at elevated temperatures) in dry biotopes and to excessive atmospheric moistening in moist habitats.

Scots pine undergrowth condition either in sphagnum bogs or on coastal rock outcrops were critical, because of the excess of moisture in the former case, and its deficit in the latter case. In such biotopes where trees are at the threshold of survival, the most important factors for their growth can be revealed against the background of local noise and their relative significance can be evaluated. According to our data, a factor for Scots pine to survive in dry and moist biotopes on the Kandalaksha Gulf coast was atmospheric precipitation (its deficit or excess).

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## Annex 5

### Report on National ICP IM activities in Sweden 2014–2015

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*The programme is funded by the Swedish Environmental Protection Agency.*

#### Introduction

The Swedish integrated monitoring programme is run on four sites distributed from south central Sweden (SE14 Aneboda) over the middle part (SE15 Kindla), to a northerly site (SE16 Gammtratten). The long-term monitoring site SE04 Gårdsjön F1 is complementary on the inland of the West Coast and has been influenced by long-term high deposition loads. The sites are well-defined catchments with mainly coniferous forest stands dominated by bilberry spruce forests on glacial till deposited above the highest coastline. Hence, there has been no water sorting of the soil material. Both climate and deposition gradients coincide with the distribution of the sites from south to north (Table 1). The forest stands are mainly over 100 years old and at least three of them have several hundred years of natural continuity. Until the 1950's, the woodlands were lightly grazed in restricted areas. In early 2005, a heavy storm struck the IM site Aneboda, SE14. Compared with other forests in the region, however, this site managed rather well and roughly 20–30% of the trees in the area were storm-felled. In 1996, the total number of large woody debris in the form of logs was 317 in the surveyed plots, which decreased to 257 in 2001. In 2006, after the storm, the number of logs increased to 433, corresponding to 2711 logs in the whole catchment. In later years, 2007–2010, bark beetle (*Ips typographus*) infestation has almost totally erased the old spruce trees. In 2011 more than 80% of the trees with a breast height over 35 cm were dead (Löfgren et al. 2014) and currently almost all spruce trees with diameter of  $\geq 20$  cm are gone.

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

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

In the following, climate, hydrology, water chemistry and some ongoing work at the four Swedish IM sites are presented (Löfgren 2015).

## Climate and Hydrology in 2014

In 2014, the annual mean temperatures were higher (1.5–2.3 °C) compared to the long-term mean (1961–1990) for all four sites. Largest deviation occurred at the northern SE16 site. Compared with the measured time series, 15 years at site SE16 and 19 years at the other sites, the temperatures in 2014 were somewhat higher at all the IM sites. The values were the highest observed for the whole measurement period with exception for SE15 being slightly higher in years 1999 and 2000. Low temperatures were observed in 2010 and 2012. The variations between years have been considerable, especially for the last five years, over 3°C. Smaller variations were found at the central site SE15 Kindla, only 1°C.

Precipitation amounts in 2014 compared to the long-term average values (1961–1990) were for SE14 Aneboda 46 mm lower and for SE16 Gammtratten in the north 109 mm lower. For site SE04 Gårdsjön precipitation amount was very high exceeding the long term mean with 332 mm while the central Swedish site SE15 Kindla only showed marginally higher value with 29 mm. In 2012, the precipitation amounts were 3–44% higher than the long-term average for the four sites, while in 2013 all sites had lower values.

The characteristic annual hydrological patterns of the catchments are for the southern sites high groundwater levels during winter and lower levels in summer and early autumn. However, at site SE15 Kindla three summer peaks were noticed with groundwater levels 0.2 m below ground surface, i.e. on similar level as in spring and autumn. The groundwater level has decisive influence on the discharge values (Fig. 1).

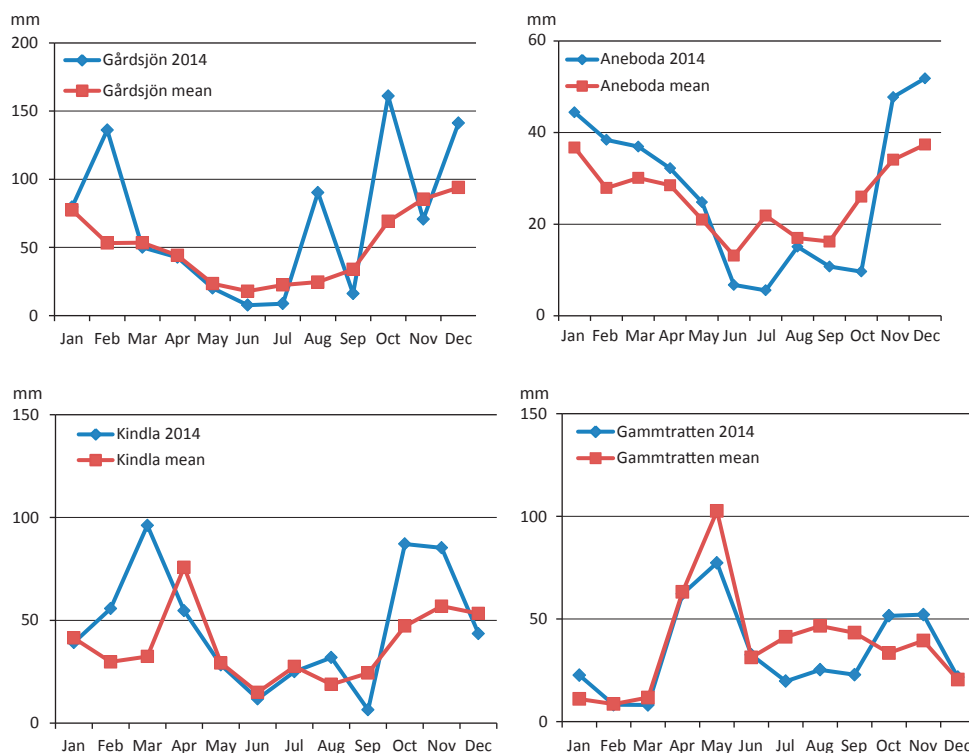


Figure 1. Discharge patterns at the Swedish IM sites in 2014 compared to monthly averages for the period 1996–2014 (mean). Note the different Y-axis scales.

In addition to precipitation, evapotranspiration affects the runoff pattern. In 2014, these patterns were fairly typical for the three sites Aneboda, Kindla and Gammtratten but high precipitation in August, October and December at SE04 Gårdsjön provided high monthly discharges and a flashy runoff pattern for late summer and autumn period. At site SE15 Kindla an early spring peak in March and high runoff in October and November pronounced the ordinary pattern for that area. For SE14 Aneboda in the south and SE16 Gammtratten in the north the patterns followed the long-term mean but with lower summer runoff than ordinary (Fig. 1).

At the two northern sites, generally snow accumulates during winter and the groundwater levels stay low furnishing low discharge. However, warm winter periods with temperatures above 0 °C have during a number of years contributed to snowmelt and runoff also during this season. As a consequence, the spring discharges have been comparably low during snowmelt, deviating from the conditions three decades ago. Apart from the early March peak at SE15 Kindla, this was not the case in 2014.

In 2014 high precipitation and runoff were observed at SE04 Gårdsjön even though interception and total evapotranspiration were high. At SE15 Kindla annual runoff 565 mm was reasonably high (Table 2), but the high precipitation (1300 mm) and throughfall (866 mm) values were out of the range compared with the national (SMHI) estimate 935 mm (sum of daily values). Hence, it is most probable that the site-specific measurements overestimated the actual conditions. A precipitation of 935 mm (national data) would result in evapotranspiration 370 mm being more in line with the long term average of ca. 450 mm. Based on the IM measurements, evapotranspiration would be 735 mm, which is out of the possible range.

Table 2. Compilation of the 2014 water balances for the four Swedish IM sites. Measured values on precipitation and throughfall at SE15 Kindla site could be biased regarding difficulties in snow collection. P – Precipitation, TF – Throughfall, I – Interception, R – Water runoff.

	Gårdsjön SE04		Aneboda SE14		Kindla SE15		Gammtratten SE16	
	mm	% of P	mm	% of P	mm	% of P	mm	% of P
Bulk precipitation, P	1294	100	633	100	1300	100	493	100
Throughfall, TF	847	65	687	109	866	67	517	105
Interception, P-TF	447	35	-55	<0	434	33	24	<0
Runoff, R	825	64	324	51	565	43	404	82
P-R	469	36	308	49	735	57	89	18

In 2014, the annual runoff made up 43–82% of the annual precipitation, which is comparable to the 40–60% found during previous years. The highest share was found at the northern site SE16 Gammtratten (82%), where partly due to a rather intense snowmelt period and cold climate during the rest of the year, yielding low evapotranspiration (18%) and consequently provided high runoff (Table 2). At site Aneboda (SE14), storm felling followed by bark beetle attacks have reduced the forest canopy cover, inducing low interception. Actually, the measured throughfall was higher than bulk precipitation. The total evapotranspiration was estimated to 308 mm, a value far lower than the long-term average 470 mm. This mirrors effects of low interception and transpiration of the reduced forest stand.

## Water chemistry in 2014

Low ion concentrations in bulk deposition (electrolytical conductivity = 1–2 mS m<sup>-1</sup>) characterise all four Swedish IM sites. The concentrations of ions in throughfall, including dry deposition, were higher at three sites. At SE16 Gammtratten, the conductivity in throughfall (1.2 mS m<sup>-1</sup>) was lower than in bulk deposition (1.4 mS m<sup>-1</sup>) indicating very low sea salt deposition and uptake of ions by the trees. At the two most southern sites, sea salt deposition provides higher ion concentrations especially

at the west coast SE04 Gårdsjön site (4.0 mS m<sup>-1</sup> in throughfall). The soil water pathways in the catchments soils are fairly short and shallow, providing rapid surface water formation from infiltration to surface water runoff. The acidity in deposition has during the last 10 years been rather similar at all sites with somewhat higher pH values (0–0.1 units) in throughfall compared with bulk deposition. However, in 2014 the two southern sites had throughfall values on 5.3 while the two northern sites only reached ca. 5.0 (Table 3).

Table 3. Mean deposition chemistry values 2014 at the four Swedish IM sites. S and N in kg ha<sup>-1</sup> yr<sup>-1</sup>.

	SE04	SE14	SE15	SE16
pH, bulk deposition	5.1	4.8	5.0	4.9
pH, throughfall	5.3	5.3	5.1	5.0
SO <sub>4</sub> -S, bulk deposition	4.6	2.4	3.4	1.4
N <sub>tot</sub> , bulk deposition	9.9	5.2	6.7	1.9

During the water passage through the catchment soils, organic acids were added and leached to the stream runoff. In the upslope recharge area, pH in the upper soil layers (E-horizon) was mainly lower than in throughfall. However, in the peat in discharge areas at SE15 Kindla and SE16 Gammtratten, pH was higher compared to throughfall while it was the opposite at SE04 Gårdsjön and SE14 Aneboda with pH 4.5 and 4.8, respectively. In the discharge areas, the buffering capacity in groundwater was fairly high with ANC over 0.04 mEq L<sup>-1</sup> and with bicarbonate (HCO<sub>3</sub><sup>-</sup>) present at Kindla and Gammtratten at average concentrations of 0.25 and 0.04 mEq L<sup>-1</sup>, respectively. The stream waters were acidic with pH values below 4.8 at all sites except Gammtratten having a pH of 5.7. The stream water buffer capacity was positive, except for at Kindla with an ANC of -0.001 mEq L<sup>-1</sup>. Anions of weak organic acids contributed to the positive ANC and bicarbonate contributed at SE16 Gammtratten.

The share of major anions in deposition was similar for sulphate, chloride and nitrate for three of the sites while chloride dominated at SE04 Gårdsjön due the proximity to the sea. In throughfall, organic anions contributed significantly at all four sites. The chemical composition changed during the passage of catchment soils and sulphate concentrations were higher in stream water compared with deposition, indicating desorption or mineralization of previously accumulated sulphur in the soils.

Besides effects on ANC and pH, the stream water chemistry is to a considerable extent influenced by organic matter. At Aneboda (SE14), the DOC concentration was high with 32 mg L<sup>-1</sup> while the other sites Gårdsjön (SE04), Kindla (SE15) and Gammtratten (SE16) showed lower values 16, 10 and 10 mg L<sup>-1</sup>, respectively. High DOC concentrations create prerequisites for metal complexation and transport as well as high organic nitrogen fluxes. The organic nitrogen concentrations in stream water ranged from 0.20 to 0.73 mg N L<sup>-1</sup>. The shares of N<sub>org</sub>/N<sub>tot</sub> were 72–96% with SE14 Aneboda having the lowest share and SE16 Gammtratten the highest. At SE14 Aneboda, the average concentration of inorganic nitrogen in stream water was 0.28 mg N L<sup>-1</sup>, which was high compared with 0.06 mg N L<sup>-1</sup> at the other sites. The high inorganic nitrogen concentrations at Aneboda are related to the forest dieback.

Total phosphorus (P<sub>tot</sub>) in bulk deposition varied between 3 µg L<sup>-1</sup> and 14 µg L<sup>-1</sup> with the highest values at SE04 Gårdsjön with influence of sea deposition. In stream water, however, P<sub>tot</sub> was highest at SE14 Aneboda with 23 µg L<sup>-1</sup> also having the highest DOC concentrations. The other sites had average P<sub>tot</sub> concentrations between 4 µg L<sup>-1</sup> and 13 µg L<sup>-1</sup> with SE16 Gammtratten being highest.

Inorganic aluminum, toxic to fish and other gill-breathing organisms, has been analyzed in soil solution, groundwater and surface waters at the IM sites. Relatively high total Al concentrations occurred in the soil solution (0.6–2.8 mg L<sup>-1</sup>) as well as



in stream water (0.24–0.7 mg L<sup>-1</sup>) at the three southern sites with low pH (4.5–4.8). At the northern site SE16 with a pH of 5.7, the total Al concentrations were low, approximately 0.2 mg L<sup>-1</sup>. Inorganic Al made up 17–45% of the total Al at the four sites, corresponding to 0.11–0.25 mg Al<sub>i</sub> L<sup>-1</sup> at the three southern sites with low pH and 0.05 mg Al<sub>i</sub> L<sup>-1</sup> at the northern site Gammtratten. Those levels are considered extremely high at the three southern sites and high in the north, according to the SEPA classification system. The priority heavy metals Pb, Cd and Hg were still accumulating in the catchment soils, while the stream concentrations were low compared with the levels causing biological effects. However, methyl mercury was still relatively high creating prerequisites for bioaccumulation.

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

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

This report 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 2015/2016 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
- A report on dynamic vegetation modelling at ecosystem monitoring and research sites
- An interim report on trend assessment for deposition and runoff water chemistry and climatic variables at ICP IM sites in 1990–2013
- National Reports on ICP IM activities are presented as annexes.



ISBN 978-952-11-4588-9 (pbk.)

ISBN 978-952-11-4589-6 (PDF)

ISSN 1796-1718 (print)

ISSN 1796-1726 (online)