Trends in ecosystem and health responses to (reductions in) long-range transported atmospheric pollutants

Comment [HWI1]: Not all pollutants have been reduced?

ICP Waters: Heleen de Wit, Øyvind Garmo, Gaute Velle, Arne Fjellheim
ICP IM: Martin Forsius, Lars Lundin
ICP Forests: Walter Seidling, and others
ICP Vegetation: Harry Harmens, Gina Mills
ICP M&M: Anne-Christine Le Gall, Jean-Paul Hettelingh, Maximilian Posch
ICP Materials: Johan Tidblad
TF Health: Marie Eve Héroux
JEG: Filip Moldan
EMEP / TF MM: Laurence Rouil
EMEP/CIAM: Markus Amann, …
EMEP/MSCW: Hilde Fagerli, … and EMEP/MSCE: Ilia Ilyin, Marina Varygina…

To be discussed in Geneva - to all ICP's

1. Read the summary, and check if the main conclusions/points from your contribution are there. Provide missing conclusions. If your contribution does not provide any conclusions about responses to air pollution, consider adding a few lines to your text.
2. Materials and methods: please add a few introductory lines about your ICP/TF H/JEG. Keep the length to one page. Look at par 2.8 (ICP Materials) for inspiration.
3. References: use references in your text, not too many, not too few. Check if your references are included in the list at the end, if not provide a separate document with your references.
4. Figures: look critically at your own figures. Too small fonts are not acceptable! Make sure that the axes and all text can be read, that lines are thick enough, etc. If possible, make such a lay-out for your figure that it can be used in powerpoints later.
5. Results: see comments in the text. In general, aim for a concise text with not too much detail (refer to papers and reports for details.) Our audience are non-experts – for one, your fellow ICP/JEG/TF H leaders should be able to understand fairly easy what your contribution is about, and national and international policymakers too. Think also about policy relevance and interpret your results in terms of deposition or exceedance of critical loads (thus, our common LTRAP language). Just add bullet points to the text – we can remove them later.
6. Do you miss anything? For instance - POPs: the only contribution on POPs so far is from ICP Waters. Is that all we have?
7. Authors: so far I have not added author names to each contribution. I suggest that we add an appendix or a foreword where all authors are acknowledged for their contribution, if you all agree. Please supply text.
8. The TF M&M and EMEP have been asked for a contribution, and hopefully we can discuss this further in Geneva. Perhaps it is possible to take some content from the Assesment Report?
9. Other missing pieces: introduction, preface, discussion. Need for lead authors.
10. New suggestion for dates: 1. Contribution from EMEP/TF MM by April 20; 2. Preface, introduction+discussion, summary by May 10; 3. Feedback on especially summary by May 30. Edited report ready by end of June.

Summary/executive summary

This report presents trends in central ecosystem and health responses to air pollutants, more specifically sulphur and nitrogen as acidifying agents, nitrogen as a nutrient, heavy metals, POPs and ozone. All bodies under the Working Group on Effects under the Convention of Lang-range Transported Pollution contributed with their specific findings for each pollutant or group of pollutants. Most focus is given to the period between 1990 and 2012.

Sulphur and nitrogen - acidification

The area exceeded by critical loads for acidification reached it maximum in 1970 for aquatic ecosystems, when 40% of all ecosystem area in Europe was exceeded. For terrestrial ecosystems, the maximum peak in area exceeded was 30% and occurred in 1980. Peak mean exceedance for terrestrial and aquatic ecosystems was 350 and 250 eq ha^{-1} yr^{-1}, respectively. Currently, the area with exceeded critical loads for terrestrial and aquatic ecosystems in Europe is 9% and 5%, respectively, with an expectation for further decrease towards 2020, while the mean acidity exceedance for Europe has declined to below 10 eq ha^{-1} yr^{-1} for terrestrial and aquatic ecosystems. However, there is considerable variation within Europe with regard to regional exceedance of critical loads.

-here, it would be logical to have a few sentences about extensive damage to forests and fish (and buildings?) in the 1970s and 1980s, but no such text is provided in the results.

Trends in forest ecosystem responses are given for crown condition (assessed by defoliation), foliar nutrients contents. Defoliation is an average below 25% for most tree species in Europe, but shows for some Mediterranean tree species an upward trend which could be a result of ozone damage. However, defoliation is not a specific response to air pollution only, and other factors such as drought may also play a role. Downward trends in foliar contents of N and S are predominant for many tree species, in addition to essential nutrients such as P. Whether these changes in foliar chemistry relate to deposition of S and N or to increased forest growth is not evident. *Is it possible to conclude with a statement on current forest condition, and whether it has benefited from reduced loads of S and N?*

Trends in aquatic ecosystems are documented for acid-sensitive lakes and streams in Europe and North America. Sulfate concentrations have decreased with on average 45-55% since 1988 as a result of decreased sulfate deposition. This has led to a widespread chemical recovery of surface waters, which opens the possibility for biological recovery of acidified surface waters. Biological records document that biological recovery of acid-sensitive waters is indeed happening, primarily as a result of improved water quality. However, species diversity in fully restored aquatic ecosystems can be much higher than is presently observed in aquatic systems that are under recovery from acidification, and full biological recovery may not be possible in most ecosystems.

Nitrogen contributes little to acidification of surface waters, as documented by catchment input-output budgets for nitrogen where less than 10% of annual inputs leach from soils to streams. There is no sign of accelerated leaching of nitrogen from catchments. For sulphur, however, there is a tendency to increasing % export, indicating a net release of previously stored SO4, particularly during the past 15 years.

With regard to effects of acidic deposition on materials, corrosion has decreased substantially to around 50% of the original values measured in 1987. In recent years, however, the improvements in corrosion are minor. The observed trends are similar for rural, urban and industrial sites, although the absolute corrosion levels are highest in industrial and lowest in rural sites.

Conclusions I-O budgets ICP IM

Key conclusions:

Comment [HWI2]: No information on North America. It seems only ICP Waters reports from North America.

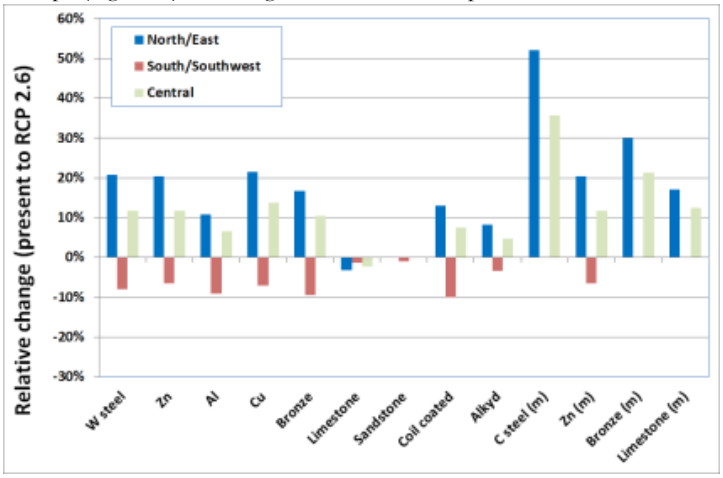
Comment [HWI3]: Appearantly, crown condition is not anymore related to acidification? Then perhaps put it somewhere else?

Comment [HWI4]: The chapter on ozone in forest indicates declining ozone concentrations. Please comment.

- ICP IM sites are now releasing sulphur that had accumulated in the past. This indicates recovery of the terrestrial systems but may delay the recovery of the surface waters.
- The more efficient retention of nitrogen than sulphur results in generally higher leaching fluxes of SO_4 than those of NO_3 in European forested ecosystems.
- Sulphate thus remains the dominant source of actual soil acidification despite the generally lower deposition inputs of S than N.
- The evaluation of ICP IM data, as well as other recent regional studies, does not show any signs of consistent and widespread regional increases in nitrate concentrations in surface waters in sensitive background areas, despite continuing N accumulation in the catchments.
- Many of these S and N retention processes are sensitive to changes in climatic variables, and would therefore be affected by future climate changes.

Input on conclusions from Johan Tidblad:

- Corrosion has decreased substantially to around 50% of the original values measured in 1987. In recent years, however, the improvements in corrosion and soiling are minor. For real cultural heritage objects made of metals the decreases are instantaneous, responding rapidly to decreasing pollution levels. For stone materials, however, there is a substantial time lag, 20 years or more, before improvements can be seen.
- The differences between polluted and non-polluted areas are not as high as in the 1980's but are still significant. At the most polluted sites 2020 targets are exceeded.
- Depending on the area, climate change can either increase or decrease the risk of corrosion. The risk is increased in North/East and Central Europe while it is decreased in South/Southwest Europe (Figure X). The magnitude of the risk depends on the chosen scenario.



- Figure X Calculated relative changes in corrosion due to SO_2 for the IPCC RCP2.6 scenario.

Comment [HWI5]: We can't have new figures in the summary. Is it possible to add this figure to the results?

-paragraph on health responses to be added

Nitrogen as a nutrient

-trends in exceedance of CLs from ICP M&M, and add a few words from results ICP IM on CLs?

- -trends in vegetation: The study based on long-term ICP IM and ICP Forests data showed that cover of oligotrophic plant species have decreased in European forest ecosystems, but diversity is still not affected by airborne N deposition.

-catchments soils continue to be enriched in N, as I-O element budgets for catchments show. No evidence for enhanced leaching of nitrate from catchment soils, as evidenced by water chemical records

Comment [HWI6]: I don't understand? Doesn't the figure below document damage from airborne N dep on species diversity?

-paragraph on health responses to be added

Heavy metals

-exceedances of CLs

-trends in records of HM in mosses

-trends in HM in soils and streamwaters

-trends in Hg in fish

--paragraph on health responses to be added?

POPs

-very little material, only something on POPs in fish

PM and ozone effects on human health

Air quality is the largest contributor to the burden of disease caused by environmental factors. The already strong evidence on the adverse effects on health of ambient air pollutants, such as particulate matter and ozone, has evolved in the last three years. Also, the monitoring and modelling of exposure to air pollution is reviewed continually. In general, indicators of exposure to the ambient air pollutants particulate matter (indicators PM_{10} and $PM_{2.5}$, indicating particulate matter with an aerodynamic diameter smaller than 10 µm and 2.5 µm, respectively) and ozone (indicator SOMO35, which stands for the sum of ozone means over 35 ppb) in the European Region have not changed substantially over the last few years. In countries in the eastern part of the Region, monitoring is very limited.

-soiling

Ozone

-vegetation

-health

-forest

Concluding remarks on where air pollution policy has been most effective, and where currently the main challenges remain

Summary for policy makers

-to be considered

Preface

At the xxth Session of the Working Group on Effects, in xxx, it was proposed that the Working Group and its Programmes prepare a Report on trends in the effects of air pollutants. Most Programmes have been in operation for 10 years or more and it is now possible to begin to identify benefits that might result from the decreases in emissions of some major air pollutants in recent years.

This report was produced by members of the Bureau of the Working Group on Effects and the Programme Centres of the international programmes of the Working Group. It was prepared at the request of the Working Group on Effects and the Executive Body of the Convention and is submitted to the xxxth Session of the Working Group for its consideration. The preparation of the report was funded by the lead countries of the international programmes, by voluntary contributions from other Parties to the Convention, and by a specific contribution from the European Commission earmarked for the production of the report.

The report is aimed at providing some of the scientific evidence for trends in long range transboundary pollution and its effects on the environment in recent years. It focuses in particular on the results collected by the international programmes operating under the Convention, bringing together their individual conclusions to provide an overview on trends for the ECE region.

The long-standing support by Prof. A. Eliassen and his colleagues from the EMEP/MSC-W is gratefully acknowledged.]

Peringe Grenfelt

Chairman, Working Group on Effects

Comment [HWI7]: Peringe should probably write this. This text was copied from the 1999 WGE trend report.

Comment [HWI8]: It'd be nice to include EMEP

Table of Contents

Contents

1. Introduction (Heleen, Max, Harry, Jean-Paul, …) (1-2 pages)

-Effect-based approach under CLTRAP, + relationship to Long-term Strategy of the Convention (to describe the "trend" in the effect based approach towards from 1987 to 2020)

- Trend assessments of environmental and health responses are a powerful tool to demonstrate if a) policy to reduce atmospheric pollutants has had its intended effect, (b) what the status is of recovery compared to a reference year or to an environmental "desired" reference state.

-aim of this trend assessment: Describe where we stand relative to "It is not over till it's over"; input to assessment report; Formal Mohaha with respect to LTS and workplan

From the Longterm strategy:

The Working Group collects, assesses and further develops environment and health related knowledge and information on: (a) The present status, long-term trends and dynamics, as well as the degree and geographical extent, of the impacts of air pollution, in particular but not exclusively its long- range transboundary impacts; (b) Exposure-response relationships for agreed air pollutants; (c) Critical loads, levels and limits for agreed air pollutants, and their links to observations; (d) The linkages between the effects of air pollution, biodiversity and the effects of changes in climate and land use.

-workplan 2014-2015, point 1.8: Improve functioning of the Working Group on Effects and EMEP and their subsidiary bodies (ICPs, task forces). Under 1.8.1:

1.8	**Improve functioning of the Working Group on Effects and EMEP and their subsidiary bodies (ICPs, task forces)**			
1.8.1	Foster integrated/thematic assessments, combining the work and output of different subsidiary bodies; identify subject areas for future integrated/thematic assessments	Continue production of integrated/thematic reports, with short summaries (brochures) providing key messages aimed at policy makers and synthesis papers in scientific journals for the science community (improving internal/external communication)	EMEP and Working Group on Effects including ICPs and other subsidiary bodies	—

2. Materials and Methods (written by each ICP/body – 1 page per icp – refer to existing reports/papers to shorten this section)

The Working Group on Effects provides information on the degree and geographic extent of the impacts on human health and the environment of major air pollutants, such as sulphur and nitrogen oxides, ozone and heavy metals. Its six International Cooperative Programmes (ICPs) and the Task Force on Health identify the most endangered areas, ecosystems and other receptors by considering damage to human health, terrestrial and aquatic ecosystems and materials. An important part of this work is long-term monitoring. The work is underpinned by scientific research on dose-response, critical loads and levels and damage evaluation. Below, each ICP and the Task Force on Health is described shortly with regard to monitoring network, main data collection and methods to calculate trends.

2.1. EMEP (1 page, including map of stations)

Emissions/deposition

Comment [HWI9]: A contribution from EMEP requested by email 12.12.2014. Rouil: no time to answer. Perhaps possible to get some figures from the Assessment report in here?
-EMEP contribution to be discussed in Geneva

2.2. ICP Modelling and Mapping (<1 page)

The ICP Modelling and Mapping (ICP on Modelling and Mapping of Critical Loads and Levels and Air Pollution Effects, Risks and Trends; ICP M&M) has as its main focus in the development of modelling and geographic mapping methodologies for the assessment of current and future air pollution related effects in Europe. Results by its Coordination Centre for Effects (CCE) are incorporated in the GAINS model of EMEP-CIAM to analyze in an integrated manner costs and benefits of abatement policies of both the LRTAP-Convention and the European Commission.

Trends of the difference between atmospheric depositions and critical loads (i.e. critical load exceedance) are based on the state of knowledge in 2013. Modelled deposition data are from EMEP MSC-W[1] and critical loads from the Coordination Centre for Effects (CCE) under the ICP Modelling & Mapping. The EMEP model (Simpson et al. 2012) produces acidic and eutrophying depositions for use in integrated assessment on a 0.50° ×0.25° (about 28×28 km²) longitude-latitude grid. In anticipation of the increased resolution of the EMEP model, National Focal Centres (NFCs) responded to a CCE call for data to update their national critical loads for the European critical load database for acidification and eutrophication during 2010–12 (see Posch et al. 2012).
Depositions of heavy metals have been computed by EMEP-MSCE (Ilyin *et al.* 2009) while critical loads and exceedances have been compiled by the CCE (Hettelingh *et al.*, 2015)

This enabled the calculation of exceedances and (terrestrial and aquatic) ecosystems at risk caused by depositions from emissions under the revised Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (UNECE 2012a,b; Reiss et al. 2012) taking also into account current legislation in 2020 (GP-CLE scenario). This emission scenario was provided by the Centre for Integrated Assessment Modelling (CIAM) of the Task Force on Integrated Assessment Modelling (TFIAM) of EMEP.

Historic emissions since 1880, used in this chapter to illustrate the long-term trend of critical load exceedances, have been derived from Schöpp et al. (2003). The historical trends of critical load exceedances (e.g. Hettelingh et al. 2013) are based on deposition patterns computed with different versions of the EMEP model. This chapter partly bears on CCE work performed under the LRTAP Convention (ICP M&M 2013; WGE 2013a,b) and for the European Environment Agency (EEA 2014a).

Exposure in a natural area for which critical loads are available is calculated as the Average Accumulated Exceedance (AAE; Posch et al. 2001; 2015), i.e. area-weighted average of the exceedance of all critical loads in an area. The AAE can be computed for any region, i.e. for all natural areas in a country, for any class of natural areas (EUNIS classification; Davies and Moss 1999). In this section a distinction is made between terrestrial and aquatic ecosystems.

2.3. JEG

JEG does not have stations – leave out, possibly. Or just describe JEG approach.

[1] Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP), Meteorological Synthesizing Centre West (MSC-W) at the Norwegian Meteorological Institute.

2.4. ICP Forests: (1 page, including map of stations)

The obvious worsening of forest condition in the 1980s over parts of Europe gave reason for the large-scale assessment of tree crown condition starting in 1985 under the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests). Soon the need to develop empirical models on cause-effect relationships became apparent and the intensive ecosystem-oriented Level II monitoring was started in 1994. Both parts of the programme, which were installed at least temporarily in 42 European countries, have produced plenty of valuable data from in total 13 surveys (Tab. 2.4.-1), all collected according to a harmonized manual (ICP Forests 2010, Ferretti & Fischer 2013).

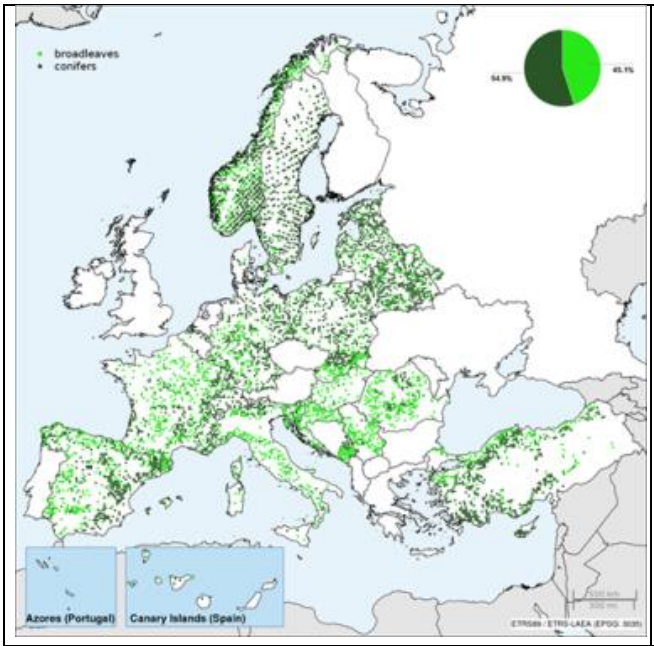


Fig. 2.4-1:Plots of the extensive Level I monitoring in 2013.

Fig. 2.4-2: Plots of the intensive Level II monitoring.

Observing air pollution impacts on forests requires both, the monitoring of crucial substance fluxes into and through the ecosystems and the recording of plant responses. To suffice the first, on-side measurements of air quality aspects, mostly by passive samplers (Chap. x on ozone), and acquisition of key pollutants like nitrogen and sulphur in the open-field and beneath the canopies are part of the intensive monitoring programme. Fate and interactions of these substances in forest soils can be described by collecting and analyzing soil solution in different depths which is also part of the Level II programme. Various responses of trees are studied: Nutrient contents of tree foliage estimated on both Level I and II reflect atmospheric and soil-related influences (Chap. x.4 on foliar). Performance parameters like crown condition on Level I and II (Chap. x.5 on crown condition) or radial stem growth on Level II are influenced by all kinds of environmental and biotic stressors. Both are unspecific and their values and developments in time have to be interpreted with care. Other ecosystem response studied on Level II (and partly Level I) plots are ground vegetation diversity and epiphytic lichens on tree stems at some sites tentatively.

Tab. 2.4-1: Surveys performed on ICP Forests plots (optional)

Survey	Assessment frequency	Survey	Assessment frequency
Crown condition	Annually	Soil condition	Every 10 years
Foliar chemistry	Every 2 years	Soil solution chemistry	Continuously
Tree growth	Every 5 years	Meteorology	Continuously
Ground vegetation	Every 5 years	Litterfall	Continuously
Ozone induced injury	Annually	Leaf area index	Occasionally (annually)
Ambient air quality	Continuously	Phenology	Several times per year
Deposition	Continuously	Epiphytic lichens	Once, pilot project

2.5. ICP Integrated Monitoring (1 page, including map of stations)

XX ICP on Integrated Monitoring Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM)…

Comment [HWI10]: This needs a bit more.

Comment [HWI11]: A short description of ICP IM

Presently there are 46 ICP IM sites from 16 countries with on-going data submission (Figure XX). The integrated monitoring of ecosystems refers to the simultaneous measurement of physical, chemical and biological properties of an ecosystem over time and across compartments at the same location. In practice, monitoring is divided into a number of compartmental subprogrammes which are linked by the use of the same parameters (cross-media flux approach) and/or same/close stations (cause-effect approach). Details are provided in the ICP IM manual (www.syke.fi/nature/icpim). For the trend assessments presented in this report, the sites have been selected based on data availability.

Figure XX. Location of the ICP IM sites *(compressed version for draft report)*

2.6. ICP Waters

ICP Waters (ICP on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes) is a programme for monitoring of the effects of acid rain and air pollution on water and water courses. Twenty countries (18 European countries, USA and Canada) participate on a regular basis and supply monitoring data on water chemistry and biology to ICP Waters databases. Sites – rivers and lakes- for ICP Waters are selected by the national focal centres and are chiefly located in catchments that are sensitive to effects of air pollution. The catchments must be without impacts from local point sources of pollution with a direct impact on water quality, for instance sewage, agriculture and industry. Currently, the database for water chemical records consists of circa 200 sites in Europe and North America with records from 10 to over 30 years. The database for biology consists of circa 50 rivers and 40 lakes in Europe with records starting during the 1980s and 1990s. The database for biology is a subset of the database for water chemistry.

Figure x. Monitoring stations in the ICP Waters programme. The stations are grouped according to region.

The water chemical database includes water chemical variables that are suitable for documenting responses to changes in deposition of sulphur and nitrogen:

1) **SO_4^{2-}** and **NO_3^-**, the acid anions of acidic deposition. Trends in the concentrations of these anions reflect recent trends in deposition (especially SO_4^{2-}) and in ecosystem response to long-term deposition (e.g., NO_3^-).
2) **Base cations:** (Ca^{2+} + Mg^{2+}) are mobilised by weathering reactions and cation exchange that neutralise acids in watersheds. Base cations will respond indirectly to changes in SO_4^{2-} and NO_3^-.
3) **Acidity,** including **pH**, **measured (Gran) alkalinity** and **calculated ANC**, reflect the outcome of interactions between changing concentrations of acid anions and base cations.
4) Concentrations of **dissolved organic carbon (DOC)** or alternatively **total organic carbon (TOC)**. These are considered as surrogates for organic acids, mostly derived through degradation of natural organic matter in catchment soils.

Concentrations of SO_4^{2-} and base cations are usually sea-salt corrected (denoted by an asterisk (SO_4^*, $(Ca+Mg)^*$), to distinguish between natural and anthropogenic emission sources.

The biological database includes time series of invertebrate data (insects, snails, zooplankton etc.) from rivers and lakes. These groups of organisms consist of species with a wide range in acid-sensitivity, have a short life-span and are therefore suitable for monitoring biological responses to changing water quality.

2.7. ICP Vegetation (1 page, including map of stations)

The ICP on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation)...

Comment [HWI12]: Some words here, and too what type of ecosystems you focus on. Vegetation sounds like forest?

In recent decades, naturally growing mosses have been used successfully as biomonitors of atmospheric deposition of heavy metals (Harmens et al., 2010, 2015). Since 1990, the European moss survey has been repeated at five-yearly intervals. Since 2005, the nitrogen concentration in mosses was also determined (Harmens et al., 2011, 2015).

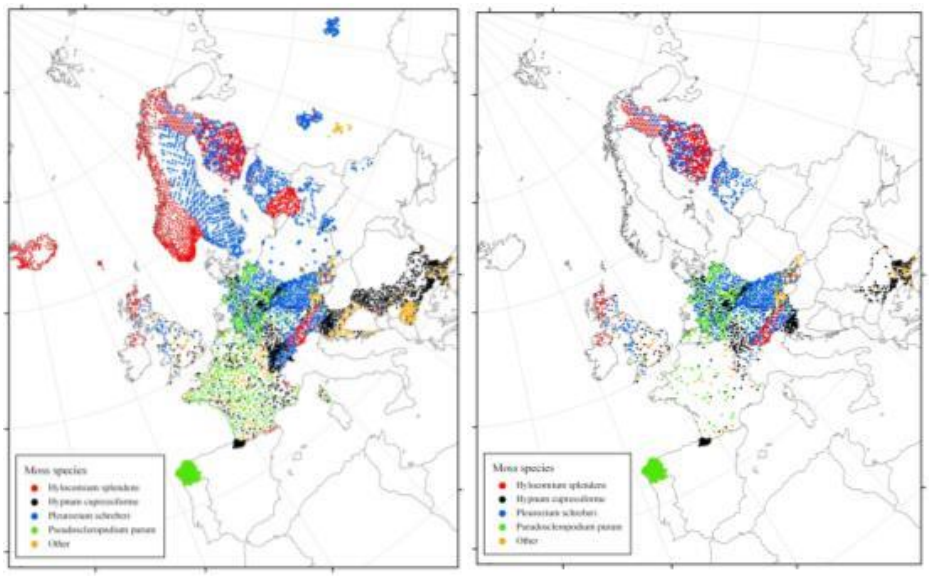


Figure 1. Moss sampling sites for determination of heavy metal (left) and nitrogen (right) concentrations in mosses in 2005 (Harmens et al., 2010, 2011).

Monitoring sites for impacts of ozone on vegetation

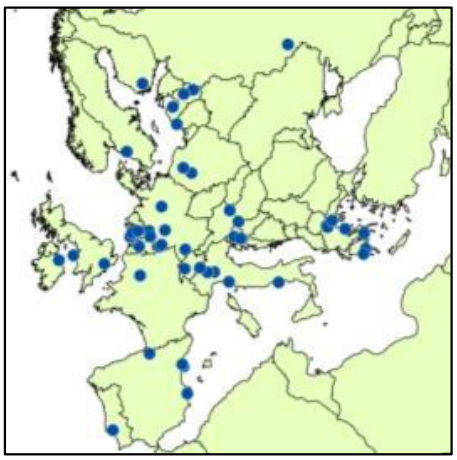


Figure 2. Sites for monitoring ozone impacts on white clover between 1996 and 2006 (Hayes et al., 2007).

Since 1990, the ICP Vegetation has been monitoring the impacts of ambient ozone on vegetation in biomonitoring experiments with species such as subterranean clover, white clover (Figure 2; Hayes et al., 2007; Mills et al., 2011), brown knapweed and French bean. The participation in those experiments varied by year has declined strongly in the last decade.

2.8. ICP Materials

The evaluation of long-term trends of corrosion and soiling is one of the main aims of ICP Materials together with the development of dose-response functions and use of results for mapping and calculation of corrosion costs at UNESCO cultural heritage sites. The monitoring programme of ICP Materials is based on racks with materials located in rural, urban or industrial sites (Figure X). Different materials are exposed and evaluated for corrosion attack and soiling including but not limited to the indicator materials carbon steel, zinc, limestone and modern glass. Exposure of materials are typically performed each third year with an exposure period of at least one year. The latest exposure was started in the fall of 2014.

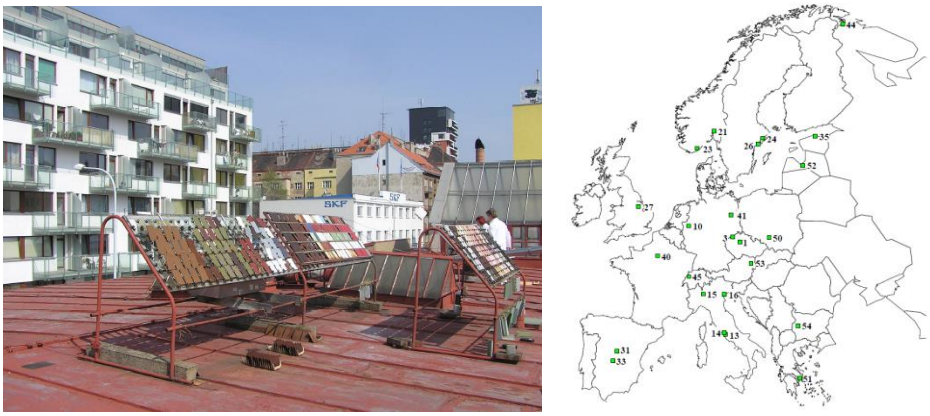


Figure X. Typical urban test site with materials on a rack (left) and map of test sites (right).

In addition to measurements of corrosion, environmental data are collected at each site as indicated in table X. The reported parameters have varied with the evolution of the programme. In the beginning focus was acidifying pollutants, especially SO_2, which is still one of the most important parameter for corrosion. Later on, focus has shifted to other pollutants, including HNO_3 and particulate matter. Further general information about ICP Materials can be found in Tidblad et al 2012.

Table X. Reported environmental data from ICP Materials test 1987-2012: X = mandatory; (X) = optional; - = not reported

Parameter	Symbol	Unit	1987-1995	1995-2001	2002-2003	2005-2012
Temperature	T	^{o}C	X	X	X	X
Relative humidity	Rh	%	X	X	X	X
Time of wetness	Tow	h	X	-	-	-
Sunshine	Sun	h	X	-	-	-
Sunshine[a]	Sun	MJ m^{-2}	X	X	X	-
SO_2 concentration	SO_2	µg m^{-3}	X	X	X	X
NO_2 concentration	NO_2	µg m^{-3}	X	X	X	X
O_3 concentration	O_3	µg m^{-3}	(X)	(X)	X	X
HNO_3 concentration	HNO_3	µg m^{-3}	-	(X)[b]	(X)	X
Precipitation: amount	Prec	mm	X	X	X	X
-: conductivity	Cond	µS cm^{-1}	X	X	X	(X)
-: pH	pH	-	X	X	X	X
-: SO_4^{2-}, NO_3^-, Cl^-	varies	mg l^{-1}	X	X	X	X
-: HN_4^+, Na^+, Ca^{2+}, Mg^{2+}, K^+	varies	mg l^{-1}	(X)	(X)	(X)	(X)
Particulate matter	PM	µg cm^{-2} $month^{-1}$	-	(X)[b]	(X)	X

[a]Calculated from sunshine hours and latitude. [b]Only data at a few test sites reported

2.9. Task Force on Health (1 page)

In order to assess exposure of the population to air pollutants and estimate the impacts on public health, the Joint Task Force on Health Aspects of Long-range Transboundary Air Pollution (TF Health) relies on data from air monitoring networks operated by national authorities.

Particulate matter and ozone are the usual indicators for health effects related to air pollution. Industries, households, and traffic produce complex mixtures of air pollutants, many of which pose risk to health. Of all of these pollutants, fine particulate matter (PM2.5) has the greatest long-term and short term effect on human health. PM2.5 shows the most consistent associations with mortality and morbidity such as lung cancer, hospitalization for cardiovascular and respiratory disease, restrictions in activity, and acute and chronic bronchitis. Ozone produces short-term effects, independent of other air pollutants such as particulate matter, on mortality and respiratory morbidity. Measurements of PM and ozone in ambient air can serve as an acceptable "proxy" for population exposure. Nonetheless, other air pollutants such SO2, NO2 can be of interest in identifying trends because they are PM precursors and contribute to PM concentrations (and related population exposure).

For particulate matter in ambient air, population exposure is reflected in the indicator reported by combining data on PM_{10} or $PM_{2.5}$ concentrations with the size of population exposed. Traditionally, there are more monitoring stations measuring PM10 than measuring PM2.5. Exposure to PM is usually calculated data from urban background locations. "Ideally, the monitoring data used to calculate the average annual PM concentrations should be collected throughout the year, for several years, to reduce bias owing to seasonal fluctuations or to a non-representative year. Care should be taken that the monitors used are not unduly influenced by a single source of pollution (i.e. a power plant, factory or highway); rather, the monitors should reflect exposures over a wide area. Although it is likely that PM data will be easily available for larger cities, residents of agglomerations and of rural areas are also exposed to PM from local industrial activity, transportation, biomass fuels, open burning and regional haze" (see WHO - Global Health Observatory Data Repository: http://apps.who.int/gho/indicatorregistry/App_Main/view_indicator.aspx?iid=1349).
Included in the calculations are national estimates which are calculated as population-weighted means of annual average concentrations in cities of PM2.5 or PM10.

The indicator reported for ozone reflects the cumulative annual exposure to ozone measured in urban background locations. SOMO35 is expressed as $\mu g/m^3$ (or ppb) × days, and can be used as an indicator for the quantification of the cumulative yearly health impacts of ozone. National estimates are calculated as population-weighted annual means of city-level accumulated maximum daily 8-hour average ozone concentration in excess of 35 ppb .

The selection of PM and Ozone as indicators, methodologies to calculate such indicators, source of data, geographical coverage, period coverage, frequency of update and data quality can be found elsewhere (WHO Regional Office for Europe, 2015).

3. Results

3.1. Sulphur and nitrogen (acidification) (ca 10-15 p)

3.1.1. Trends in atmospheric concentrations and deposition of S and N 1990-2012 for Europe (and North America) (max 2 p)

Comment [HWI13]: To be added

-graph of mean annual concentrations of SO4 and NOx in Europe from 1990-2012 (or from 1980 or 1970, if possible), including trend
-grap of mean annual deposition of S and N from 1990-2012 (or from 1980 or 1970, if possible).

Text – on % decrease in deposition and concentrations, which relate to reduce emissions.
For instance:

Sulphate

Comment [HWI14]: Example from ICP W report 106

The decrease in sulphur emissions in both Europe and North America have caused a substantial decrease in the atmospheric deposition the last decades, and this has been reported in several assessments and publications (i.e. EMEP, 2004; IJC 2008; Vestereng, 2007; Sickles, 2007). These reductions are naturally also reflected in precipitation chemistry. All the sites in Europe and 97% of the sites in northeastern North America show a decreasing trend from 1990-2008, with an average reduction of 56% and 37% respectively (**Error! Reference source not found.**).

-please supply a draft text and figures

3.1.2. Trends in exceedances of CLs for acidification (CCE) (max 2 p)

Comment [HWI15]: add nrs for 2012 in the text?

The risk of acidification in Europe has diminished significantly between 1990 and 2010. The broad areas at risk of high exceedances (red and orange shading) in 1990 exhibit relatively low exceedances (blue shading) in 2010 (Figure CCE-1).

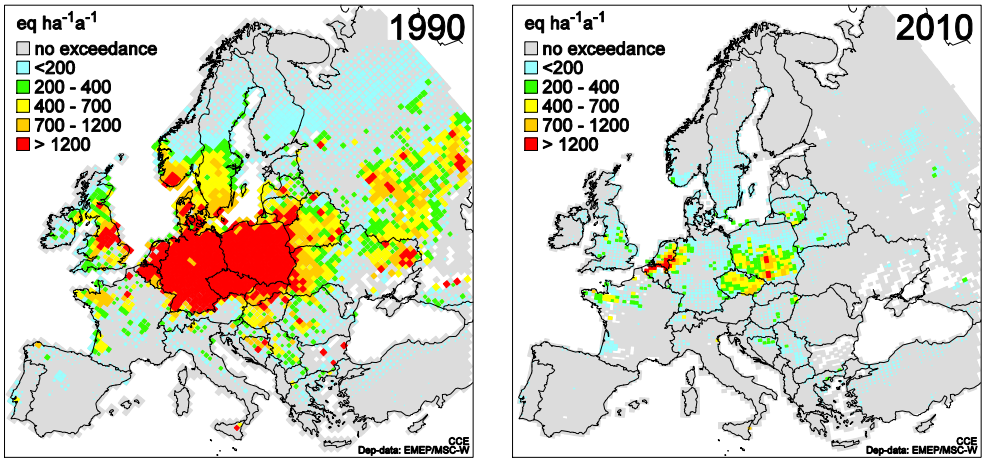


Figure CCE-1: Exceedances (AAE) of the critical loads of acidity in 1990 (left) and 2010 (right) in Europe (Deposition data from MSC-W; European critical loads data from the CCE)

Assuming the GP-CLE scenario to be implemented as of 2010, Figure CCE-2 shows that the percentage of the European terrestrial (red) and aquatic ecosystem (blue) area of which critical loads for acidification are exceeded, attain peaks of 30% and 40%, respectively, in 1980. The AAE peaks also occur in 1980. However, compared to the area exceeded, a reversed order can be noted, i.e. the AAE on terrestrial ecosystems (350 eq $ha^{-1}yr^{-1}$) exceeds the AAE on surface waters (250 eq $ha^{-1}yr^{-1}$) in 1980.

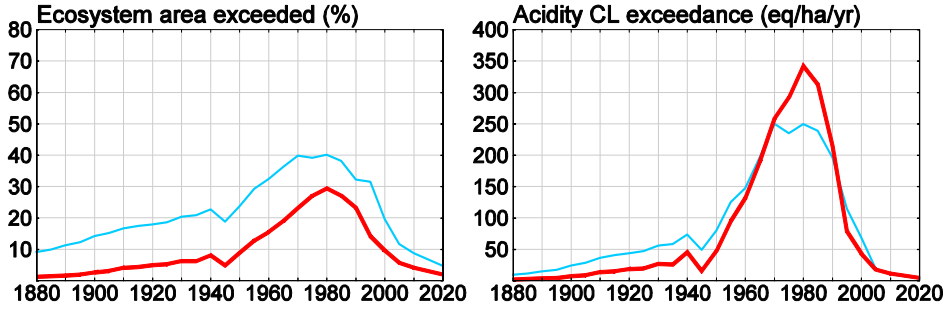


Figure CCE-2: Temporal development of the area where acidity critical loads are exceeded (left; % of ecosystem area) and average accumulated exceedance (AAE) (right; eq ha^{-1} yr^{-1}) of critical loads of acidity (red: all ecosystems, blue: surface waters). Future exceedances are based on the implementation of agreed legislation (temporal emission and deposition data from EMEP-CIAM and EMEP- MSC-W; European critical loads data from the CCE)

A projection towards 2020 illustrates that the area at risk for surface waters is about 5% of catchments for which critical loads for surface waters were submitted. Note that the area of surface waters at risk is relatively higher than the area at risk including terrestrial ecosystems (about 3% of European ecosystems). While exceedances have markedly decreased since 1980, it may still take decades for recovery from acidification to be established (Hettelingh et al. 2015).

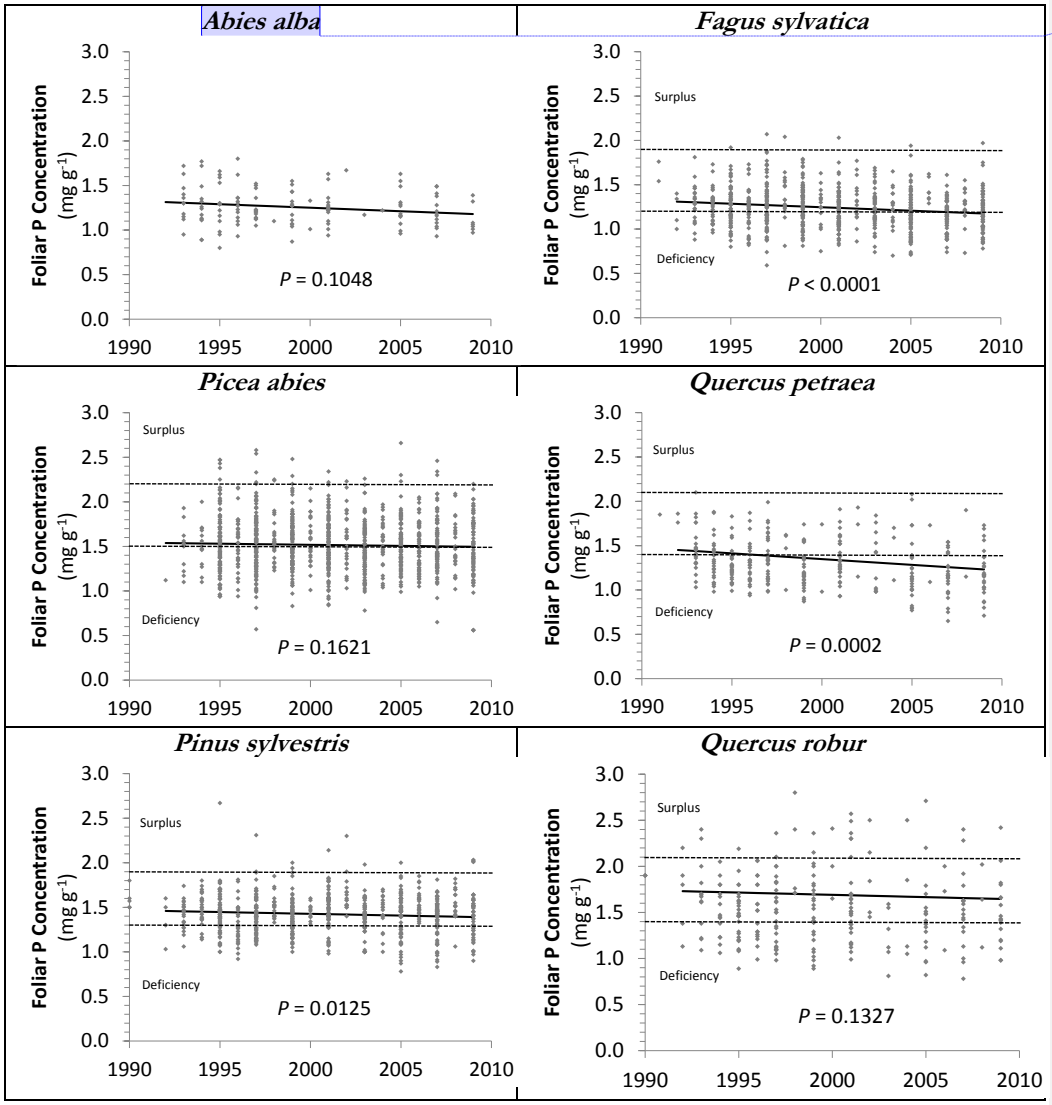
3.1.3. Trends in forest responses (ICP Forests) (max 2 p)

3.1.3.1. Foliar Element Contents

Foliar nutrient concentrations have been monitored in ICP Forests intensive monitoring (Level II) plots from the early 1990s. The objectives have been to assess the possible changes in the concentrations of the main pollutants (sulphur and nitrogen) but also how these possibly affect forest tree nutrition in general, that might in turn be reflected in tree health. Here we analysed the nutritional status of the main European tree species using data collected during 1992-2009 on Level II plots. Tree selection, leaf collection, and foliar analysis were carried out according to the guidelines provided by the ICP Forests manual on sampling and analysis of needles and leaves (Rautio et al. 2010, Rautio & Fürst 2013). To detect temporal trends, linear mixed models under consideration of plot and country as random factors were used (Jonard et al. 2015).

Of the 22 significant temporal trends that were found in foliar nutrient concentrations, 20 were decreasing and two were increasing (Table X). Even though both N and S concentrations in many species show decreasing trends, worryingly many essential nutrients are decreasing also. Perhaps the most alarming trend is the clear deterioration in P nutrition during the past two decades in some of the main tree species (Table X and Fig. X). Increased tree productivity, possibly resulting from high N deposition and from the global increase in atmospheric CO_2, has led to higher nutrient demand by trees. However, soil nutrient supply has not always been sufficient to meet the demand of faster growing trees. As tree nutrient status exerts a tight control on net ecosystem productivity, this deterioration in tree nutrition could have a strong impact on the response of forest ecosystems to climate change.

Comment [HWI16]: Can you highlight a relation with air pollution? Even it is not there, something should be said.



Comment [HWI17]: Give english name in parenthesis

Figure X. Temporal trends in foliar phosphorus concentration of current-year leaves for the main tree species in Europe. Dashed lines are thresholds separating the deficiency, normal and surplus ranges according to Mellert & Göttlein (2012); *p* values < 0.05 indicate whether the linear trends (solid line) are significant or not.

Table X. Linear temporal trends in foliar concentrations for the main tree species in Europe. Direction of slope is given as "+" or "-", degree of significance: $p < 0.1$: **(+)** or **(-)**, $p < 0.05$: **+** or **-**, $p < 0.01$: **++** or **--**, $p < 0.001$: **+++** or **---**.

Tree species	Leaf/needle age	Mass	Foliar concentrations (mg g^{-1})					
			N	P	S	Ca	Mg	K
Fagus sylvatica	current year	+++	-	---	---	-	---	
Quercus petraea	current year		---	---	---	-		-
Quercus robur	current year	(+)						
Abies alba	current year		--				+	
Picea abies	current year	+++			---		(+)	--
Pinus sylvestris	current year			-	---			
Abies alba	1-year-old			-				
Picea abies	1-year-old		-	---	--			---
Pinus sylvestris	1-year-old					+++		

3.1.3.2. Crown condition

Defoliation assessed according to Eichhorn et al. (2010) is the most frequently referred feature of crown condition of forest trees. Its increase is a rather sensitive, but unspecific response parameter indicating stress. Apart from direct and indirect effects of air pollutants, defoliation is influenced by biotic damaging agents or adverse weather conditions (e.g. climatic drought, Seidling 2007). Its yearly plot-specific response characteristic over time (and even more at tree level) is usually highly variable (not shown). However, when averaged over many plots temporal developments can be observed (Fig. x.1).

Deleted: Crown condition of forest trees as assessed according to Eichhorn et al. (2010) is a sensitive, but unspecific response variable to environmental stress.

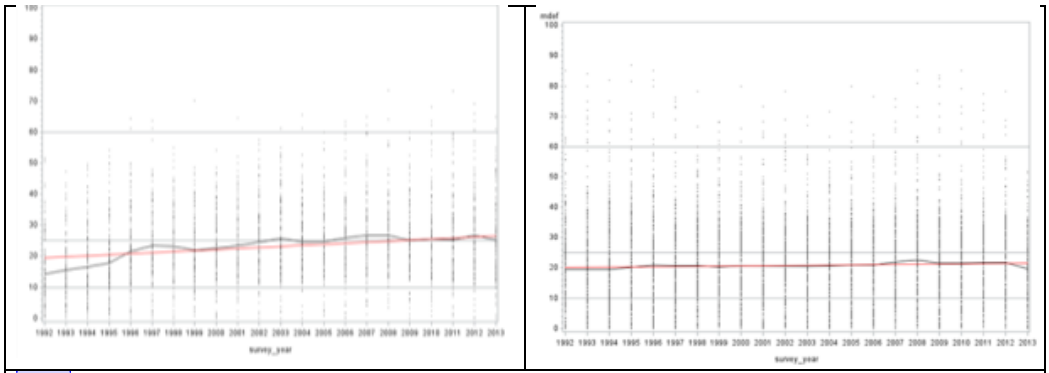


Fig. x: Examples for trends (regional Sen's slopes; minimum length of time span: 20 years, red line) and developments of annual means (bold line) of crown condition of Mediterranean deciduous oaks (left) and Norway spruce (right) at the Level I sites. Points represent annual plot means; for clarity these are not interconnected from year to year. Thin horizontal lines denote traditional damage thresholds.

Comment [HWI18]: The font is too small, the figure is very difficult to read.

Comment [HWI19]: So, an upward trend in crown condition is good? This is confusing. What is on the y-axis? Defoliation?

All three groups of Mediterranean tree species reveal highly significant positive trends in defoliation (Sen's slope with Mann-Kendall test, Tab. x.1), indicating a deterioration of crown condition. The nemoral oak species show the most prominent increase, while European beech reveals only a moderate increase. For the two coniferous species, mainly distributed in the nemoral and boreal part of Europe, only a small (Fig. 1b) or even no trend (Tab. x1) is found.

Tab. x.1: Statistics for crown condition (defoliation) of European main tree species or species groups; *: *Quercus cerris, Q. pubescens, Q. frainetto, Q. pyrenaica*, **: *Quercus coccifera, Q. ilex, Q. rotundifolia, Q. suber*, ***: *Pinus pinaster, P. halepensis, P. pinea*.

Tree species (group)	*Mann-Kendall Tau*	Regional Sen's slope	P	Overall mean [%]
Mediterranean deciduous oaks*	0.205	+ 0.333	< 0.0001	23.05
Mediterranean evergreen oaks**	0.216	+ 0.263	< 0.0001	21.71
Mediterranean lowland pines***	0.205	+ 0.333	< 0.0001	19.26
Quercus robur et petraea	0.221	+ 0.400	< 0.0001	24.58
Fagus sylvatica	0.134	+ 0.200	< 0.0001	19.21
Picea abies	0.098	+ 0.075	<0.0001	20.79
Pinus sylvestris	-0.006	0.0000	0.3430	18.18

On the driver side different developments occurred across Europe: While an increase in ozone concentrations is found in certain regions in the Mediterranean basin (Sicard et al. 2013), there has been a considerable reduction in SO2 air concentrations and deposition especially in central Europe. Without respective additional statistical analyses, it is hard to disentanglecause-effect relationships. However, the observed increase in defoliation in the Mediterranean area coincides with increased ozone concentrations. Apart from air pollutants, tree crown condition is influenced by biotic damaging agents and other abiotic factors like weather conditions. For instance, common beech is influenced by drought (Seidling 2007). New research based on larger sets of soil, foliar and defoliation data from the ICP Forests Level I network and modelled climate and deposition data has revealed species-specific predictor importance for nitrogen load (Veresoglou et al. 2013, Vitale et al. 2014).

Please add one concluding remark on the trends in crown condition in European forest, and how these trends possibly connect to air pollution.

Comment [HWI20]: Please give english names

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3.1.4. Trends in input-output budgets of S and N (ICP IM) (max 2 p)

Annual input-output budgets for sulphate (SO_4) and total inorganic nitrogen (TIN = NO_3 + NH_4) in the period 1990–2012 were calculated for a selection of 18 IM sites (Vuorenmaa et al. 2014). In addition, annual output fluxes for organic nitrogen were calculated for 16 sites during the period 1990–2012, according to data availability. Total deposition of sulphate (meq m^{-2} yr^{-1}) i.e. the input of wet and dry deposition to the catchment was estimated from bulk deposition (open area) and throughfall (forest stands) measurements. Total deposition of inorganic nitrogen (TIN) was calculated using the same method. Total deposition of N to the forest floor is dependent on wet and dry deposition, and net exchange of material with the vegetation. Because of the strong impact of canopy processes, bulk deposition measurements for TIN were also reported. Annual total deposition fluxes to the catchments were calculated as the sum of monthly values. Output fluxes of TIN (meq m^{-2} yr^{-1}) from the catchments were calculated as the product of measured catchment discharge and ion concentrations. Annual runoff water element fluxes were calculated by summing mean monthly fluxes, obtained from monthly mean water flux and monthly mean solute concentration.

At each site trends in fluxes were analysed using the non-parametric Mann-Kendall test applied to annual data. The magnitude of trend was estimated by the Theil-Sen slope estimation method. The unit of the slope estimate for yearly based data is meq m^{-2} yr^{-1} for fluxes. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis.

The % release or % retention of sulphur and nitrogen in catchments was calculated as follows In order to quantify the retention or release of sulphur and nitrogen in the catchment, the % release or % retention was calculated, as follows: a percent net export (pne) was calculated. The percent net export was defined as: pne = (output–deposition)*100/deposition. Positive pne values indicate release and negative pne values indicate retention in the catchment. For sulphur, total deposition estimate (bulk + throughfall) was used. In the case of nitrogen, bulk deposition in open area was generally larger than throughfall deposition (a surrogate for total deposition) and therefore bulk deposition measurements were used as N deposition estimates in the pne calculations.

A statistically significant downward trend ($p < 0.05$) of total sulphur deposition from 1990 to 2012 was observed at all studied ICP IM sites (Table 3.1.5). As a response to decreased S deposition, sulphate fluxes in runoff have decreased at 15 out of 18 sites, being significant at 67% of the sites (Vuorenmaa et al. 2014). Moreover, a weak decreasing trend ($p < 0.10$) was observed at two sites. Bulk deposition of nitrogen also decreased at almost all sites (17 out of 18), being significant at 71% of the sites. Total deposition of nitrogen, indicating N fluxes through the canopy to the forest floor, decreased in 13 sites, being significant at 54% of the sites. Total deposition of N decreased less (mean annual change -0.70 meq m^{-2} a^{-1}) than that of bulk deposition (mean annual change -1.02 meq m^{-2} a^{-1}).

In contrast to sulphate, total inorganic nitrogen (TIN) fluxes in runoff showed mixed response with both decreasing and increasing trends. Statistically significant decreasing trends were observed at five sites and increasing trends at two sites (DE01, Forellenbach, Germany and SE14, Aneboda, Sweden). The significant increasing trends for these two sites are probably due to excess N mineralization and increased NO_3 leaching, resulted from forest damage and dieback in the areas due to storm logging and bark beetle infestation.

Table 3.1.5 Trends of annual input (deposition) and output (runoff water) fluxes and percent net export (pne) for sulphate (SO_4) and total inorganic nitrogen (TIN = NO_3 + NH_4) at studied ICP IM catchments in 1990–2012. A statistically significant trend ($p < 0.05$) of annual change (meq m^{-2} yr^{-1} for fluxes, % yr^{-1} for pne) is indicated in bold and a potential trend ($p < 0.10$) is indicated in italics.

Comment [HWI21]: This text is long and quite technical. Please reduce to two pages.

Comment [HWI22]: "Percent net export" is a bit had to understand, even for a co-author on the paper. The difference between inputs and outputs, normalized with regard to input (=deposition).: (catchment export – catchment input)/ catchment input. A positive value means that there is release of the element, a negative value means retention.
Could you call it % release or % retention, and stay away from percent net export?

Comment [HWI23]: I wonder if this table is necessary. Figures are better than tables.

Catchment	Data	Total SO_4 deposition	SO_4 output	SO_4 pne	TIN bulk deposition	TIN total deposition	TIN output	TIN pne
		(meq m^{-2} yr^{-1})		(% yr^{-1})	(meq m^{-2} yr^{-1})			(% yr^{-1})
AT01, Zöbelboden	1994-2012	**-1.53**	**-1.04**	*0.88*	-0.17	-0.43	*1.21*	1.15
CZ01, Anenske Povodi	1990-2012	**-6.44**	1.06	**6.30**	**-1.07**	0.90	**-0.13**	*-0.15*
CZ02, Lysina	1990-2012	**-10.05**	**-7.08**	**8.07**	**-1.89**	**-1.49**	**-0.76**	**-0.71**
DE01, Forellenbach	1991-2012	**-2.78**	**-1.08**	**10.15**	**-1.24**	0.31	**2.95**	**5.03**
EE02, Saarejärve	1995-2012	**-1.90**	-0.66	**7.58**	-0.09	-0.16	0.18	1.02
FI01, Valkea-Kotinen	1990-2012	**-1.16**	-0.52	*1.88*	**-0.36**	-0.05	-0.01	0.04
FI03, Hietajärvi	1990-2012	**-0.58**	**-0.33**	**0.82**	-0.17	0.07	**-0.02**	-0.02
IT01, Renon-Ritten	1993-2012	**-1.66**	0.05	**2.92**	**-1.23**	-0.74	-0.01	0.00
LT01, Aukstaitija	1994-2012	**-2.04**	**-1.49**	**8.64**	**-1.14**	**-1.81**	-0.02	-0.07
LT03, Zemaitija	1995-2012	**-3.13**	1.26	**8.05**	-1.14	-1.23	0.03	*0.12*
LV01, Rucava	1994-2009	**-1.48**	-5.03	-1.52	**-2.82**	*-2.03*	1.26	**1.26**
LV02, Zoseni	1994-2009	**-1.06**	**-6.13**	-8.28	**-2.14**	**-2.56**	**-0.28**	-0.15
NO01, Birkenes	1990-2012	**-3.45**	**-3.59**	**2.02**	-0.58	**-1.16**	-0.15	-0.11
NO02, Kårvatn	1990-2012	**-0.42**	*-0.39*	0.63	0.16	0.15	0.00	-0.10
SE04, Gårdsjön	1990-2012	**-3.86**	**-4.46**	1.03	**-1.43**	**-1.92**	0.05	**0.12**
SE14, Aneboda	1996-2012	**-1.58**	**-3.16**	4.06	**-0.91**	0.25	**0.24**	**0.62**
SE15, Kindla	1996-2012	**-1.39**	**-3.62**	-4.35	**-0.79**	**-0.79**	**-0.03**	-0.04
SE16, Gammtratten	1999-2012	**-0.44**	*-0.94*	-0.34	**-0.72**	**-0.30**	*-0.04*	*-0.15*

Sulphate budgets showed increasing percent net exports (pne) at majority of the sites, indicating a net release of previously stored SO_4, particularly during the past 15 years (Fig. 3.1.5, Table 3.1.5). This process has taken place both in high and low sulphur deposition areas. In the selected set of ICP IM sites (Fig. 3.1.5a), median values for S pne exhibited a significant increase ($p < 0.01$, 2.3 % yr^{-1}) in 1990-2012. A net release of stored SO_4 is considered to act as a H^+ source at many ICP IM sites (Forsius et al. 2005), and SO_4 remains the dominant source of actual soil acidification despite the generally lower input of S than N in European forested ecosystems. Several processes, including desorption and excess mineralisation, regulate the long-term response of soil S, and a differentiation is necessary for assessing the effects of emission reductions on acidification recovery and for predictions of the future responses. In general, many of these S retention/release processes are also sensitive to changes in climatic variables, and would therefore be affected by climate change.

Nitrogen is generally the growth-limiting nutrient in forest ecosystems, and the uptake of available N compounds is efficient. In contrast to sulphur, nitrogen deposition is usually retained in boreal terrestrial ecosystems; typically < 10% is leached in runoff, mostly as NO_3. The percent net export (pne) of nitrogen generally ranged between -98% and -80% at the studied ICP IM sites during the 2000s (Fig. 3.1.5b), indicating a strong retention of N in the catchment, and 50% of the sites exhibited increase in net retention. Correspondingly, median values for TIN pne exhibited a significant decrease ($p < 0.05$, -0.11 % yr^{-1}) i.e. increase in net retention in 1990-2012 (Fig. 3.1.5b). Although nitrogen has played a rather minor role in the acidification in the past, its relative importance is increasing because N emissions have decreased much less than sulphur emissions (Helliwell et al. 2014). Continued high nitrogen deposition can result in nitrogen saturation of terrestrial ecosystems, and excess NO_3 leaching to surface waters (e.g. Macdonald et al. 2002, Oulehle et al. 2012). So far, there have been no signs of consistent and widespread regional increases in nitrate concentrations in sensitive freshwaters in Europe (Wright et al. 2001, Garmo et al. 2014, Helliwell et al. 2014). However, nitrogen continues to accumulate in catchment soils and vegetation. Nitrogen saturation may require many decades to occur, at least at levels of N deposition typical for Europe (Wright et al. 2001). Climate change will likely impact mineralization of organic

nitrogen and leaching of organic matter, and could thus increase the risk for elevated N loss from watersheds.

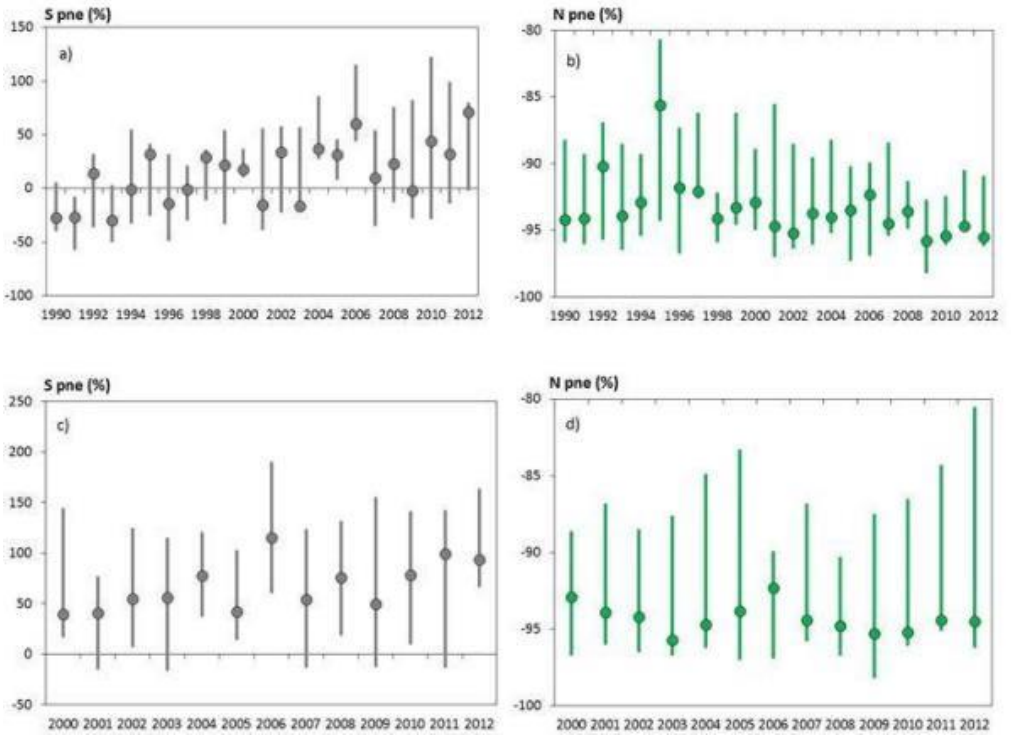


Figure 3.1.5. Release (negative values) and retention (positive) values of sulphate (SO_4) and total inorganic nitrogen (TIN) calculated as the difference between catchment inputs and outputs, divided by catchment inputs in %. Circle shows median, and the lines show the 25% and 75% percentile of annual values between 1990 and 2012, for 8 forested ICP IM sites. (from Vuorenmaa et al. 2014).

Comment [HWI24]: Is it possible to refer to a paper for most of the technical information here? I find 'S pne' a bit hard to understand – why not write 'Ratio of catchment input to catchment output for S and N (in %)'. Why show both 1990-2012 and 2000-2010? Please pick the graph with the longest period.

Deleted: Percentiles (25%, median 50%, 75%) of

Deleted: percent net export (pne, %) of sulphate (SO_4) and total inorganic nitrogen (TIN) for the IM sites CZ01, CZ02, DE01, FI01, FI03, NO01, NO02, SE04 in 1990-2012 (a and b, respectively) and for the sites AT01, CZ01, CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16 in 2000-2010 (c and d, respectively). DE01 and SE14 were omitted from the calculation of pne for TIN due to excess N mineralization after Norway spruce (Picea abies) dieback due to a bark beetle attack in 1996–1997 and storm logging / bark beetle attack in 2005–2009, respectively. pne = (output–deposition)*100/deposition

3.1.5. Trends in water chemistry and biology (ICP Waters) (max 2 p)

Water chemistry has responded strongly to reductions in sulfur deposition. Figure x shows regional mean annual concentrations of non-marine SO4 in four regions, expressed in % of the 1988 regional mean SO4 concentration. Despite differences in deposition loadings, climate and catchment characteristics (forested, peatlands, mountainous, lakes and rivers), all regions show essentially the same pattern: a 45 to 55% reduction of SO4 since 1988. The exception is the non-glaciated catchments in North America, where sulfate in rain adsorps to the soil instead of leaching out. A large sulfate adsorption capacity is typical of non-glaciated areas, and can result in high leaching of sulfate when the capacity is exceeded, even after a substantial reduction in sulphur deposition.

The decrease in sulfate is associated with an increase in surface water pH. The most significant increase in pH is documented in Central Europe, from 5.3 to 5.8. In Northern Europe, pH increased from 5.8 to 6.0, while in unglaciated areas in North America, pH increased from 5.6 to 6.1. There is no trend in pH in surface waters from glaciated areas in North America. The more variable responses in pH between regions are because there are more factors than only SO4 deposition that drive pH, for instance soil buffer capacity, acidification history and climate. Still, the increase in pH documents widespread chemical recovery of surface waters, which opens the possibility for biological recovery of acidified surface waters. More details on trends in surface water chemistry are found in Garmo et al. (2014) and De Wit et al. (2015).

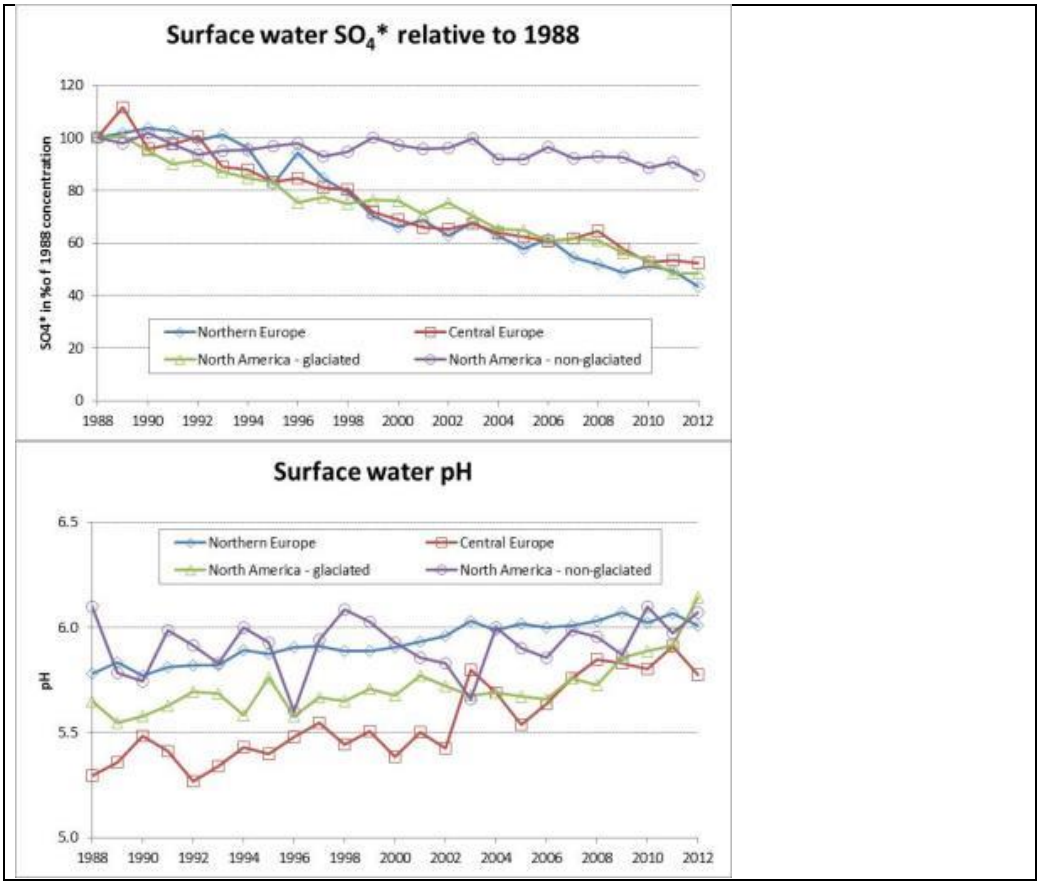


Figure x *Trends in surface water chemistry at ICP Waters sites 1988-2012. Shown are the mean concentrations of non-marine sulphate (SO4*) and pH at 22 sites in northern Europe, 21 sites in central Europe, 37 sites in glaciated areas of*

eastern North America, and 7 sites in non-glaciated areas of eastern North America. (Source: De Wit et al. (2015))

Trends in biology

An example of relations between sulfate in deposition, sulfate in surface waters and associated water chemistry and biological responses is given for the severely acidified lake Saudlandsvann in southwest Norway, where monitoring started in the 1970s (Text Box 1). While biological responses to deteriorating water quality related to acidification can be immediate, biological recovery related to improved water quality is usually more slowly. The main driver for biological change during acidification is the toxic effect of water chemical components, leading to disappearance of acid-sensitive species. Under the recovery process, when water quality is no longer as critical, biological responses also depend on the physical environment, for instance dispersal and colonisation ability of different species.

Comment [HWI25]: Remove? It is a bit different from the rest.

Monitoring of biological recovery follows national designs, which makes quantitative integration of biological recovery similar to chemical recovery challenging. The table below summarizes biological responses to improved water quality in acid-sensitive lakes and rivers in a number of European countries, for organism groups varying from fish, plant plankton (photosynthetic algae, phytoplankton), animal plankton (zooplankton), sediment-dwelling animals (zoobenthos) to water plants.

The national contributions (Czech Republic, Finland, Norway, Switzerland) to the documentation of time trends in biological recovery differ considerably in time span of records, targeted groups of biota, and type of variable considered. All countries report evidence of chemical and biological recovery. The best documentation of improved water quality is where the longest time series (over 30 years) were available (Norway, Finland) or where acid deposition has been reduced most strongly (Czech republic). Biological time series are usually shorter than chemical time series and do not show an equally consistent recovery as for water quality. Comparison with reference sites suggests that species diversity in fully restored aquatic ecosystems could be much higher than is presently observed in aquatic systems that are under recovery from acidification (Fjellheim & Raddum 1995).

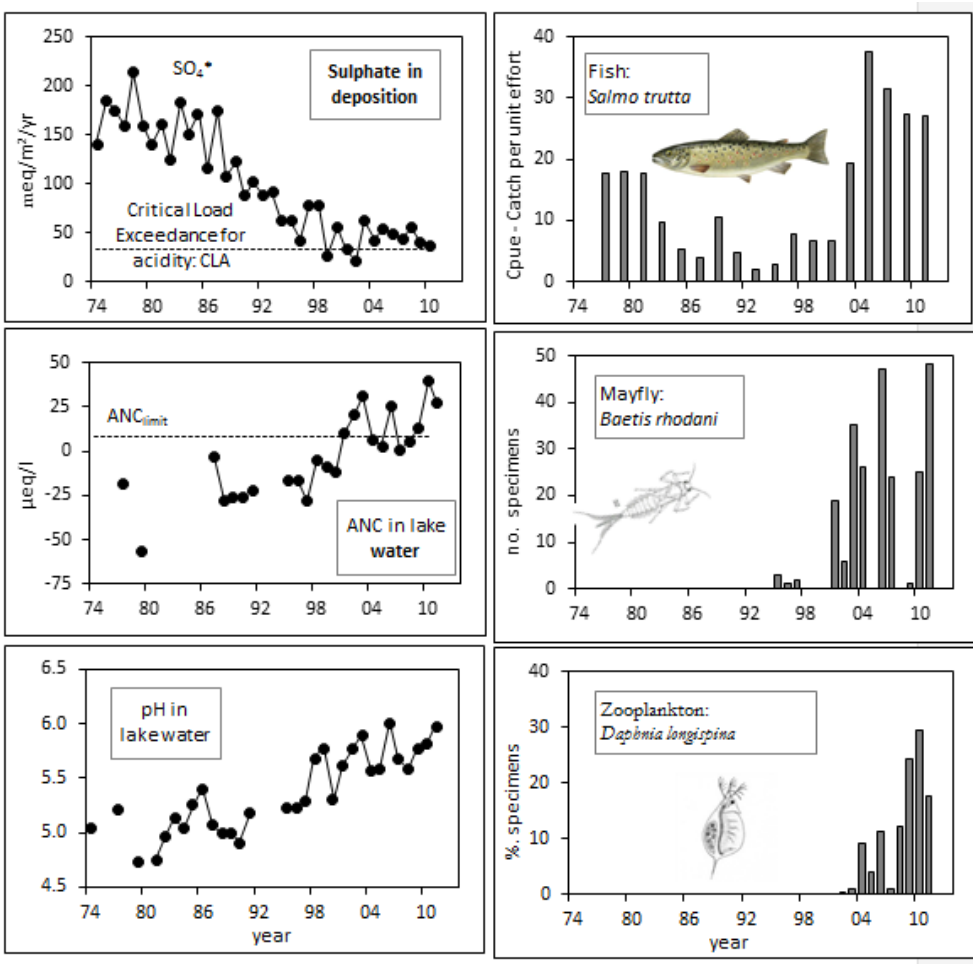
***Table x**. Summary of findings from national reporting on biological recovery. Colour coding for trends: , only positive trends; mixture of positive and no trend;. no trends. Source: De Wit et al. (2015), Skjelkvale and De Wit (2011)*

Region	Country	Water body	Biota	Biological parameter	Period	Trends
Nordic	Norway	5 rivers	Zoobenthos	Acidification index, Biodiversity, Acid-sensitive organisms	1982-2013	
	Sweden	8 lakes	Phytoplankton	Species number, abundance, richness	1988-2008	
			Zoobenthos	Species number, abundance	1988-2008	
	Finland	21-30 lakes	Fish	Abundance, Population structure	1985-2012	
		29 lakes	Zoobenthos	Communities	1985-2001	
		30 lakes	Periphyton, phytoplankton	Communities	1985-2001	
Central Europe	Czech Republic	8 lakes	Phytoplankton	Species number, abundancel	1999-2011	
			Zooplankton	Species number, abundance	1999-2011	
			Zoobenthos, Nepomorpha	Species number, abundance	1999-2011	
			Macrophytes	Abundance	2004-2010	
	Germany	lakes, streams	Zoobenthos	Species number, abundance acidification index	1982-2010	
	Switzerland	4 lakes	Zoobenthos	Species number, abundance	2000-2011	
	(Alps)	3 rivers	Zoobenthos	Species number, abundance	2000-2011	

Text box 1: Biological recovery – an example from Norway

Recovery from acidification at Lake Saudlandsvatn, Norway. As sulfur deposition has decreased, the acid neutralising capacity (ANC)and pH have increased in the lake, and the populations of three sensitive species, i.e. brown trout, the mayfly *Baetis rhodani* and the planktonic crustacean *Daphnia longispina* have begun to recover (modified from Hesthagen et al. 2011 Sci Tot Env).

Data from T. Hesthagen, NINA (fish), NIVA (water chemistry), NILU (deposition),UiB (invertebrates)



3.1.6. Trends in corrosion (ICP Materials)

Corrosion has decreased substantially to around 50% of the original values measured in 1987 (Tidblad et al 2014). In recent years, however, the improvements in corrosion and soiling are minor. Taking carbon steel as an example, several different one-year exposures have been performed during the period 1987-2011. Figure X shows corrosion of carbon steel exposed at two selected test sites, one industrial test site in the Czech Republic (Kopisty) and one rural test site in Sweden (Aspvreten), representing two extremes of corrosion attack. Worth noting is that even for the originally "clean" site (Aspvreten), there has been substantial improvement in corrosion, resulting from improved air quality, during this period.

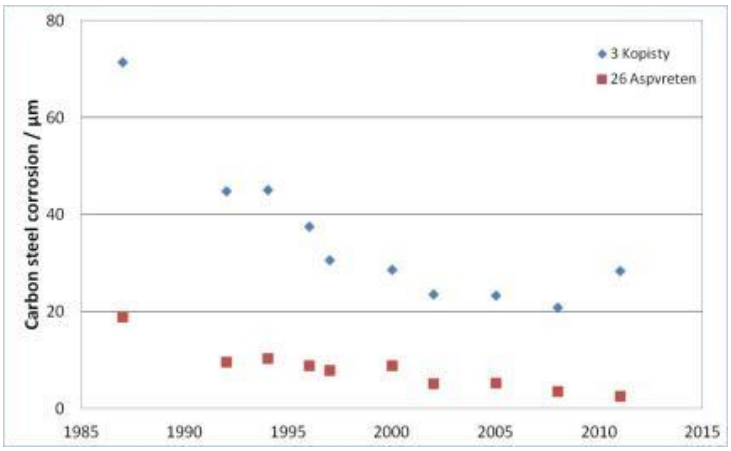


Figure X. Trends of carbon steel at two selected test sites

When grouping the sites into categories of rural, urban and industrial (Figure X) differences in corrosion between polluted (industrial) and non-polluted areas are still evident. There is also a difference between urban and rural areas but it is worth noting that improvements have resulted in average values in urban areas in the recent exposure lower than the average values in rural areas for the first exposure in 1987.

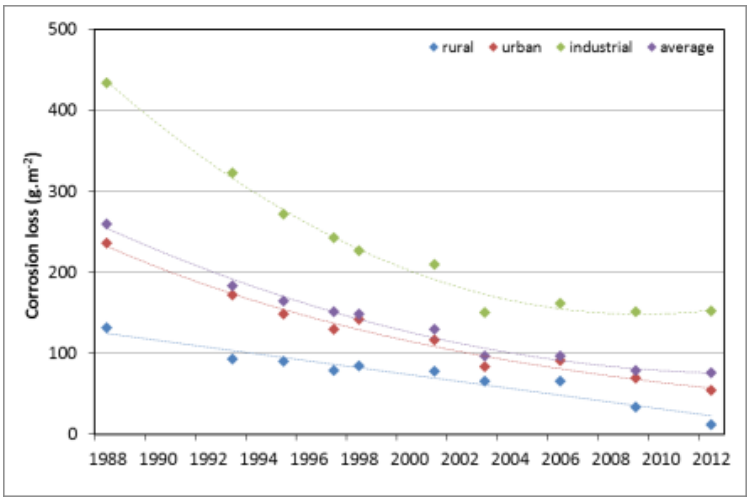


Figure X Trends in carbon steel yearly corrosion loss based on results from 20 test sites.

For real cultural heritage objects made of metals the decreases are instantaneous, responding rapidly to decreasing pollution levels. For stone materials, however, there is a substantial time lag, 20 years or more, before improvements can be seen.

The popularity of building with Portland limestone, particularly in the UK, has led to many research projects measuring the rate of weathering of Portland limestone, both in exposure trials and on building surfaces. Figure X shows some of these results. The studies on weathered surfaces were carried out at St. Paul's Cathedral in London between 1980 and 2012. The early results showed a mean erosion rate for six locations of around 50 μm year^{-1} between 1980 and 1985. This was considerably lower than the long term average from the period from 1718 to 1987 of 100μm year^{-1} but still very high considering that the SO_2 concentrations had fallen by 90% between 1960 and 1985.

A further set of measurements (Inkpen et al 2012) was made in 2000 and these showed that there had been a reduction in the rate of erosion to 25 μm year^{-1} during the period 1990 to 2000 (see Figure X). In the same period the atmospheric SO_2 concentration had continued to decline, but the rate of change in the weathering appears to be greater with a 'time-lag' of 15 – 20 years between the reduction in SO_2 and the benefit being seen in the existing building stone.

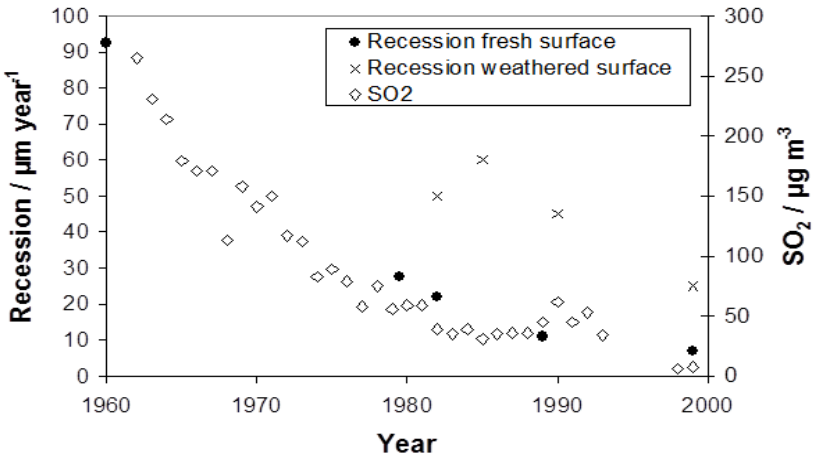


Figure X. Surface recession rates of limestone and SO_2 concentration at St Pauls Cathedral.

3.1.7. Prediction of future trends in recovery/exposure (JEG) (max 2 p)

Filip Moldan suggests : JEG deals with limited number of trends which are modelled into the future. One way to write this is by case studies. Alternative way is to describe which trends are modellable and what the key outcomes frrom JEG are. But in that case it would probably be better to place it as new heading 3.8 and include whatever of 3.1 – 3.6 JEG can contribute to when it comes to predicting future.

Comment [HWI26]: I would like to hear from JEG in Geneva exactly what contribution they can give.

3.2. Nitrogen as a nutrient (ca 5-10 p)

3.2.1. Trends in deposition (EMEP)

-I leave this for now, but of course reference can be made to 3.1.1, in stead of repeating what is said there.

3.2.2. Trends in exceedances of CLs (CCE/TF M&M)(max 1 p)

The risk of eutrophication, both in terms of magnitude and geographical distribution, diminishes from 1990 to 2010 (Figure CCE-3). However, it is obvious that critical loads of nutrient nitrogen remain exceeded in all European countries, while areas with relatively high exceedances (red or orange shading) in 2010 occur in most of the countries.

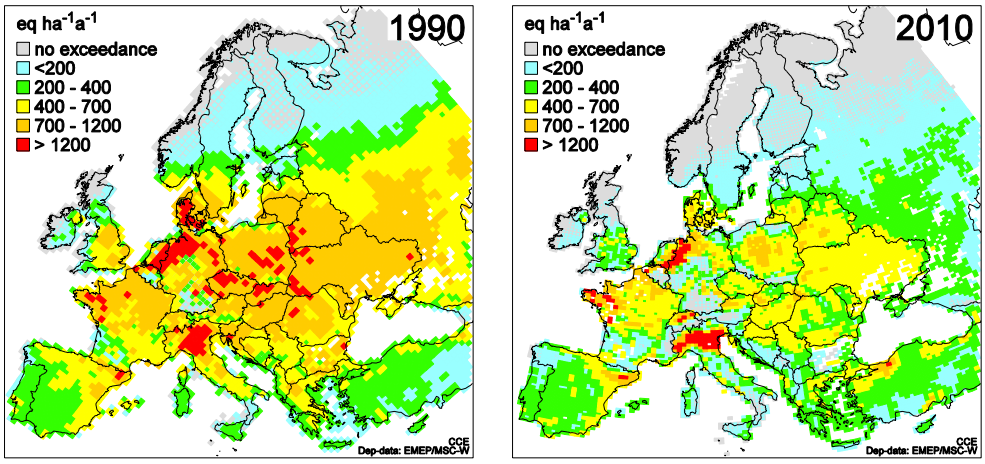


Figure CCE-3: Exceedances (AAE) of the critical loads of nutrient nitrogen in 1990 (left) and 2010 (right) in Europe. (Deposition data from MSC-W; European critical loads data from the CCE)

The area with high exceedances (red and orange shading) of critical loads of nutrient nitrogen is significantly larger in 1990 in comparison to 2010. On the other hand, the area at risk in the lower range of exceedances (green and blue shading) is larger in 2010 in comparison to 1990. This implies that abatement policies have been successful, although lower exceedances in 2010 still imply that damage to structure and function of ecosystems will occur. Compared to the magnitudes of exceedances in 1990, damage caused by exceedances in 2010 may occur at a later point in time.

Figure CCE-4 illustrates that areas where critical loads of nutrient nitrogen are exceeded, continue to remain a serious issue under GP-CLE emissions. Eutrophication affects about 55% of the European terrestrial ecosystem area in year 2020 (target year for the revised Gothenburg Protocol) with an AAE of about 150 eq $ha^{-1}yr^{-1}$.

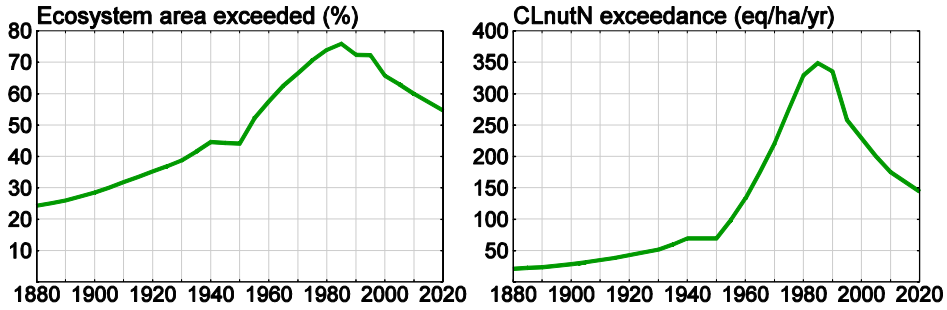


Figure CCE-4: Temporal development of the area where nutrient N critical loads are exceeded (left; % of ecosystem area) and average accumulated exceedance (AAE) (right; eq ha^{-1} yr^{-1}) of nutrient N critical loads (all ecosystems). Future exceedances are based on the implementation of agreed legislation. (temporal emission and deposition data from EMEP-CIAM and EMEP- MSC-W; European critical loads data from the CCE)

Aquatic area at risk of eutrophication is not shown here due to the lack of European coverage. However, recent findings (De Wit and Lindholm 2010) suggest that eutrophication of surface waters is of increasing concern.

3.2.3. Trends in exceedances of critical loads at ICP IM sites

Calculation of trends in the exceedance of critical loads

The Steady-State Water Chemistry (SSWC) model embedded into the First-order Acidity Balance (FAB) model (Henriksen and Posch 2001, Posch et al. 2012) was used to calculate critical loads for 18 ICP IM catchments for which observations of runoff volume and water chemistry were available for the years evaluated (Holmberg et al. 2013). Mass balance critical loads of nutrient nitrogen $CL_{nut}N$ (eq ha^{-1} yr^{-1}) were calculated for the same 18 catchments. In addition to the mass balance critical loads of nutrient N, empirical critical loads of nutrient N, $CL_{emp}N$, were also assigned to a total of 83 vegetation plots at 37 ICP IM sites. To study the connection between the calculated critical loads and empirical impact indicators, concentrations and fluxes of S and N in surface waters were derived for the catchments with available data. Total inorganic nitrogen (TIN) was estimated as TIN = NO_3+ NH_4.

The deposition estimates were generated with the source-receptor matrices derived from the EMEP/MSC-W unified atmospheric dispersion model (Amann et al., 2010) using the NAT2000, COB2020, Low*2020, MID2020, High*2020 and MFR2020 emission scenarios. The NAT2000 scenario represents historic emissions for the year 2000 and the COB2020 scenario national economic projections for 2020 as reported by the countries under the LRTAP Convention. The Low*2020 and High*2020 scenarios refer to the ambition level of the reductions, so that with lower reductions the scenario Low*2020 leads to more deposition than High*2020. MFR2020 assumes all available abatement technologies being implemented by 2020. The depositions were available for 50 km×50 km grid cells covering Europe.

Critical loads for acidification and eutrophication and their exceedances were determined for a selection of ICP IM sites (Holmberg et al. 2013). The level of protection of these sites with respect to acidifying and eutrophying deposition was estimated for 2000 and 2020 (Table 3.2.2). In 2020 more sites were protected from acidification (67%) than in 2000 (61%). However, due to the sensitivity of the sites, even the maximum technically feasible emission reductions scenario would not protect all sites from acidification. In 2000, around 20% of the IM sites were protected from eutrophication. In 2020, under reductions in accordance with current legislation, about one third of the sites would be protected, and at best, with the maximum technically feasible reductions, half of the sites would be protected from eutrophication.

Table 3.2.2 Average exceedances for ICP IM sites not protected from acidification ($AExCL_A$) and eutrophication ($AExCL_{nut}N$, $AExCL_{emp}N$) and percentage of sites protected with different deposition scenarios.

	Total nr of sites	NAT2000	COB2020	Low*2020	MID2020	High*2020	MFR2020
$AExCL_A$ (eq $ha^{-1}yr^{-1}$)	18	987	494	421	392	354	310
Acidification protection %	18	61 %	67 %	67 %	67 %	67 %	67 %
$AExCL_{nut}N$ (eq $ha^{-1}yr^{-1}$)	18	625	369	284	286	277	230
Eutrophication protection %	18	22 %	28 %	28 %	33 %	39 %	39 %
mass balance critical load							
$AExCL_{emp}N$ (eq $ha^{-1}yr^{-1}$)	37	578	349	238	221	177	154

Comment [HWI27]:
-this is essentially about exceedance in the future. We might leave that out in our report, haven't decided yet.

We have structured our report according to pollutant. Here, exceedances of CL with regard to acidity AND nutrient nitrogen are presented. Consider changing this contribution in a way that it fits the structure of this report.
Also, provide text to link your bit to the paragraph above.
Use the paragraph above as inspiration for simplifying your text, and reducing the length with 25%

Eutrophication protection % empirical critical load	37	19 %	35 %	38 %	41 %	41 %	54 %

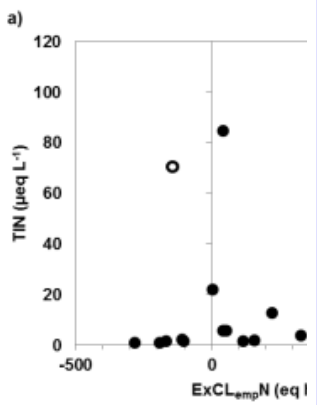
Key conclusions:

- Current emission reduction plans would decrease the exceedance of critical loads of S and N at the ICP IM sites significantly. However, particularly regarding N, the exceedance would remain high even assuming maximum technically feasible emission reductions.
- ICP IM sites with estimated CL exceedances of S and N also showed elevated S and N leaching in surface waters according to the empirical monitoring data. This increases confidence in the European-scale critical loads mapping used in integrated assessment modelling to support emission reduction agreements.

Comment [HWI28]: I deleted this text because it is about model validation, not about trends (of course it is one of the contribution of ICP IM to the convention, but it is of less relevance in a trend report, and we need to keep the length of the report to a minimum).
-or, if you can rephrase this so it is relevant for trends, you can keep it in. (this figure looks a bit like the plot of biodiversity loss vs exceedance of CLs, but there trends are reported, here it's only catchment TIN export, no trends).

Deleted: Data

Deleted: from the intensively monitored ICP IM sites provide a connection between modelled critical thresholds and empirical observations, and thus an indication of the applicability of critical load estimates for natural ecosystems. Across the sites, there was good correlation between the exceedance of critical loads for acidification and key acidification parameters in runoff water, both with annual mean fluxes and concentrations. There was also evidence of a link between exceedances of critical loads of nutrient nitrogen and nitrogen leaching (Fig. 3.2.2). The collected empirical data of the ICP IM thus allow testing and validation of key concepts used in the critical load calculations. ¶
¶



¶
Figure 3.2.2. Eutrophication empirical impact indicators in relation to exceedance of critical loads of N at ICP IM sites. The x-axes show exceedances of empirical critical loads of nutrient N ($ExCL_{empN}$, NAT2000 deposition). The y-axes show annual mean concentrations (a) and fluxes (b) of TIN ($=NO_3 + NH_4$) in runoff. Negative exceedance values indicate that the critical loads are not exceeded. Open circles indicate catchments with inputs of N from sources other than deposition. TIN flux for site DE01 (1373 eq ha^{-1} yr^{-1}) is outside axis range and not shown in graphs (based on Holmberg et al. 2013).¶

3.2.4. Trends in ground vegetation species cover and diversity (ICP IM) (max 2 p)

Long-term monitoring data from 28 forest sites belonging to the ICP IM and ICP Forests networks with a total of 1,335 permanent forest floor vegetation plots was analysed to detect temporal trends in vascular plant species cover and diversity (Dirnböck et al. 2014). The cover of every vascular plant species was estimated in the percentage of the total sampling unit area or as ordinal abundance classes. The latter were transformed to their class means percentage of cover. In total, 646 species were recorded. The exceedance of the CL for eutrophication ($ExCL_{emp}N$) was estimated as the measured throughfall N deposition minus the empirical CL for each site. The long-term average of the annual canopy throughfall deposition of inorganic N was used. In addition, modelled inorganic N to deposition forests was used for 1995, a reference year roughly in the middle of the time series with vegetation observations. Deposition estimates were available from the EMEP/MSC-W unified atmospheric dispersion model. At the study sites, throughfall deposition of inorganic N ranged from 0.6 to 20.2 kg N $ha^{-1}yr^{-1}$. Empirical CL were assigned according to the respective EUNIS habitat type. For the detection of significant temporal changes of species cover, Generalized Linear Mixed Models (GLMM) were applied. Trends in diversity were analysed using the number of vascular plant species and beta diversity. Overall changes in the species numbers and their relation with $ExCL_{emp}N$ were tested with a Linear Mixed Model (LMM), where the study plots were nested within study sites (for details see Dirnböck et al. 2014).

Chronic nitrogen deposition poses a threat to biodiversity as a result of an eutrophication of sensitive ecosystems. Excess N may favour a few plant species causing competitive exclusion and, in the long run, loss of less competitive species (Suding et al. 2005, Hautier et al. 2009). Many local to regional studies have shown that chronic N deposition leads to a shift in the species composition of the forest floor and eventually to diversity loss (e.g. Bobbink et al. 2010). However, so far there has been no unequivocal evidence that nitrogen deposition is a broad scale driver behind the eutrophication signal in forest plant communities, as it is in other ecosystems such as e.g. grasslands (Stevens et al. 2010).

Long-term monitoring data from 28 forest sites from the ICP Integrated Monitoring and ICP Forests Programme was used to analyse temporal trends in species cover and diversity (Dirnböck et al. 2014, Figure 3.2.3). Main findings of this study were that the cover of plant species that prefer nutrient-poor soils (oligotrophic species) decreased the more the measured N deposition exceeded the empirical critical load (CL) for eutrophication effects (p-value = 0.002). The observed response was the first detection of a N deposition effect on vascular plants of forest floor vegetation in a European-wide long-term monitoring data set. Contrary to species cover changes, neither the decrease of species richness (alpha and gamma diversity) nor of homogeneity (beta diversity) correlated with nitrogen CL exceedance in the dataset.

Key conclusions:

- The study based on long-term ICP IM and ICP Forests data showed that cover of oligotrophic plant species have decreased in European forest ecosystems, but diversity is still not affected by airborne N deposition.
- Estimated critical loads are very useful to describe the sensitivity of forest floor vegetation to N deposition.
- The use of critical load exceedances is particularly suitable to revealing the eutrophication signal of N deposition. It is superior to N deposition alone, which is ignoring the differences in sensitivity among ecosystems.

Comment [HWI29]: I don't understand? Doesn't the figure below document damage from airborne N dep on species diversity?

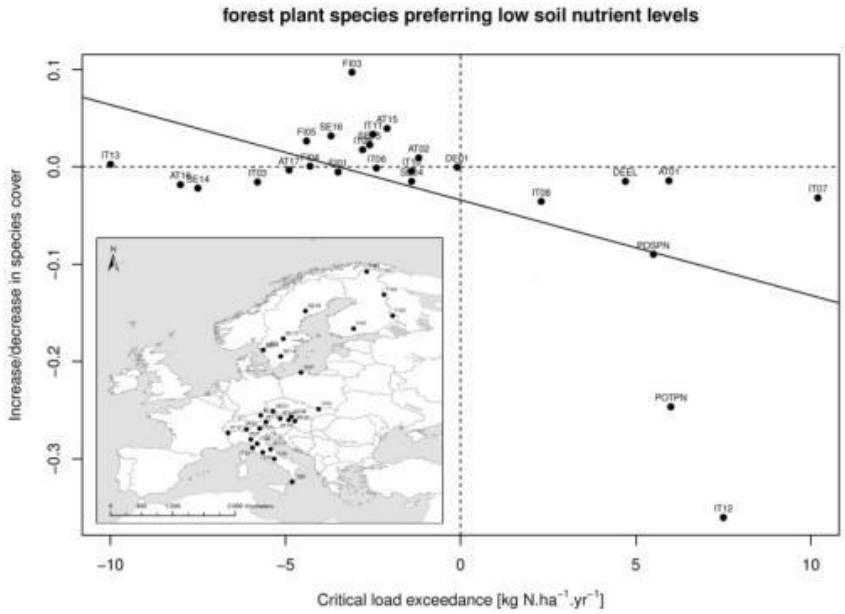


Figure 3.2.3: Forest plant species that prefer low soil nutrient levels have decreased during the last 10-50 years in 28 ICP IM and ICP Forests sites across Europe owing to the exceedance of the nitrogen critical loads. The y-axis indicates the strength of the cover change of all oligotrophic species in a study site (negative values indicate decrease, positive values increase). The critical load exceedances are shown as the difference between the N deposition and the empirical critical load (negative values indicate no exceedance, positive values exceedance) (based on Dirnböck et al. 2014).

Comment [HWI30]: The fonts in this figure are very small.

3.2.5. Other responses? (ICP Forest/ICP Vegetation) (max 2 p)

Temporal trend (2005 – 2010) in nitrogen concentrations in mosses

Comment [HWI31]: This gives the situation at two points in time, which is a bit thin for a trend analysis. Should it be included?

Between 2005 and 2010, the average median nitrogen concentration in mosses has declined by 5% in Europe (Harmens et al., 2015), which is similar to the 7% decline in modelled deposition reported by EMEP for EU27 (Fagerli et al., 2012).

3.2.6. Trends in N leaching/ catchment input-output budgets (ICP W+ICP IM)

Nitrate in surface waters has been highly variable since the late 1980s. Figure x shows regional mean annual concentrations of nitrate in three regions, expressed in % of the 1988 regional mean NO3 concentration. There is no clear regional pattern in NO3 concentrations in lakes and streams. Interannual and inter-site variation dominates rather than long-term trends. However, in those sites where trends are found, declines in nitrate are more common than increases (Garmo et al. 2014).

Comment [HWI32]: ICP IM: do you have relevant text to give here? Perhaps refer to par 3.1.4 in some way, on I-O budgets for N?

Comment [HWI33]: Perhaps add a few words about effects of N in surface waters.

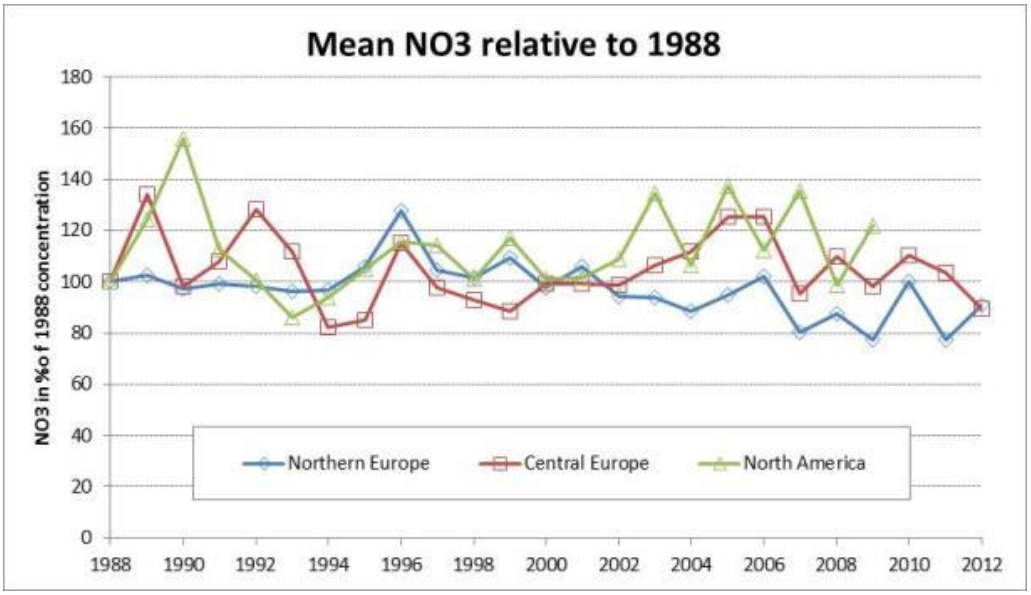


Figure x *Trends in surface water chemistry at ICP Waters sites 1988-2012. Shown are the mean concentrations of NO3 at 22 sites in northern Europe, 21 sites in central Europe and 37 sites in eastern North America. (for 2010-2012, no data available for North America). (Source: De Wit et al. (2015))*

3.3. Heavy metals (ca 5-10 p)

3.3.1. Trends in concentrations and deposition of HMs (EMEP) (max 2 p)

--graph of mean annual concentrations and deposition of HMs in Europe from 1990-2012

Text – on % decrease in deposition and concentrations, which relate to reduce emissions.
Example of text for SO4, to be adapted for HMs.

The decrease in sulphur emissions in both Europe have caused a substantial decrease in the atmospheric deposition the last decades, and this has been reported in several assessments and publications (i.e. EMEP, 2004; IJC 2008; Vestereng, 2007; Sickles, 2007). These reductions are naturally also reflected in precipitation chemistry. All the sites in Europe and 97% of the sites in northeastern North America show a decreasing trend from 1990-2012, with an average reduction of 56% and 37% respectively (**Error! Reference source not found.**).

-please supply a draft text and figures

3.3.2. Trends in exceedances of CLs (CCE/TF M&M) (max 2 p)

The area and magnitude of the exceedances in 1990 and 2010 of critical loads of cadmium, lead and mercury are shown in Figures CCE-5, CCE-6 and CCE-7 respectively. The trends of these exceedances between 1990 and 2010 are shown in Figure CCE-8.

Comment [HWI34]: The background should be highlighted – what is the critical limit? Which ecosystems?

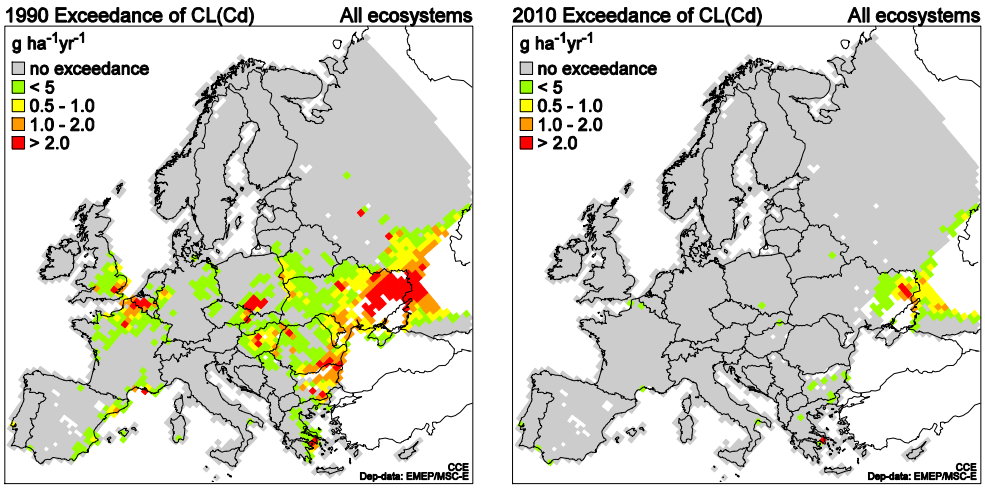


Figure CCE-5: Exceedances (AAE) of the critical loads of cadmium in 1990 (left) and 2010 (right) in Europe. (Deposition data from EMEP-MSCE; European critical loads data from the CCE)

The area at risk of atmospheric deposition of cadmium exceeding its critical load, occuring in many countries in 1990, is limited to scattered regions in 2010 mostly in the eastern part of Europe. However, it should be noted that cadmium input from agricultural practices are not included in these calculations. Limiting computations of the risk of Cd to that caused by atmospheric deposition alone reveals trends between 1990-2010 of the area at risk as well as magnitudes of critical load exceedance that are close to nil (Figure CCE-8, left top and bottom graphs).

The area at risk and magnitude of the exceedance of critical loads of Pb evolves from very high in the whole of Europe in 1990 to relatively low but still occurring in all countries in Europe (Figure CCE-6). The trend of the exceedances since 1990 moves in a downward direction leaving about 10% of the ecosystem area with an exceedance of about 1 g $ha^{-1}yr^{-1}$ (Figure CCE-8, centre top and bottom graphs respectively) in 2010.

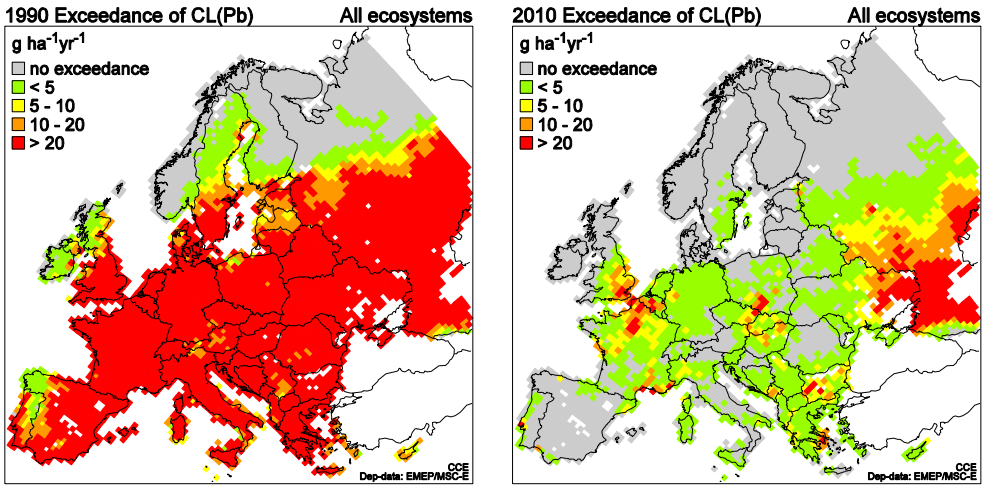


Figure CCE-6: Exceedances (AAE) of the critical loads of lead in 1990 (left) and 2010 (right) in Europe. (Deposition data from EMEP-MSCE; European critical loads data from the CCE)

Figure CCE-7 and CCE-8 confirm that excessive deposition of mercury remains an issue of concern in 1990 as well as in 2010 with about 44% and 25% of the area at risk of exceedance of the critical loads of Hg respectively.

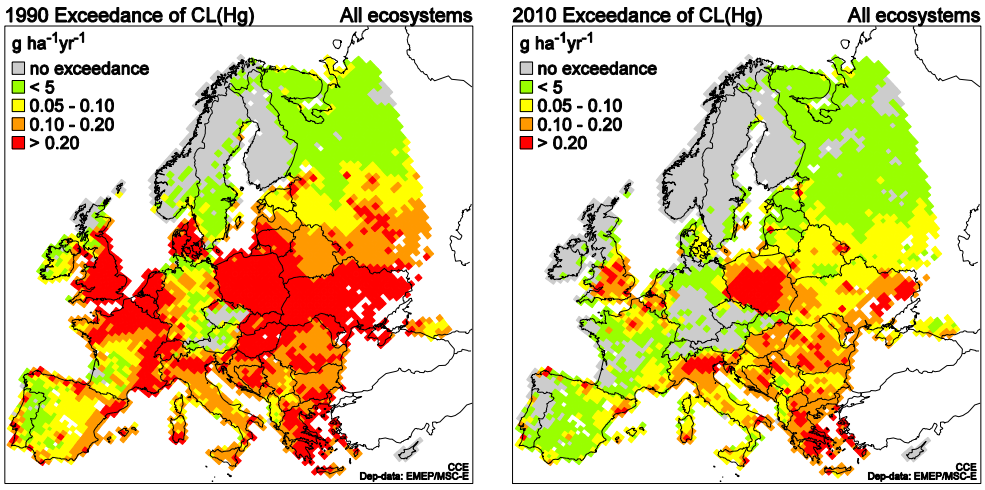


Figure CCE-7: Exceedances (AAE) of the critical loads of mercury in 1990 (left) and 2010 (right) in Europe (Deposition data from EMEP-MSCE; European critical loads data from the CCE)

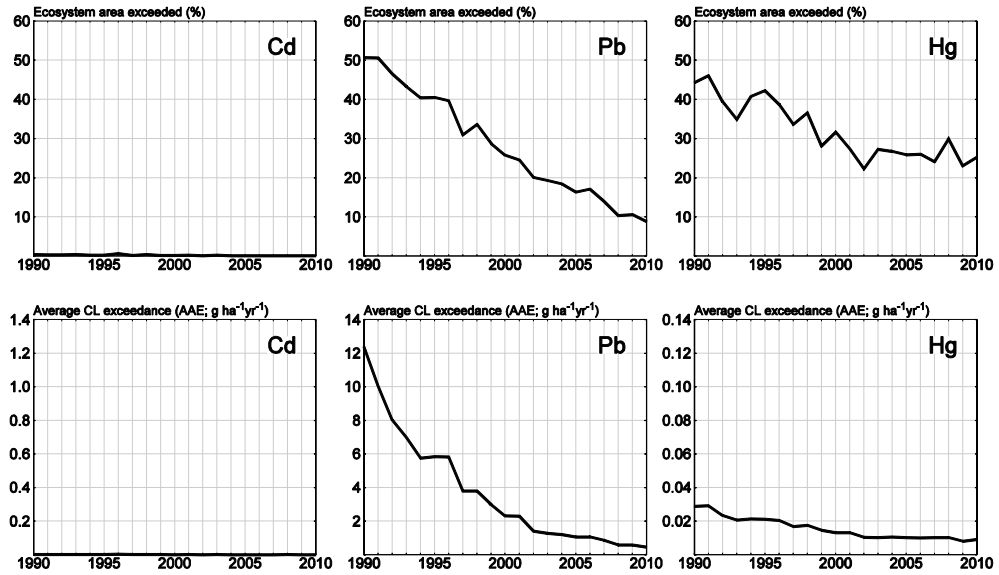


Figure CCE-8: Temporal development of the area (top; % of ecosystems) where critical loads of cadmium (left), lead (centre) and mercury (right) are exceeded and the magnitudes of the average accumulated exceedance (AAE) (bottom; g ha^{-1} yr^{-1}) of the critical loads of these metals respectively (Deposition data from EMEP-MSCE; European critical loads data from the CCE)

3.3.3. Temporal trend (1990 – 2010) in heavy metal concentrations in mosses (max 2 p)

In 2010, the lowest concentrations of metals and nitrogen in mosses were generally found in northern Europe, whereas the highest concentrations were observed in (south-)eastern Europe for metals and the rest of Europe for nitrogen (Harmens et al., 2015). Since 1990, the metal concentration in mosses has declined the most for lead (77%), followed by vanadium (55%), cadmium (51%), chromium (43%), zinc (34%), nickel (33%), iron (27%) arsenic (21%, since 1995), mercury (14%, since 1995) and Cu (11%), as shown in Figures 3 and 4. For lead and cadmium, the decline is similar to those reported by EMEP for the modelled deposition across Europe, i.e. 74% and 51% for lead and cadmium respectively (Figure 1). The 14% decline in Hg between 1995 and 2010 was lower than the decline (27%) in EMEP modelled deposition across Europe. For mercury the decline was lower than for cadmium and lead due to hemispheric transport of mercury across the globe, resulting in a considerable contribution of mercury pollution from other continents to mercury deposition in Europe (Travnikov et al., 2012).

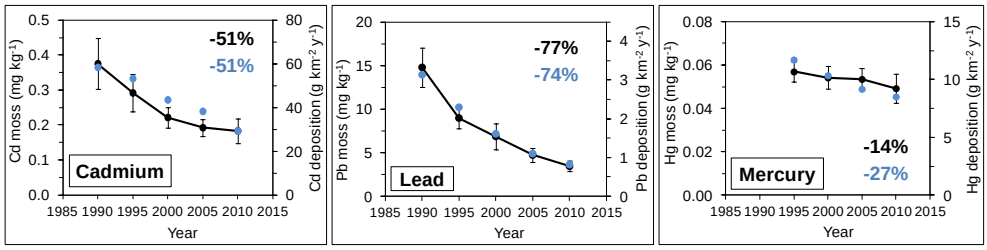


Figure 3. Average median metal concentration in mosses for countries that reported metal data for at least four survey years since 1990; some countries reported three survey years since 1995 for mercury (Harmens et al., 2015). The blue dots in the graphs show the decline in deposition across Europe as modelled by EMEP (Travnikov et al., 2012). The black and blue values in the graphs represent the percentage decline between 1990 (1995 for mercury) and 2010 for the moss concentrations and EMEP modelled depositions respectively.

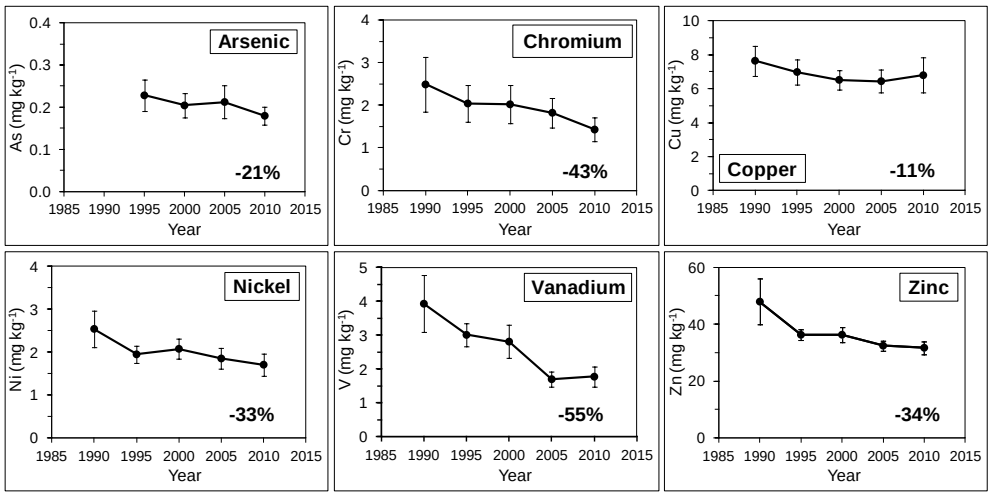


Figure 4. Average median metal concentration in mosses for countries that reported metal data for at least four survey years since 1990 (1995 for arsenic; Harmens et al., 2015). The values in the graphs represent the percentage decline between 1990 (1995 for arsenic) and 2010.

Comment [hh35]: This figure could be omitted.

3.3.4. Temporal patterns in soil and stream water mercury, lead and cadmium chemistry

Comment [HWI36]: The length of 3.3.4 is now 3 pages. I think it can be reduced to two pages.
Also, see my emails with feedback on this text.

Data reported to the ICP IM Programme Centre were used for the evaluation of temporal changes and trends of heavy metals in soil and runoff at ICP IM sites across Europe. Trends of metal concentrations at ICP IM sites are features included in subprogrammes for soil chemistry and runoff water chemistry. Reported data in soil chemistry and runoff from European sites were evaluated to test for the occurrence of temporal changes/trends in metal concentrations. At Swedish ICP IM sites (Aneboda, Gårdsjön, Kindlahöjden and Gammtratten) soil samples have been collected with regular intervals over the last decades and were tested for temporal trends in more detail.

Reported soil chemistry data at ICP IM sites outside Sweden show a sampling frequency between two and three (at a few occasions four) times between 1994-2011. Within soil profiles there were only one value from each soil depth reported at each time, except from Sweden where six samples from soil plots (50 * 50 m) taken every 5-10 years. Temporal changes of metal concentrations were estimated by simple linear regression between sampling year and metal concentrations for each site. For soil metal concentrations, values from soil layers (litter+organic layer, 0-5 cm depth, 10-20 cm depth, 30-50 cm depth, 50- cm depth) were used for the determination of temporal changes. At the four Swedish ICP IM sites, linear regressions were used to determine temporal trends of soil metal concentrations in the FH-layer, and mineral soil at 0-5 cm and 10-20 cm depths.

For soil chemistry, Sweden excluded, temporal changes at each ICP IM site and soil depth were attributed with a symbol indicating either an increase (+) or decrease (-) in metal concentration over time. Strength of the changes were indicated with either one (+/-), two (++ / --) or three (+++ / ---) symbols. Higher number of symbols means greater strength ($R^2 < 0.3$; $0.3 < R^2 < 0.7$; $R^2 > 0.7$).

In stream water, annual metal concentration means were used as dependent variables in regression analysis. At ICP IM sites outside Sweden temporal trends in stream water metal concentrations could not be evaluated properly, since the reported data covered short sampling periods (2008-2010). At Swedish ICP IM sites, samples were collected and analyzed every month from 1994 to 2013.

Time-trends in soil metal concentrations

European data

Metals in soil and runoff are to a large degree dependent on long-term and long-range atmospheric transport, and the main CLRTAP priority has been on mercury (Hg), lead (Pb) and cadmium (Cd) (Bringmark et al. 2013). Significant changes at the ICP IM sites were estimated from the proportion of changes (symbols + decrease and - decrease) and strength in significance indicated by number of symbols. Equal proportion indicated no change. In the forest floor layer (litter + organic topsoil) trends for Hg at four sites revealed the results of 2 sites with + of 4 sites giving total trends = 0.5, Pb (13 / 21 = 0.62) or Cd (13 / 20 = 0.65) (Table 3.3.5). In the upper mineral soil layer (0-5 cm), significant increases of Hg (4 / 4 = 1) and Pb (12 / 12 = 1) could be observed over time while the temporal change for Cd was weaker (6 / 9 = 0.67). There were more indications of increasing than decreasing trends on Cd, however not significant.

Table 3.3.5. Temporal trends based on the proportion of number indicating increasing (+) and decreasing (-) temporal changes in Hg, Pb and Cd in soil concentrations at 2-28 ICP IM sites located in up to 8 countries from the Mediterranean to Boreal region. Number of sites for each element and soil layer would be the sum of + and -.

Soil layer, cm	Hg		Pb		Cd	
	+	-	+	-	+	-
Litter + organic topsoil	2	2	13	8	13	7
5	4	0	12	0	6	3
10-20	2	1	9	9	12	7
30-50	0	6	15	13	8	6
50-	1	1	9	6	9	3

Swedish data

For Swedish ICP IM sites Pb concentrations in the forest floor (F+H-horizons) decreased between 1994 and 2011 in sites Aneboda, Gårdsjön and Kindla. For Cd, the content decreased in site Aneboda and Gårdsjön (Fig. 3.3.5). Mercury concentrations, on the other hand increased over the same period in sites Gårdsjön and Kindlahöjden. In mineral soil layers, at a depth between 0 and 5 cm Pb increased in sites Gårdsjön and Gammtratten where also Cd increased, as well as in site Kindla (Fig. 3.3.5). In Kindla, also Hg concentration increased in the upper mineral soil horizon. In deeper mineral soil layers (10-20 cm), Pb concentrations increased significantly at all Swedish IM-sites (Fig. 3.3.5). The Hg concentration also showed increased values in the mineral soil at 10-20 cm depth in site Kindla. No significant changes in Cd concentrations could be seen at any site in the lower parts of the mineral soil.

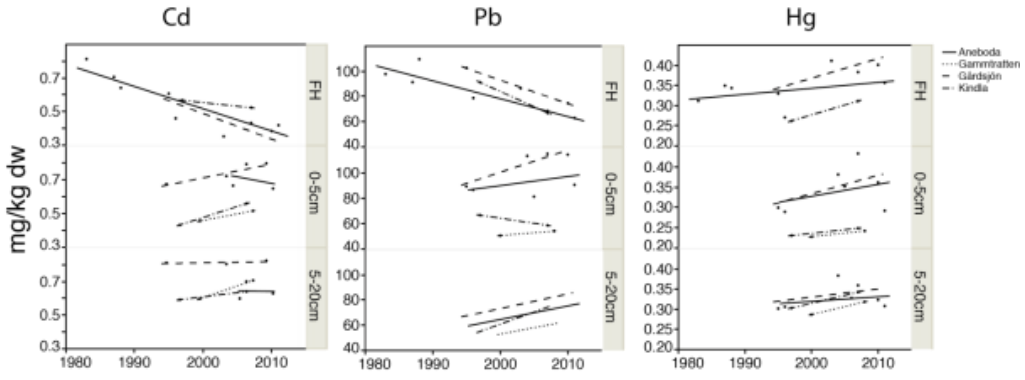


Figure 3.3.5. Soil metal concentrations at the four ICP IM sites in Sweden (Aneboda, Gammtratten, Gårdsjön, Kindla).

Time trends in stream water metal concentrations

For Swedish IM sites almost 20 year records of Pb, Cd and Hg concentrations could be evaluated. Cadmium concentrations decreased at sites Kindla and Gårdsjön, while no significant changes were observed at the other two sites. Regarding trends for Pb and Hg at the Swedish sites no significant trends were observed, except for Hg concentrations that increased at Gårdsjön.

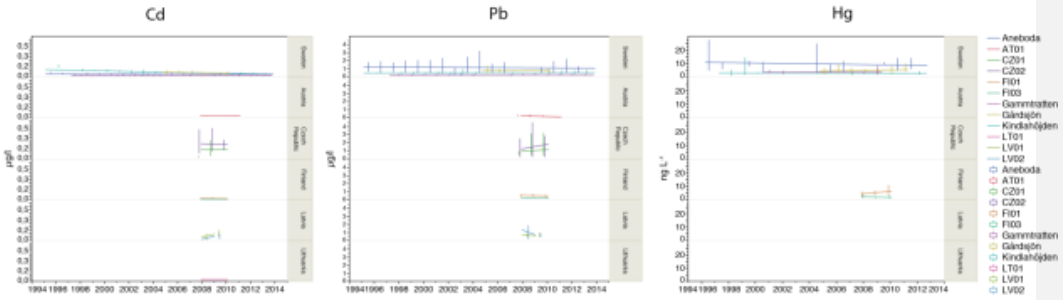


Figure 3.3.5_2. Stream water Cd and Pb (mg L^{-1}) and Hg (ng L^{-1}) concentrations at European ICP IM sites in six countries. Error bars indicate 10th and 90th percentiles of annual metal concentrations from each site.

Comment [HWI37]: I suggest to leave out the sites where you don't have enough data to do a trend analysis. Look at figure above to improve the figure. Can you leave out all the site ID's? Just refer to a report instead?

Key conclusions:

- The trends in soil Pb and Cd concentrations in the forest floor showed decreases meaning that deposited elements now are transferred to deeper soil layers but still accumulation is ongoing considering the total soil profile. Toxic elements still occurred in the soil at elevated contents.
- Mercury, Hg concentrations, are increasing in the forest floor with hazardous effects on biological activity and provide extended possibilities for methylation and release to surface waters. Hg is also transferred to deeper soil layers with ongoing accumulation and connected impacts.

The trends in stream water concentrations of heavy metals were limited, but for Cd and Pb downward trends were found similar to the top soil layers, which document less release to surface waters. Hg may show opposite conditions as trends on increasing surface water concentrations have been observed.

3.3.5. Trends in mercury in fish (ICP Waters)

Aquatic biota in northern freshwater ecosystems contain elevated concentrations of Hg, related to anthropogenic emissions of Hg to the atmosphere (Driscoll et al. 2013). In North America and Fennoscandia, fish Hg concentrations exceed limits advised for human consumption (0.3-0.5 μg Hg g^{-1} wet weight) in thousands of lakes and rivers. Adverse behavioral effects on fish have been found above 0.5 μg Hg g^{-1} ww, while sublethal effects on for instance reproduction have been found below 0.2 μg Hg g^{-1} ww Depew et al. (2012). Global emissions of Hg are currently increasing (Pirrone et al. 2010). A compilation of multi-annual studies of Hg levels in terrestrial, freshwater and marine biota in polar and circumpolar areas in North America and Scandinavia, under coordination of the Arctic Council, suggests that no trends and rising trends of Hg dominate (Riget et al. 2011)). However, only a few time series for freshwater fish were included. Increases in concentrations of Hg in freshwater fish from the 1990s onwards have been documented in Sweden (Akerblom et al. 2012), Norway (Fjeld et al. 2009) and Ontario (Gandhi et al. 2014), although this rising trend is not found in all regions and for all fish species. Despite reduced Hg emissions in Europe and North America, there is little evidence to suggest that Hg contamination of fish is beginning to decline.

3.4. POPs (no more than 5 p)

Comment [HWI38]: Is there nothing more on POPs?

3.4.1. Trends in concentrations and deposition of POPs (EMEP) (max 1 p)

--graph of mean annual concentrations and deposition of POPs in Europe from 1990-2012

Text – on % decrease in deposition and concentrations, which relate to reduced emissions.
Example of text for SO4, to be adapted for POPs.

The decrease in sulphur emissions in both Europe have caused a substantial decrease in the atmospheric deposition the last decades, and this has been reported in several assessments and publications (i.e. EMEP, 2004; IJC 2008; Vestereng, 2007; Sickles, 2007). These reductions are naturally also reflected in precipitation chemistry. All the sites in Europe and 97% of the sites in northeastern North America show a decreasing trend from 1990-2012, with an average reduction of 56% and 37% respectively (**Error! Reference source not found.**).

-please supply a draft text and figures

3.4.2. Trends in POPs in fish (ICP Waters) (2 p)

The ICP Waters report on POPs (Fjeld et al. 2005) included trends in polychlorinated biphenyls (PCBs) in fish in Swedish reference lakes since the 1960s until the 1990s. This Swedish time series was extended to the year 2012 and published by Nyberg et al. (2014), presenting times series of PCBs in perch, pike, and Arctic char, in nine lakes.

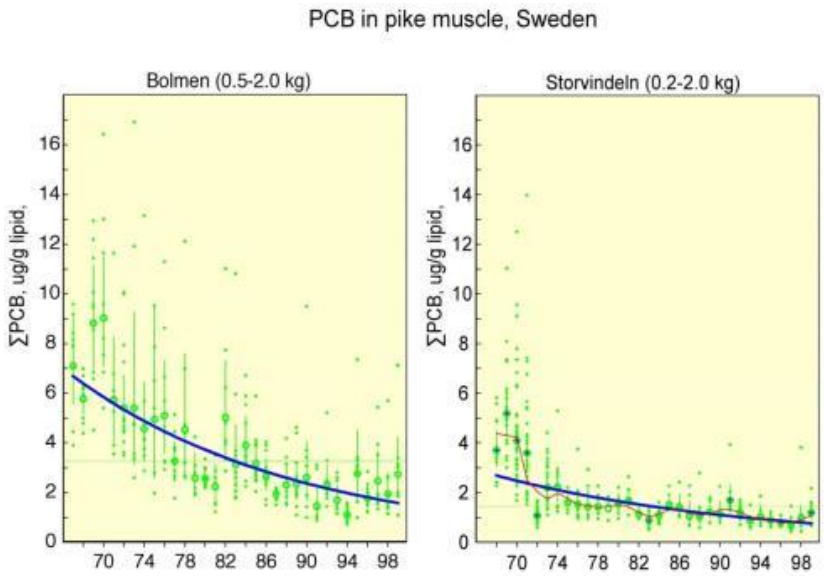


Figure x. *Example of two time series of PCBs in pike muscle (µg/g lipid) from Lake Bolmen (southern Sweden) and Lake Storvindelen (northern Sweden). Source: Fjeld et al., 2005; Nyberget et al., 2014.*

The key results of the Nyberget study are as follows: Overall, PCB concentrations in fish were decreasing, but this was not consistent for all types of PCBs across all lakes and species. The concentrations of PCBs were generally decreasing by about 3–8 % per year in both pike and Arctic char. No trend was observed for the perch time series, but this was most likely due to the short duration of these time series and because monitoring in perch started after the steep decrease, during the 1980s and 1990s, observed for pike and Arctic char.

The results are put into context by Nyberg et al. (2014) in the following way (text almost copied directly from journal): In a review on temporal trends of PCBs in arctic biota, Riget et al. (2010) found a mean annual decrease for the PCB CB-153 of 1.2 % based on all the time series analyzed in the review (40 in total), which was somewhat lower than the decrease found in the time series in Nyberg et al. (2014). A number of the time series presented in Riget et al. (2010) started in the 1990s, later than the time series in Nyberg et al. (2014), with lower concentrations and less marked decreases. The decrease over time, seen for concentrations of PCBs in both freshwater and marine fish in Sweden, mirrors the measures taken (e.g., bans and restrictions) to reduce PCBs in the environment. Gewurtz et al. (2010) found similar results for various fish species in boreal Canada: a steep decrease in PCBs in the 1970s and 1980s, levelling out in the mid-1990s, most probably as a result of bans and restrictions. Thus, long-term monitoring records of PCBs in fish in boreal Sweden, boreal Canada and Arctic regions all document a decrease in PCBs in fish.

3.5. PMs (ca 5 p)

3.5.1. Trends in concentrations (and deposition?) (EMEP) (max 1 p)

--graph of mean annual concentrations (and deposition?) of PMs in Europe from 1990-2012.
Is it important to discern between urban and rural sites? Just asking.

Text – on % change in concentrations, which relate to emissions.
Example of text for SO4, to be adapted for PMs.

The decrease in sulphur emissions in both Europe have caused a substantial decrease in the atmospheric deposition the last decades, and this has been reported in several assessments and publications (i.e. EMEP, 2004; IJC 2008; Vestereng, 2007; Sickles, 2007). These reductions are naturally also reflected in precipitation chemistry. All the sites in Europe and 97% of the sites in northeastern North America show a decreasing trend from 1990-2012, with an average reduction of 56% and 37% respectively (**Error! Reference source not found.**).

-please supply a draft text and figures

3.5.2. Trends in health responses, PM2.5 PM10. (TF Health) (max 2 p)

Particulate matter is a mixture with physical and chemical properties that vary by location. Biological components, such as allergens and microbes, are also found in particulate matter. The health effects of particulate matter are well documented. They are due to both short-term (hours, days) and long-term (months, years) exposure and include: respiratory and cardiovascular morbidity, such as aggravation of asthma, respiratory symptoms and an increase in hospital admissions; and mortality from cardiovascular and respiratory diseases and lung cancer (WHO Regional Office for Europe, 2013; Loomis et al., 2013).

Figures 1 and 2 show the average levels of exposure to PM_{10} and PM2.5, respectively, for 2012 (or the most recent year of data available) for Member States of the WHO European Region. The population-weighted country-level average background PM10 exposure in urban or suburban areas varied from 8.7 $\mu g/m^3$ to 71.0 $\mu g/m^3$. The most recent data on PM10 shows that in 2012 from 31 countries with available data only eleven presented values below the WHO air quality guideline value of 20 $\mu g/m^3$. A variation in exposure levels of twofold to threefold was observed between cities in some countries. For PM2.5, also in 2012 (or the most recent year available), the levels varied from 4.6 $\mu g/m^3$ to 50.4 $\mu g/m^3$. The data for PM2.5 shows that in 2012 from 27 countries where data were available only seven presented values below the WHO air quality guideline value of 10 $\mu g/m^3$.

Figures 3 and 4 show trends in population exposure for PM10 and PM2.5, respectively. In general, population-weighted average exposure to PM_{10} and $PM_{2.5}$ in all cities of the Region for which data are available has not changed substantially over the last few years. The number of monitoring stations, however, has increased over the years, especially for $PM_{2.5}$. In 2012, the PM_{10} and $PM_{2.5}$ data from regular population-relevant monitoring were available, respectively, for 479 cities in 30 countries and 300 cities in 26 countries. In European cities where particulate matter is monitored, 75.4% and 94.0% of people experience annual levels exceeding the WHO air quality guideline for PM_{10} (20 $\mu g/m^3$) and $PM_{2.5}$ (10 $\mu g/m^3$), respectively (yearly average values, WHO Regional Office for Europe, 2006). This gives rise to a substantial risk to health. For 28.6% of urban residents, the EU limit value for PM_{10} (40 $\mu g/m^3$) was exceeded in 2012.

For both particulate matter and ozone, ground-level monitoring is very limited in countries in eastern Europe, the Caucasus and central Asia, due to the small number of monitoring stations. Monitoring needs to be improved in many countries to assess population exposure and assist local authorities in establishing plans for improving air quality.

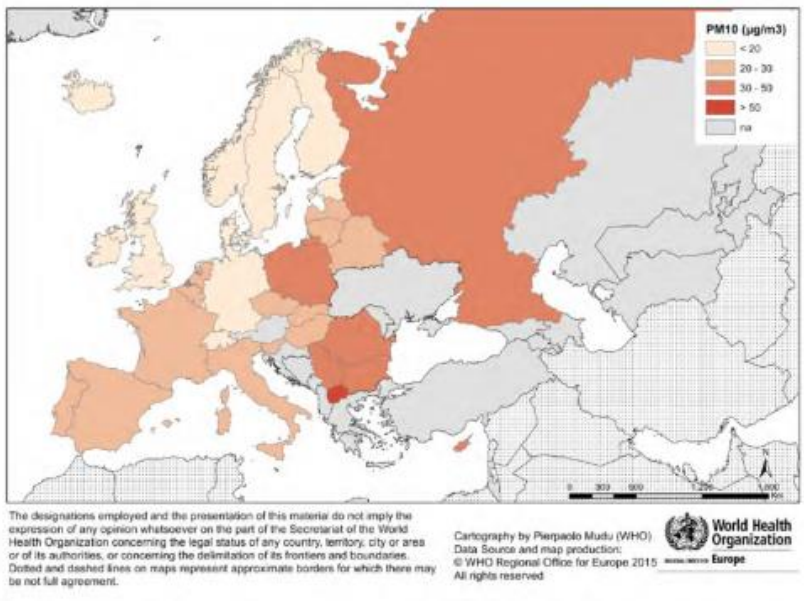


FIGURE 1. Population weighted annual mean PM_{10}, for 2012 or the last year available *(compressed version for draft report)*

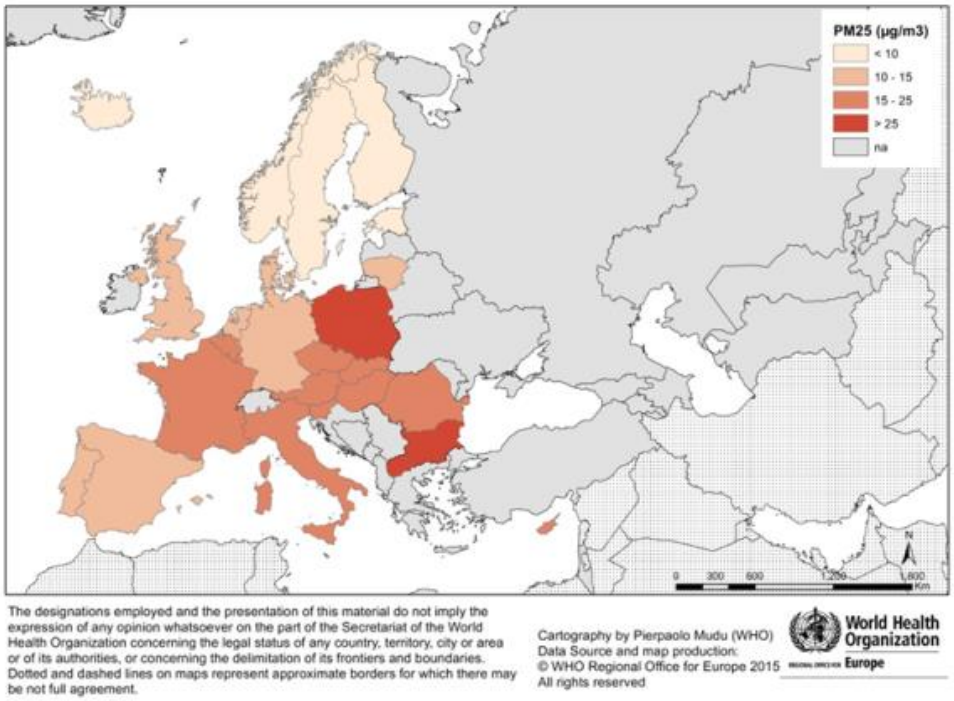


FIGURE 2. Population weighted annual mean $PM_{2.5}$, for 2012 or the last year available *(compressed version for draft report)*

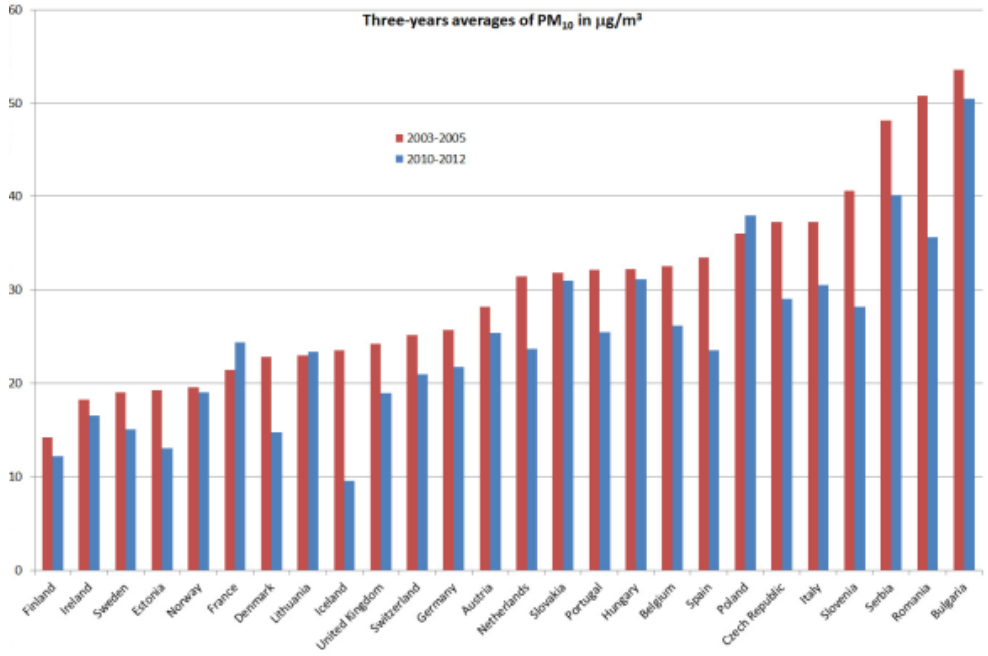


FIGURE 3. Population weighted annual mean PM_{10}, three-year averages, for 2003-2005, and 2010-2012.

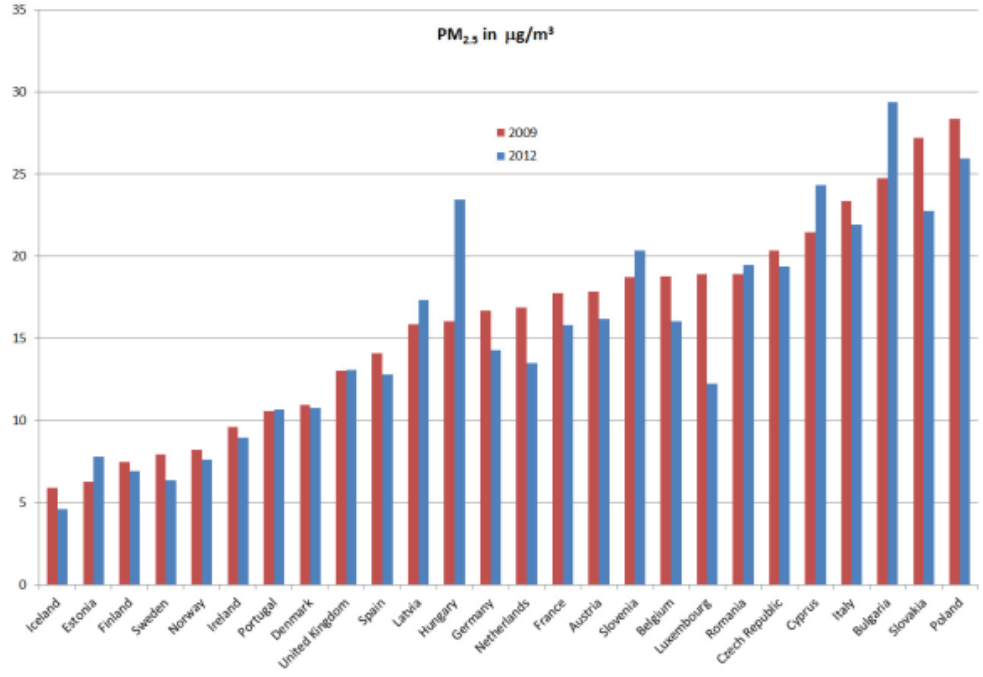


FIGURE 4. Population weighted annual mean $PM_{2.5}$, one-year averages, for 2009 and 2012.

3.5.3. Trends in soiling (TF Materials)

Evaluation of soiling of modern glass started later than the corrosion measurements and the first trend exposure was performed in 2005-2006. The range of haze variation is similar for the three exposure campaigns (Figure X). Minimum values are 0.9% (Svanvik) for 2005-2006, 1.0% (Chaumont) for 2008-2009 and 1.0 % (Svanvik) for 2011-2012. Maximum values are found for Athens (8.9% in 2005-2006, 10.5% in 2008-2009 and 10.1% in 2011-2012). The average haze (for sites where measurements for the three exposures are available) is comparable for the three years: 2.9 ± 1.9 % (2005-2006), 3.0 ± 2.2 % (2008-2009) and 3.1 ± 2.3 % (2011-2012).

Regarding the observed trends, haze increases significantly in Bottrop, Casaccia, Venice, Stockholm and Katowice. It decreases for Prague, Paris and Vienna and remains constant in Kopisty, Rome, Milan, Oslo, Birkenes, Aspvreten, Madrid, Toledo, Lahemaa, and Athens. For the other sites, it is difficult to draw a single trend: haze increases from 2005-2006 to 2008-2009 but decreases for the last exposure in Berlin and Svanvik, whereas it decreases from 2005-2006 to 2008-2009 and increases in 2011-2012 for Chaumont. In summary, by considering all the sites, haze does not show a clear trend. The majority of sites display similar haze values during the three campaigns.

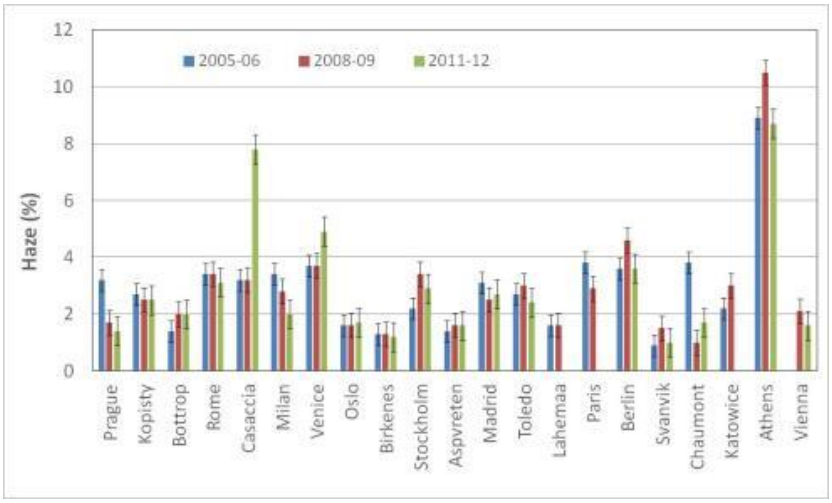


Figure X: Comparison of Haze (in %) during the three extensive campaigns (2005-2006; 2008-2009; 2011-2012).

Figure X shows predicted values of haze for different types of test sites. In contrast to corrosion, there are no "official" target values for haze, but a value of 1% was considered for this analysis. This corresponds to the threshold where the deposit leads to visual nuisance detected by human eyes. Figure X displays the annual evolution of haze predicted by the dose-response function based on a multi-linear regression. Further development of the dose-response function involved using neural networks (Verney-Carron et al., 2012). Input environmental data are measured environmental parameters for the 4 sites: Athens, Katowice, Paris and Chaumont, each corresponding to a particular type of environment (traffic, industrial, urban and rural, respectively). The threshold is exceeded after 90 days for the traffic site, 110 days for the industrial one and 130 days for the urban sites, whereas it is reached after a year for the rural site. As for corrosion there is a clear difference between polluted and non-polluted areas.

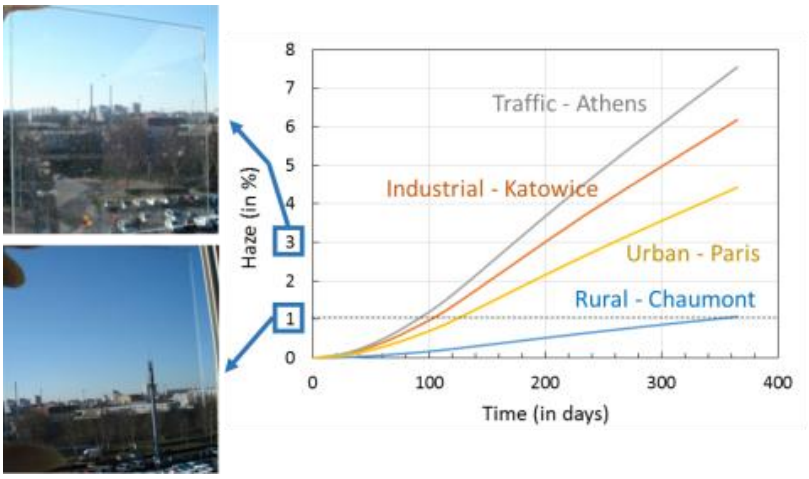


Figure X. Predicted haze (in %) as a function of time for 4 types of sites: rural, urban, industrial and traffic. Input environmental data for the 2008-2009 campaigns were selected (Lombardo et al., 2014). Visual aspects are shown on 2 pictures: Katowice after 1 year (haze = 3%), upper image and Chaumont (haze = 1%), lower image.

3.6. Ozone (ca 5-10 p)

3.6.1. Trends in concentrations (EMEP) (max 2 p)

-also trends in precursors? NOx, VOC? Possible to cross-reference to other paragraphs?

3.6.2. Trends in ozone concentrations, fluxes into leaf pores and effects (1999 – 2010) (ICP Vegetation) (max 2 p)

Despite a more than 30% reduction in European emissions of ozone precursors during the last two decades, a decline in ozone levels is generally not seen at EMEP ozone monitoring stations (Torseth et al., 2012; Simpson et al., 2014). Rural background data over 1990 – 2010 show a decrease in the highest levels and a corresponding increase in the very low levels at sites in the UK, the Netherlands and some other countries, but no clear trends in for example Switzerland or Austria. Whereas the median ozone concentration has hardly changed, peak concentrations (above >95th percentile) have declined and background concentrations (lower percentiles) have risen in many places (Simpson et al., 2014). Reduced precursor emissions might well be masked by large inter-annual variations in ozone, caused by, for example climate, weather or biomass burning events.

Comment [HWI39]: Please clarify what kind of vegetation they are located in. Not forest?

Statistical analysis of the ICP Vegetation ozone data showed that in recent years the proportion of hourly ozone concentration in the lowest and highest ozone categories has declined significantly ($P < 0.10$), whereas the proportion in the category 20 - 39 ppb has increased significantly; the proportion in the category 40 - 59 ppb has not changed (Table 1). These results confirm the general trend observed across Europe, i.e. background ozone concentrations have risen whereas peak concentrations have declined.

Table 1. Trends (1999 – 2010) in ozone concentrations at ICP Vegetation monitoring sites.

Ozone concentration	Europe	Sites showing European trend
0-19 ppb	Decline	Tervuren (BE), Seibersdorf (AT)
20-39 ppb	Increase	Östad (SE), Ascot (GB), Tervuren (BE), Giessen (DE)
40-59 ppb	None	All, except increase in Seiberdorf (AT)
>60 ppb	Decline	Ljubljana (SI)

No temporal trends were found for the 24 hr mean and daylight mean ozone concentrations (Table 2). This is in agreement with trends reported for mean and median ozone concentrations at EMEP monitoring sites. However, night time mean and daily minimum ozone concentration have increased (0.27 ppb per year) across Europe, although only significantly in Tervuren, Belgium. Despite a decline in the ozone concentrations of 60 ppb or higher, the average European daily maximum ozone concentration has not changed, although a decline was reported for Ljubljana, Slovenia.

Table 2. Trends* (1999- 2010) in ozone concentrations and leaf fluxes at ICP Vegetation monitoring sites**.

Country	Site	24 hr mean	Daylight mean	Night mean	Daily max	Daily min	AOT40[a]	POD_3 IAM[b]
Belgium	Tervuren	None	None	Increase	None	Increase	None	None
Slovenia	Ljubljana	None	None	None	Decline	None	Decline	None
European mean		**None**	**None**	**Increase**	**None**	**Increase**	**None**	**None**

* The non-parametric Mann-Kendall test was applied.

** Data are shown for sites with at least one significant trend (P < 0.10). No significant trends for any of the variables were observed for Östad (Sweden), Ascot (UK), Giessen (Germany), Seibersdorf (Austria) and Pisa (Italy).

AOT40[2] and ozone fluxes into the leaf pores (POD_3IAM[3]) provide an indication of ozone risk to vegetation. It should be noted that ozone flux provides a biologically more meaningful assessment of ozone risk (Mills et al., 2011). Despite a European decline in ozone concentrations of more than 60 ppb, AOT40 did not show any trend, apart from a decline in Slovenia (0.6 ppm h per year), similar to the trend observed for the daily maximum ozone concentration (decline of 1.2 ppb per year).

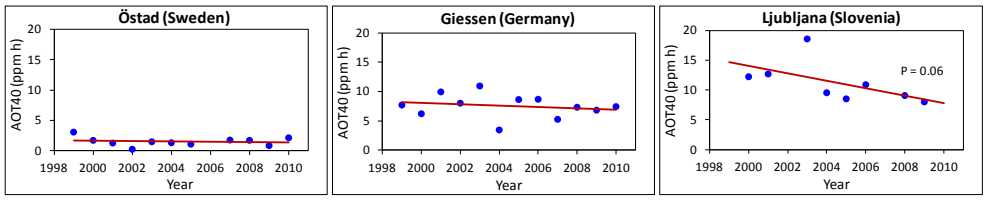


Figure 5. Trend in AOT40 at three selected sites in Europe. A significant trend (P = 0.06), a decline of 0.6 ppm h per year, was found only in Ljubljana (Slovenia).

Concentrations much lower than 40 ppb contribute to the ozone flux. The ozone flux into leaves showed no trend between 1999 and 2010, indicating that risk of ozone-induced effects on wheat has not changed. Previous analysis had shown no significant trend in the impact of ozone on leaf damage in white clover between 1999 and 2006 (Hayes et al., 2007). Considering the annual variation in ozone concentrations due to climate variation, much longer time series are probably required to detect temporal trends in ozone concentrations and effects on vegetation across Europe.

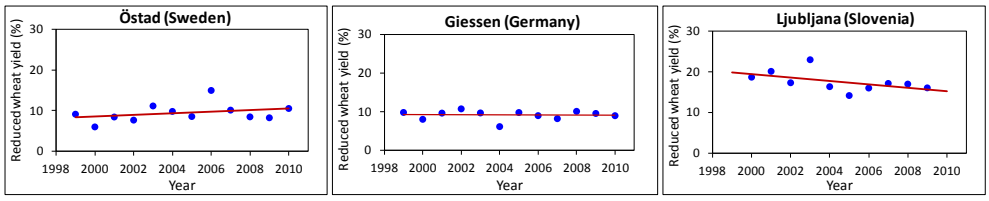


Figure 6. Reduction (%) in wheat yield due to ozone fluxes in recent years compared to pre-industrial ozone fluxes of a POD_3IAM of zero (corresponding to ozone concentrations in the range of 10 – 15 ppb), assuming no soil water limitation. No significant trends were found.

The lack of trends in ozone flux into the leaf pores means that there is also a lack of trends in the effects of ozone on wheat yield (Figure 6). On average over the period 1999 - 2010, wheat yield was the least reduced by ozone in the area of Seibersdorf (7%), followed by Östad, Tervuren and Giessen (9%) and was most reduced in the area of Llubljana (18%), assuming no soil water limitation. A rise in

[1] AOT40 = Sum of differences between hourly mean ozone concentration (in ppb) and 40 ppb for each hour when the concentration exceeds 40 ppb, accumulated during daylight.
[3] POD_3IAM = Phytotoxic Ozone Dose above a flux threshold of 3 nmol m^{-2} s^{-1}, accumulated during daylight hours. Parameterisation is based on wheat for application in integrated assessment modelling (IAM). Modelling includes a simplistic representation of soil moisture within the EMEP model.

the background concentration in a relatively low ozone year, for example 2009, would result in additional wheat yield reduction of about 2% at most sites and ca. 3% in Ljubljana.

Future trends in ozone impacts on crop yield

Comment [hh40]: Could be omitted.

When the revised Gothenburg Protocol scenarios (Amann et al., 2013) were applied to estimate yield losses for a generic crop, based on parameterisation of the wheat flux model for application in integrated assessment modelling (POD_3IAM), it was calculated that ambient ozone levels in the base year 2005 reduce crop yield by 10.7% on average compared to pre-industrial ozone levels in EU27 + Norway + Switzerland (Figure 7). With full implementation of the revised Gothenburg Protocol, crop yield loss is predicted to decline to 8.8% in 2020 and 8.2% in 2030. Hence, crops will remain at risk of adverse effects of ozone in the near future. It remains unclear how emission controls in Europe may be offset by global background ozone increases, by changes in longer-lived ozone precursors such as methane or by changes in chemical processing or transport driven by future shifts in climate. Applying the latest climate change scenarios, surface ozone concentrations are predicted to decline in future in Europe and North-America, with the magnitude of decline depending on scenario, whereas an increase is expected in South Asia. Limiting atmospheric methane growth is becoming more important when emissions of other ozone precursors are controlled (HTAP, 2010; Wild et al., 2012).

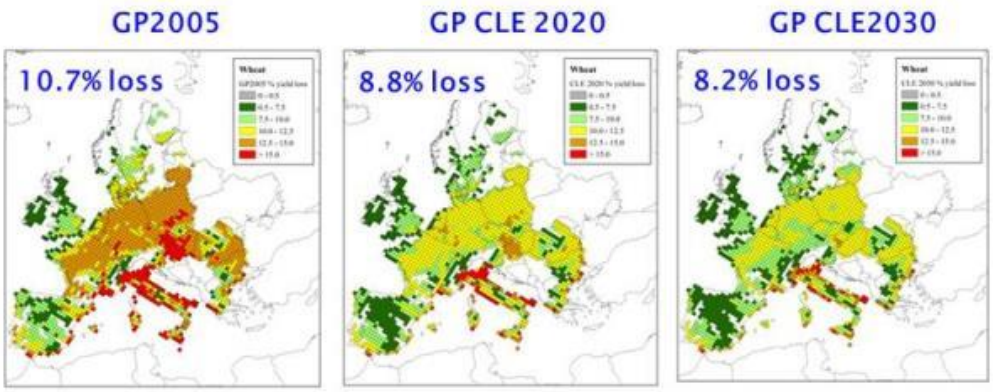


Figure 7. Calculated percentage yield losses due to ozone effects on wheat per 50 km x 50 km EMEP grid square in 2005, 2020 and 2030 for the wheat growing areas of the EU27+CH+NO for the base year (2005) of the revised Gothenburg Protocol (GP) and with implementation of its current legislation (CLE) for 2020 and 2030. The percentage yield loss was calculated using the generic ozone flux model for application in integrated assessment modelling (POD_3IAM; LRTAP Convention, 2015). The EMEP model includes a simplistic representation of soil moisture (Simpson et al., 2012).

Conclusions

- The implementation of air pollution abatement strategies in Europe in recent decades has contributed to the general decline in heavy metal concentrations in Europe. The slower implementation of air pollution abatement policies in parts of eastern and south-eastern Europe has likely contributed the relatively high levels of heavy metal pollution in this area.
- Few trends have been observed at ICP Vegetation monitoring sites between 1999 and 2010 regarding ozone concentrations and risk of ozone impacts on vegetation. Time series much longer than 12 years are required to distinguish significant long-term trends from inter-annual variability in ozone concentrations due to climate variation.

- Whereas peak concentrations of ozone have declined in recent decades in some (but not all) parts of Europe, an increase in background concentrations at the same time has contributed to no change in median or average ozone concentrations across Europe. The rise in background concentrations can contribute significantly to impacts of ozone on vegetation.
- Ozone pollution in the future is critically dependent on changes in regional emissions and global transport of ozone precursors.

3.6.3. Ground level ozone concentrations and exposures (ICP Forests) (max 2 p)

Comment [HWI41]: Is there a relation to crown condition here? Possible to cross-reference?

Comment [HWI42]: Marcus Schaub [a], Matthias Haeni [a], Marco Ferretti [b], Elena Gottardini [c], Vicent Calatayud [d]
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[b] TerraData environmetrics, Via L. Bardelloni 19, I-58025 Monterotondo M.mo (GR), Italy
[c] Research and Innovation Centre, Fondazione Edmund Mach (FEM), Via E. Mach 1, 38010 San Michele all'Adige, Italy
[d] Fundacion Centro de Estudios Ambientales del Mediterraneo (CEAM), Parque Tecnologico, Paterna, Spain

Tropospheric ozone (O3) concentrations from passive samplers have been monitored according to harmonized methodologies on ICP Forests intensive monitoring (Level II) sites, starting in the year 2000. The objective of measuring the concentrations of ozone is to contribute to a better understanding of the actual exposure of European forest ecosystems to air pollutants. In particular, we aim to (i) quantify ozone concentrations over the course of the vegetation period (April-September), (ii) estimate the related ozone exposures of forest ecosystems, and (iii) detect temporal and spatial trends across Europe.

Passive sampling is the standard method for ozone concentration measurements adopted by ICP Forests (Schaub et al., 2010) and was verified by means of specific tests carried out in comparison with conventional monitors. It has been proven to be a valuable method at remote sites (e.g. Sanz et al., 2007; Gottardini et al., 2010; Hůnová et al., 2011) where the availability of electric power is often limited. By means of passive sampling, the determination of ambient air concentrations can be achieved at relatively low costs and with sufficient accuracy at the very forest site. Here, we analyzed the temporal and spatial trends for i) ozone concentrations (reported as volume:volume, in parts per billions, ppb) and ii) ozone exposure (reported as ozone Accumulated Over a Threshold of 40 ppb, AOT40) for the 2000-2013 period on Level II sites across Europe.

The results presented here are based on 18'362 measurements from 214 sites and the following 20 countries: Austria (AUT), Belgium (BEL), Cyprus (CYP), Czech Republic (CZE), Estonia (EST), France (FRA), Germany (DEU), United Kingdom of Great Britain (GBR), Greece (GRC), Hungary (HUN), Ireland (IRL), Italy (ITA), Latvia (LVA), Luxembourg (LUX), Poland (POL), Romania (ROU), Slovakia (SVK), Slovenia (SVN), Spain (ESP) and Switzerland (CHE). In these countries, methods have been applied according to the ICP Forests Manual, Part XV on Monitoring Air Quality (Schaub et al., 2010). For quality assurance, only data measurements within the period from 1 April until 30 September that are higher than 5 ppb and lower than 140 ppb (plausibility check) have been considered. As the exposure time of passive samplers differed from time to time and among sites, mean calculations were weighted according to exposure time. For trend analyses, the Sen's slope method (Sen 1968) plus Xuebin Zhang's (Zhang 2000) and Yue-Pilon's (Yue 2002) pre whitening approaches to determining trends in climate data have been applied according to Bronaugh (2013).

April-September mean ozone concentrations ranged from 23 to 64 ppb. A decreasing south-north gradient across Europe is apparent with the highest concentrations being measured in Italy, southern Switzerland, Czech Republic, Slovakia and Greece (Figure 1). An overall trend analyses, including all data from 20 countries and 2000-2013 reveals a significant decrease of 0.5 ppb per year (n = 18'362; p = 0.000) (Figure 2). When considering only sites with a data coverage of at least 6 years and 120 days (66%) from 1 April until 30 September, site-specific trend analyses did not reveal any uniform pattern across Europe.Ozone exposures in terms of AOT40 (EU Directive 2008/50 CE) have been assessed according to Ferretti et al. (2012).Mean AOT40 for 2000-2013 ranged from 2 to 67 ppm h. The AOT40 threshold of 5 ppm h set to protect forests from adverse ozone effects was exceeded on 75% of the plots from 80% of the participating countries.

Conclusions

ICP Forests ozone concentration data from in situ passive samplers reveal an overall decreasing trend of 0.5 ppb ozone per year over the period 2000 to 2013. This slight decrease matches the findings in EMEP (2014) where 6-months modeled maximum values decreased by 0.1 - 0.5 ppb/year for the April-September period in most of Europe during 2000 to 2012. A number of studies of tropospheric ozone trends have been published in the last years, as summarized in Tørseth et al. (2012) and Simpson et al. (2014). A fairly consistent picture has been found by Logan et al. (2012), Parrish et al. (2012) and Derwent

et al. (2013), with a flattening or even reduction in the ozone levels, most pronounced in summer. EEA (2014b) however, reports that measured ground-level ozone concentrations have reduced only marginally or have even increased due to long-range transport of pollutants from outside Europe. The differing outputs from various trend reports demonstrate the difficulty of modeling ozone concentration trends, which underlines the great value of long-term air pollution measurements at the very forest site, also in view of model validation. According to EEA (2013) up to 2009, approximately 250 sites and 10 countries had continuous ozone monitoring data for the past 11 years. The ICP Forests database for ozone concentration contains data from 214 forested sites and 20 countries. Data series from 81 sites, however, could not be considered for trend analyses as their data coverage was smaller than 6 years. It is therefore crucial to extend the data series of ozone concentrations on the already established sites.

Measurement of air pollutants in forests is important in order to evaluate the risk for vegetation and to document spatial patterns, temporal variability, and trends in areas not covered by conventional air quality monitoring networks. The presented results demonstrate that passive sampling represents a cost-effective and reliable method. Given the dense coverage of 214 forest monitoring sites from 20 countries where ozone measurement is carried out together with several other measurements on forest health, growth, nutrition, biodiversity and climate, the potential of the ICP Forests ozone data set is unique. Follow-up studies will focus on trend analyses based on more extended data series, and studies on the relationship between ozone, ozone-induced symptoms, tree health, and growth. Ozone data may be combined with extensive meteorological data series to be applied and tested for ozone flux modeling, in comparison with the respective EMEP outputs.

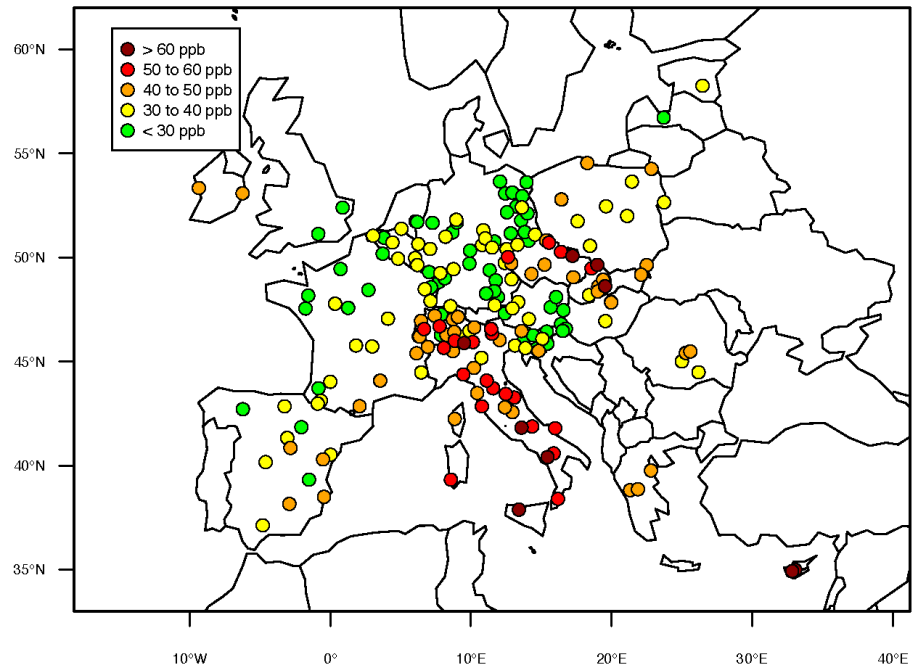

Figure 1. Mean ozone concentration classes from passive samplers on 214 plots during 2000-2013.

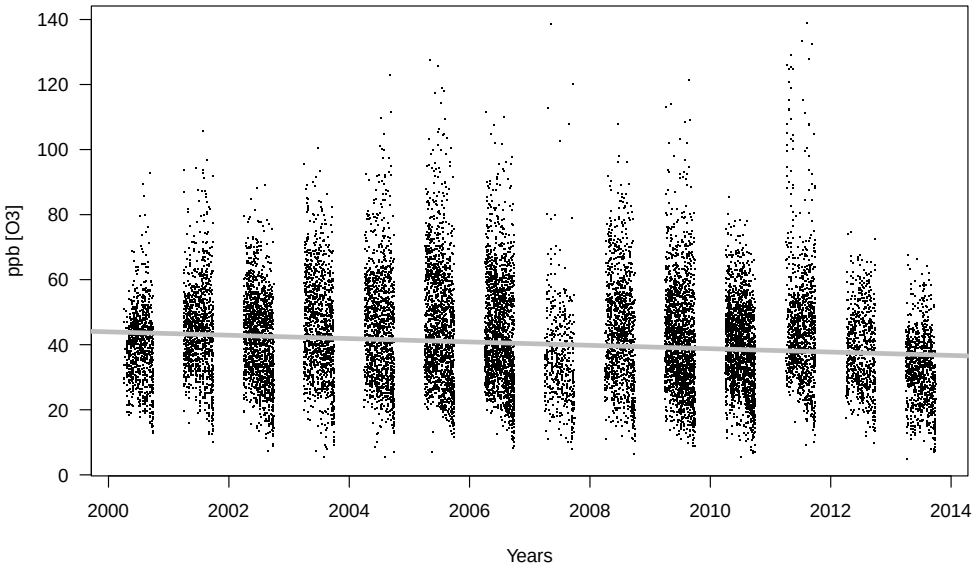

Figure 2. Scatter plot for ozone concentration from passive samplers exposed in 20 countries from 2000 until 2013 with a significant decrease of 0.5 ppb/year (n=18'362; p=0.000).

3.6.4. Health responses (SOMO35) (TF Health) (max 2 p)

Comment [HWI43]: Maire-Eve: Here we will include trends in annual mean, population exposure represented by SOMO35. We will also include a summary of latest information on evidence of health effects from ozone exposure.

There is evidence that short-term exposure to ozone is associated with morbidity (adverse effects on pulmonary function and lung permeability, lung inflammation, respiratory symptoms, and increased use of medication) and mortality. These effects appear to be independent of the effects of other air pollutants, such as particulate matter. Evidence on the effects of long-term exposure to ozone is accumulating; several cohort analyses have been published on long-term exposure and mortality (WHO Regional Office for Europe, 2013).

The indicator SOMO35, expressed as μg/m³ (or ppb) × days, can be used to quantify the cumulative yearly health impacts of ozone. At this time, there is no convincing evidence of a threshold for an effect on mortality at the population level from exposure to ozone; there is, however, substantial uncertainty about the magnitude of health effects from exposure to ozone at low concentrations (WHO Regional Office for Europe, 2013). Therefore, the quantification of possible effects of daily exposure to ozone on mortality is feasible only when ozone concentrations are sufficiently high and estimates are reliable – that is, above 70 μg/m³ (35 ppb). For this reason, the indicator SOMO35 is used here.

Error! Reference source not found. shows the average levels for SOMO35 (in μg/m³ × days) for the most recent year of data available (2012) for Member States of the WHO European Region. Mean SOMO35 values varied by country from 438 μg/m³ × days to 7474 μg/m³ × days. The data for O3 shows that in 2012 from 28 countries where data were available six presented values above 6000 mg/m3/days. Figure 6 shows the trend in SOMO35 levels between 200-2002 and 2010-2012. In general, the indicator values increased slightly during the period 2000–2012 in the WHO European Region Member States for which data were available. In most countries, there was a significant increase in the indicator values for the year 2003, most likely due to the unusually hot summer.

Ozone data, as part of regular monitoring, were available for 426 cities in 28 countries in 2012. The coverage of urban populations varied from 14.6% to 59%.

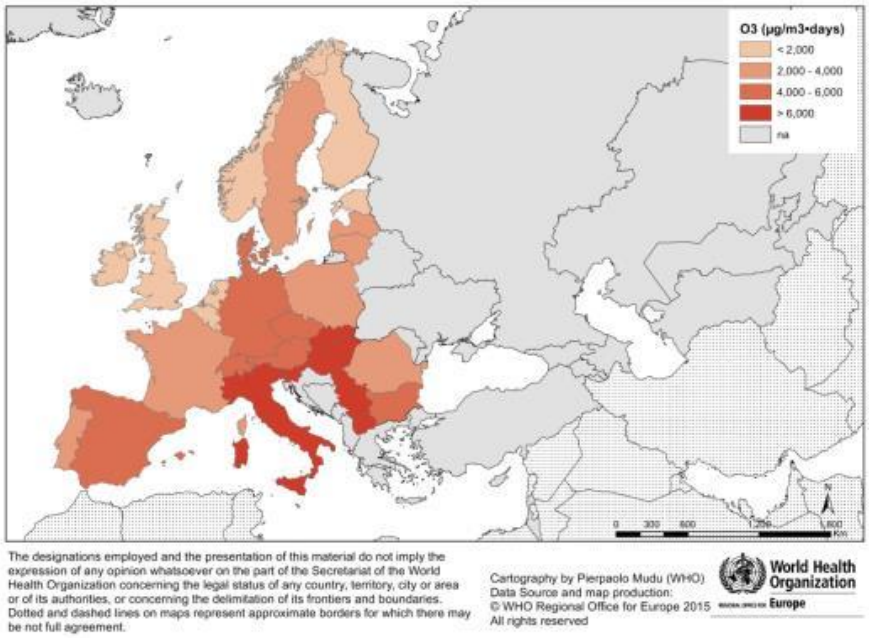

FIGURE 5. Population weighted annual mean SOMO35, 2012

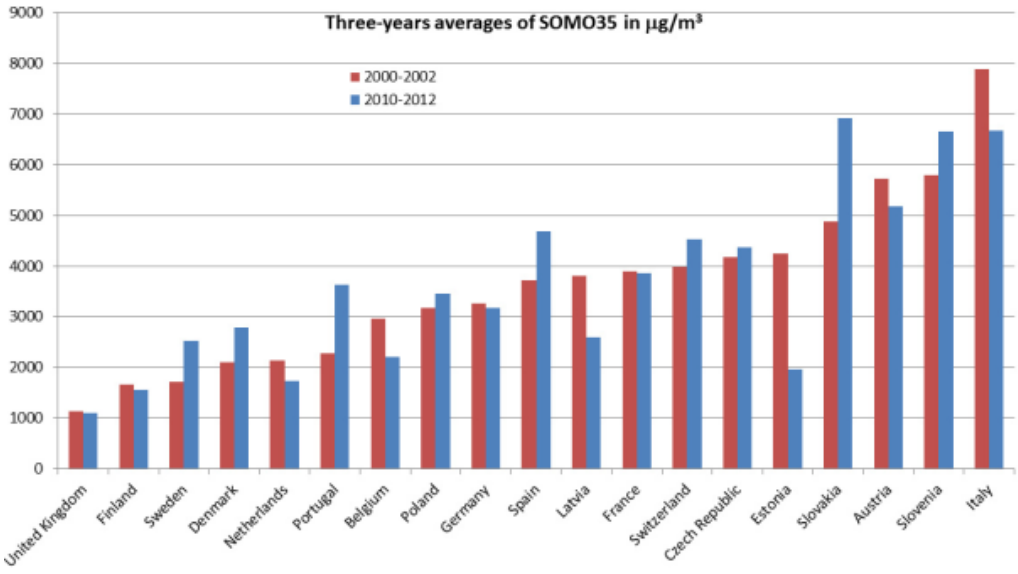

FIGURE 6. Population weighted annual mean SOMO35, three-year averages, for 2000-2002 and 2010-2012

4. Discussion/conclusions

-should address the future (time delay for recovery…, the role of climate…)

- include text on uncertainty and quality assurance in chapter 4. Consideration of uncertainty (e.g. robustness of exceedance calculations) and quality assurance (regarding measurement methods; refer to ICP manuals etc.)

-should focus on first 2 questions of assessment report – achievements, further improvements (where does the 'future' come in?), and should highlight remaining challenges /poor coverage of certain areas/responses

- Q1: what has been achieved
- Q2: further improvement in air quality and deposition that are possible? Some pollutants need to be regulated at hemispheric scale (e.g. ozone, mercury)
- Q3: co-benefits with climate change mitigation?

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6. Appendix A

The following expert contributed… X, Y,Z from ICP Waters, A, B, C from ICP Forests (crown condition), and D, E, F for foliar contents (ICP Forests) etc.