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

Summary/executive summary Max 4 pages.

This report documents trends in environmental and health responses to long-range transported air pollution, primarily focusing on from 1990 to 2012. Some longer-term trends and some prognoses given scenarios for future emissions and climate are also included. Air pollutants included in the report are sulphur and nitrogen as acidifying agents, nitrogen as a nutrient, ground-level ozone, particulate matter (PM), heavy metals and persistent organic pollutants (POPs). Most trends are reported for Europe, as data collection is concentrated there, in addition to some data from North America. So far, too limited data has been collected from EECCA (Eastern Europe, Caucasus and Central Asia) countries to provide trend assessments.

The documented trends stem primarily from work done under the bodies of the Working Group on Effects of the LRTAP Convention, i.e. ICP Integrated Monitoring, ICP Forests, ICP materials, ICP Modelling and Mapping, ICP Vegetation, ICP Waters, JEG Dynamic Modelling and the Task Force on Health. EMEP and AMAP also contributed to the report.

The main aim of the report is to document whether air pollution policies have been effective, by documenting obtained progress and lack there-of, gaps in the data and scientific understanding, and identify remaining challenges. It aims to provide support for policy assessment and development under the Long-range Transboundary Air Pollution (LRTAP) Convention.

Acidification

Extensive damage to ecosystems, related to acidifying effects of atmospheric deposition of sulphur and nitrogen, was observed in the 1960s and 1970s. Examples are forest die-back in parts of Central Europe, and large-scale extinction of fish populations in Scandinavia. The wide-spread concern for ecosystem damage related to acid deposition was a key driver for initiation of air pollution policy, monitoring of ecosystems and development of models to assess ecosystem damage related to air pollution.

Emission of the air pollutants sulphur and nitrogen, their long-range transport and deposition in Europe and eastern North America reached a maximum in the late 1970s, following decades of increase during industrialisation and urbanisation. In Europe, deposition of sulphur has decreased with 70% to 90% since 1980, while nitrogen deposition decreased with around 25% since 1990, in large part due to emission abatement measures adopted the LRTAP Convention. Trends in sulphur and nitrogen in throughfall in European forests are by and large consistent with the documented decreases of atmospheric deposition, although there is regional variation.

In Europe, the area exceeded by critical loads for acidification reached its maximum around 1970 for aquatic ecosystems, when 40% of all ecosystem area was exceeded. For terrestrial ecosystems, the maximum peak in area exceeded was 30% and occurred around 1980. Peak mean exceedance for terrestrial and aquatic ecosystems was 350 and 250 eq ha^{-1} yr^{-1}, respectively, but has now declined to below 10 eq ha^{-1} yr^{-1} for terrestrial and aquatic ecosystems. Currently, the area with exceeded critical loads for terrestrial and aquatic ecosystems in Europe is 9% and 5%, respectively, with an expectation for a further decrease towards 2020. However, exceedance of critical loads for acidification varies greatly within Europe.

In acid-sensitive lakes and streams in Europe and North America, sulphate concentrations have decreased on average 45-55% since 1988 as a result of a decrease in sulphate deposition. This has led to a widespread chemical recovery of surface waters, i.e. pH and acid-neutralizing capacity have increased. Biological recovery of acid-sensitive waters is also occurring, primarily as a result of improved water quality. However, aquatic systems that are under recovery from acidification still have lower species diversity than pristine aquatic ecosystems. Full biological recovery may not be possible in some

ecosystems. Inter-annual variations in water chemistry related to variations in acid deposition and climate is larger than the expected improvement in water chemical status in 2020. Thus, effects of climate variability and change are expected to offset and delay chemical and biological recovery of acid-sensitive waters.

Some catchments now release sulphur that has accumulated in the past, possibly delaying the recovery of the surface waters. The more efficient soil retention of nitrogen than sulphur results in generally higher leaching fluxes of sulphate than those of nitrate in European forested ecosystems. For most regions, sulphate remains the dominant source of soil acidification despite the greatly reduced deposition inputs of sulphur. Nitrogen contributes little to acidification of surface waters, because in many regions less than 10% of annual inputs leach from soils to streams. There is no sign of consistent and widespread regional increases in nitrate concentrations in surface waters in sensitive areas, despite continuing nitrogen accumulation in the catchments.

In European forests, defoliation records reveal a slight, but significant, deterioration of crown condition, except for Scots pine. Reduced sulphur deposition, reflected in a decline in foliar sulphur concentrations, is expected to lead to reduced soil acidification and improved conditions for trees. Therefore, it seems unlikely that the reduced crown condition is directly linked to reduced sulphur deposition in forests. High nitrogen deposition, however, has been shown to be negatively correlated with crown condition in three common forest species in Europe. Ozone, drought, insect gradations, and other biotic and abiotic factors also have potential adverse effects on needle and leaf biomass. Thus, the deterioration of forest crown condition in European forest may be related to interlinked climatic factors and deposition.

Corrosion of the indicator materials carbon steel, zinc and limestone has decreased substantially to around 50% of the reference values measured in the late 1980's, mainly as a result of reduced sulphur inputs. Since 2000, however, the improvements in corrosion and soiling are minor. For cultural heritage objects made of metals, positive responses to decreasing pollution levels are instantaneous. For limestone, however, a porous material, there is a substantial time lag of 20 years or more, before improvements can be seen.

While not much additional reduction in sulphur deposition may be expected, damage related to acidification remains an issue for surface waters, materials and buildings. Effects of climate change may set back effects of reduced sulphur deposition within the next decades.

Nitrogen as a nutrient

Nitrogen deposition in Europe has declined less than for sulphur, roughly 25% since the late 1970s. The total change in observed wet deposition of oxidised nitrogen for the 1990-2012 period is 22%. The comparable number for reduced nitrogen is 13%. However, nitrogen deposition remains high and leads to enrichment of soils, as generally more than 90% of all nitrogen deposition to forested catchments is retained. There is no evidence of widespread increases in leaching of nitrate to surface waters. For some tree species, foliar nitrogen concentrations in forests have decreased, possibly as a response to declining nitrogen deposition. A significant reduction of foliar phosphorous concentrations has been observed too, possibly a result of climate change, changed allocation patterns within trees, or dilution effects due to higher overall productivity caused by CO_2 and/or nitrogen fertilization effects.

Species cover of oligotrophic plant species (i.e. those thriving in low nitrogen conditions) has decreased in European forests where critical loads for nitrogen are exceeded, while species diversity has not changed. Thus, certain plant species in forests are negatively affected by nitrogen deposition, a demonstration of the eutrophication effect on nitrogen. The use of critical load exceedances is particularly suitable to

demonstrate the eutrophication signal of nitrogen deposition. The eutrophying impact of N deposition on plant species has been documented also in other ecosystems, such as grasslands and heathlands.

The area with exceedances of critical loads of nutrient nitrogen has declined from 72 to 60% between 1990 and 2010. Still, about 55% of the European terrestrial ecosystem area will not be protected from eutrophication in 2020, i.e. has exceeded its critical loads, given emissions reductions under the revised Gothenburg Protocol.

Ozone

Trends in ozone concentrations vary with site type (e.g. urban, rural, forest), time period and season studied, and calculated metric (e.g. annual or seasonal means, low or high percentiles, maximum or minimum values). Trends might well be masked by large inter-annual variations caused by for example climate and weather conditions, hence long time series (at least a few decades) are often required to identify ozone trends. Whereas peak concentrations of ground-level ozone have been reduced since 1990 in parts of Europe, a reduction of the average ozone concentration during the summer has not been observed at rural monitoring stations, despite a more than 30% reduction in ozone precursor emissions in Europe. This is because of less ozone titration by NO_x emissions close to source regions (e.g. urban areas), and the increasing precursor emissions in for example Asia and methane emissions over large parts of the Northern Hemisphere. More recently, since 2000 a small average decrease of 0.35 ppb ozone per year was measured at forested sites across Europe using passive samplers, associated with a decline in the accumulated ozone over a threshold of 40 ppb (AOT40) during the growing season at some (especially Italian) but not the majority of sites (no significant change at 80% of the sites). Between 1999 and 2010, no clear trends were found at monitoring sites regarding the risk of adverse impacts of ozone on crop yield, whether based on AOT40 or based on the biologically more relevant accumulated ozone flux (POD = Phytotoxic Ozone Dose).

Health effects from ozone exposure appear to be independent of the effects of other air pollutants, such as PMs. At country level, there is no clear trend in the risk of population exposure to ozone for the period of 2000–2012 in Europe. In general, the indicator of population exposure to ozone (indicator SOMO35, which stands for the sum of ozone means over 35 ppb (70 µg/m3)) has not changed substantially. In most countries, there was a significant increase in the indicator values for the year 2003, most likely due to the unusual hot summer, which resulted in longer ozone episodes with high peaks. Both human health and vegetation (including crops) remain currently at considerable risk of adverse impacts of ozone.

Ozone pollution in the future is critically dependent on changes in regional emissions and global transport of ozone precursors. Further ozone pollution abatement requires measures at the global scale to reduce emissions of ozone precursors, including methane. With full implementation of the revised Gothenburg Protocol, wheat yield loss is predicted to decline from 10.7% in 2005 to 8.8% in 2020 and 8.2% in 2030 in Europe. Hence, sensitive crops and natural ecosystems will remain at risk of significant 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 increases is becoming more important when emissions of other ozone precursors are controlled.

Particulate matter (PM)

Extensive monitoring of indicators of population exposure to PM (indicators PM_{10} and $PM_{2.5}$: particulate matter with an aerodynamic diameter smaller than 10 µm and 2.5 µm, respectively) started only after 2000. Existing evidence on the adverse effects on health of ambient air pollutants has been complemented through new studies and knowledge and hence has been strengthened further in the last years. Monitoring of aerosol mass in background areas since 2002 shows a clear decrease in average concentrations per year, of PM_{10} and $PM_{2.5}$, of circa 2.5% for PM_{10} and circa 3% for $PM_{2.5}$.

By contrast, indicators of population exposure to PM (indicators PM_{10} and $PM_{2.5}$) in populated areas in the Europe have not changed substantially since monitoring has been implemented. An overall decrease of $PM_{2.5}$ and PM_{10} emissions and PM_{10} concentrations across Europe has been reported during that same period, and that trend may partially be attributable to the emission reductions initiated under the Convention. However, in European cities where PM is monitored, 75.4% and 94.0% of people experience annual levels exceeding the WHO Air Quality Guidelines (AQG) for PM10 and PM2.5, respectively. In addition, compliance with the EU limit values remains an issue across Europe, where 21 % of the EU-28 urban population is living in areas where the EU daily limit value for PM10 concentrations was exceeded in 2012. However, in countries in the eastern part of the Europe, more monitoring is required to properly quantify exposure and the impacts to health from air pollution and document trends.

With regard to soiling of glass, which is most sensitive to particulate pollutants in addition to gaseous pollutants, improvements are expected to be limited if the PM_{10} concentration remain at the current level. Glass soiling data are only available since 2000 and show no increasing or decreasing trends.

Heavy metals

Reductions in total deposition of lead, cadmium and mercury between 1990 and 2012 in Europe were 78%, 53% and 23%, respectively. For lead and cadmium, these trends are primarily driven by reductions of anthropogenic emissions of lead and cadmium in Europe. For mercury, secondary emission sources and emission sources outside of Europe lead to a lower decline in deposition than in anthropogenic emissions. Hemispheric transport of mercury across the globe results in a considerable contribution of mercury pollution from other continents to mercury deposition in Europe.

Mosses are bio-indicators for atmospheric deposition of heavy metals to ecosystems. Between 1990 and 2010, the metal concentration in mosses declined 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%). For lead and cadmium, the decline is similar to those reported by EMEP for the modelled deposition across Europe. The 14% decline in Hg between 1995 and 2010 was lower than the decline in EMEP modelled deposition across Europe.

Lead and cadmium in upper soil layers in remote sites in Sweden showed decreases between 1994 and 2011, consistent with the trends observed in mosses and deposition. However, lead and cadmium are being transferred to deeper soil layers as documented by increasing concentrations in those layers, indicating that accumulation is ongoing in the total soil profile. Contrary to the mercury trend in mosses, concentrations of mercury increased in the forest floor and in deeper soil layers. In stream waters, hardly any trends in cadmium, lead and mercury were found in the last two decades. Heavy metals continue to leach from soils to surface waters, with no clear relation to trends in deposition.

Mercury concentrations in freshwater fish show a mixture of trends, from downward in Sweden since 1965, and rising since the 1990s in Sweden, Norway and Canada although not for all fish species and not for all investigated regions. Mercury in many fish species remains above limits advised for human consumption in Scandinavian countries.

Exceedances of critical loads of cadmium and lead have declined since 1990. For lead, the area at risk in Europe declined from 67% to 20% between 1990 and 2010, and also the magnitude of the exceedance of critical loads dropped drastically. Still, exceedances for lead occurred in all countries in Europe in 2010. For cadmium, atmospheric deposition excluding other (agricultural) inputs, is computed not to exceed critical loads of cadmium on a broad European scale in 2010. For mercury, high deposition remains an issue of particular concern, as between 1990 and 2010, the ecosystem area at risk of exceedance of mercury critical loads declined only from 69% to 56%.

Thus, except for mercury, air pollution policy for heavy metals has been effective in reducing emissions, deposition and accumulation rates in the environment. However, heavy metal accumulation in soils continues, with possible consequences for export to aquatic ecosystems. There is still a long-term risk of harmful effects to human health and ecosystems by the atmospheric deposition of lead and mercury in certain areas in Europe.

POPs

Trend analysis of air concentrations and deposition fluxes from 1990 to 2012 for POPs included in the EMEP monitoring and modelling programs (B[a]P, PCDD/Fs, PCB-153 and HCB), using a on combination of modeling and monitoring data, showed noticeable decreases of atmospheric pollution within the EMEP region. The largest decrease of air concentrations was estimated for PCBs and HCB (80 – 90%), while lowest decrease was obtained for PAHs (about 30%). It was found that largest reduction of atmospheric POP pollution took place in early 1990's, while after 2005 the reduction of atmospheric POP pollution diminished or even switched to a small increase (for B[a]P). The analysis showed strong seasonal variation of POP pollution levels (the difference between summer and winter concentrations can be as much as an order of magnitude). Model assessment indicates that, in spite of generally low annual mean B[a]P air concentrations over the major part of the EMEP countries, there are areas where EU target value for B[a]P, equal to 1 ng/m^3 (Directive 2008/50 EC), is exceeded.

Little information has been assembled on levels and trends of POPs, originating from atmospheric transport, in environmental media and on effects. Some declining trends of selected POPs in remote surface waters document a decline in PCBs in fish in boreal Sweden, boreal Canada and Arctic regions. A pilot study on POPs in mosses was conducted, but trends can only be assessed in the future provided this type of investigation proofs to provide useful information.

POPs levels in air, biota and humans in the Arctic show generally decreasing trends in air and biota for most POPs covered by the Conventions. Levels of POPs in the blood of Arctic residents have also generally declined over the past 20 to 30 years. This includes significant falls in levels of DDT and polychlorinated biphenyls (PCBs). However, levels of some POPs in human blood still remain higher in some Arctic populations than in most general populations in North America and Europe. Trends in levels of POPs that have emerged in recent years as chemicals of concern in the Arctic, including some regulated brominated flame retardants show a more mixed pattern, with some time-series still showing increasing trends, whereas in others levels appear to have peaked in the early 2000s.

For the Arctic, controls on pollutants have proven effective; however, there is a need for more timely controls on chemicals of emerging concern. Recognizing that long-range transport remains the most significant source of Arctic contamination, it is recommended that [Arctic] states continue to show leadership on international pollution control, and where necessary ratify existing Conventions as soon as possible. A need for consideration of additional unilateral, regional and global actions to control pollutants of emerging concern is recognized.

Achievements and remaining challenges from the conclusions at the end.

Comment [HW1]: Here inputs from Peringe and Rob are necessary. Suggestion from Harry: From Harry: *You could also look at long-term strategy of the Convention to find confirmation for text below (see comments made by Richard Ballaman regarding Assessment Report).*

Summary for policy makers

-to be considered

Preface

After signing the Convention on Long-range Transboundary Air Pollution in 1979 and the establishment of the Working Group on Effects soon thereafter, co-ordinated long term monitoring of environmental effects became an important issue for the Convention. Monitoring programmes were established throughout the 1980s organised in the form of so called International Co-operative Programmes (ICPs). For 25 years and more five ICPs have been running on Waters, Forest, Material, Integrated Monitoring and Vegetation each with a programme chair and coordination centre. In addition, a special Task Force (later ICP Modelling and Mapping) was established in connection with the introduction and implementation of the critical loads concept. This ICP has together with its centre - Coordination Centre for Effects – over the years formed a central resource for coordination of several effects activities. Finally a Task Force on Health was established in 1997 as a joint effort with WHO in order to meet the needs to of assessing effects to human health.

The ICP programmes have, during the 25-30 years they have been running, served as important sources of information regarding observations, trends and assessment of effects but also as a basis for research. In particular the ICPs have contributed to understanding processes and causes of the various effects and their relations to other stresses as well estimating future development in relation to control scenarios.

One important task for Working Group on Effects is to investigate the outcome of the control measures undertaken within the UN ECE region. This trends report puts together as a common activity between the Working Group on Effects, with gratefully acknowledged support from centres under EMEP (European Monitoring and Evaluation Programme), a comprehensive picture of the outcomes of the measures undertaken since 1990 as well as in some cases also expected future development. It will also serve as an input to the broader Assessment Report that is under preparation for the Convention.

In this connection, I want to point at two particularly important activities without which the trends analyses and this report would not have been produced. First, I want to mention the long-term commitment by Parties to take part in the programmes, and support monitoring activities that deliver the data upon which this report is based, including issues regarding quality, harmonisation and reporting. This participation has given an increased and common understanding of the problems related to long-range transported air pollution, and their solutions. Second, I want to highlight the work by the ICP centres and the support to these centres that is provided by their host countries. The subsidiary programmes to the WGE are only financed to a very limited extent by central LRTAP Convention funds, and this support is voluntary.

Even if we see improvements in terms of decreased emissions and atmospheric concentrations, there is still a need to monitor air pollution effects and through directed research activities estimate future effects. The recovery of damaged systems will most often take long time and sometimes also need mitigation. I am therefore seeing a continuous support to the monitoring programmes as a key driver for further actions.

Over the 30 years period that the ICPs have been running, a tremendous amount of data has been produced. This report has only to a limited extent been able to make use of this information. Since the data in many ways are unique, it is important to both make sure that the data are safely and systematically stored but also that the data are made available to larger extent than so far, for scientific research and evaluation to communities outside the WGE community. This is joint challenge for the WGE and for the entire Convention.

Peringe Grennfelt

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At the 33rd session of the Working Group on Effects, in 2014, it was proposed that the Working Group and its Programmes prepare a Report on trends in the effects of air pollutants. This report was produced by the Programme Centres of the international programmes of the Working Group and members of the Bureau of the Working Group on Effects. It was prepared at the request of the Working Group on Effects and the Executive Body of the Convention and is submitted to 34th Session of the Working Group on Effects in September 2015, 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.

-list of contributors, acknowledgements

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List of contributors

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The following have contributed to the report by reviewing specific chapters (Sabine Augustin, Isaura, Gunnar Skotte, Martin Forsius, Gudrun Schütze) and the entire report (Richard F. Wright, Rob Maas)

Comment [HWI2]: To be updated.

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AMAP The Arctic Monitoring and Assessment Programme (AMAP) contribution was prepared by Simon Wilson[1] based on information compiled by members of the AMAP expert groups on persistent organic pollutants, mercury and human health and their assessments of data gathered under the AMAP pan-Arctic trends and effects monitoring programme. The contribution of all scientists and experts who provide data and support AMAP assessment activities is gratefully acknowledged, as are the national monitoring programs of the eight Arctic countries that form the basis for the AMAP.

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1. Introduction

1.1. Discovering detrimental effects of transboundary air pollution

During the 1970s and 1980s, alarming observations were made of forest die-back as a consequence of air pollution in Central Europe (Schutt & Cowling 1985; Ulrich 1990). In Scandinavia and North America, fish populations disappeared during the 20th century from lakes and rivers because acidification created toxic water chemical conditions (Beamish 1976; Hesthagen et al. 1999). High concentrations of heavy metals (Holy et al. 2009) and Persistent Organic Pollutants (POPs) (Wania & Mackay 1996) were measured in remote ecosystems. Heavy metals and POPs have been shown to have detrimental effects on ecology and human health (Brown et al. 1984; Jorgenson 2001). Damage of air pollution on building materials (Tidblad et al. 1991) and cultural heritage (Van Grieken et al. 1998) were observed.

The common cause for the ecosystem and health damages was long-range transported air pollutants, which had impacts far from their emission sources.

1.2. A successful integration of science and policy to combat effects of air pollution

Pushed into the public sphere, the scientific alarms about health and ecosystem damage related to air pollution lead to the initiation of the Convention on Long–range Transboundary Air Pollution (LRTAP Convention) in 1979. Under this Convention, its Parties developed regulations to curb atmospheric pollution in two regions: Europe and North America. These regulations have taken the form of eight protocols with three revisions (ECE 2004a). The development of these policies has been based on scientific knowledge, gathered and compiled within the two scientific pillars of the Convention, the Cooperative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe (EMEP - – European Monitoring and Evaluation Programme) and The Working Group on Effects (WGE). EMEP focuses on the emission, chemical transformations, dispersion and deposition of atmospheric pollutants on a European scale. The WGE focuses on the monitoring and modelling of effects of air pollutants and deposition on terrestrial and aquatic ecosystems and the effects of direct exposure to human health, cultural heritage, building materials and vegetation. Components include acidifying pollutants (sulphur and nitrogen compounds), eutrophying pollutants (nutrient nitrogen compounds), ozone, particulate matter (PM), heavy metals and persistent organic pollutants (POPs).

Established in 1980, the WGE offers an arena where policymakers meet scientists and exchange information on the causes and consequences of atmospheric pollution. Seven technical groups were established under the WGE between 1985 and 1997 to cover the whole range of air pollution impacts on the environment and human health. These International Cooperative Programmes (ICPs) are ICP Waters, ICP Materials, ICP Forests, ICP Vegetation, ICP Modelling and Mapping, ICP Integrated Monitoring and the Task Force on Health. In addition the Joint Expert Group on Dynamic Modelling (JEGDM) was formed as a forum to discuss specific issues related to dynamic modelling.

In 2004 the Convention celebrated its 25th anniversary, at the occasion of which a booklet was published presenting an accessible overview its work (ECE 2004b). All Convention reports and publications are available at the website …..

1.3. The work under the LTRAP Convention today

Today, the ICPs have conducted more than twenty years of monitoring, soil and water biochemical process analysis and modelling. Acidification and of eutrophication of terrestrial and aquatic ecosystems is well documented, in part through results of the monitoring and modelling networks of the ICPs, which cover most parts of Europe and, in some cases, North America.

Thresholds for depositions (critical loads) and concentrations were developed to support European policies with knowledge on required reductions of emissions, and hence deposition and ambient

concentrations, to protect the environment. An important milestone of this so-called "effect-oriented policy support" was achieved through the establishment of the second sulphur protocol in 1994 (ECE, 2004a). This protocol focused on the protection of terrestrial and aquatic ecosystems from acidification. Thresholds have been developed since to protect human health (ozone, fine particular matter, and heavy metals), vegetation (ozone) and terrestrial ecosystems (nitrogen).

An important characteristic of the work of the WGE is the analysis and documentation of trends in effects of sensitive indicators to air pollution for ecosystems, materials and public health. Trends are compiled through long-term systematic monitoring of ecosystem and health indicators carried out year after year, following monitoring and modelling protocols and guidelines agreed between scientists from participating countries across Europe and North America. Long-term time series of ecosystem and environmental changes are indispensable for scientific understanding of response to changes in deposition and climate. For policymakers, trends in sensitive indictors of air pollution provide valuable information as to whether policies to reduce emissions to the atmosphere have had the intended effects.

1.4. Objectives and organisation of this report

This report comprises trend analyses by monitoring and modelling groups under the WGE for roughly the past twenty years, presenting developments in forests, vegetation, catchments, lakes and rivers, building materials and cultural heritage in relation to air pollution. Trends in indicators for the risk of air pollutants on human health are also included. Trends in air pollution loads and concentrations of pollutants are presented by EMEP.

The trends demonstrate damage, recovery and lack of change in ecosystem conditions and health, illustrating whether emission policies have had their intended effect or whether more work remains necessary. Also, the contributions highlight the presence of factors other than air pollution with potential relevance for health and ecosystem responses. This work takes into account 20+ years of progress in monitoring and modelling atmospheric pollution effects and is the result of long-standing scientific collaboration across Europe and North America. It is a demonstration of how the WGE contributes to understanding and assessing the implementation of the air pollution regulation under the LRTAP Convention.

This report has been structured according to pollutant, in the subsequent order that the different protocols were developed, i.e. acidification (sulphur and nitrogen) (the 1985 Helsinki Protocol the 1994 Oslo Protocol and the 1988 Sofia Protocol), nitrogen as a nutrient, ozone (the 1991 Geneva Protocol on VOCs), heavy metals (the 1998Aarhus Protocol), POPs (the 1998 Aarhus Protocol). In 1999, the first multi-pollutant protocol was developed on acidification, eutrophication and ground-level ozone (the 1999 Gothenburg Protocol) and its revised version (2012) now also includes PM.

The bodies under the WGE have contributed with results on trends in concentrations and effects under each particular pollutant, in addition to contributions from EMEP/TFMM (Task Force on Measurement and Modelling) on deposition and/or atmospheric pollutants. AMAP (Arctic Monitoring and Assessment Programme) contributed to the chapter on POPs. AMAP is the group of the Arctic Council responsible for monitoring and assessment of pollution- and climate change-related issues in the circum-Arctic region. AMAP has had a long-term collaboration with the LRTAP bodies on effects monitoring and modelling.

This report has as its primary aim to document trends in environmental (ecosystems and build-up environment) and health responses from 1990 onwards, from the work done under the bodies of the WGE, thereby also focusing on obtained progress and lack there-of, gaps in the data and scientific understanding, and identify remaining challenges.

2. Materials and Methods

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

The Convention on Long-range Transboundary Air Pollution was the first international legally binding instrument to deal with problems of air pollution on a broad regional basis. One of the eight protocols under the Convention was the 1984 Geneva Protocol on Long-term Financing of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). This Protocol secures the funding of the EMEP centers, and through this a harmonized monitoring network has been developed by the EMEP Chemical Coordinating Centre. Parties to the Convention are required to provide EMEP with information regarding emissions as well as observations related to atmospheric transport of air pollutants, which is the basis for the EMEP monitoring network. To monitor the regional scale concentrations and deposition of air pollutants, a network of sites was established in the 1970ies (Figure 1). The sites are located to avoid influence by local emission sources and the methods employed are tailored to provide comparable results between the sites. The data are used to monitor spatial and temporal trends, and are used in developing atmospheric chemistry-transport models which are used to derive source-receptor relationships. For a comprehensive description of the EMEP observations, we refer to Tørseth et al, 2012.

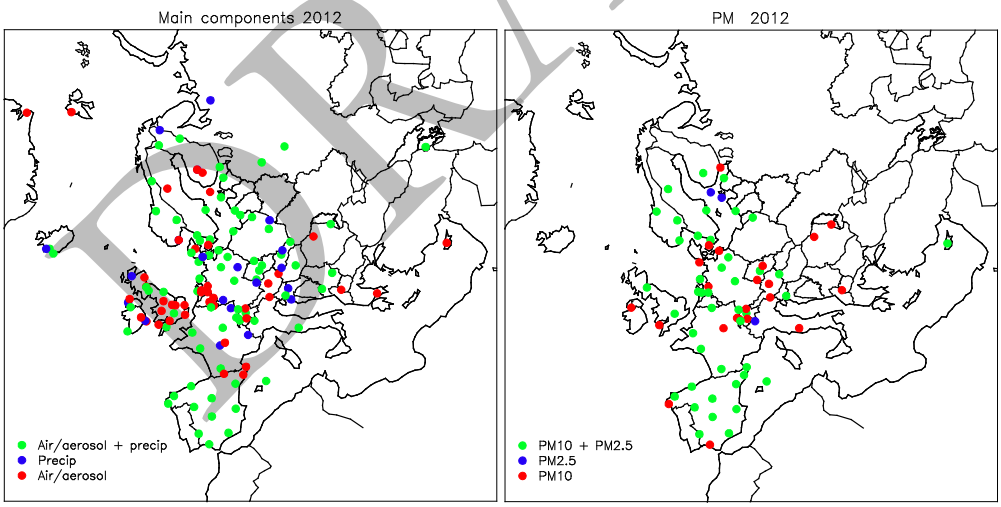

Figure 1: EMEP sites in operation in year 2012, for main components, precipitation and particulate matter.

2.2. ICP Forests

The 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 (level I; Figure 2) under the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests). While the large scale Level I monitoring aim at the detection of large scale patterns and trends of tree vitality, the need emerged for more intensive monitoring on selected sites, aiming at the study of the underlying processes, and develop empirical models on cause-effect relationships. Therefore, the intensive ecosystem-oriented monitoring (Level II; Figure 3) was initiated in 1994. Both monitoring networks were at least temporarily installed on in 42 UNECE countries and have produced an extensive, highly valuable from in total 13 surveys (Figure 2), all collected according to a harmonized manual (ICP Forests 2010).

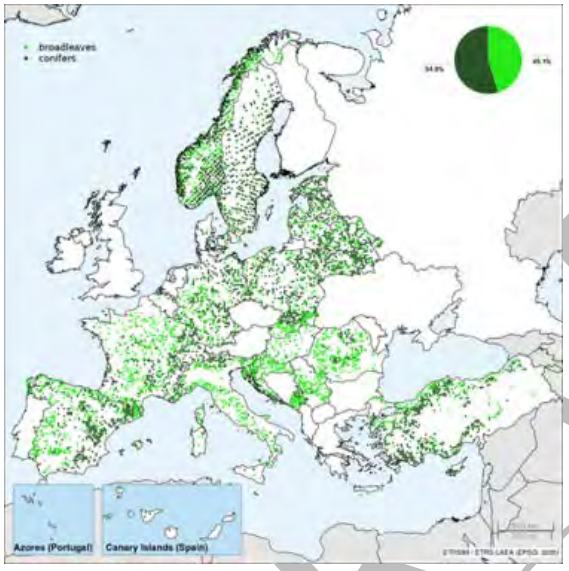

Figure 2: Plots of the Level I monitoring in 2013

Figure 3: Plots of the Level II monitoring in 2009

Observing air pollution impacts on forests requires monitoring of fluxes into and through forests, and recording of soil and vegetation responses. With regard to fluxes, on-site measurements of air quality and deposition of key pollutants such as nitrogen and sulphur are performed in the open-field and beneath the canopies. Fate and interactions of these substances in forest soils are described by analysing soil solution sampled at various depths and calculating fluxes. Nutrient contents of tree foliage, which reflect atmospheric and soil-related influences, are investigated. Response variables like crown condition or radial stem growth are influenced by a variety of environmental and biotic stressors. Other ecosystem responses studied are diversity and composition of ground vegetation and epiphytic lichens.

Table 1: Surveys performed on intensive and extensive ([1]) or only extensive ([2]) ICP Forests monitoring plots and their assessment frequencies

Survey	Frequency	Survey	Frequency	Survey	Frequency
Crown condition[1]	Annual	**Deposition**[2]	Continuous	**Litterfall**[2]	Continuous
Foliar chemistry[1]	Bi-annual	**Soil condition**[1]	Every 10th year	**Leaf area index**[2]	Occasionally (annual)
Tree growth[2]	Every 5th year	**Soil solution chemistry**[2]	Continuous	**Phenology**[2]	Several times per year
Ground vegetation[2]	Every 5th year	**Meteorology**	Continuous	**Epiphytic lichens**[2]	Once, pilot project
Ozone-induced injury[2]	Annual	**Ambient air quality**[2]	Continuous		

2.3. ICP Integrated Monitoring

The multi-disciplinary Integrated Monitoring programme (ICP IM) has as overall aim to determine and predict the state and change of terrestrial and freshwater ecosystems in a long-term perspective with respect to the impact of air pollutants, especially nitrogen and sulphur but also extending to heavy metals.

The ICP IM sites are located in undisturbed areas, mainly in natural parks or comparable protected areas. The focus of the programme is on detecting and modelling of long-term ecosystem impacts of air pollutants and climate change, and their interactions. Most of the sites are forested catchments, where data of both terrestrial and surface water ecosystem compartments is collected. Integrated monitoring thus refers to the simultaneous measurement of physical, chemical and biological properties of an ecosystem over time and across compartments at the same location. Since the sites are not influenced by local site management, high-quality time-series can be obtained and complex interactions between the impacts and ecosystem compartments can be investigated. 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 on the comprehensive monitoring programme are provided in the ICP IM manual (www.syke.fi/nature/icpim). Presently there are 46 ICP IM sites from 16 countries with on-going data submission (Figure 4). For the trend assessments presented in this report, the sites have been selected based on data availability.

Figure 4: Location of the ICP IM sites

2.4. ICP Materials

The main aims of ICP Materials are evaluation of long term trends in corrosion and soiling (deposition of pollutants on surfaces), 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 material test racks located in rural, urban or industrial sites (Figure 5). 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. Exposures 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.

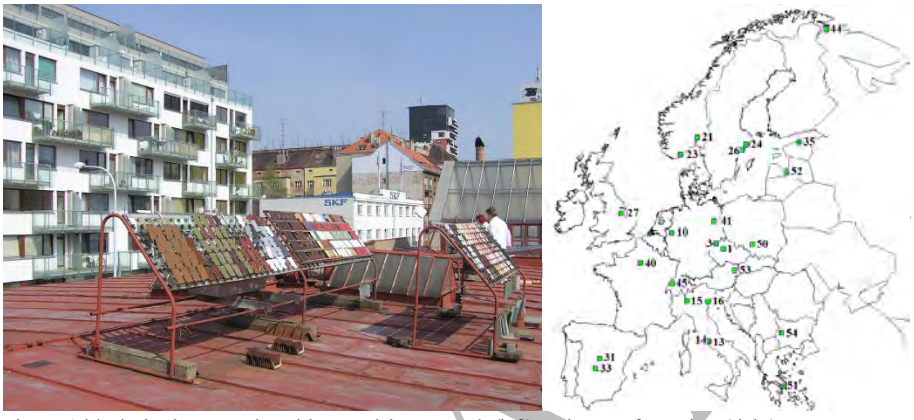

Figure 5: Typical urban test site with materials on a rack (left) and map of test sites (right)

Environmental data are also collected at each site as indicated in (Table 2). Both the test sites and the reported parameters have varied with the evolution of the programme. In the beginning, focus was on acidifying pollutants, especially SO_2, which is still one of the most important parameters 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 2: Reported environmental data from ICP Materials tests 1987-2012: M = mandatory; O = optional; - = not reported

Parameter	Symbol	Unit	1987-1995	1995-2001	2002-2003	2005-2012
Temperature	T	^{o}C	M	M	M	M
Relative humidity	Rh	%	M	M	M	M
Time of wetness	Tow	h	M	-	-	-
Sunshine	Sun	h	M	-	-	-
Sunshine[a]	Sun	MJ m^{-2}	M	M	M	-
SO_2 concentration	SO_2	µg m^{-3}	M	M	M	M
NO_2 concentration	NO_2	µg m^{-3}	M	M	M	M
O_3 concentration	O_3	µg m^{-3}	O	O	M	M
HNO_3 concentration	HNO_3	µg m^{-3}	-	O[b]	O	M
Precipitation: amount	Prec	mm	M	M	M	M
-: conductivity	Cond	µS cm^{-1}	M	M	M	O
-: pH	pH	-	M	M	M	M
-: SO_4^{2-}, NO_3^-, Cl^-	varies	mg l^{-1}	M	M	M	M
-: NH_4^+, Na^+, Ca^{2+}, Mg^{2+}, K^+	varies	mg l^{-1}	O	O	O	O
Particulate matter	PM	µg cm^{-2} $month^{-1}$	-	O[b]	O	M

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

2.5. ICP Modelling and Mapping

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 the development of modelling and mapping methodologies for the assessment of current and future air pollution related effects in Europe. Results from its Coordination Centre for Effects (CCE) are incorporated in the GAINS model of EMEP-CIAM to analyse in an integrated manner costs and benefits of abatement policies of both the LRTAP Convention and the European Commission.

Trends of the differences between atmospheric depositions and critical loads (i.e. critical load exceedances) are based on the state of knowledge in 2013. Modelled deposition data are from EMEP MSC-W[1] and critical loads from the CCE. The EMEP model (Simpson et al. 2012) produces acidifying and eutrophying, i.e. sulphur (S) and nitrogen (N), 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. 2015a).

Critical loads are aimed to protect an endpoint which, following the definition of critical loads, could be broadly indicated as "…specified sensitive elements of the environment…". In terrestrial ecosystems this could include "tree growth" or "species diversity", while in aquatic ecosystem the endpoint could be "fish population". Human health can be an endpoint that is affected through changes of the quality of terrestrial or aquatic ecosystems. Finally, in critical loads modelling, "critical limits" (see De Vries *et al.*, 2015) are needed with values identified to protect an endpoint. Critical limits are based on recommendations made in the Mapping Manual, and specific limit values can vary between countries that submit data. The ratio of base cations to aluminium in the soil solution and Acid Neutralizing Capacity are generally chosen to protect terrestrial and aquatic ecosystems respectively, against acidification. For eutrophication values for acceptable nitrogen concentrations in the soil solution are used. For heavy metals both human and ecosystem health health are used as endpoint (De Vries *et al.* 2015; Hettelingh, 2015a; Slootweg *et al.* 2007; Hettelingh *et al.* 2006; Slootweg *et al.* 2005). Health effects can be addressed through (a) terrestrial ecosystems via drinking water (Cd, Pb, Hg), (b) idem via food (Cd) or (c) in aquatic ecosystems via fish (Hg). Critical loads of heavy metals for protecting ecosystems are based on varying limits for Cd, Pb in both aquatic and terrestrial ecosystems, also in including limits for Hg in the latter.

The European critical load database, held at the CCE and incorporated in the GAINS model at the International Institute for Applied Systems Analysis, enabled the calculation of exceedances for (terrestrial and aquatic) ecosystems at risk caused by depositions from emissions that are regulated 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.

[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.

Historic emissions since 1880, used by the ICP M&M 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 section 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 (e.g., the EUNIS classification; Davies and Moss 1999).

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2.6. ICP Vegetation

ICP Vegetation (International Cooperative Programme on the Effects of Air Pollution on Natural Vegetation and Crops) focusses on monitoring and modelling the impacts of ozone on (semi-)natural vegetation and crops, and the development of ozone critical levels for vegetation (the latter also for trees in collaboration with ICP Forests). Modifying influences of other pollutants such as nitrogen and modifying influences of climate change on the impacts of ozone are also studied. In addition, ICP Vegetation monitors the concentration of heavy metals, nitrogen and to a limited extent POPs in mosses as an indication of atmospheric deposition of these compounds to vegetation.

Monitoring sites

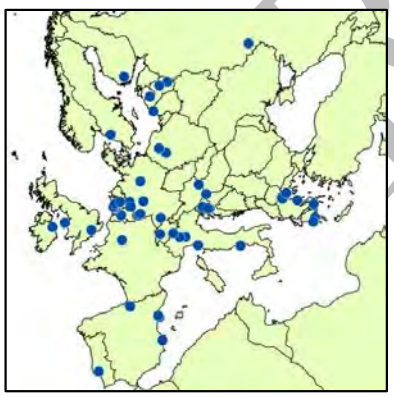

Figure 6: Impacts on white clover between 1996 and 2006 (Hayes et al., 2007)

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 1; Hayes et al., 2007; Mills et al., 2011), brown knapweed and French bean. The participation in those experiments varied by year and has declined in recent years. Recently, the ICP Vegetation has developed a smart-phone App for recording incidences of ozone-induced injury on vegetation (http://icpvegetation.ceh.ac.uk/record/index).

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) and in 2010 a pilot study was included for **POPs** (Harmens et al., 2013).

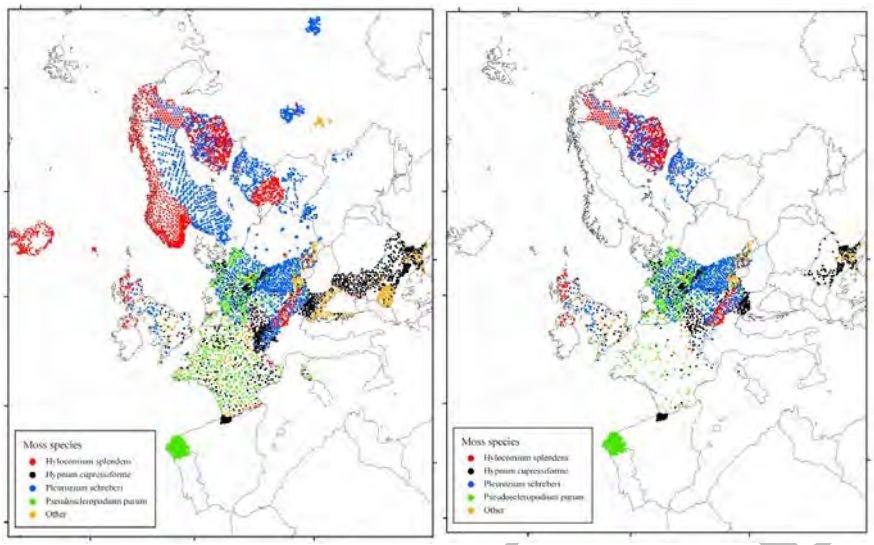

Figure 7: Sampling sites for determination of heavy metal (left) and nitrogen (right) concentrations in mosses in 2005 (Harmens et al., 2010, 2011).

2.7. 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 8: Monitoring stations in the ICP Waters programme. The stations are grouped according to region.

The chemistry 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 (zoobenthos: community of bottom-dwelling species such as insects and snails) 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.

2.8. JEG DM

The Joint Expert Group on Dynamic Modelling (JEG DM) was set up in 1999 with the aim to provide a discussion forum for experts on modelling under the Convention, and whose interest is to use dynamic models to predict air pollution effects on ecosystems and materials. In addition, JEG aims to summarise and develop knowledge based on dynamic modelling for the benefit of WGE.

An important issue in JEG DM is to introduce time dimension into modelling critical loads calculations. Critical loads do not consider temporal aspects of critical load exceedances and expected damage or recovery of ecosystems and materials, once the critical loads no longer are exceeded. In contrast, the dynamic modelling contains per definition time aspect and provides information on when any specific effects, such as lake acidification, changes in plant community and other eutrophication-related problems, will occur. Thus, JEG DM contributes to extending the concept of critical loads by including information about the dynamics of the future air pollution effects in the work of WGE.

The JEG DM meets annually which typically attract 20 – 30 modelling experts from several ICPs and other Convention bodies. The meetings are regularly reported to WGE and the agenda is set according to WGE work plan. Two other principal functions of JEG DM are to analyse outcomes of the Calls for data and directions for future Calls; and to keep the work of WGE connected with dynamic modelling carried out outside the work of Convention, most importantly within various EU projects. The JEG DM has been dependent on voluntary contribution from DEFRA in UK and from Swedish EPA. Currently the JEG DM is chaired by Sweden.

2.9. Task Force on Health

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.

PM and ozone are the key indicators that are widely measured for health effects related to air pollution. Industries, agriculture, households, and traffic produce complex mixtures of air pollutants, many of which pose risks to health. Of all of these pollutants, PM10 and fine PM (PM2.5) are associated with the greatest long-term and short-term effects on human health. PM2.5 shows the most consistent associations with mortality and morbidity, such as cardiovascular and respiratory disease, acute and chronic bronchitis, lung cancer and restrictions in daily activity. Ozone produces short-term effects, independent of other air pollutants such as PM, on mortality and respiratory morbidity. In addition, evidence for the long-term health effects of ozone is accumulating (WHO Regional Office for Europe, 2013). Measurements of PM and ozone in ambient air are serving as an acceptable "proxy" for population exposure. Nonetheless, other air pollutants such SO2, NO2 are of interest in identifying trends as well, due to the growing evidence for the independent direct health effect of NO2 and SO2. These air pollutants are also PM precursors and contribute to PM concentrations (and related population exposure).

For PM in ambient air, population exposure is reflected by the indicator reported by combining data on PM10 or PM2.5 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 using data from urban background locations. For the calculation of the average annual PM concentrations, consistent monitoring data collected throughout the year, for several years, are desirable to reduce the bias arising due to seasonal fluctuations or due to a year with pollutant concentrations being very different from the average concentrations observed in other years, i.e. a non-representative year. PM data is generally available for larger cities due to a good spatial coverage of air monitoring networks. However, in many agglomerations and in rural areas, the monitoring network is either less dense or only in a planning stage (Brauer et al., 2012; WHO, 2011). The monitors used to assess population exposure should ideally be located such that they are not unduly influenced by a single source of pollution (i.e. a power plant, factory or highway), but that they rather reflect exposures over a wide area (WHO, 2011).

National estimates, which are calculated as means of annual average concentrations of PM2.5 or PM10 in cities, are included in the calculations. Population data are used for weighting and for estimating the share of urban population covered by the indicator. The indicator reported for ozone reflects the cumulative annual exposure to ozone measured in urban background locations. The indicator SOMO35, expressed in this report as µg/m3 × days, can be used to quantify the cumulative yearly health impacts of ozone. The exposure parameter is the sum of excess of maximum daily 8-hour averages over the cut-off of 70 µg/m³ (35 ppb) calculated for all days in a year. The term SOMO35 is proposed as a name for this indicator of cumulative annual exposure (WHO Regional Office Europe, 2008). National estimates are calculated as annual means of city-level accumulated maximum daily 8-hour average ozone concentrations in excess of 35 ppb. As for PM, population data is used for weighting and for estimating the share of urban population covered by the indicator. 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, 2015b).

2.10. Arctic Monitoring and Assessment Programme (AMAP)

The Arctic Monitoring and Assessment Programme (AMAP) is the Arctic Council's group responsible for monitoring and assessment of pollution- and climate change-related issues in the circum-Arctic region. Since its establishment in 1991, AMAP has delivered and a series of scientific assessments covering issues relevant to this report including acidification and Arctic Haze (most recent update in 2006), mercury (most recent update in 2011) and POPs in the Arctic. These assessments are intended to support sound, science-based policy-making.

AMAP has been active in the work to establish, develop and implement international agreements that aim to reduce the impact of long-range transported contaminant on Arctic ecosystems, including impacts on health of Arctic human populations. In this context, AMAP has a mandate to support the implementation of the UN ECE LRTAP Convention, and in particular its Protocols on Heavy Metals and POPs as well as the Gothenburg Protocol. The AMAP area partly overlaps with the area covered by the EMEP, and AMAP and EMEP have worked closely together over the past 30 years to coordinate monitoring, quality assurance, and data management activities. A number of air monitoring stations in northern Europe deliver data to both AMAP and EMEP.

The map (Figure 9) shows the AMAP region and its trend monitoring network for POPs and mercury in air, environmental media (primarily based on monitoring of freshwater and marine fish, seabirds and marine mammals), and locations of recent AMAP human health bio-monitoring studies.

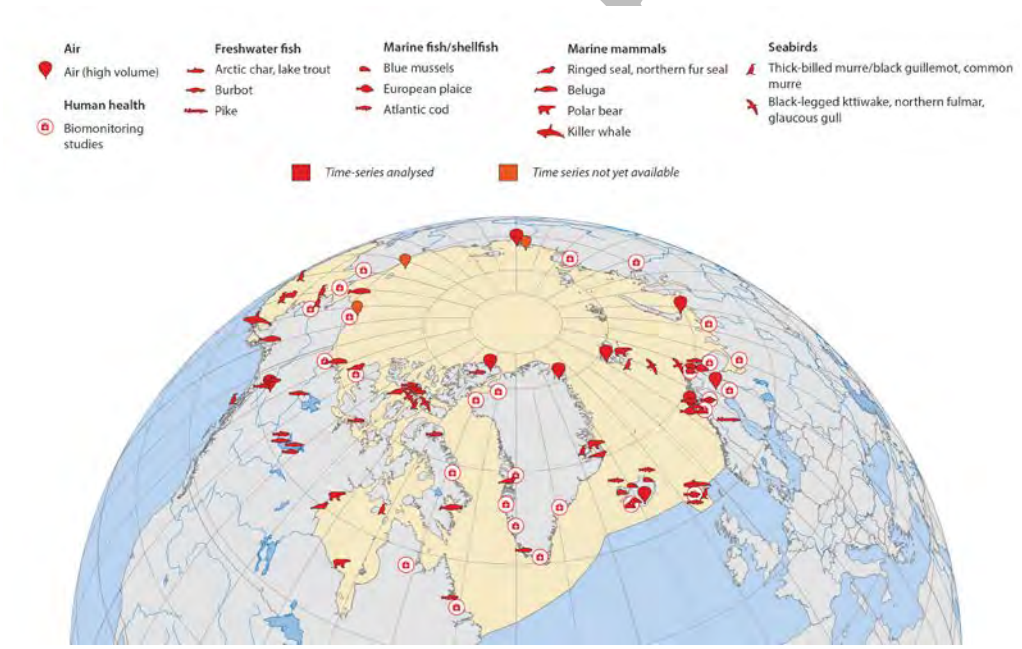

Figure 9 AMAP's trend monitoring network for contaminants in air and biota and locations of recent AMAP human health bio-monitoring studies.

3. Results

3.1. Sulphur and nitrogen (acidification)

3.1.1. Trends in atmospheric concentrations and deposition of S and N

3.1.1.1. Trends in atmospheric concentrations and wet deposition of S and N (EMEP)

European-scale harmonized monitoring of atmospheric composition was initiated in the early 1970s, and the activity has generated a comprehensive dataset (available at www.emep.int) which allows the evaluation of regional and spatial trends of air pollution during a period of nearly 40 years (Tørseth et al, 2012). Here, we present an overview of the observed and modelled trends in wet deposition and air concentrations in Europe from 1990-2012. A more detailed analysis of these trends will be presented in the EMEP TFMM assessment, which is currently being prepared and planned to be published in 2016.

Dry deposition is not measured on a large temporal and spatial scale in Europe, and to assess the trends in dry- and thus total deposition, only modelled results can be used. Trends in air concentration is however a good proxy for trends in dry deposition. Modelled total deposition is used in the calculation of trends in exceedances of critical loads for acidification and nutrients (see 3.1.6 and 3.2.6).

In Figure 10, the observed and modelled average concentrations in air and wet deposition of sulphur show a substantial decrease. Over 90% of the sites show significant (Mann Kendall, $p<0.05$) decreases, while there are no sites with increases. There is a larger decrease in the precursor gas (SO_2) than in sulphate. The mean annual reduction of SO_2 is 3.8% and 4.1% (Sen slope relative to 1990) in observed and modelled concentrations, respectively. For sulphate in aerosols, the modelled trend is slightly larger than in trend in the observations, with 3.2% and 2.6% reductions per year, respectively. For sea-salt corrected wet deposition of sulphate, there are similar differences in the modelled and observed trend, with 3.8% and 2.9% change per year in wet depositions respectively. Thus, the average total change of wet deposition for the whole period from 1990-2012 is 87% and 66% in model results and observations respectively. The trend is somewhat larger in observed concentrations of sulphate in precipitation (72%), in contrast to the modelled results, which is the same in concentration and wet deposition. The annual variations in observed precipitation amounts are quite considerable for some sites, which increases the uncertainty in the trend estimate.

For observed and modelled oxidized nitrogen species there is a clear decreasing trend (Figure 11), though less substantial than for sulphur. Significant decreases are observed at about 50% (Mann Kendall test, $p<0.05$) of the sites depending on component, and a somewhat higher percentage of modelled sites with significant trend. The mean annual reduction of NO_2 is similar for modelled and observed concentrations, 1.2% and 1.4% (Sen slope relative to 1990), but for sum of nitrate (HNO_3+NO_3) in air the difference between measured and modelled trends is larger, and the observations are lower than the modelled concentrations in the early 1990s. This is also the case for wet deposition. The model results give a larger downward trend (2.1% per year) than in observations (1.0% per year). The total change in wet deposition of oxidised nitrogen for the 1990-2012 period is 48% and 22%, respectively, for model results and observations. The average reduction in the observed nitrate concentrations in precipitation is 31%, while 51% for model results, for 1990 to 2012.

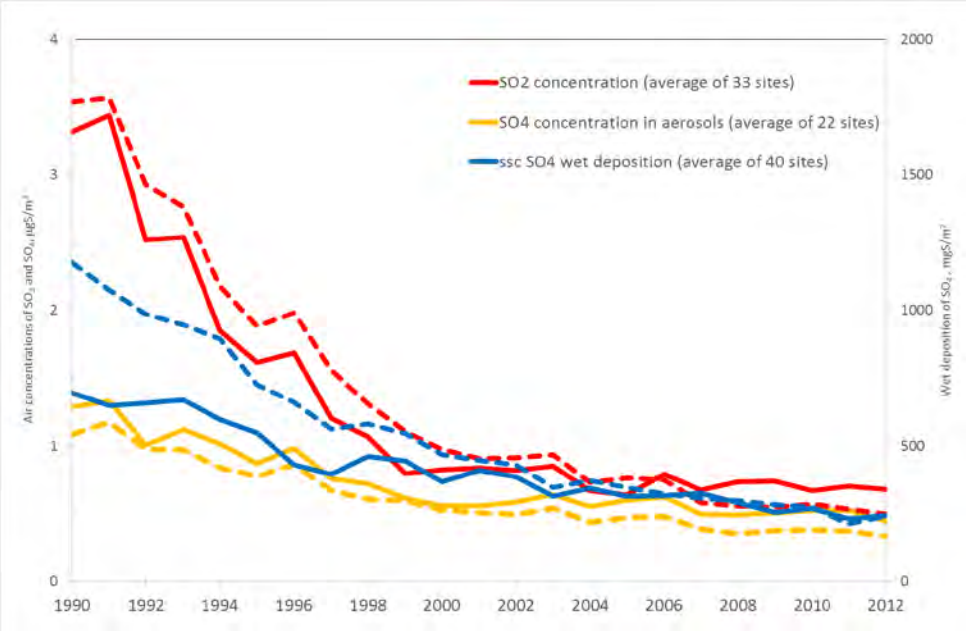

Figure 10: The observed and modelled annual average concentration in sulphur components in air and wet deposition at EMEP sites with measurements for at least 75% of the time period, 1990-2012. Solid lines are observations while the dotted lines are EMEP model results for the same sites

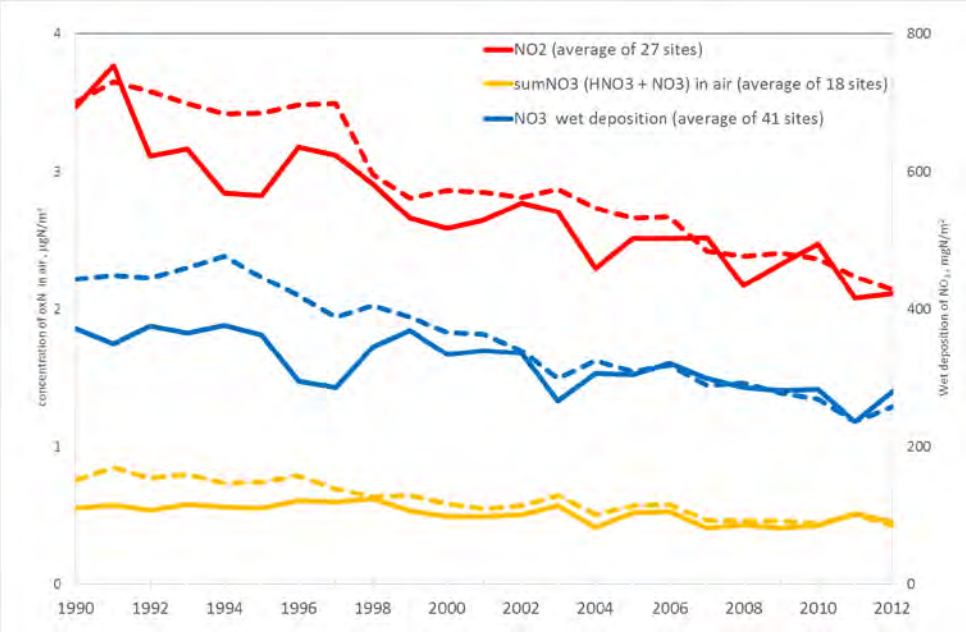

Figure 11: The observed and modelled annual average concentration in oxidized nitrogen components in air and wet deposition at EMEP sites with measurements for at least 75% of the time period, 1990-2012. Solid lines are observations while the dotted lines are EMEP model results for the same sites.

Reduced nitrogen species also show a quite clear decrease for concentrations in air and in wet precipitation (Figure 12). However, there is a rather large variability in the observations, possibly due to changes in regional and local agricultural activities throughout the period. The decrease in observed concentration is significant (Mann Kendall test, $p<0.05$) at more than 60% of the sites in both air and precipitation, for wet deposition there are only 40% with significant trend probably due to larger variability when including precipitation effect. The model and observations of ammonium in precipitation show similar mean annual decreases with 1.3% and 1.1% reductions (Sen slope relative to 1990), respectively. But the trend is less for wet deposition compared to concentrations and the differences between model and observations increase when including precipitation effect, 1.2% and 0.6% reduction in wet deposition for model and observations respectively. The total change in wet deposition of reduced nitrogen for the 1990-2012 period is then 26% and 13%, respectively.

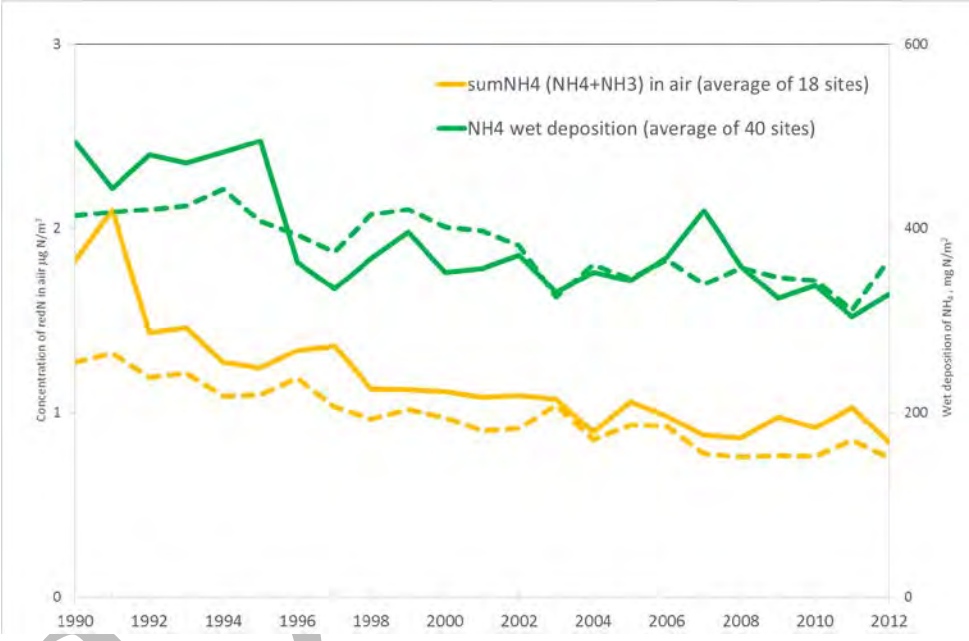

Figure 12: The observed and modelled annual average concentrations in reduced nitrogen components in air and wet deposition at EMEP sites with measurements for at least 75% of the time period, 1990-2012. Solid lines are observations while the dotted lines are EMEP model results for the same sites.

3.1.1.2. Atmospheric deposition of inorganic nitrogen and sulphate to forests (ICP Forests)

Assessing the effects of the deposition of the main pollutants sulphur (S) and nitrogen (N) on forest ecosystems is one of the objectives of the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests). For this purpose, bulk precipitation (BD) and throughfall (TF) have continuously been sampled with harmonized methods (ICP Forests 2010) on the intensively monitored forest plots and nearby open field sites of the Level II plot network.

TF of sulphate sulphur (SO_4^{2-}-S) and inorganic nitrogen (NO_3^--N and NH_4^+-N) were analysed for temporal trends for sets of plots with continuous measurements from 2001 to 2010 (10 years). Trend

analyses for individual plots were carried out with the partial Mann-Kendall (PMK) tests (Libiseller & Grimvall, 2002) and Sen's (1968) slope calculated based on monthly deposition data as described by Waldner et al. (2014) using Marchetto (2013). Relative trends slopes (% yr-1), were calculated by dividing Sen's slope through the mean.

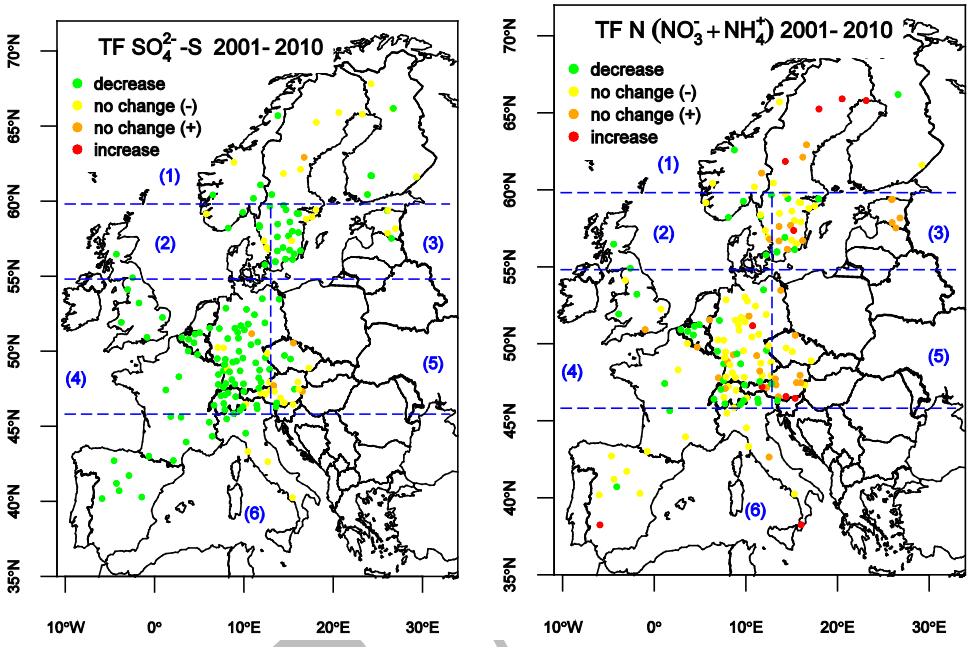

Figure 13: Trend of sulphate sulphur (SO_4^{2-}-S) and inorganic nitrogen (NO_3^--N and NH_4^+-N) in throughfall deposition determined with PMK on plots with continuous measurements (at least 9 years with >300 days of sampling) from 2001 to 2010. Non-significant positive and negative changes are indicated with 'no change (+)' and 'no change (-)'. Displayed in blue are delineation lines that were defined to assign each plots to a region (1: North, 2: Central NW, 3: Central NE, 4: Central SW, 5: Central SE and 6: South).

For individual plots, significant decreasing trends were prevailing, especially for SO_4^{2-}-S (Figure 13). The overall decreasing trends for inorganic N and SO_4^{2-}-S deposition were about 1% and 4% per year, respectively, during the period 2001 to 2010 (Figure 14). Trend slopes were typically in the range from -5% yr^{-1} to +2% yr^{-1} for N compounds and from -10% yr^{-1} to +2% yr^{-1} for SO_4^{2-}-S. The strongest decreasing trends for SO_4^{2-}-S deposition during the 10 year period were found in northern central Europe from Belgium and the Netherlands to Germany. For N compounds the region of strongest decreasing trends extended further to Switzerland, France, Italy, Czech Republic, Slovakia, and Denmark.

Some individual plots sites with significant increasing trends for N deposition were observed in the Nordic countries and the southern part of Europe.

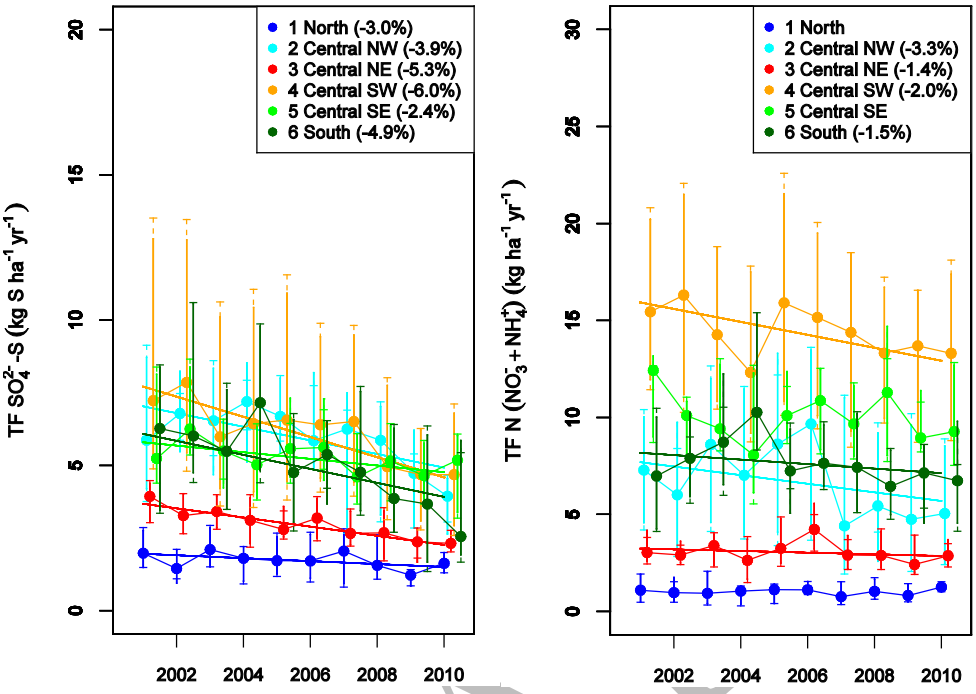

Figure 14: Median and quartile range of inorganic nitrogen (NH4+-N + NO3--N) and sulphate (SO42--S) throughfall deposition of ICP Forests Level II plots with continuous measurements (at least 9 years with >300 days of sampling) from 2001 to 2010 grouped into six regions as delineated and enumerated on Figure 1 (1: North, 2: Central NW, 3: Central NE, 4: Central SW, 5: Central SE, 6: South). A relative trend slope (% yr-1) resulting from a linear mixed effect model (fixed effect: year centered to 2005, random effect: country/plot) are shown for plots grouped according to these regions.

In general, decreasing trends in measured sulphate and inorganic N deposition are in good agreement with the general trends in the EMEP estimates of S and N emission and deposition. However, given the high temporal variability of S and N deposition, long time series are required to correctly assess the presence of significant trends.

3.1.2. Trends in forest responses (ICP Forests)

3.1.2.1. Crown condition

Forest health is assessed by means of tree defoliation (Eichhorn et al. 2010) a relative estimate of the amount of foliage in tree crowns. An increase in defoliation is a sensitive, but unspecific response parameter indicating stress. Trees showing values higher than 25% defoliation are generally considered as damaged. The plot-specific response is often highly variable from year to year, however, when defoliation data are averaged over many plots and years, trends can be found (Figure 15, Table 3).

Applying statistics (regional Sen's slope with Mann-Kendall test) reveal highly significant trends of increasing defoliation for all three groups of Mediterranean tree species, indicating a deterioration of crown condition over time. In the temperate zone, pedunculate and sessile oak together show the most prominent increase of all species, while common beech reveals only a moderate increase. A smaller, but

still significant, trend is found in Norway spruce, and no trend at all in Scots pine. Both conifers are mainly distributed in the temperate and boreal part of Europe.

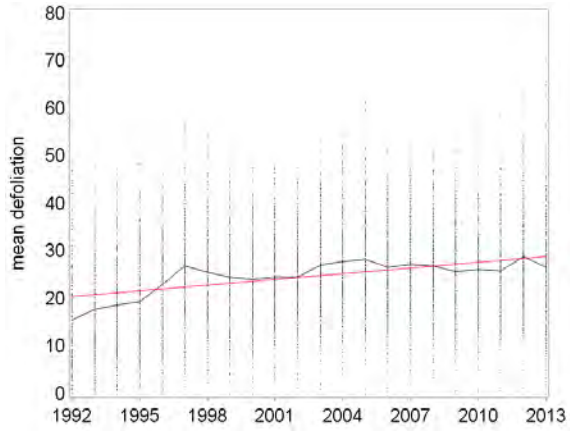

Figure 15: Plot mean defoliation (points) and the defoliation trend (regional Sen's slope, red line) for pedunculate and sessile oak 1992-2013. Black line connects annual means

Table 3: Statistics for defoliation of European main tree species or species groups; *: Turkey oak, downy oak, Hungarian oak, Pyrenean oak, **: kermes oak, holm oak, ballota oak, cork oak, ***: maritime pine, Aleppo pine, stone pine.

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
Pedunculate/sessile oak	0.221	+ 0.400	< 0.0001	24.58
Common beech	0.134	+ 0.200	< 0.0001	19.21
Norway spruce	0.098	+ 0.075	<0.0001	20.79
Scots pine	-0.006	0.0000	0.3430	18.18

Tree defoliation is influenced by many other factors, such as biotic damaging agents and weather conditions (Scidling 2007, Carnicer et al. 2010; Ferretti et al. 2014, de Vries et al. 2014). Responses to air pollution appears to be pollutant- and species-specific. While field evidence of O_3 effects on defoliation is ambiguous (e.g. Ferretti & Bussotti 2007; Šrámek et al. 2014), research based on data from the ICP Forests network has shown a substantial importance of nitrogen load as predictor for defoliation in certain tree species (Veresoglou et al. 2014, De Marco et al. 2014, Vitale et al. 2014) and thoughfall N deposition correlates with a higher proportion of common beech and Norway spruce trees with defoliation >25%, while the effect is opposite for Scots pine (Ferretti et al. 2015).

3.1.2.2. Foliar Element Contents

Foliar nutrient concentrations have been monitored at Level II plots starting in the early 1990s. The objectives have been to assess the possible changes in the concentrations of the main pollutants (sulphur and nitrogen) and how these affect forest tree nutrition. Here, the nutritional status of the main tree species was evaluated using data collected from 1992-2009. Sampling was carried out according to Rautio et al. (2010) and trends analysed by linear mixed models under consideration of plot and country as random factors (Jonard et al. 2015).

Similar to the decreasing trends in N and S concentrations in many species, many essential nutrients are decreasing also, while leaf mass tends to increase. The most alarming trend is the deterioration in P nutrition during the past two decades in some of the main tree species (Table 4, Figure 16). Increased tree productivity, possibly resulting from a fertilization effect of N deposition and of rising atmospheric CO_2, has led to higher nutrient demands. Soil nutrient supply may not always been sufficient to meet the demand of faster growing trees, however, changed within-tree P allocation patterns are discussed as causes as well. As tree nutrition exerts a tight control on net ecosystem productivity, this deterioration in tree nutrition could have a strong impact on the response of forest ecosystems in the future.

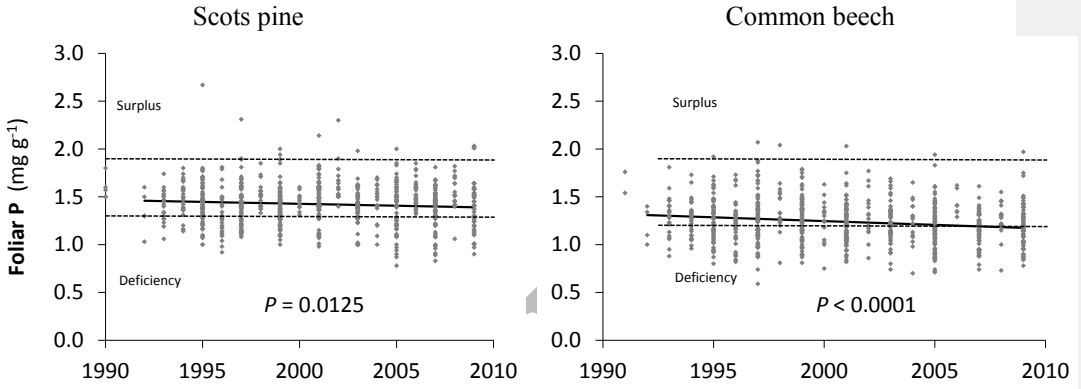

Figure 16: Temporal trends in foliar phosphorus concentration of current-year leaves for Scots pine and Common beech. Dashed lines: thresholds separating deficiency, normal and surplus ranges after Mellert & Göttlein (2012); $p < 0.05$ indicate significance.

Table 4: Linear temporal trends in foliar concentrations for main tree species. Sign of slope: '+' or '-', significance: p < 0.1: (+) or (-), p < 0.05: + or -, p < 0.01: ++ or --, p < 0.001: +++ or ---.

Tree species	needle age	Mass	Foliar concentrations (mg g^{-1})					
			N	P	S	Ca	Mg	K
Common beech		+++	-	---	---	-	---	
Sessile oak			---	---	---	-		-
Pedunculate oak		(+)						
Silver fir	current year		--				+	
Norway spruce	current year	+++			---		(+)	--
Scots pine	current year			-	---			
Silver fir	1-year-old			-				
Norway spruce	1-year-old		-	---	--			---
Scots pine	1-year-old					+++		

3.1.3. Trends in input-output budgets of S and N (ICP IM)

Mass balance budgets integrate information about the complex chemical and biological recovery processes that govern the retention or release of sulphur and nitrogen compounds and regulate acid production and buffering, in both the terrestrial and aquatic portions of catchments in the ecosystem. In order to assess the long-term trends in retention and release of S and N in the European forested catchments, annual input (bulk and throughfall deposition) and output (catchment runoff) fluxes 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 ICP IM sites, according to data availability (Vuorenmaa et al. 2014). The percent net export (pne), used in

this chapter, illustrates the ratio of catchment input to catchment output for S and N (in %). A positive value indicates release and a negative value indicates retention in the catchment.

Sulphur deposition significantly decreased at all 18 ICP IM sites from 1990 to 2012 (Vuorenmaa et al. 2014). As a response to decreased S deposition, sulphate fluxes in runoff decreased at 15 out of 18 sites, being significant at 67% of the sites. Bulk deposition of nitrogen also decreased at almost all sites (17 out of 18), being significant at 71% of the sites. In contrast to sulphate, total inorganic nitrogen (TIN) fluxes in runoff showed mixed responses. Statistically significant decreasing trends were observed at five sites and increasing trends at two sites. The significant increasing trends were probably due to excess N mineralization and increased NO_3 leaching, from forest damage and associated dieback related to storm logging and bark beetle infestation.

Sulphate budgets showed a net release of previously stored SO_4 at majority of the sites, particularly during the past 15 years (Figure 17). This process has taken place both in high and low sulphur deposition areas. 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.

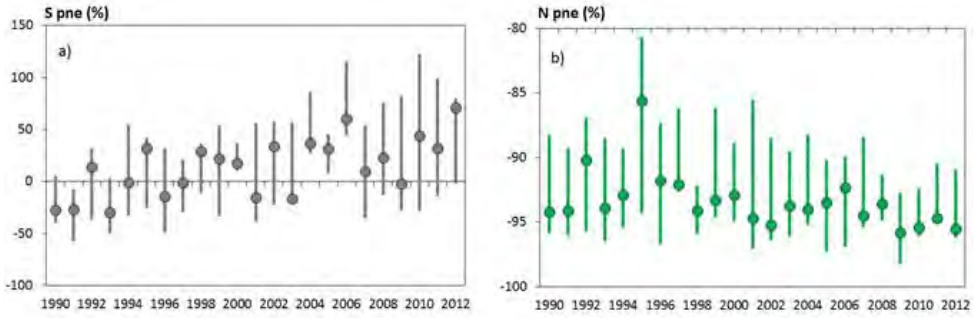

Figure 17: Percentiles (25%, median 50%, 75%) of percent net export (pne, %) of sulphate (SO_4) and total inorganic nitrogen (TIN) for 8 IM sites in 1990-2012 (a and b, respectively)

The uptake of N is efficient in forest ecosystems, and nitrogen deposition is effectively retained in boreal terrestrial catchments. The percent net export (pne) of TIN generally ranged between -98% and -80% at the studied ICP IM sites during the 2000s (Figure 17), indicating a strong retention of nitrogen in the catchment, and 50% of the sites exhibited increase in net retention. This is in agreement with other studies, which show that 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 deposition continues to accumulate in catchment soils and vegetation. Nitrogen saturation and excess NO_3 leaching to surface waters 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.

3.1.4. Trends in water chemistry and biology (ICP Waters)

Water chemistry has responded strongly to reductions in sulfur deposition. Figure 18 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 (i.e. not covered by ice during the last glacial period) catchments in North America, where sulfate in rain still adsorbs to the soil. A large sulfate adsorption capacity is typical for deeply-weathered soils in non-glaciated areas, and can result in high leaching of sulfate when the capacity is exceeded, even after a substantial reduction in sulphur deposition. Few regions show a regional trend in nitrate concentration (see 3.1.4), implying that its relative importance as an acidifying anion has increased. However, sulphate is still the dominating acid anion for the majority of sites (Skjelkvåle and De Wit, 2011).

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 glaciated areas in North America, pH increased from 5.6 to 6.1. There is no trend in pH in surface waters from non-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).

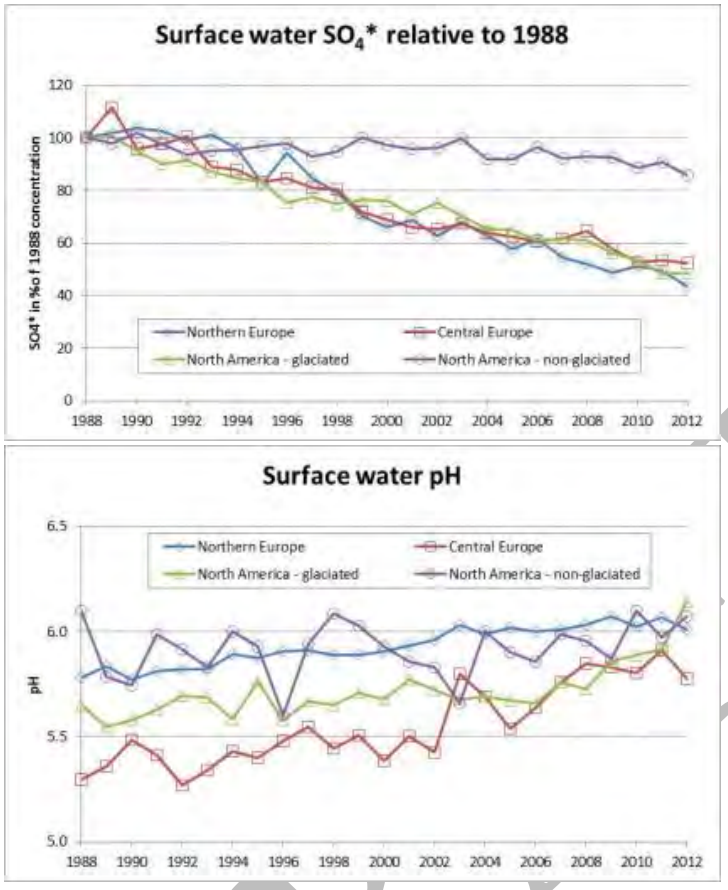

Figure 18: Trends in surface water chemistry at ICP Waters sites 1988-2012. Shown are the mean concentrations of non-marine sulphate (SO4*) and pH at 22 sites in northern Europe, 21 sites in central Europe, 37 sites in glaciated areas of eastern North America, and 7 sites in non-glaciated areas of eastern 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 (Figure 18). 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.

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 5: 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	

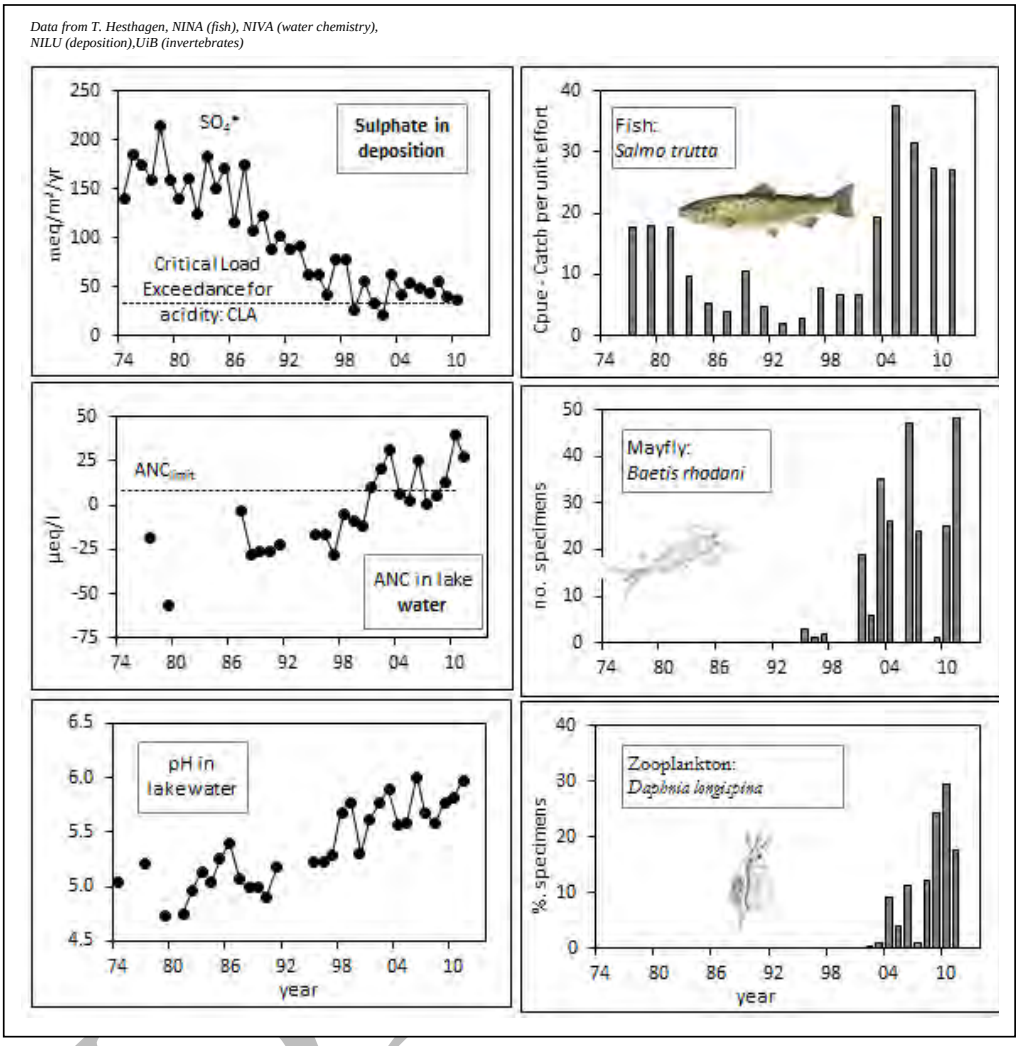

Figure 19: Biological recovery – an example from Norway. Recovery from acidification at Lake Saudlandsvatn, Norway. As sulfur deposition has decreased, the acid neutralising capacity (ANC) and pH have increased in the lake, and the populations of three sensitive species, i.e. brown trout, the mayfly *Baetis rhodani* and the planktonic crustacean *Daphnia longispina* have begun to recover (modified from Hesthagen et al. 2011 Sci Tot Env).

The chemical recovery is expected to proceed slowly as demonstrated by the increase in observed ANC and its trend prognosis in Europe for the near future (Figure 20). The expected increase in ANC is small compared to the year-to-year variations in measured values caused by fluctuations in deposition and climatic conditions. Predictions show that approximately 1/3rd of the European ICP Waters sites will still have ANC lower than the critical limit in 2020 (Garmo and De Wit 2015). A changing climate is therefore likely to affect biological recovery from acidification.

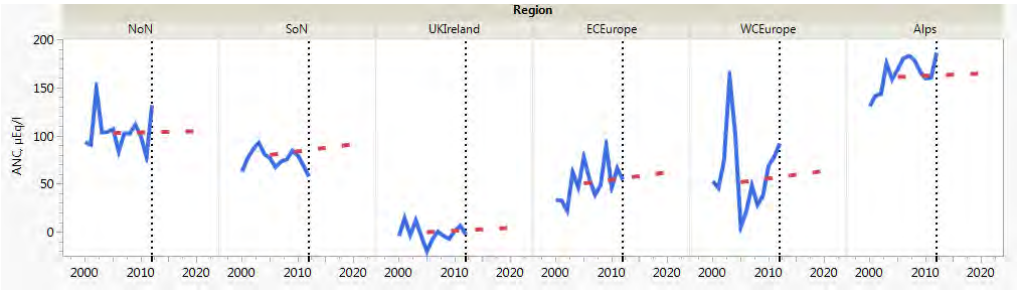

Figure 20: Observed mean (blue) and modelled (red) acid neutralizing capacity (ANC) in European regions between 2000 and 2020. Regions are northern Nordic (NoN), southern Nordic (SoN), the UK and Ireland, East-Central Europe (EC Europe), West-Central Europe (WC Europe) and the Alps (see also 2.7).

3.1.5. 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 have been minor. Taking carbon steel as an example, several different one-year exposures have been performed during the period 1987-2011. Figure 21 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. It is worth noting that even for the originally "clean" site (Aspvreten), there has been substantial improvement in corrosion, resulting from improved air quality, during this period.

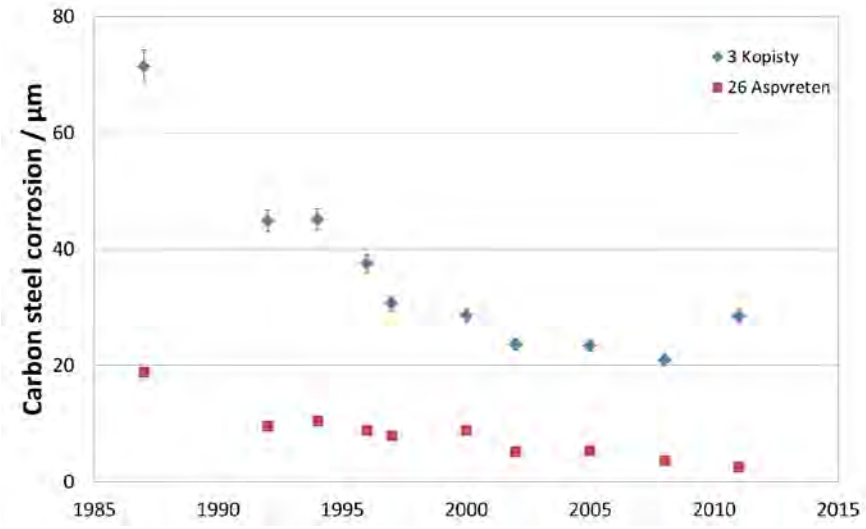

Figure 21: Trends of carbon steel at two selected test sites.

Differences in corrosion between polluted (industrial) and non-polluted areas are still evident. There is also a difference between urban and rural areas, but due to improvements in air quality recent average

values in urban areas are now in fact lower than the average values for rural areas from 1987 (Figure 21).

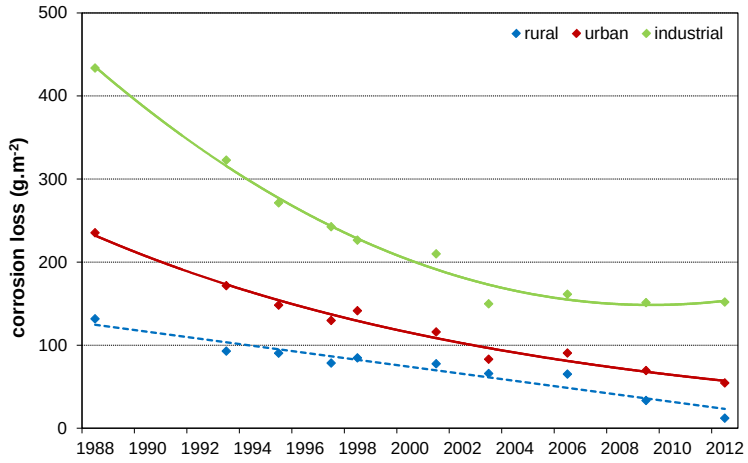

Figure 22: Trends in carbon steel yearly corrosion loss based on results from 20 test sites.

For cultural heritage objects made of metals the changes are instantaneous, responding rapidly to decreasing pollution levels. For stone materials, however, there is a substantial time lag of 20 years or more before improvements can be seen. This is related to the porous structure of the material, and therefore its ability to maintain pollutants in its fabric or structure for a significant period of time after the air pollutants have disappeared.

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 measured on fresh surfaces and on building surfaces measured by so called micro-erosion meters inserted into the building fabric. Figure 23 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 of 100 µm year^{-1} for the period from 1718 (when the balustrade was completed) to 1987, 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. 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 23). 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, stone, related to the porous structure of the stone material as explained above.

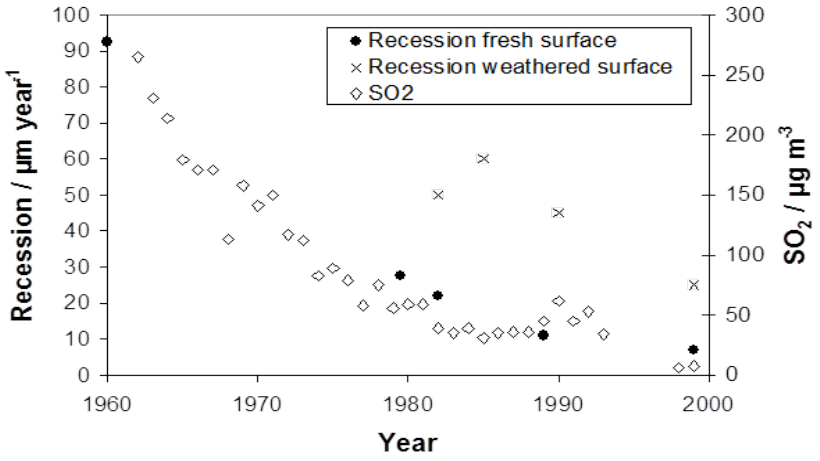

Figure 23: Surface recession rates of limestone and SO_2 concentration at St Paul's Cathedral. The surface recession of fresh surfaces is measured on standard samples while recession on weathered surfaces is measured on the actual building using micro-erosion meters.

The guidance document on 'health and environmental improvements using new knowledge, methods and data' (ECE/EB.AIR/124) estimates that future materials corrosion (% area at significant risk) will decrease from an EU-28 average of 47% in 2005 to 17% in 2020. This is a significant improvement but there are still several countries where the geographical area at risk is more than 50%. The scenarios used in the guidance document might be influenced by the possible effect of climate change as a confounding factor, see Figure 24. Depending on the locations, 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 24). The magnitude of the risk depends on the chosen scenario.

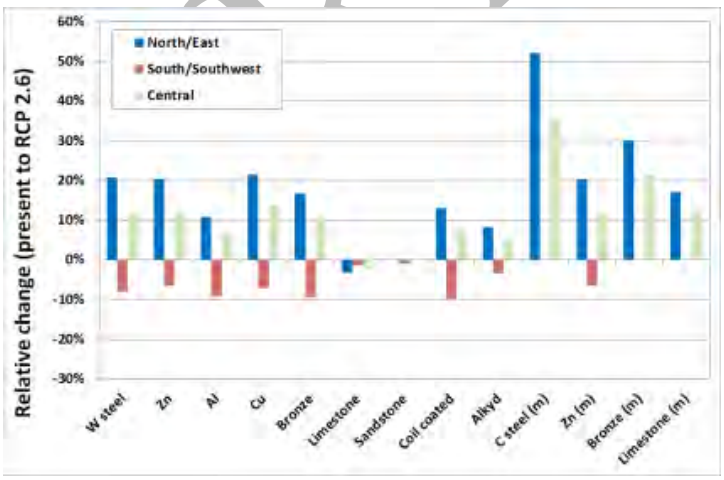

Figure 24: Calculated relative changes in corrosion due to SO_2 for the IPCC RCP2.6 scenario. Calculations are based on dose-response functions for the SO_2 dominating situation for most materials and dose-response functions for the multi-pollutant situation, indicated by (m).

The data in figure Figure 24 is based on calculations using dose-response functions that include the combined effect of possible changes of temperature and relative humidity, from the period 1986-2005 to the period 2081-2100, in different geographical areas as determined by the IPCC RCP2.6 scenario.

3.1.6. Trends in exceedances of CLs for acidification (ICP M&M - CCE)

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 25).

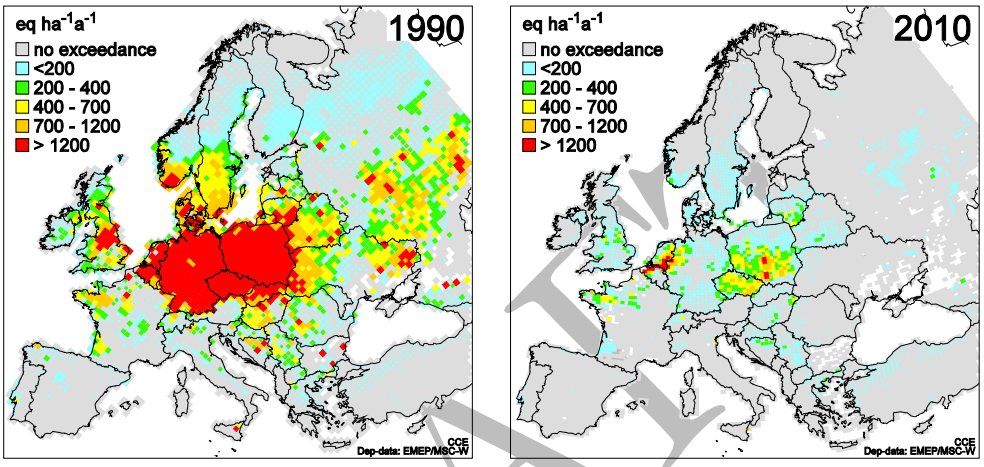

Figure 25: Exceedances (AAE) of the critical loads of acidity in 1990 (left) and 2010 (right) in Europe. Note that the size of the colour shading in a grid cell indicates the % of the ecosystem area at risk. The ecosystem area can vary widely among grid cells. (Deposition data from MSC-W; European critical loads data from the CCE)

Assuming the GP-CLE scenario to be implemented as of 2010, Figure 26 shows that the percentage of total (red) and of the aquatic ecosystem (blue) area of which critical loads for acidification are exceeded, attained peaks of 30% and 40%, respectively, in 1980. Note that the area of aquatic ecosystems is significantly smaller than that of terrestrial ecosystems. The AAE peaks also occurred in 1980. However, compared to the area exceeded, a reversed order can be noted, i.e. the AAE on terrestrial ecosystems (350 eq $ha^{-1}yr^{-1}$) exceeded the AAE on surface waters (250 eq $ha^{-1}yr^{-1}$) in 1980.

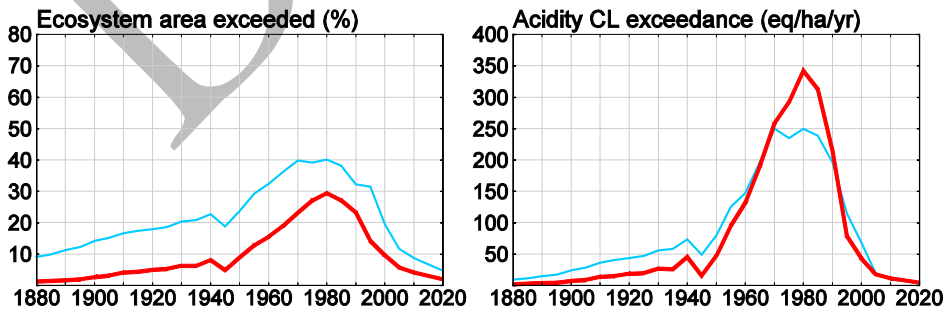

Figure 26: Temporal development of the area in Europe where acidity critical loads are exceeded (left; % of ecosystem area) and average accumulated exceedance (AAE) (right; eq ha^{-1} yr^{-1}) of critical loads of acidity (red: all ecosystems, blue: surface waters). Future exceedances are based on the implementation of agreed legislation (emission and deposition data from EMEP-CIAM and EMEP- MSC-W; 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 are available. 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 of aquatic and terrestrial ecosystems to be established (Hettelingh et al. 2015b).

The area exceeded by critical loads for acidification reached its 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 for acidification.

3.1.7. Trends in exceedances of CLs for acidification at ICP IM sites (ICP IM)

Data from the intensively monitored ICP IM sites provide a connection between modelled critical thresholds and empirical observations, and thus a means of testing the applicability of critical load estimates for natural ecosystems. Critical loads of acidification were calculated for 18 ICP IM catchments (Holmberg et al. 2013). The level of protection of these sites with respect to acidifying deposition was estimated for 2000 and 2020 (Table 6). The NAT2000 scenario represents historic emissions for the year 2000 while the MID2020 and MFR2020 scenarios are based on economic projections by the PRIMES model. The scenario MFR2020 assumes that all available abatement technologies are being implemented by 2020. 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. 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, supporting the use of CL exceedances as measures of acidification (Holmberg et al. 2013).

Table 6: Average exceedances for ICP IM sites not protected from acidification ($AExCL_A$) and percentage of sites protected with different deposition scenarios.

	Total nr of sites	NAT2000	MID2020	MFR2020
$AExCL_A$ (eq $ha^{-1}yr^{-1}$)	18	987	392	310
Acidification protection %	18	61 %	67 %	67 %

3.1.8. Time delays in damage and recovery (JEG)

The protocols of the LTRAP Convention are based on the underlying assumption that reductions in emissions of air pollutants will decrease harmful effects to ecosystems, materials and human health. The trends in effects are in many cases delayed, sometimes by years to decades, from the changes in concentrations and deposition of air pollutants. Sulphur deposited and stored in soil may take years to decades to leach out (Par 3.1.3), thus delaying the response to reduced S deposition (Church et al. 1990). Similarly most terrestrial ecosystems store a large fraction of N from deposition, and ecosystem effects may be first apparent after years of decades of excess N deposition (Par. 3.2.2 and Par 3.2.6). Recovery of

soils and surface waters may be delayed while base cation stores are replenished after decades of leaching to increase acid-neutralizing capacity (Wright et al., 2005). Similar delays may occur in effects of air pollutants on human health (Par 3.4.2), as health effects may reflect cumulative exposure. Time delays vary widely between effects, ecosystems and indicator organisms.

"Dynamic models" take into account time delays and thus can help explain observed trends in effects, why, for example, biological recovery lags behind (Par 3.1.4), despite the fact that emissions of sulphur have been reduced by >80%. Dynamic models can be used to project future trends in effects, given scenarios of future emissions (Wright et al. 2005; Helliwell et al. 2014). They are thus tools useful for policymakers. Dynamic modelling can help assess, for example, how much faster ecosystems will recover if emissions are decreased over a period of 10 years, or if they are decreased over a period of 50 years. By introducing the time dimension the critical load becomes the "target load", in which the time and duration of implementation is considered in addition to the amount of deposition (Jenkins et al. 2003).

So far, target loads have not been used in protocols (and revisions) of the Convention. Dynamic models are available to calculate target loads for surface waters and forest soils, and are under development for ground vegetation in semi-natural ecosystems.

3.2. Nitrogen as a nutrient

3.2.1. Trends in deposition (EMEP)

For trends in deposition and atmospheric concentrations of reduced and oxidized nitrogen species, reference is made to Paragraph 3.1.1.

3.2.2. Trends in ground vegetation species cover and diversity (ICP IM)

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 with a total of 1,335 permanent forest floor vegetation plots was used to analyse temporal trends in species cover and diversity (Dirnböck et al. 2014, Figure 27).). The exceedance of the CL for eutrophication ($ExCL_{empN}$) was estimated as the measured throughfall N deposition minus the empirical CL for each site. Trends in diversity were analysed using the number of vascular plant species and beta diversity. 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.

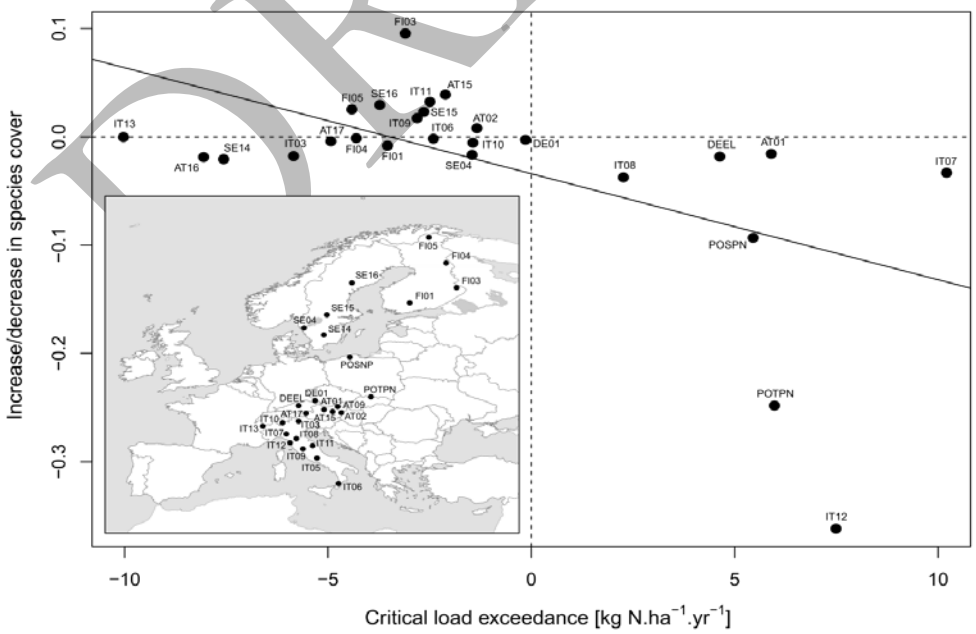

Figure 27: Forest plant species that prefer low soil nutrient levels have decreased during the last 10-50 years in 28 ICP IM and ICP Forests sites across Europe owing to the exceedance of the nitrogen critical loads. The y-axis indicates the strength of the cover change of all oligotrophic species in 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).

3.2.3. Trends in nitrogen in mosses (ICP Vegetation)

Between 2005 and 2010, the average median nitrogen concentration in mosses 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).

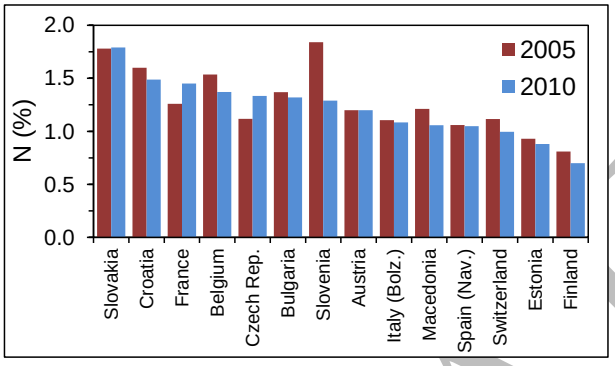

Figure 28: Median total nitrogen concentration in mosses in 2005 and 2010. In Italy, mosses were sampled in Bolzano region (Bolz.) and in Spain in Navarra regions (Nav.).

3.2.4. Trends in nitrogen leaching/ catchment input-output budgets (ICP W)

Atmospheric deposition of nitrogen may lead to enhanced leaching of N species to surface waters in natural and semi-natural ecosystems, and impact freshwater biology in the form of shifts in algal communities and increased productivity (De Wit and Lindholm, 2010).

Nitrate in surface waters has been highly variable since the late 1980s. Figure 29 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, except for lakes in the southern Alps where NO3 in surface water correlates with deposition of nitrogen (Rogora et al., 2012). Elsewhere, interannual and inter-site variation dominates rather than long-term trends. In those sites where trends are found, declines in nitrate are more common than increases (Garmo et al. 2014).

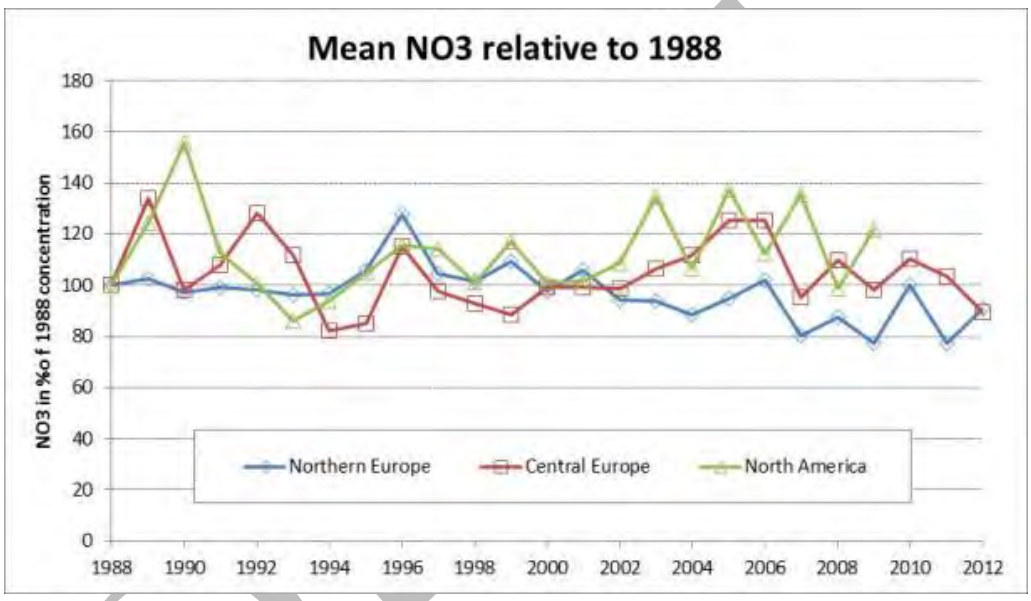

Figure 29: *Trends in surface water chemistry at ICP Waters sites 1988-2012. Shown are the mean concentrations of NO3 at 22 sites in northern Europe, 21 sites in central Europe and 37 sites in eastern North America. (for 2010-2012, no data available for North America). (Source: De Wit et al. (2015))*

Trends on total inorganic nitrogen (TIN=NO_3 + NH_4) fluxes in runoff and retention and release of TIN in the catchments at ICP IM sites showed mixed response with both decreasing and increasing trends. Trends were rarely significant and declines on TIN fluxes were more common than increases. Nitrogen deposition is effectively retained in the catchment; at a majority of the sites < 10% of TIN deposition was leaching in runoff (paragraph3.1.3). Data from the ICP IM sites have shown evidence of a link between N deposition, exceedances of critical loads of nutrient nitrogen and inorganic nitrogen leaching (see paragraph 3.1.7, Holmberg et al. 2013).

Thus, trends in inorganic nitrogen concentrations and fluxes do not show a general increasing pattern, despite continued accumulation and retention of atmospheric inorganic nitrogen in catchment soils.

3.2.5. Trends in exceedances of critical loads for eutrophication (ICP IM)

Mass balance critical loads of nutrient nitrogen $CL_{nut}N$ (eq ha^{-1} yr^{-1}) were calculated for 18 ICP IM catchments (Holmberg et al. 2013). In addition to the mass balance critical loads of nutrient N, empirical critical loads of nutrient N, $CL_{emp}N$, were assigned to a total of 83 vegetation plots at 37 ICP IM sites. The NAT2000 scenario represents historic emissions for the year 2000 while the MID2020 and MFR2020 scenarios are based on economic projections by the PRIMES model. The scenario MFR2020 assumes that all available abatement technologies are being implemented by 2020. In 2000, around 20% of the IM sites were protected from eutrophication. In 2020, at best, with the maximum technically feasible reductions, only half of the sites would be protected from eutrophication.

Table 7: Average exceedances for ICP IM sites not protected from eutrophication ($AExCL_{nut}N$, $AExCL_{emp}N$) and percentage of sites protected with different deposition scenarios.

	Total nr of sites	NAT2000	MID2020	MFR2020
$AExCL_{nut}N$ (eq $ha^{-1}yr^{-1}$)	18	625	286	230
Eutrophication protection % (mass balance critical load)	18	22 %	33 %	39 %
$AExCL_{emp}N$ (eq $ha^{-1}yr^{-1}$)	37	578	221	154
Eutrophication protection % (empirical critical load)	37	19 %	41 %	54 %

Evidence of a link between exceedances of critical loads of nutrient nitrogen and nitrogen leaching was found at the ICP IM sites (Figure 30). The collected empirical data of the ICP IM thus allow testing and validation of key concepts used in the critical load calculations.

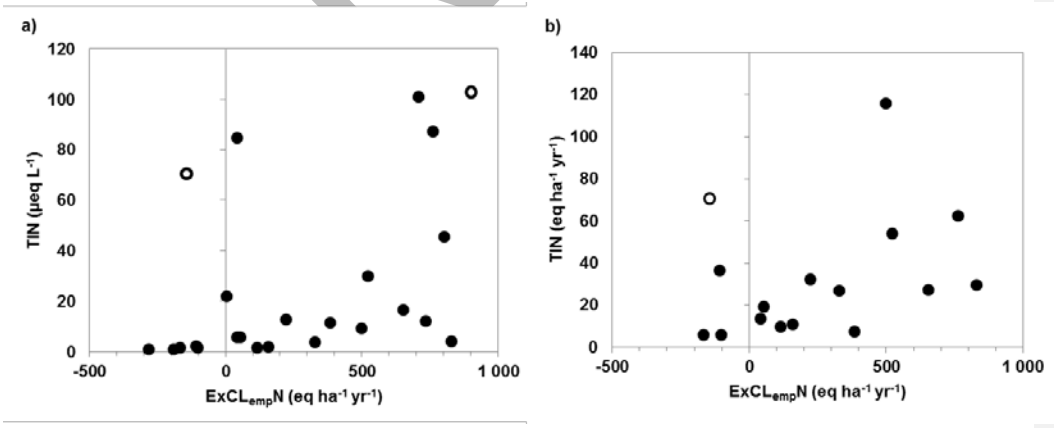

Figure 30: Eutrophication empirical impact indicators in relation to exceedance of critical loads of N at ICP IM sites. The x-axes show exceedances of empirical critical loads of nutrient N ($ExCL_{emp}N$, NAT2000 deposition). The y-axes show annual mean concentrations (a) and fluxes (b) of TIN (=NO_3 + 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).

3.2.6. Trends in exceedances of CLs (ICP M&M - CCE)

The risk of eutrophication, both in terms of magnitude and geographical distribution, is computed to diminish from 1990 to 2010 (Figure 31). However, it is obvious that critical loads of nutrient N remain exceeded in all European countries, while areas with relatively high exceedances (red or orange shading) in 2010 occur in most of the countries.

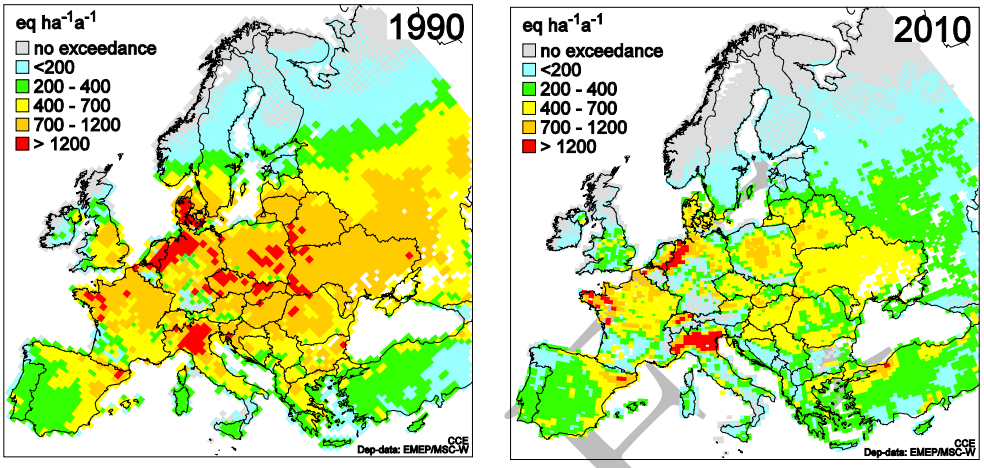

Figure 31: Exceedances (AAE) of the critical loads of nutrient N in 1990 (left) and 2010 (right) in Europe. Note that the size of the colour shading in a grid cell indicates the % of the ecosystem area at risk. The ecosystem area can vary widely among grid cells. (Deposition data from MSC-W; European critical loads data from the CCE)

The area with high exceedances (red and orange shading) of critical loads of nutrient N was 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) was larger in 2010 in comparison to 1990. This implies that abatement policies have had important positive effects, although lower exceedances in 2010 still imply that damage to structure and function of ecosystems may occur. Compared to the magnitudes of exceedances in 1990, damage caused by exceedances in 2010 may occur at a later point in time.

Figure 32 illustrates that areas where critical loads of nutrient N are exceeded, continue to remain a serious issue under GP-CLE emissions. Eutrophication is computed to affect 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 $ha^{-1}yr^{-1}$.

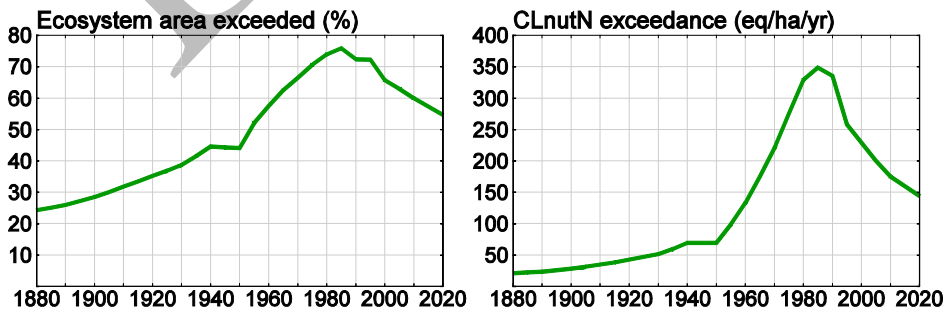

Figure 32: Temporal development of the area in Europe where nutrient N critical loads are exceeded (left; % of ecosystem area) and average accumulated exceedance (AAE) (right; eq ha^{-1} yr^{-1}) of nutrient N critical loads (all

ecosystems). Future exceedances are based on the implementation of agreed legislation. (Emission and deposition data from EMEP-CIAM and EMEP- MSC-W; 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.

3.2.7. Factors affecting recovery/delay in recovery (JEG)

Dynamic modelling of ecosystem response to changing N deposition is often complicated by the lack of observed statistically significant trends over the time span of available observations. Conceptually, the commonly used models of N leaching such as MAGIC, VSD or ForSAFE respond to changes in N input by changing soil-, ground- or runoff water NO3 concentrations. Consequently also the models of vegetation change (e.g. Veg, PROPST) respond by predicting changes in plant communities. However, common rates of change in atmospheric N deposition are typically slow and typically even the ecosystems most sensitive to N deposition contain equivalent of hundreds to thousands of years of current annual N deposition. The large pool of nitrogen in the soil and in vegetation is dynamic with internal N circulation typically several fold larger than the external annual input. Incoming "new" nitrogen is by and large incorporated into the existing N pool. The ecosystem change could thus be very significantly delayed by slow rate of the change in the large total N pool caused by relatively slow trends in N input. Further complication in modelling and/or detecting changes in vegetation is due to importance of other factors such as light. In forests, silviculture practices can be the main driver of plant species composition and thereby complicating detection of changes (Dirnböck et al., 2014). The situation is somewhat easier for e.g. grasslands (Stevens et al., 2010). Critical load (CL) estimates traditionally take the precautionary principle, i.e. assuming that deposition above the critical tolerable level eventually will cause undesirable change, without answering the question when. Such an approach is adequate to the task of mapping consequences of N emissions scenarios, because it provides calculations of the extent of damage due to CL exceedance – provided enough time (Figure 32). It is, however, important to recognize that the damage may occur much later than in the year for which exceedance is calculated and that there are other important factors (climate change and land management being the two most important ones) which could further complicate future predictions.

The trend data from surface waters and from the integrated monitoring sites (Par 3.2.4) indicate that leaching rates of N from N-poor terrestrial systems respond fast to decreasing deposition, in contrast to a slow response to increasing deposition. Similar responses were found in the experimental catchment manipulation project NITREX, where N inputs were decreased in roof experiments or increased by direct additions (Emmet et al., 1998).

3.3. Ozone (ca 5-10 p)

3.3.1. Trends in concentrations (EMEP, TFMM) (max 2 p)

Within the EMEP TFMM assessment, trends in various ozone metrics have been calculated for EMEP and Airbase sites for different time periods. Figure 33 shows the measured annual 95-percentile of daily max ozone during 1990-2012 for four selected sites together with the modelled values, using the EMEP MSC-W model (Simpson et al., 2012). The estimated Sen's slopes are also plotted. For CZ03 (Kosetice) and GB36 (Harwell) a good agreement between modelled and measured values are seen, both showing a marked downward trend. For AT30 (Pillersdorf bei Retz) and DE02 (Waldhof) the results indicate a clear correlation between modelled and measured data while the model shows a stronger reduction than the measurements.

These examples reflect a more general pattern in Europe, as seen in Figure 34. This figure shows the modelled trends (1990-2012) with the results from the EMEP stations on top. Note that for clarity, the significance criteria for the stations were changed from p=0.05 to p=0.2 in this plot. For several of the sites the results were not significant on a p=0.05 level. In the UK, Scandinavia and a few other sites the magnitude of modelled trends agree with the observed trend, while for Germany, Switzerland and Austria the measured trends are generally smaller (smaller reductions) than predicted by the model. Also in the last decade (2002-2012), peak ozone levels have dropped in most regions (-0.2 ppb y^{-1} to -0.3 ppb y^{-1} on average), but the reductions are generally smaller than expected from model predictions and not statistically significant at many sites.

Differences between model results and measurements may reflect the confounding effects from titration of ozone by NO_2 and the inter-annual variability in weather regimes. Whether the difference between models and measurements also point to systematic deficiencies in the models, is an open question which should be studied further.

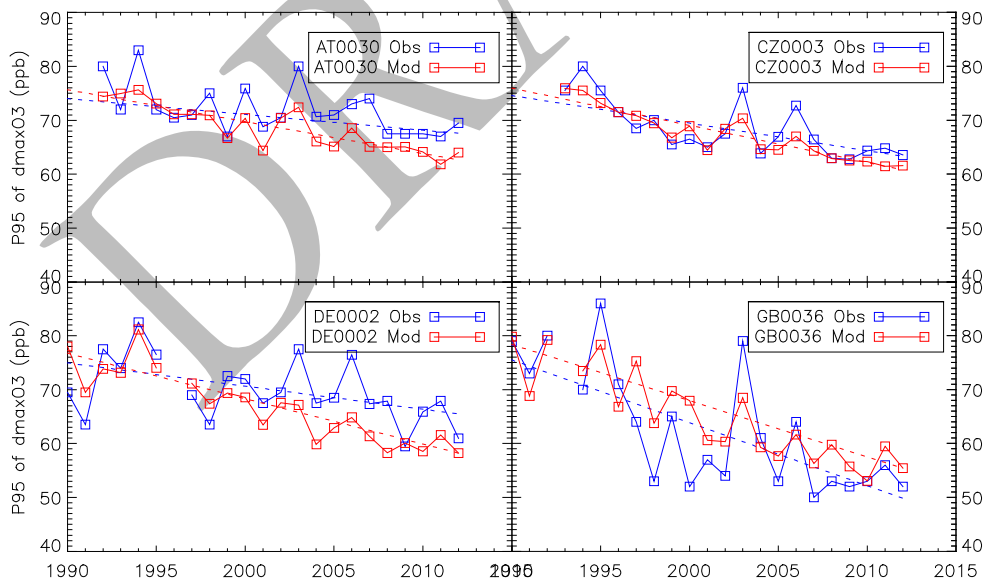

Figure 33: Observed and modelled trend in the annual 95-percentile of daily max ozone 1990-2012 for four EMEP sites. The dotted lines show the significant Sen's slope (p=0.05) for the observed and modelled values, respectively. AT0030: Austria, Pillerdorf bei Retz; CZ0003: Czech Republic, Kosetice; DE0002: Germany, Waldhof; GB0036: United Kingdom, Harwell.

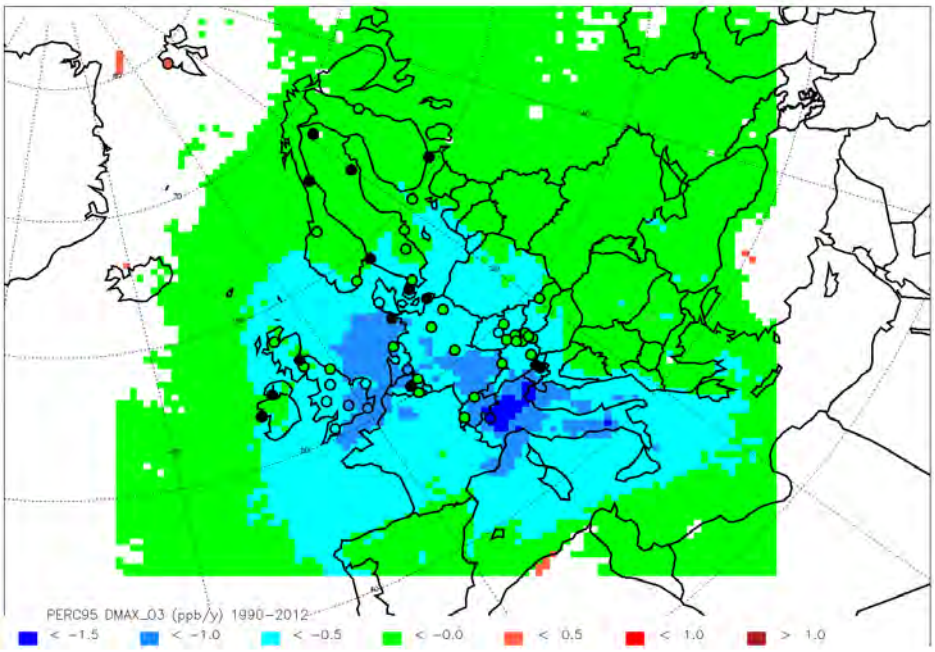

Figure 34: Sen's slopes (ppb year^{-1}) of the modelled annual 95 percentiles of the daily max ozone concentration for the period 1990-2012 with the results from EMEP monitoring data on top (circles). The colors indicate the magnitude of the trend. Filled, black circles indicate non-significant slopes. Note that the significance criteria for the stations were changed from p=0.05 to p=0.2 in this plot.

Using measured rural background EMEP data over 1990–2010, Tørseth et al. (2012) found a decrease in the highest levels and a corresponding increase in the low percentiles in the UK, Netherlands and some other sites, but no trends in Switzerland or Austria. These data were re-analysed in Simpson et al. (2014), showing that these changes from increasing ozone at low percentiles to decreasing ozone at high is very site-specific, but with the most dramatic changes being those at > 95th percentiles and hardly any change in the median ozone concentrations.

3.3.2. Ground level ozone concentrations and exposures (ICP Forests)

Tropospheric ozone (O_3) concentrations measured by passive samplers have been monitored according to harmonised methodologies on ICP Forests intensive monitoring (Level II) sites (Schaub *et al.,* 2010), starting in 2000. This method has been proven to be valuable at remote sites with sufficient accuracy (Sanz *et al.*, 2007; Gottardini *et al.*, 2010; Hůnová *et al.*, 2011). The objective of measuring the concentrations of O_3 is to (i) quantify O_3 concentrations over the course of the vegetation period (April-September), (ii) estimate the related O_3 exposures of forest ecosystems, and (iii) detect temporal and spatial trends across Europe. Here, we analysed the temporal and spatial trends for i) O_3 concentrations (reported as parts per billions, ppb) and ii) O_3 exposure (reported as O_3 Accumulated Over a Threshold of 40 ppb, AOT40) for the period 2000-2013.

The results given are based on 29,356 measurements from 203 sites in 20 countries (Figure 35). For quality assurance, only 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. Calculations of mean concentrations were weighted according to exposure time. Trend analysis was performed according to Sen (1968), using the R package (Bronaugh, 2013).

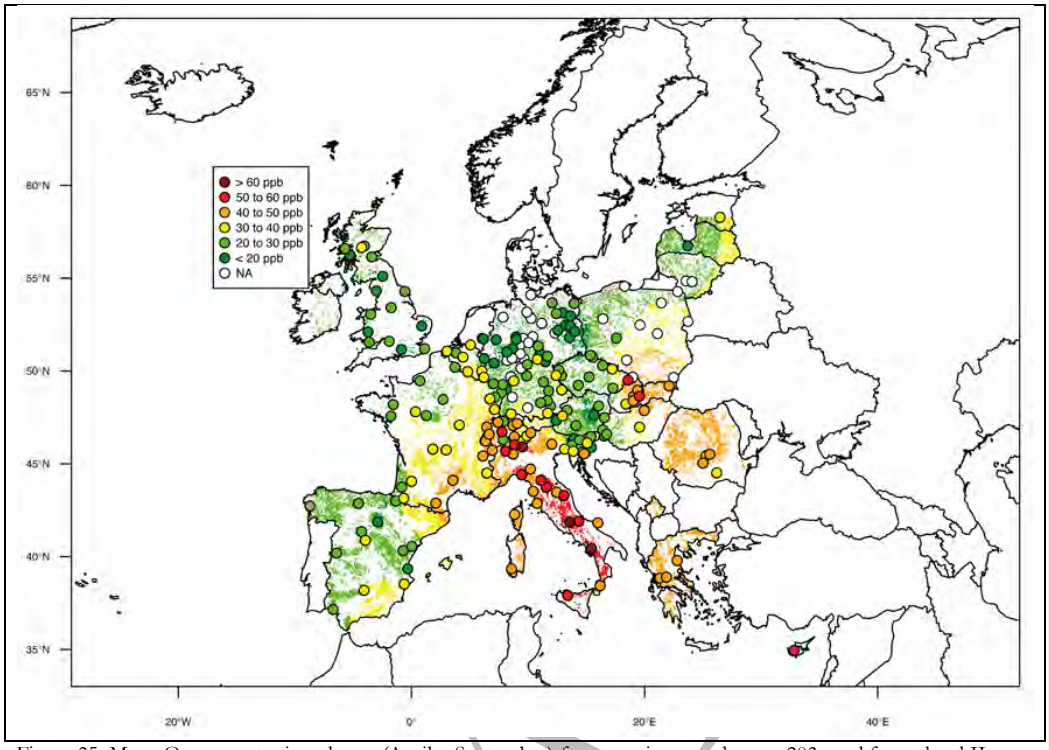

Figure 35: Mean O_3 concentration classes (April – September) from passive samplers on 203 rural forest level II plots during 2000-2013.

Between April and September mean O_3 concentrations ranged from 19 to 64 ppb. A decreasing south-north gradient is apparent with the highest concentrations being measured in Italy, southern Switzerland, Czech Republic, Slovakia and Greece (Figure 35). An overall trend analyses reveals a significant decrease of ca. 0.35 ppb per year (Figure 36). When considering only sites with a data coverage of at least 4 years and 120 days per year (66%), 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.* (2012a). 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 O_3 effects was exceeded on 75% of the plots. The trend analyses for 65 plots (in Germany, France, Spain, Switzerland and Italy) with a data coverage of at least 8 years for AOT40 revealed an overall decrease of 0.074 ppm h per year ($P < 0.1$), with a significant decline observed at some (primarily Italian) but not at the majority of sites (no change at 80% of the sites).

As summarized in Tørseth *et al.* (2012) and Simpson *et al.* (2014), a number of studies of tropospheric ozone trends have been published in the last years and a fairly consistent picture is found with a flattening or even reduction in the ozone levels, most pronounced in summer (Logan *et al.* 2012, Parrish *et al.* 2012, Derwent *et al.* 2013). However, these studies also demonstrate the complexity of modeling ozone concentrations. As noted by EEA (2009), the lack of long-term O_3 data is a major obstacle for identifying significant trends in Europe today, which underlines the great value of the ICP Forests data from in situ measurements for model validation and to detect long-term effects on forest ecosystems.

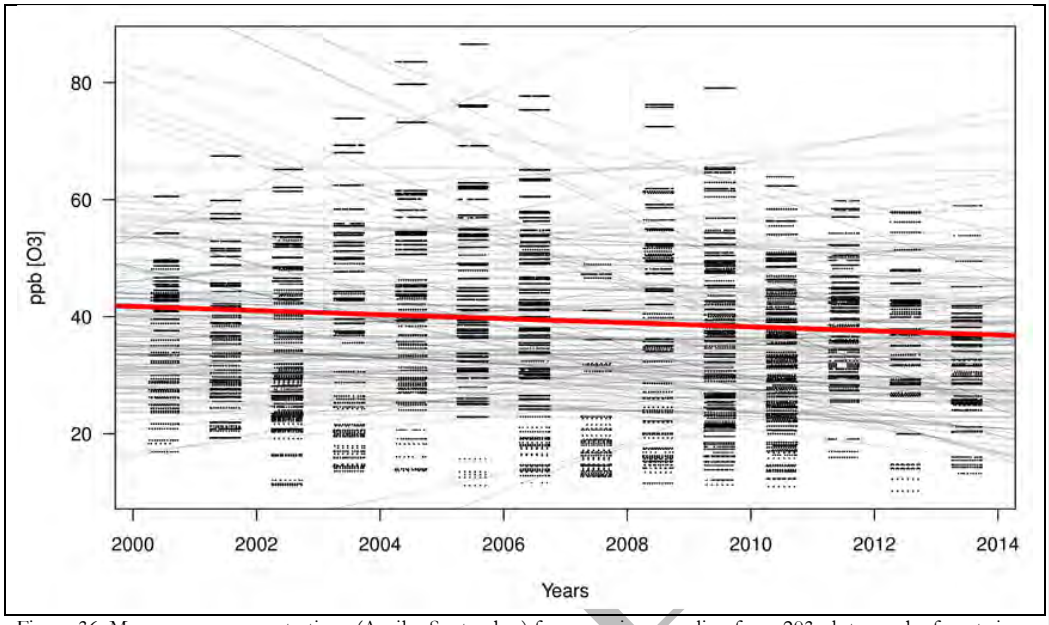

Figure 36: Mean ozone concentrations (April – September) from passive sampling from 203 plots nearby forests in 20 countries from 2000-2013, showing a significant decline of 0.35 ppb y-1 (n = 29,356; p = 0.000).

3.3.3. Trends in ozone concentrations, fluxes into leaf pores and effects (ICP Vegetation)

From seven ICP Vegetation sites in seven European countries, sufficient hourly ozone concentration data were available (for at least seven out of the twelve years) to assess trends in ozone metrics between 1999 and 2010. These sites were: Östad (Sweden), Ascot (UK), Tervuren (Belgium), Giessen (Germany) Seibersdorf (Austria), Pisa (Italy) and Ljubljana (Slovenia). The proportion of hourly ozone concentration in the lowest and highest ozone categories has declined ($p < 0.10$), whereas the proportion in the category 20 - 39 ppb has increased; the proportion in the category 40 - 59 ppb has not changed in the months June, July and August (Table 8). These results confirm the general trend observed across Europe, i.e. background ozone concentrations have risen whereas peak concentrations have declined (see 3.3.1).

Table 8: Trends (1999 – 2010) in ozone concentrations at ICP Vegetation monitoring sites during the months June, July and August.

Ozone concentration	European trend	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 Seibersdorf (AT)
≥60 ppb	Decline	Ljubljana (SI)

No temporal trends were found for the 24 hr mean and daylight mean ozone concentrations (Table 9). This is in agreement with trends reported for mean and median ozone concentrations at EMEP monitoring sites (Torseth et al., 2012; Simpson et al., 2014). However, night time mean and daily minimum ozone concentrations have increased (0.27 ppb per year) across Europe, although only significantly (at $p<0.1$) in Tervuren, Belgium. Despite a decline in the hourly ozone concentrations of 60

ppb or higher for the months June, July and August, the average European daily maximum ozone concentrations have not changed, although a decline was reported for Ljubljana, Slovenia. Whereas the 95th percentile of the hourly daylight ozone concentration showed for Ljubljana and Pisa, no trend was found at other sites, resulting in no significant European trend. In contrast, the 5th percentile of the hourly daylight ozone concentration showed a significant increase (0.38 ppb per year; p = 0.001), although such for Seibersdorf, Ljubljana and Pisa.

Table 9: 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).

[a] AOT 40 from seven sites, [b] POD3IAM from five sites (not including Ascot (UK) 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 as it takes into account the varying effects of climatic conditions, soil moisture and phenology on the amount of ozone taken-up by the leaf pores (LTRAP Convention, 2015; Mills et al., 2011). Despite a European decline in ozone concentrations of more than 60 ppb, AOT40 did not show any trend (Table 9, Figure 37) , 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).

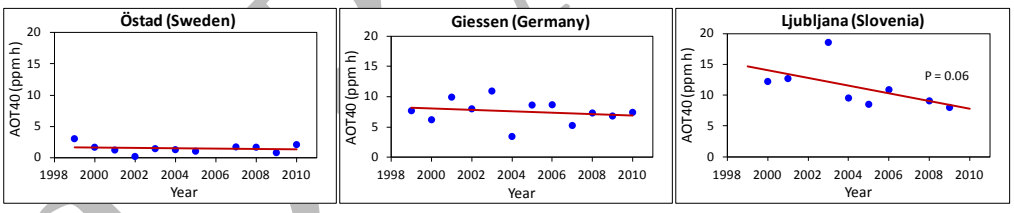

Figure 37: Trend in AOT40 at three selected sites in Europe. A significant trend (P = 0.06), a decline of 0.6 ppm h per year, was found only in Ljubljana (Slovenia).

Concentrations much lower than 40 ppb contribute to the ozone flux. The ozone flux into leaves showed no trend between 1999 and 2010 (Table 9), indicating that risk of ozone-induced effects on wheat has not changed (Figure 38). 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). In recent years (averaged over 1999 – 2010), modelled wheat yield was the least reduced (compared to pre-industrial 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.

[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).

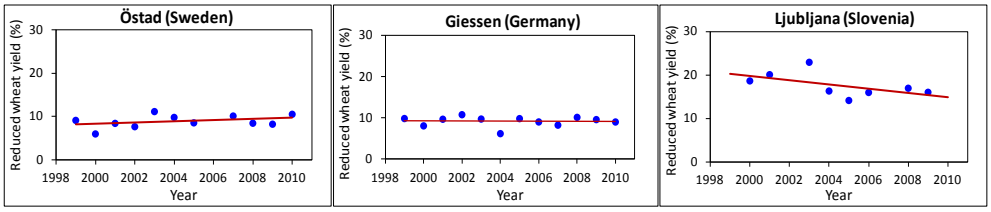

Figure 38: Reduction (%) in wheat yield due to ozone fluxes in recent years compared to pre-industrial ozone fluxes of a POD_3IAM of zero (corresponding to ozone concentrations in the range of 10 – 15 ppb), assuming no soil water limitation. No significant trends were found.

Future trends in ozone impacts on crop yield

When the revised Gothenburg Protocol scenarios (Amann et al., 2012) 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 39). 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 2030. Hence, crops will remain at risk of significant adverse effects of ozone in the near future.

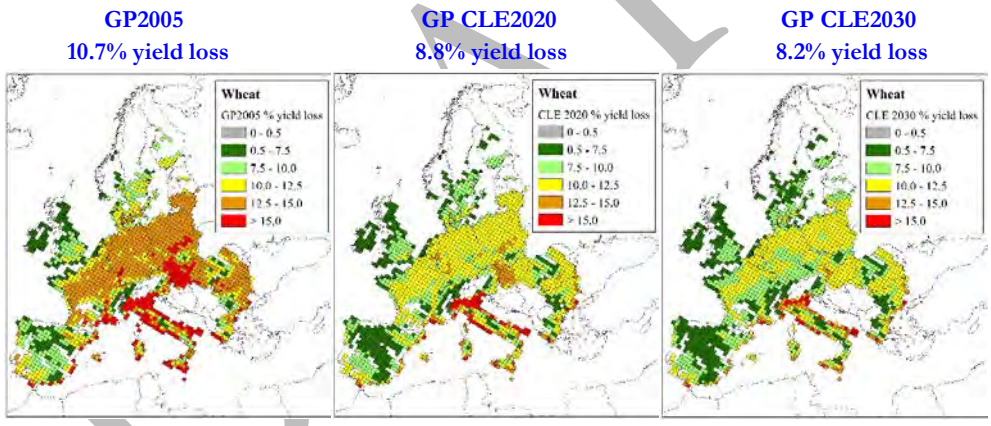

Figure 39: Calculated percentage yield losses due to ozone effects on wheat per 50 km x 50 km EMEP grid square in 2005, 2020 and 2030 for the wheat growing areas of the EU27+CH+NO for the base year (2005) of the revised Gothenburg Protocol (GP) and with implementation of its current legislation (CLE) 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).

3.3.4. Trends in population exposure and health responses of ozone (SOMO35) (TF Health)

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 PM. 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 (i.e. concentration below which no adverse health effects are observed) 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 the 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, and recommended based on the current state of scientific knowledge, 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.

Figure 40 shows the average levels for SOMO35 (in μg/m³ × days) for the most recent year of data available (2012) for the 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 ozone show that in 2012 in 28 countries, for which data were available, six presented values above 6000 mg/m3/days. Figure 41 shows the trend in SOMO35 levels between 2000-2002 and 2010-2012. At country level, there is no clear trend visible for the period of 2000–2012 in the WHO European Region Member States. 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%.

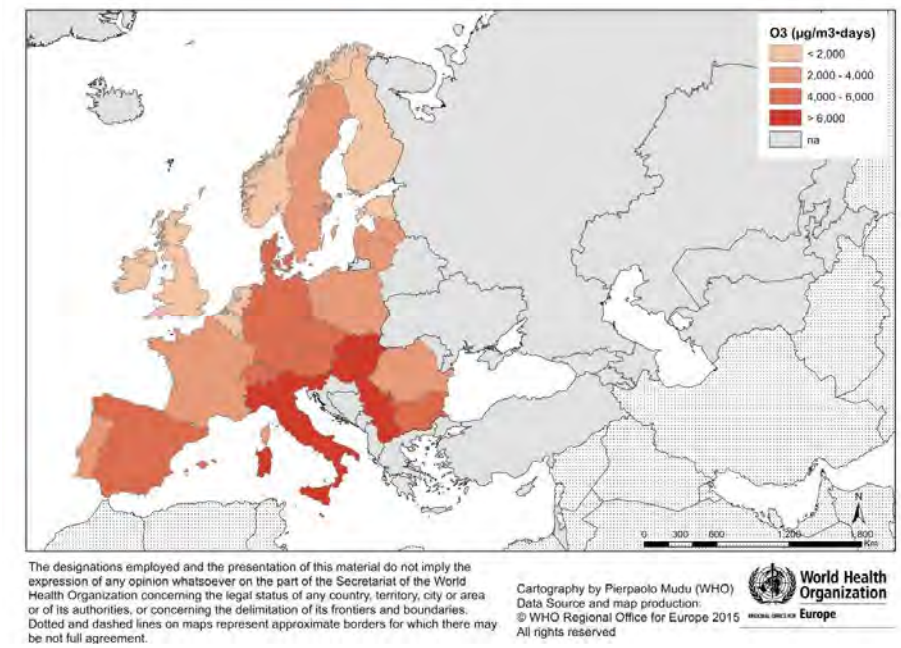

Figure 40: Population weighted annual mean SOMO35, 2012

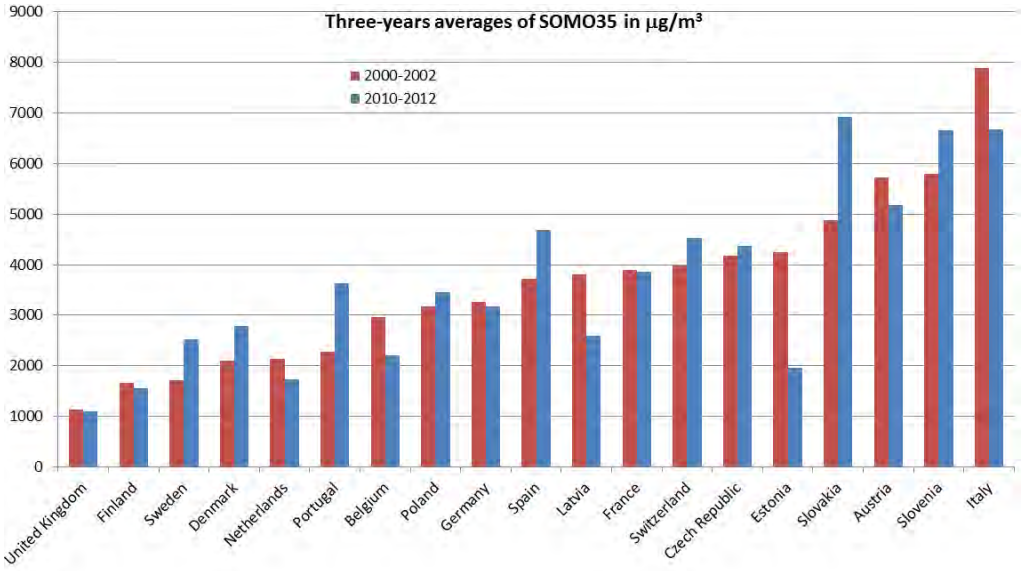

Figure 41: Population weighted annual mean SOMO35, three-year averages, for 2000-2002 and 2010-2012

3.4. PMs

3.4.1. Trends in concentrations (EMEP)

Aerosol mass has been part of the EMEP program for much shorter period than the sulphur and nitrogen components and a trend analysis is only possible for the last decade . In Figure 42, the observed and modelled average concentrations of PM_{10} and $PM_{2.5}$ since 2002 are presented, showing a decrease in both size fractions. Although the model underestimates the absolute concentrations, the trends in observations and model results are quite similar. About 50% of the sites show significant decreases (with Mann Kendall test, $p<0.05$) in both size fractions. The model and observations show similar decreases with 2.6% and 2.4% reduction per year for PM_{10}. There is a larger reduction in $PM_{2.5}$ due to the higher relative impact of anthropogenic emissions; the model shows a decrease of 3.5% per year versus 3.0% per year in the observations.

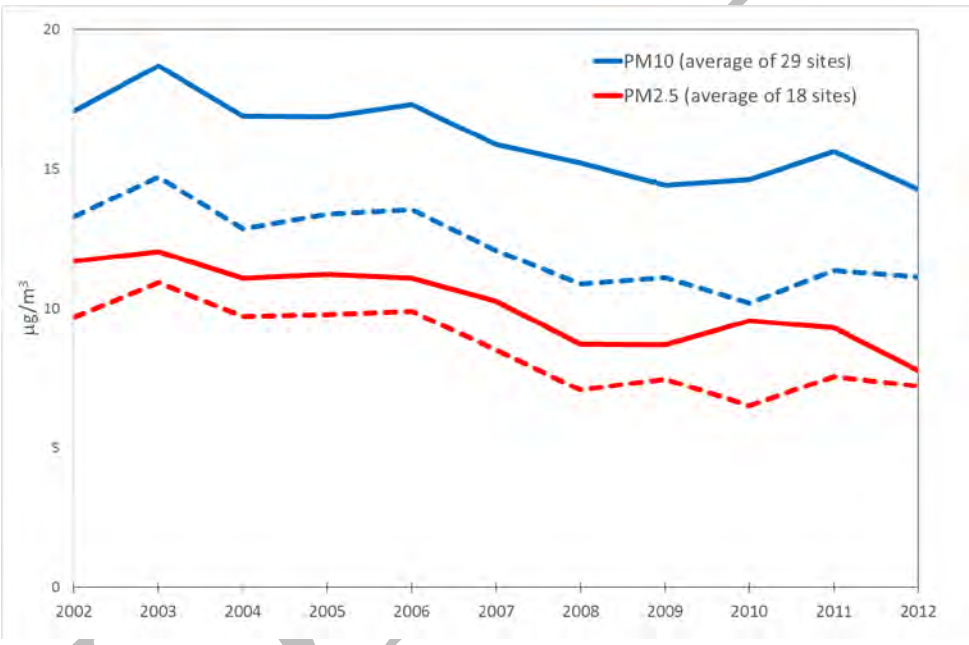

Figure 42: The observed and modelled annual average concentrations in PM_{10} and $PM_{2.5}$ at EMEP sites with measurements for at least 75% of the time period, 2002-2012. Solid lines are observations while the dotted lines are EMEP model results for the same sites.

3.4.2. Trends in health responses, PM2.5 and PM10 (TF Health)

PM is a mixture with physical and chemical properties that varies by location and time. The PM mixture also contains biological components, such as allergens and microbes. The health effects of PM 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 and respiratory symptoms; and an increase in hospital admissions and mortality from cardiovascular, cerebrovascular and respiratory diseases, and lung cancer (Loomis et al., 2013; WHO Regional Office for Europe, 2013).

Figure 43 and Figure 44 show the average levels of exposure to PM10 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 of background PM10 exposure in urban or suburban areas varied between 8.7 µg/m³ and 71.0 µg/m³. The most recent data on PM10 exposure shows that in 2012,

among 31 countries with available data, only 11 reported values below the WHO AQG value of 20 μg/m³. Variation in exposure levels of two- 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 μg/m³ to 50.4 μg/m³. The data for PM2.5 show that in 2012 (or the most recent year of data available), among 27 countries for which data were available, only seven reported values below the WHO AQG value of 10 μg/m³. In European cities where PM is monitored, 75.4% and 94.0% of the population experienced in 2012 annual average levels exceeding the WHO AQG for PM10 (20 μg/m³) and PM2.5 (10 μg/m³), respectively (WHO Regional Office for Europe, 2015a). For 28.6% of the urban residents, the EU limit value for PM10 (40 μg/m³) was exceeded in 2012. The exposure levels for PMs document a substantial risk to health.

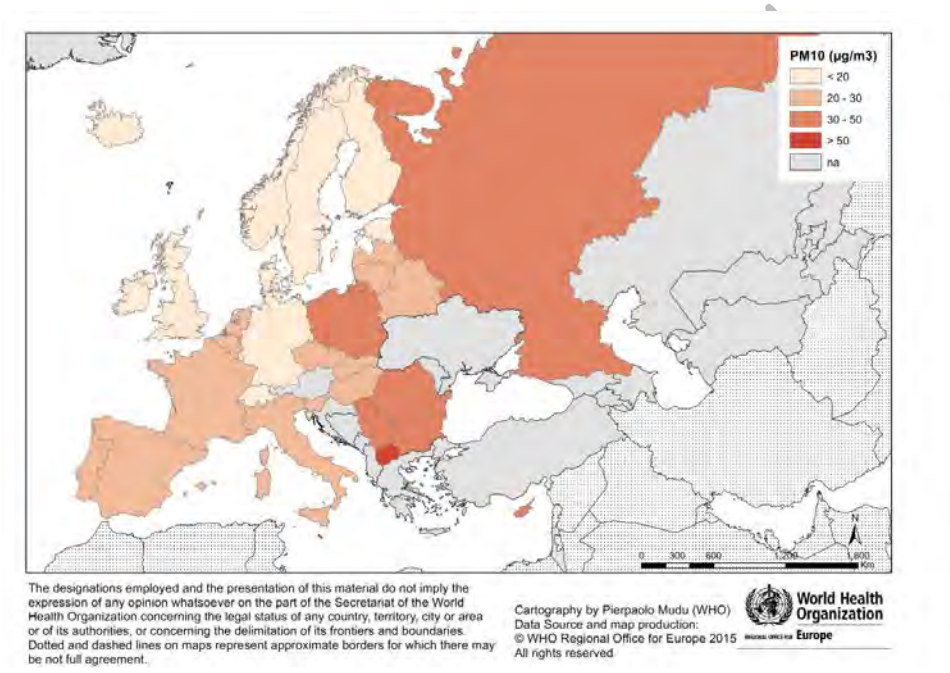

Figure 43: Population weighted annual mean PM10, for 2012 or the last year available

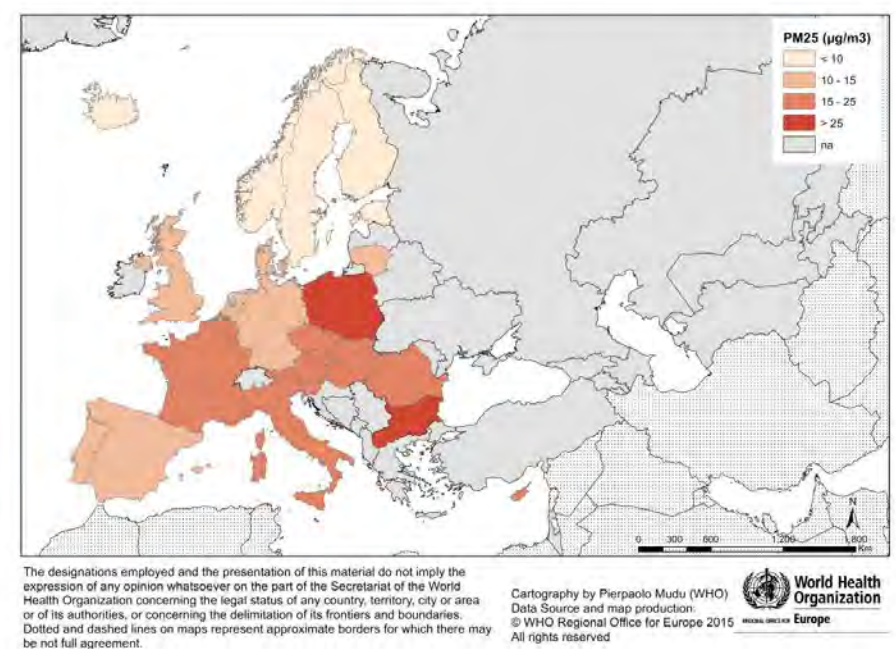

Figure 44: Population weighted annual mean $PM_{2.5}$, for 2012 or the last year available

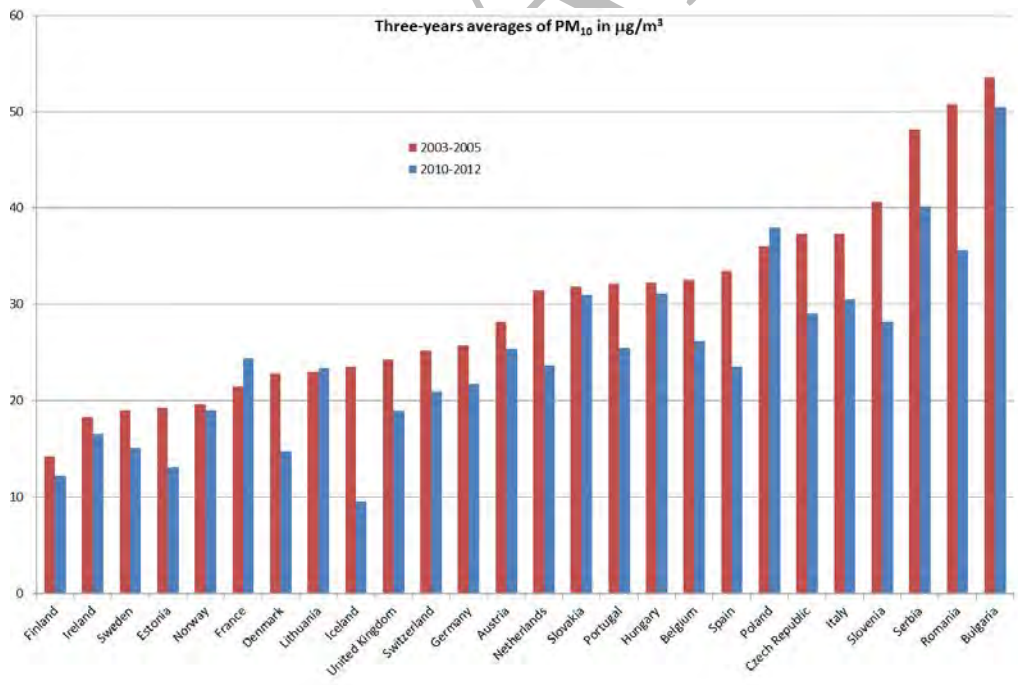

Figure 45: Population weighted annual mean PM_{10}, three-year averages, for 2003-2005, and 2010-2012.

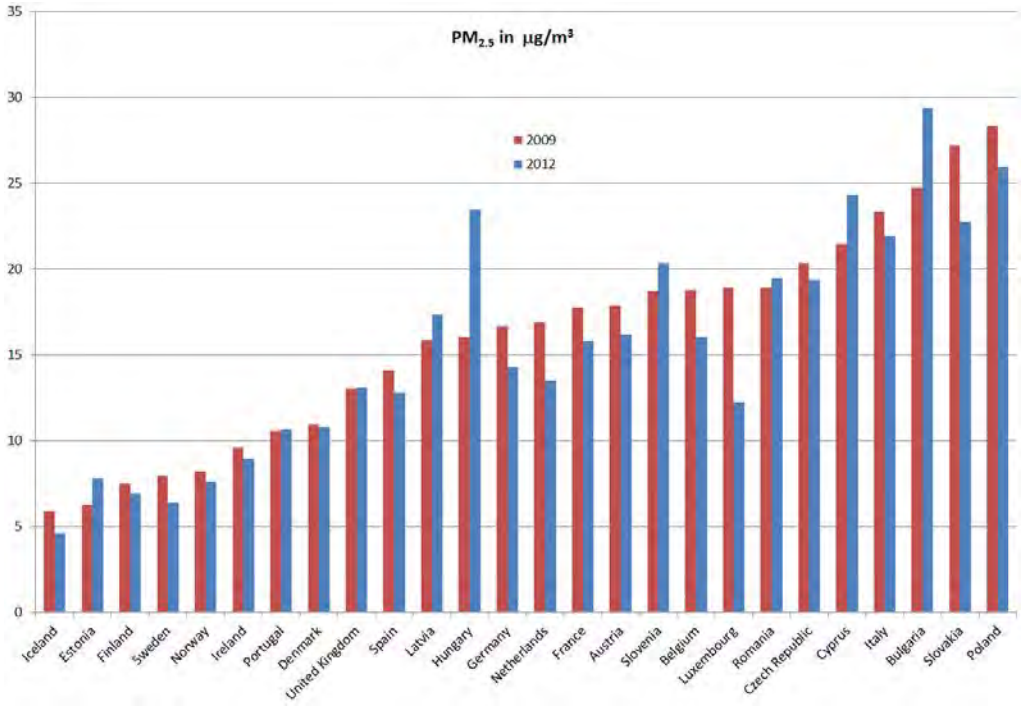

Figure 46: Population weighted annual mean PM_{10}, three-year averages, for 2003-2005, and 2010-2012.

Figure 45 and Figure 46 show trends in population exposure for three-year averages, for PM10 and PM2.5, respectively. In general, the population-weighted average exposure to PM2.5 in all cities of the European Region has not changed substantially, , but a decreasing trend can generally be noted for PM10. However, it is important to note that the number of monitoring stations has increased over the years, especially for PM2.5. In 2012, PM10 and PM2.5 data from regular population-relevant (urban background, residential, commercial and mixed areas) monitoring were available, respectively, for 479 cities in 30 countries and 300 cities in 26 countries.

3.4.3. Trends in soiling (ICP Materials)

Evaluation of soiling of modern glass started with the first trend exposure performed in 2005-2006. Two subsequent trend exposures were carried out in 2008-2009 and 2011-2012. The range of haze variation measured is similar for the three exposure campaigns (Figure 49). 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 has increased significantly in Bottrop, Casaccia, Venice, Stockholm and Katowice. It has decreased for Prague, Milan, Paris and Vienna and has remained constant in Kopisty, Rome, Oslo, Birkenes, Aspvreten, Madrid, Toledo, Lahemaa, and Athens. For the other sites, it is difficult to observe a trend: haze increased from 2005-2006 to 2008-2009 but decreased for the last exposure in Berlin and Svanvik, whereas it decreased from 2005-2006 to 2008-2009 and increased in 2011-2012 for Chaumont. In summary, when considering all the sites, haze does not show a clear trend. The majority of sites display similar haze values during the three campaigns.

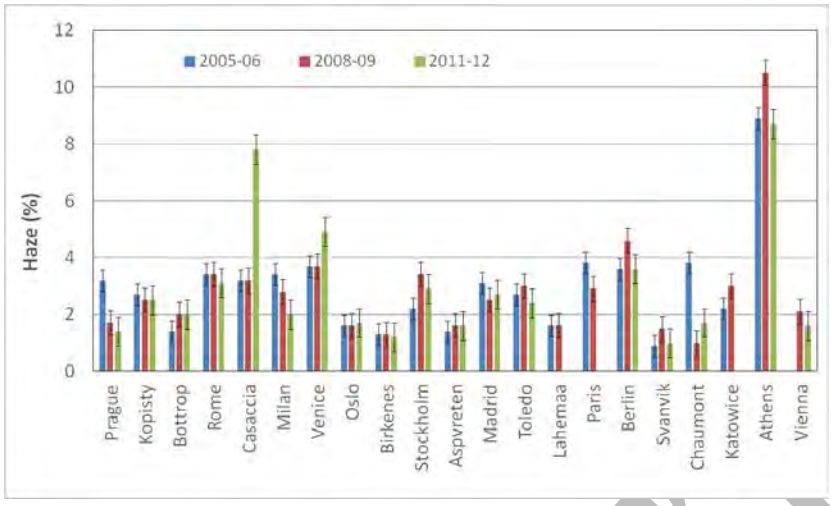

Figure 47: Comparison of Haze (in %) during the three extensive campaigns (2005-2006; 2008-2009; 2011-2012).

Figure 50 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% is considered to correspond to the threshold where the deposit leads to visual nuisance detected by human eyes. Figure 50 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). The input 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. There is a clear difference between polluted and non-polluted areas.

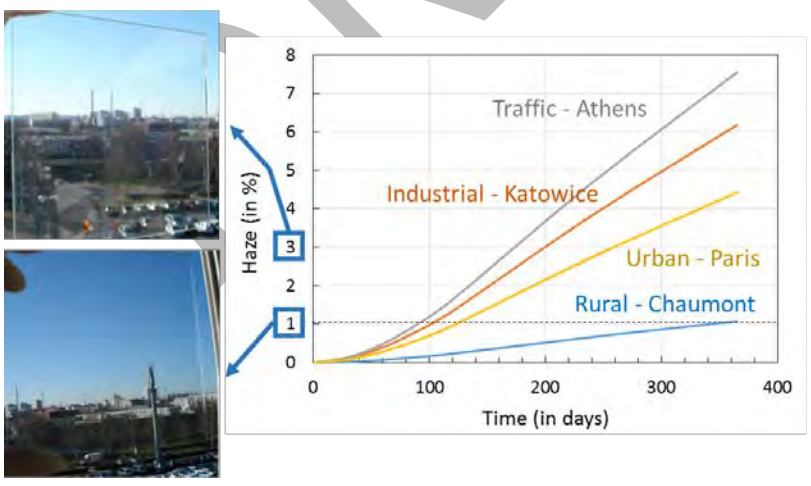

Figure 48: Predicted haze (in %) as a function of time for 4 types of sites: rural, urban, industrial and traffic. Input environmental data for the 2008-2009 campaigns were selected (Lombardo et al., 2014). Visual aspects are shown on 2 pictures: Katowice after 1 year (haze = 3%), upper image and Chaumont (haze = 1%), lower image.

3.5. Heavy metals (ca 5-10 p)

3.5.1. Trends in deposition of heavy metals in the EMEP region (EMEP)

Long-term trends of total deposition and air concentrations of lead, cadmium and mercury were analyzed for the EMEP region and for particular countries for the period from 1990 to 2012. The methodology of the trend analysis was elaborated by MSC-E (https://wiki.met.no/emep/emep-experts/start). Reduction of deposition and concentrations occurred at different rates for the period, therefore the trends were non-linear.

The main characteristics of long-term trends of deposition in the EMEP region are summarized in Table 10. As demonstrated, lead was characterized by the highest, and mercury by the lowest magnitude of total reduction and average rate of reduction, respectively. Rates of deposition reduction of lead and mercury were higher in the beginning and lower in the end of the two-decade period, while the relative rate of cadmium annual decline remained constant.

Table 10. Main characteristics of long-term deposition trends of lead, cadmium and mercury for the period 1990-2012 in the EMEP region

Heavy metal	Type of deposition	Total reduction, %	Average reduction, % per year	Maximum reduction, % per year	Minimum reduction, % per year
Pb	Averaged for EMEP	78.3	6.4	6.8	5.9
	Forests*	80.7	7.2	7.7	6.9
	Natural**	70.6	5.4	5.9	4.5
Cd	Averaged for EMEP	52.8	3.2	3.2	3.2
	Forests*	57.2	3.8	3.8	3.8
	Natural**	37.9	2.1	2.1	2.1
Hg	Averaged for EMEP	23.4	1.2	3.5	0.4
	Forests*	23.4	1.2	3.3	0.4
	Natural**	15.6	0.8	2.9	0.2

* Temperate coniferous forests, temperate deciduous forests, Mediterranean needleleaf forests, Mediterranean broadleaf forests; ** - Non-forested natural surfaces (grasslands, semi-natural surfaces, wetlands, tundra and inland waters)

Time series of deposition were not smooth (Figure 51). Irregularities of time series were explained by annual variability of meteorological conditions and emission sources.

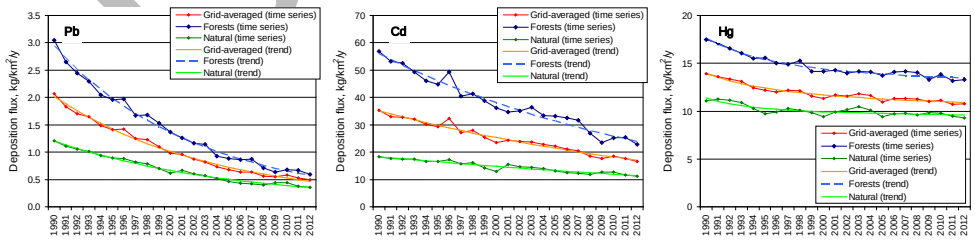

Figure 49: Deposition fluxes of lead (left), cadmium (middle) and mercury (right) for 1990-2012

Long-term trends of total deposition were linked with long-term tendencies of emissions. The reduction of anthropogenic emission of lead and cadmium for the period 1990- 2012 was 90% and 60%, respectively (EMEP trend report, in preparation). However, total deposition reduction to the EMEP

region was smaller (Table 10). This is because of significant contribution of natural emission and re-suspension of previously deposited metals (so-called secondary sources) to deposition in the EMEP region. Reduction of the sum of anthropogenic and secondary emission sources within the region made up 80% for lead and 57% for cadmium, which was very close to the reductions of deposition, because most of lead or cadmium emitted from the EMEP region was deposited within the region.

The reduction of mercury deposition in the region was lower (23%) compared to that of lead and cadmium. The main reason for this is hemispheric transport of mercury across the globe, resulting in a considerable contribution of from other continents to mercury deposition in Europe (Travnikov et al., 2012). This contribution expressed in absolute terms did not change much over the period (Figure 52).

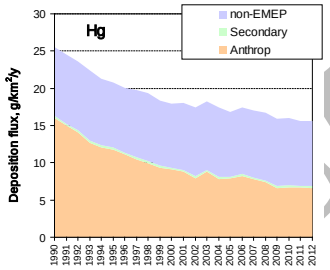

Figure 50: Mercury deposition fluxes caused by anthropogenic and secondary sources within EMEP region and by non-EMEP sources

For grid cells where background measurement stations are located, trends could be analysed based on both observed and modelled information. On average, mean annual reduction of observed and modelled concentrations and wet deposition amounted to about 7-9% for lead and about 4-8% for cadmium (Figure 53). Corresponding values of total reduction related to the entire period, were 80-87% for lead and 60-84% for cadmium. Reduction of modelled and measured mercury air concentrations was low (less than 0.5%) (Figure 53). Decline of observed mercury wet deposition was higher (about 3% per year) than that of modelled levels. However, the range of reductions of observed mercury deposition was quite high and fully overlapped the range for modelled values.

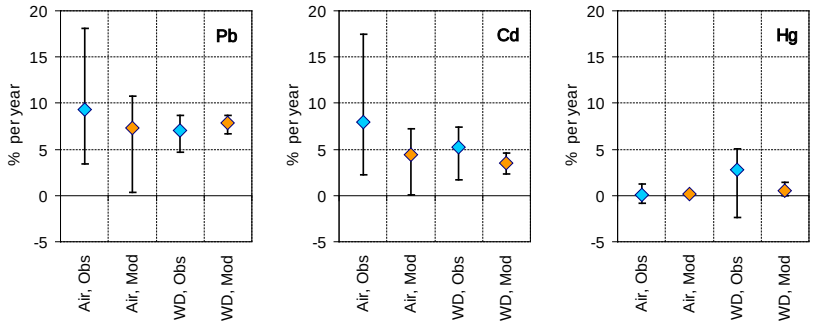

Figure 51: Mean annual reduction rates for modelled and observed air concentrations and wet deposition fluxes of lead (left), cadmium (middle) and mercury (right) over selected EMEP monitoring stations. Whiskers mean range of average reduction rates among stations

3.5.2. Temporal trend in heavy metal concentrations in mosses (ICP Vegetation)

In 2010, the lowest concentrations of metals in mosses were generally found in northern Europe, whereas the highest concentrations were observed in (south-)eastern Europe (Harmens et al., 2015). Since 1990, the average median 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 copper (11%), as shown in Figure 54 and Figure 55. For lead and cadmium, the decline is similar to that reported by EMEP for the modelled deposition across Europe, i.e. 74% and 51% respectively (Figure 54). The 14% decline in mercury between 1995 and 2010 was lower than the decline (27%) in EMEP modelled deposition across Europe.

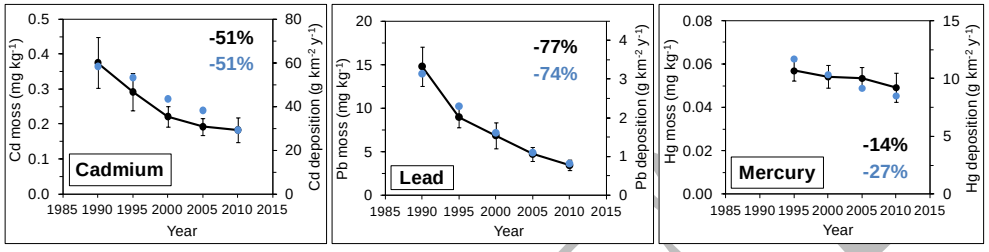

Figure 52: Average median metal concentration (± one standard error) in mosses for countries that reported metal data for at least four survey years since 1990; some countries reported three survey years since 1995 for mercury (Harmens et al., 2015). The blue dots in the graphs show the decline in deposition across Europe as modelled by EMEP (Travnikov et al., 2012). The black and blue values in the graphs represent the percentage change (i.e. a decline) between 1990 (1995 for mercury) and 2010 for the moss concentrations and EMEP modelled depositions respectively.

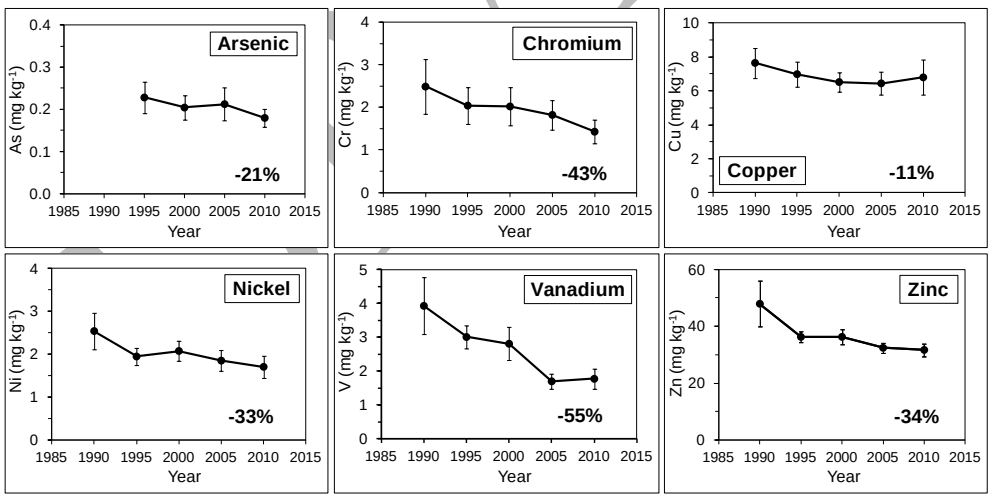

Figure 53: Average median metal concentration in mosses (± one standard error) for countries that reported metal data for at least four survey years since 1990 (1995 for arsenic; Harmens et al., 2015). The values in the graphs represent the percentage change (i.e. a decline) between 1990 (1995 for arsenic) and 2010.

3.5.3. Temporal patterns in soil and streamwater mercury, lead and cadmium (ICP IM)

Metals in soil and stream water in remote ecosystems 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). Temporal changes in metal concentrations were tested in soil and stream water at European IM-sites. At Swedish ICP IM sites (Aneboda, Gårdsjön, Kindla and Gammtratten) soil and stream water samples have been collected with regular intervals over the last decades and there metal concentrations were tested for temporal trends in more detail.

Time-trends in soil metal concentrations

European data

For soil chemistry, Sweden excluded, temporal changes (1994 – 2011) at each ICP IM site and soil depth (litter+organic layer, 0-5 cm depth, 10-20 cm depth, 30-50 cm depth, 50- cm depth) were attributed with a symbol indicating either increases (+) or decreases (-) in metal concentration over time. The proportion in the number of increasing changes (+) and decreases (-) were tested by the binomial z-test and equal proportions indicated no change. In the forest floor layer (litter + organic topsoil) both increases and decreases could be detected for Hg, Pb or Cd (Table 10). In the upper mineral soil layer (0-5 cm), significant increases were observed for Pb over time while the temporal change for Cd and Hg was weaker. There were more indications of increasing than decreasing trends on Cd in deeper mineral soil layers, however not significant.

Table 11: Temporal trends[a] 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.

Soil layer, cm	Hg[b]		Pb[b]		Cd[b]	
	+	-	+	-	+	-
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**

a Number of sites for each element and soil layer adds up to the sum of + and - for each element.
b the significance of temporal trend in soil metal concentrations is based on the proportion of +/- tested with the binomial z-test (Decrease: proportion ≤ 0.5, increase: proportion > 0.5). Significant trends are indicated in bold numbers.

Swedish soil data

For Swedish ICP IM sites Pb and Cd concentrations in the forest floor (F+H-horizons) decreased between 1994 and 2011 in sites Aneboda, Gårdsjön and Kindla (Kindla only Pb)(Figure 56). Mercury concentrations, on the other hand increased over the same period. In mineral soil layers, a general increase were observed for all metals (Figure 56). No significant changes in Cd concentrations could be seen at any site in the lower parts of the mineral soil.

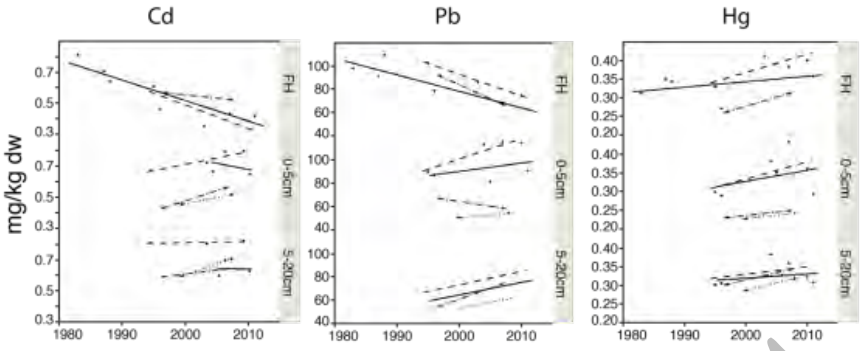

Figure 54: Soil metal concentrations at the four ICP IM sites in Sweden (Aneboda (solid line) , Gammtratten (dotted line), Gårdsjön (dashed line), Kindla (dotdashed line)).

Time trends in stream water metal concentrations

For Swedish IM sites almost 20 year records of Pb, Cd and Hg concentrations could be evaluated (Figure 57). 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.

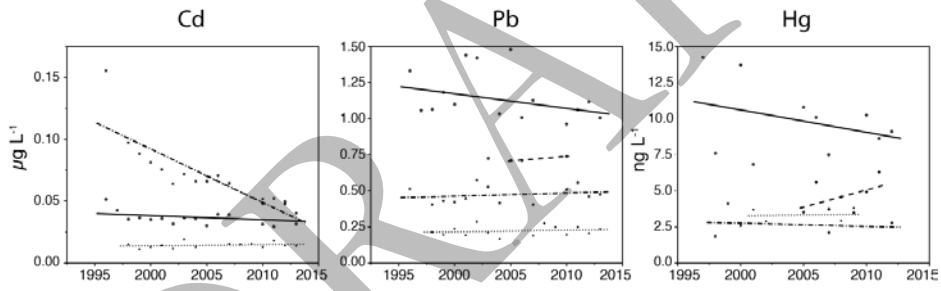

Figure 55: Stream water Cd and Pb (µg L^{-1}) and Hg (ng L^{-1}) concentrations at at the four ICP IM sites in Sweden (Aneboda (solid line) , Gammtratten (dotted line), Gårdsjön (dashed line), Kindla (dotdashed line)).

The main conclusion from the monitoring data on Pb, Cd and Hg in soils and streamwaters are the following. Soil Pb and Cd concentrations in the forest floor showed decreases, implying that deposition of these elements has been reduced. However, the increases of Pb and Cd in deeper soil layers show that deposited elements now are being transferred to deeper soil layers, and indicate that accumulation is ongoing considering the total soil profile. Concentrations of Hg increased in the forest floor, with potentially hazardous effects on biological activity, increased potential for methylation and transport to surface waters. Concentrations of Hg in deeper soil layers have increased, demonstrating continued soil accumulation.

Streamwater concentrations of Cd, Pb and Hg showed few trends, except for Cd where declines were found at two sites. Heavy metals continue to leach from soils to surface waters, with no clear relation to trends in deposition.

3.5.4. 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. An almost 20% decline in Hg in fish was found for Swedish freshwater fish for the period 1965-2012, without a consistent regional pattern (Akerblom et al. 2014). However, from the 1990s and a decade onwards there is evidence of increases in concentrations of Hg in freshwater fish 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.

3.5.5. Trends in exceedances of CLs (ICP M&M - CCE)

The area and magnitude of the exceedances in 1990 and 2010 of critical loads of cadmium (Cd), lead (Pb) and mercury (Hg) are shown in Figure 58, Figure 59 and Figure 60, respectively. The trends of these exceedances between 1990 and 2010 are shown in Figure 61. The maps and trends are related to critical loads addressing effects on ecosystems and human health, the latter related to the protection of drinking water and food quality (Table 12, effect nr 1 to 4).

Table 12: Overview of indicators used in the computation of critical loads (adapted from chapter 5.5 in the Modelling and Mapping Manual; www.icpmapping.org).

Receptor Ecosystem	*Endpoints*	*Heavy metals of concern*	*Land cover types to be considered*	*Indicator/ critical limit*	*Effect number**
Terrestrial	*Human health effects*	Cd, Pb, Hg	All ecosystems	Total concentration in soil water below the rooting zone(to protect ground water)	1
		Cd	Arable	Content in food, fodder and crops	2
		Cd	Grassland	Content in grass and animal products (cows, sheep)	
	Ecosystem functions	Cd, Pb	Arable land, grassland, non-agricultural	Free ion concentration in view of effects on soil micro-organisms, plants and invertebrates	3
		Hg	Forest soils	Total concentration in humus layer in view of effects on soil micro organisms and invertebrates	
Aquatic	*Ecosystem functions*	Cd, Pb	Freshwaters	Total concentration in view of effects on algae, crustacea, worms, fish, top predators	4
	Human health	Hg	Freshwaters	Concentration in fish	5

Source: Hettelingh et al., 2015a

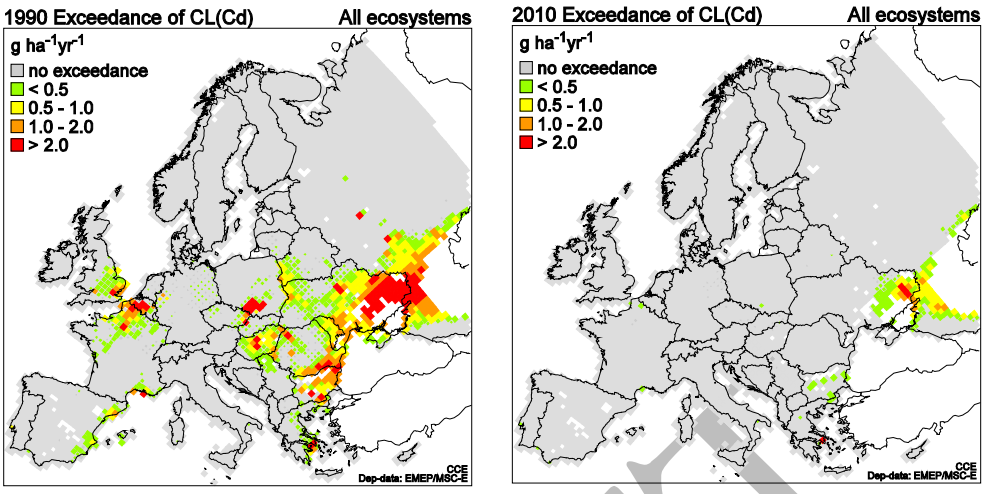

Figure 56: Exceedances (AAE) of the critical loads of cadmium in 1990 (left) and 2010 (right) in Europe. Note that the size of the colour shading in a grid cell indicates the % of the ecosystem area at risk. The ecosystem area can vary widely among grid cells. (Deposition data from EMEP-MSCE; critical loads data from the CCE)

Safe levels of mercury in fish for human consumption are not considered in these assessments, because other approaches are needed for such risk assessments (see modelling and mapping manual (LRTAP Convention, 2015). Critical concentrations of Hg in rainwater related to critical limit in fish (Table 12, Effect number 5) have been exceeded in assessments made for 2000 in grid cells located in northern Europe in particular in nearly grid cells in the year 2000 (Slootweg et al. 2007), but trends have not been assessed since.

The area at risk of atmospheric deposition of Cd exceeding its critical load occurred in many countries in 1990 and was limited to scattered regions in 2010 mostly in the eastern part of Europe. However, it should be noted that Cd inputs from agricultural practices are not included in these calculations. Limiting computations of the risk of Cd to that caused by atmospheric deposition alone reveals a trend of ecosystem area at risk fom about 4% in 1990 to close to nil in 2010. The trend in AAE magnitudes of critical load exceedance also converge to nil in 2010 (Figure 58, 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 in 2010 (Figure 59). The trend of the exceedances since 1990 moves in a downward direction from about 67% to about 20% of the ecosystem area with an exceedance of about 2 g ha^{-1} yr^{-1} in 2010 (Figure 61, centre top and bottom graphs respectively) in 2010.

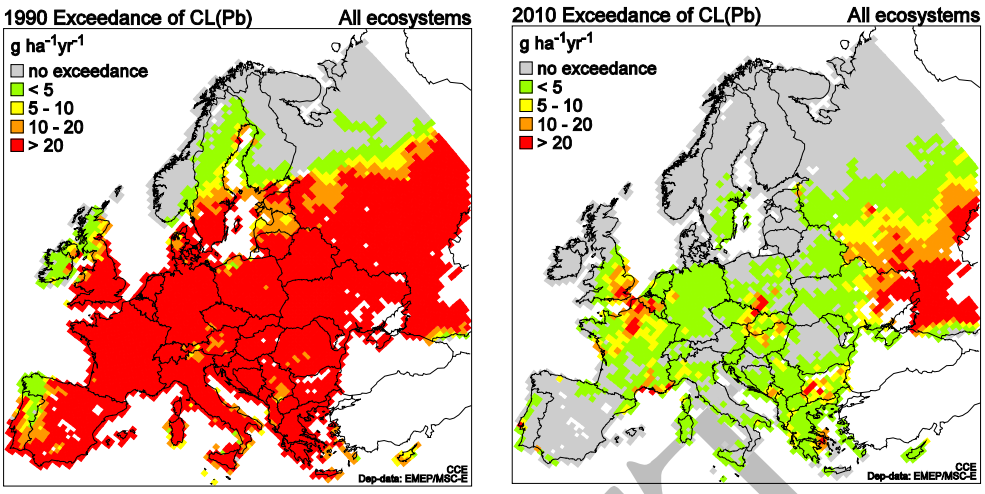

Figure 57: Exceedances (AAE) of the critical loads of lead in 1990 (left) and 2010 (right) in Europe . Note that the size of the colour shading in a grid cell indicates the % of the ecosystem area at risk. The ecosystem area can vary widely among grid cells. (Deposition data from EMEP-MSCE; critical loads data from the CCE)

Figure 60 and Figure 61 confirm that excessive deposition of mercury remains an issue of concern in 1990 as well as in 2010 with about 69% and 56% of the ecosystem area at risk of exceedance of the critical loads of Hg respectively.

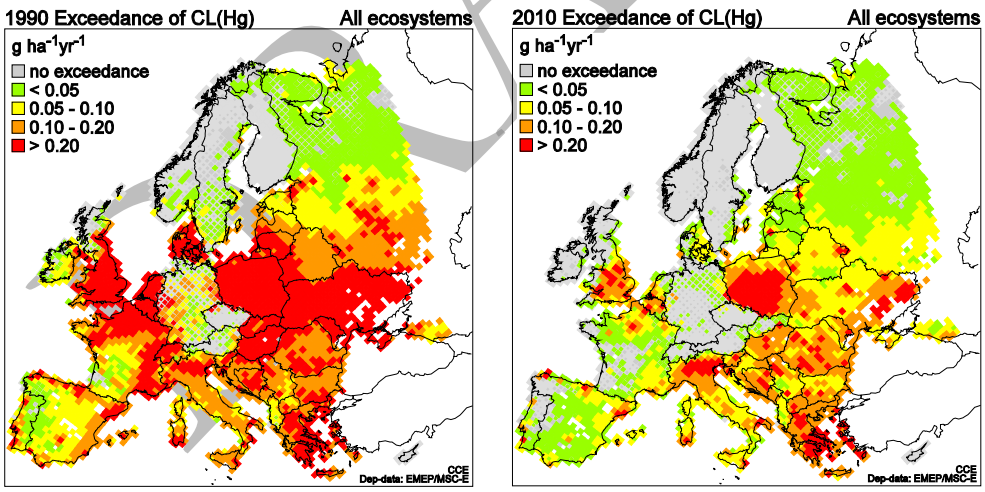

Figure 58: Exceedances (AAE) of the critical loads of mercury in 1990 (left) and 2010 (right) in Europe Note that the size of the colour shading in a grid cell indicates the % of the ecosystem area at risk. The ecosystem area can vary widely among grid cells. (Deposition data from EMEP-MSCE; critical loads data from the CCE).

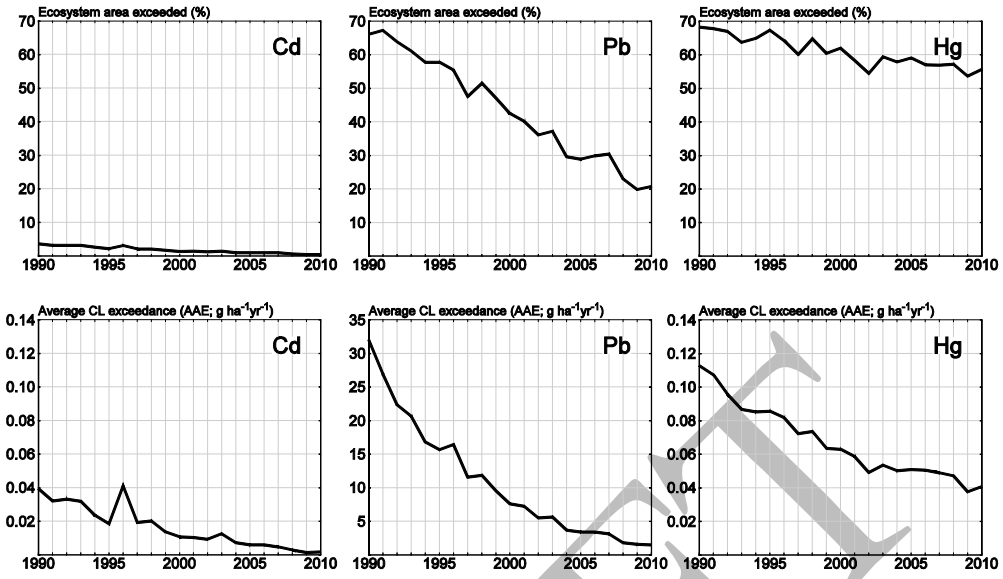

Figure 59: Temporal development of the area (top; % of ecosystem area) where critical loads of cadmium (left), lead (centre) and mercury (right) are exceeded and the magnitudes of the average accumulated exceedance (AAE) (bottom; g ha^{-1} yr^{-1}) of the critical loads of these metals, respectively (Deposition data from EMEP-MSCE; critical loads data from the CCE)

Figure 61 shows that the average critical load exceedance varies over time. The deposition of heavy metals, used in the evaluation of critical load exceedances and areas at risk, is calculated for each year separately between 1990 and 2010. These deposition calculations are performed on the base of emission data and meteorological information for particular years. Meteorological conditions in each year are not the same: precipitation amounts, wind patterns, temperature and other parameters vary from year to year around long-term climatic mean values. This variability of meteorological conditions affects long-term simulations of pollution levels of heavy metals, resulting in irregularities in the downward trend of pollution levels and exceedances over the EMEP region. Additionally, these irregularities can be higher in particular areas of the EMEP domain (e.g., countries, provinces etc.) compared to those obtained for the entire EMEP region.

3.6. POPs

3.6.1. Trends in concentrations and deposition of POPs in the EMEP region (EMEP)

Evaluation of trends in atmospheric POP pollution levels in the EMEP region was performed for the period from 1990 to 2012 (Gusev et al., 2015). Long-term changes of air concentrations and deposition fluxes of B[a]P (B[a]P belongs to PAHs and is used as an indicator PAH), PCDD/Fs, PCB-153 (indicator PCB congener), and HCB were analyzed using the methodology elaborated by MSC-E (https://wiki.met.no/emep/emep-experts/start). The proposed methodology allows singling out main and seasonal components of the considered time series of pollution levels. Both measured and modelled values of atmospheric POP pollution levels were used in the trend analysis. In particular, for the places, where the monitoring stations were located, modelled and measured data were used in combination. For other parts of the EMEP region trends in pollution levels were characterized entirely by the modelling.

Analysis of temporal variation in pollution levels, based on the modelling results showed that maximum reduction of pollution in the EMEP region was for HCB (over 90%), followed by PCB-153 and PCDD/Fs. The lowest decline, about 30%, was estimated for B[a]P. For all considered POPs, maximum changes occurred in early 1990-s, whereas after 2000 the rates of pollution reduction declined or almost leveled off (Figure 60).

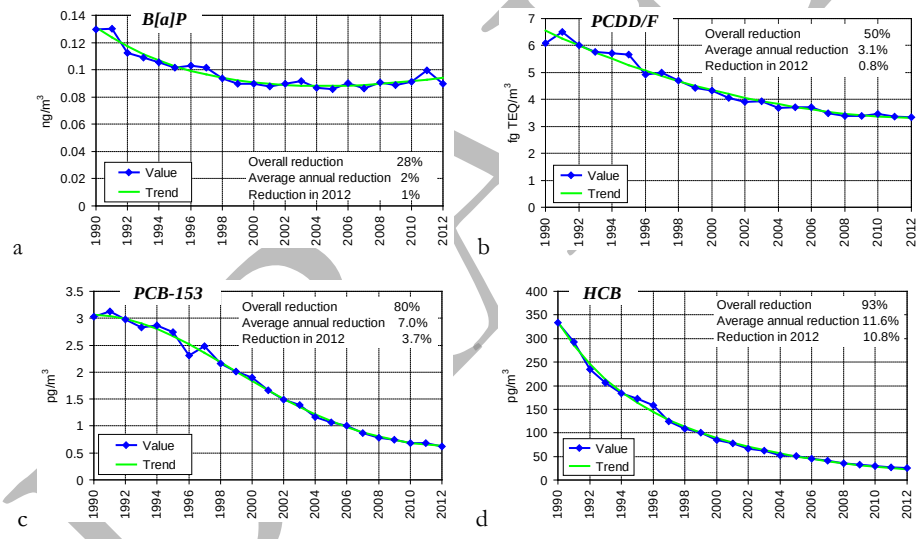

Figure 60. *Trends in annual mean air concentrations of (a) B[a]P, (b) PCDD/Fs, (c) PCB-153 and (d) HCB in the EMEP region for the period from 1990 to 2012. Negative reduction denotes increase of air concentrations. Green line indicates estimated trend and blue line – annual mean air concentrations. Reduction rates refer to main trend component. (Source: Gusev et al., 2015)*

Evaluation of trends in B[a]P air concentrations indicated that for most of the EMEP countries, the decline in pollution levels until the year 2005 was changed to an increase, which is also confirmed by measurements of the EMEP monitoring sites. A similar tendency can also be seen in temporal variations of B[a]P deposition fluxes to various ecosystems that is

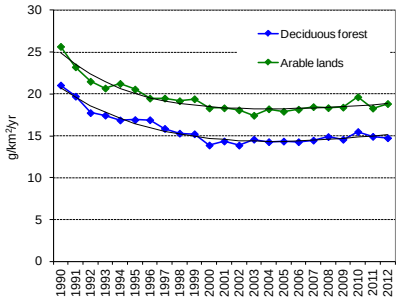

Figure 61: *Trends of B[a]P deposition to deciduous forest and arable lands within the EMEP region (Source: Gusev et al., 2015).*

important information for the evaluation of harmful effects of POP pollution.
As seen from Figure 61, reduction of B[a]P deposition to deciduous forests and arable lands is estimated to about 25% for the period 1990 – 2012. However, the trend switched from downward to upward around 2005. The increase in deposition fluxes was found to be statistically significant at 90% confidence level.

Seasonal variability of concentrations for some POPs is an important characteristic in the evaluation of long-term trends. In particular, intra-annual variations of PAH concentrations can reach an order of magnitude that needs to be considered in course of trend analysis. The example of B[a]P air concentration trend in the EMEP region is shown in Figure 62. Due to substantial seasonal variability, B[a]P air concentration in winter can exceed several times the annual average concentration, which needs to be taken into account in the evaluation of exposure.

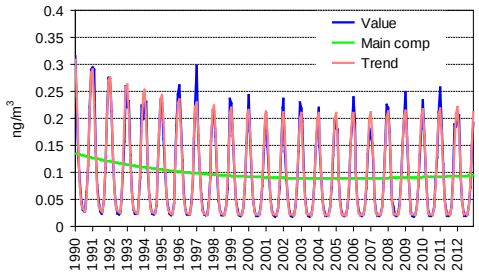

Figure 62: *Trend of monthly mean B[a]P air concentrations in the EMEP region for the period from 1990 to 2012.* ***(Source: Gusev et al., 2015).***

Trends of POP air concentrations in the EMEP region were also analyzed for the particular places where the monitoring sites were located. Comparison of estimates of trends for B[a]P, HCB and PCB-153, performed on the basis of model data and measurements, shows that model predictions generally capture the observed long-term tendencies in changes of pollution levels.

POP air concentrations in the EMEP region significantly varied between and within countries. Particularly, modelling results show that in spite of generally low annual mean B[a]P air concentrations, there are areas in some of the EMEP countries where EU target value for B[a]P (equal to 1 ng/m³ (Directive 2008/50 EC)) are exceeded. The number of people living in these areas is shown in Figure 63.

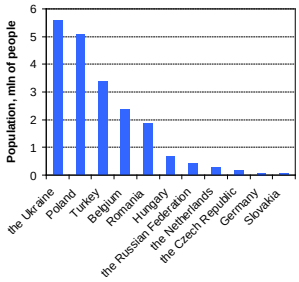

Figure 63: *Number of people in regions where B[a]P air concentrations exceed EU target value of 1 ng/m³ in EMEP countries (Source: Shatalov et al, 2015)*

3.6.2. Trends of POPs in arctic air (AMAP)

The AMAP air monitoring network includes 4 stations where (active) air sampling has been employed to monitor POPs g for up to 20 years. Three of these (Stórhöfði on Iceland; Zeppelin at Ny-Ålesund on Svalbard; and Pallas in Arctic Finland) are also part of the EMEP network; the other is the Alert station, on Ellesmere Island, Canada. POPs monitoring has also been performed at station Nord on Greenland since 2009. Other POPs air monitoring sites (including Dunai Island and Valkarkai in Arctic Russia, and Tagish/Little Fox Lake in the Yukon, Canada) have monitored POPs in past years, but lack long-term continuity of measurements and therefore do not have associated long time-series datasets. AMAP is currently working to establish POPs monitoring at the Tiksi and Amderma stations in Russia. AMAP air monitoring data are compiled in the AMAP atmospheric thematic data centre (TDC) at the Norwegian Institute for Air Research (NILU) in Norway, which also serves the EMEP.

Results of AMAPs air monitoring for POPs are being compiled in a new POPs trends assessments due to be published in 2016; preliminary results of this work have been published in a technical report (AMAP, 2014), which also includes results of short-term air monitoring at Little Fox Lake (August 2007 – 2009) and Valkarkai (2008 - 2009). Findings reported in AMAP, 2014 include the following:

- Most POPs that have been banned for extended period of time (>20-30 years) in developed countries, e.g. DDTs, aldrin, dieldrin, endrin (the "drins"), PCBs and chlordanes, are now showing slower rates of decline in Arctic air, indicating that they are approaching steady state with other environmental media and secondary sources now dominate.

- HCB and PCBs are showing increasing trends in Arctic air at certain locations which may be related to revolatilization from the open ocean due to sea ice loss or glacial melting in recent years as a result of warming.

- PBDEs (penta- and octa-BDEs) are significantly declining in air at Pallas and Zeppelin. However, no decline was observed at Alert which may be related to influence from nearby military site and generally much higher usage of these brominated flame retardants in North America compared to Europe and Russia.

- PFOS precursors MeFOSE and EtFOSE showed non-changing and declining trends, respectively; reflecting the voluntary phase-out of the production of PFOS, PFOA, and PFOS-related products. However, particle phase PFOS and PFOA are not showing any decline in Arctic air at Zeppelin.

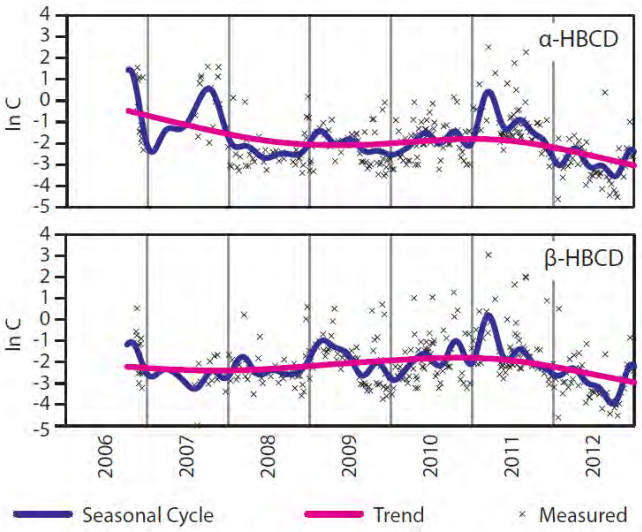

Figure 64: Air concentrations of α- and β-HBCD at Zeppelin (Source: AMAP, 2014).

- α-HBCD (which is not addressed in the POPs Protocol) is showing a declining trend in Zeppelin air (Figure 64).

3.6.3. Trends of POPs in biota in the arctic (AMAP)

As part of an ongoing assessment of POPs trends in biota, AMAP have statistically analysed more than 1500 time-series of different POPs compounds obtained for the species/locations shown in Figure 9. The majority of these time series concern substances addressed under the CLRTAP POPs Protocol. Trend analyses were performed for the entire time-series and for the period since 2000. The full set of results for these analyses will be presented in an AMAP assessment report due to be published later in 2016; a summary of results relevant to the Stockholm Convention, which also covers most of the POPs addressed under the CLRTAP POPs Protocol can be found in AMAP (2014).

Aggregated results illustrated in Figure 65 show that:

- Downward trends constitute the majority of statistically significant trends in Arctic biota for POPs addressed under the CLRTAP.
- Many of the time-series begin decades before the CLRTAP POPs Protocol was adopted and therefore likely reflect the impact of control measures that were introduced at the national level, in the 1980s and 1990s in particular, in most Arctic countries.
- Comparing trends for the period after 2000 with those for time-series starting earlier than 2000, the proportion of significant (log) linear trends is lower and the number of time-series exhibiting fluctuating trends or no significant trends greater – as expected given that longer time-series are generally more powerful for trend detection.
- Results for specific compounds or compound groups show that increasing trends are most evident in the results for PBDEs and HBCD and for PFOS. Controls on these substances at both the national and inter- national level were not widely introduced before the late 1990s/2000s. For these substances, the increasing trends are no longer so apparent if the time-series for the period after 2000 are considered and, for PBDE-47 and PFOS in particular the proportion of decreasing trends increases (see AMAP, 2014).

Interpretations of monitoring data often associate observed trends in biota with changes in emissions in an overly-simplistic manner. Changes in food-web structure, and in feeding habits and diet can strongly affect levels in biota (including humans). Trends in levels in air and biota can reflect changes in environmental processes – a number of which can be associated with climate change and variability. Nonetheless, consistency in results from a large number of trend studies, over a large geographical extent, and involving different matrices indicates that global controls on emissions are responsible for at least a part of the observed development.

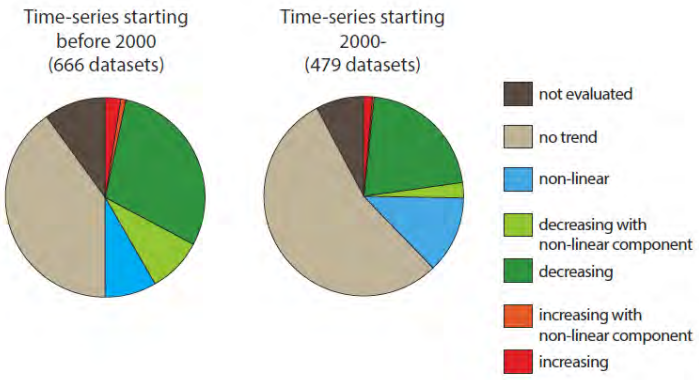

Figure 65: Aggregated results of statistical analyses of time-series of POPs in Arctic biota for entire time-series (left) and time-series since 2000 (right) (Source: AMAP, 2014).

3.6.4. Trends of POPs in humans in the arctic (AMAP)

AMAPs most recent (2015) assessment of human health in the Arctic updates previous AMAP assessments of contaminant effects on Arctic human populations and includes results from bio-monitoring studies around the Arctic, some of which are now suitable for evaluation of trends in POPs levels. Few POPs have significant emissions sources in the Arctic, thus presence of these contaminants in the Arctic is evidence of long-range transport. Diet is the main route of exposure to POPs in Arctic

communities, and bio-magnification of certain POPs in Arctic food-webs has been identified as an important factor in the exposure pathways. Particularly high exposures are found in some (indigenous) population groups that rely on marine mammals as part of their traditional diet.

AMAP human bio-monitoring studies for POPs are based mainly on blood monitoring, with breast milk studies also performed in some countries. Although a number of repeated studies have been performed in different regions of the Arctic over the past decades, results of these can be difficult to compare, especially if complicating factors such as dietary change are not taken into account. Multi-year time series are available for POPs (and metals) from three Arctic communities (Nunavik in Canada and Nuuk and Disko Bay on Greenland) (see Figure 66). For other Arctic areas, results of POPs monitoring in different 5-yearly periods have been compared (e.g., Figure 65)

The AMAP 2015 human health assessment report (AMAP, 2015a) findings include the following:

- Human exposure and levels of POPs in human tissues are declining in many regions of the circumpolar Arctic, although the decline is not uniform.
- Despite the significant decreases in concentrations of many POPs, including PCBs and DDT, measured in blood of Arctic human populations, levels remain elevated in some Arctic populations (in particular Inuit in parts of Canada and Greenland) relative to populations in other parts of the world due to dietary factors.
- Current levels of some contaminants therefore remain a concern and blood guidance levels are still exceeded in some regions.
- Some brominated flame retardants and perfluorinated contaminants have now been measured in human populations living in the Arctic. Contaminants of emerging concern, several of which are not yet internationally regulated, are thus being transported to the Arctic and contaminating human populations.

Climate change may affect how contaminants cycle in the Arctic and may also have indirect effects, for example on food-web structures (feeding habits and dietary changes in relation to availability of local species, etc.) that may impact future concentrations of contaminants in wildlife and humans.

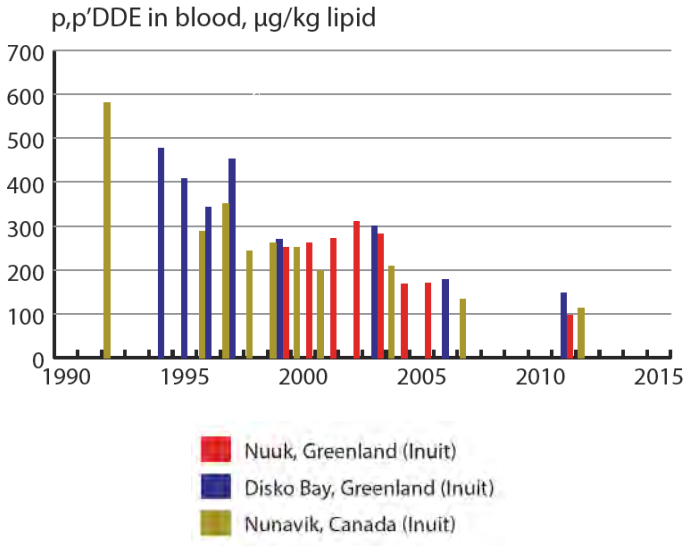

Figure 66: Multi-annual measurements of DDE in human blood from Nunavik (Canada), and Nuuk and Disko Bay (Greenland) (Source: AMAP 2015a).

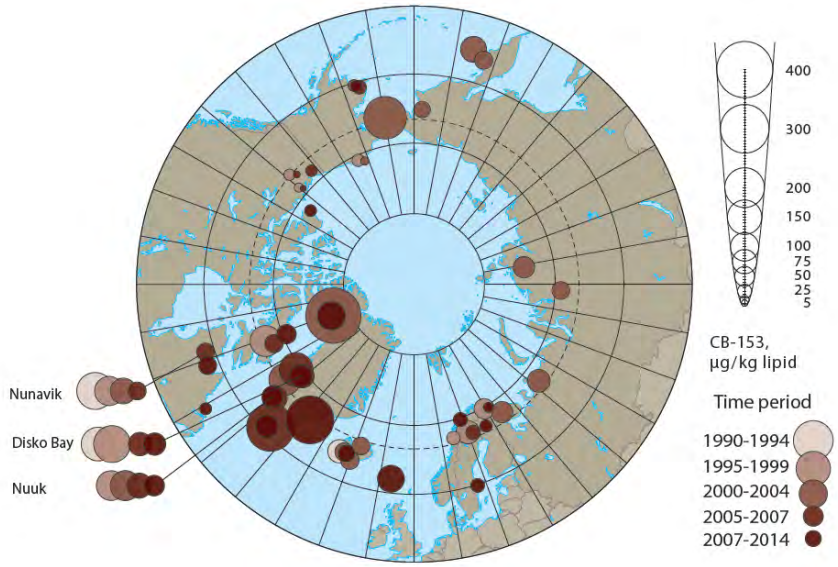

Figure 67: PCB concentrations in blood (Finland – breast milk) of mothers, pregnant women and women of child-bearing age during different time periods. (Source: AMAP 2015a).

3.6.5. Trends in POPs in fish (ICP Waters)

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 (Figure 68). 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.

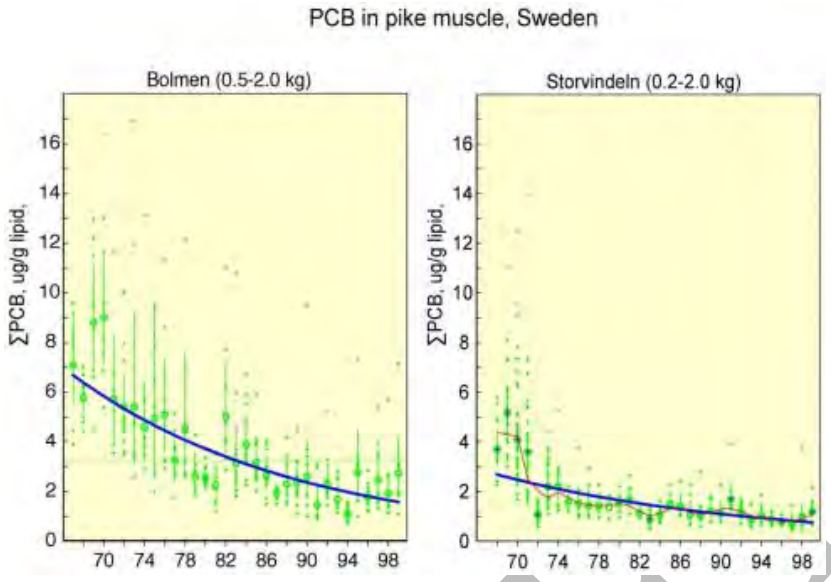

Figure 68: *Example of two time series of PCBs in pike muscle (µg/g lipid) from Lake Bolmen (southern Sweden) and Lake Storvindelen (northern Sweden) for the years 1967/1968 - 1999. Source: Fjeld et al., 2005; Nyberget et al., 2014.*

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.

4. Discussion

The signs of forest dieback, extinct fish populations and damage to cultural heritage as a consequence of high deposition and atmospheric concentrations of sulphur and nitrogen in the 1970s and 1980s created a large public concern and awareness around 'acid rain'. The response was the establishment of the Convention on Long-Range Transboundary Air Pollutants (LRTAP). Over the past 40 years the Protocols under the Convention have led to substantial decreases in the emissions of air pollutants, most notably sulphur and to a lesser extent nitrogen, over large regions of Europe and North America. Deposition of sulphur and nitrogen in Europe has declined with roughly 70% to 90%, and 25%, respectively, since the late 1970s (Torseth et al. (2012)). Health effects of long-range transported air pollution gained interest somewhat later than the ecosystem effects, but are currently in high public and policy awareness.

Acidification

Tree conditions in European forests (as indicated by defoliation status) have declined significantly for all dominate tree species except Scots pine (Par 3.1.2.1), despite the decrease in acid deposition. Crown condition is an unspecific indicator of ecosystem stress. The extremely high concentrations of atmospheric SO_2 common in Central Europe during the 1970s and 1980s were apparently responsible for the forest dieback in the region (Kandler & Innes 1995). Below critical concentrations of atmospheric SO_2 concentrations, crown condition may be controlled by a set of factors such as drought, insect damage and ozone (Sicard & Dalstein-Richier 2015) (Reich, 1987; Sandermann, 1996; Takemoto *et al.*, 2001 Today, there is little evidence that that forest crown condition is strongly, adversely impacted by acidifying effects of sulphur and nitrogen deposition only. Among contributing factors, however, nitrogen deposition has been identified, together with biotic damaging agents and weather conditions.

In three of six tree species, foliar N concentrations have declined, reflecting the declining trend in deposition of nitrogen in recent decades (3.1.2.2). Some of the decline might be driven by enhanced forest growth related to nitrogen deposition and climate change (de Vries et al. 2014; Thomas et al. 2010). The areal exceedance of critical loads for acidification of terrestrial ecosystems has been reduced to below 10% for Europe in 2010, while the mean acidity exceedance has declined from its peak value of 250 eq ha^{-1} during the 1970s to circa 10 eq ha^{-1} (3.1.6). Locally, exceedances may be much higher. Decades of sulphur deposition and sulphate leaching have depleted stores of base cations in forest soils (Kirchner & Lydersen 1995), presenting a possible delay factor for recovery from acidification. Additionally, catchment input-output budgets for sulphur indicate that in some catchments, outputs have begun to exceed inputs (Vuorenmaa et al., 2014), which presents a delay factor for chemical recovery of surface waters.

Trends in surface water chemistry show a widespread and consistent recovery from acidification, which is primarily driven by the reductions in sulphur deposition (Par. 3.1.4 ; (Garmo et al. 2014)). Records of biological recovery are fewer and less consistent, partly because the time series are shorter and have less geographical coverage. Additionally, for many organisms there are chemical thresholds which must be achieved before re-colonisation can begin. Biological recovery also is subject to delays due to factors such as dispersal mechanisms and proximity to viable populations. For many freshwater ecosystems full chemical recovery is still a long way off, and thus biological recovery is also delayed. As levels of acid deposition decline, year-to-year variations in deposition and climate become increasingly important as a driver of water chemistry, especially in ecosystems where acid deposition is at or near the critical load.

Nutrient nitrogen

Nitrogen deposition in both Europe and eastern North America has declined somewhat in the past 30 years (Par 3.1.1), but to a much lesser extent than sulphur (Torseth et al. (2012)(Waldner et al., 2014). Semi-natural and natural terrestrial ecosystems, including most forests retain most of the nitrogen

deposited from the atmosphere, as nitrogen is typically the growth-limiting nutrient. Usually less than 10% leaches to surface waters (Vuorenmaa et al., 2014)(Watmough et al. 2005).

Decades of enhanced nitrogen deposition have led to large accumulation of nitrogen in terrestrial ecosystems , and as yet there are no widespread signs that this has led to increased leaching of nitrogen to surface waters; i.e. no widespread signs of "nitrogen saturation" (Garmo et al., 2014). Evidence from forest monitoring of soil solution also suggests that nitrogen is effectively retained in soils. At nitrogen deposition levels below 10 kg N ha^{-1} yr^{-1}, the efficient soil retention of nitrogen leads to nearly negligible N leaching, while S is on average leached out at the same level as S deposition in European forested ecosystems. In those ecosystems, sulphate thus remains the dominant source of soil acidification despite the greatly reduced deposition inputs of sulphur. At N deposition levels above 10 kg N ha^{-1} yr^{-1}, it depends on rate of N retention and the level of S deposition whether the leaching fluxes of sulphate are higher than those of nitrate or the other way around (De Vries et al., 2007).

The increased nitrogen deposition and retention has altered the species composition of ground vegetation due to nutrient enrichment (eutrophication) (Stevens et al., 201, Dirnböck et al., 2014). In addition, evidence of significant effect of N deposition on defoliation (e.g. De Marco et al. 2014; Vitale et al. 2014; Ferretti et al. 2015) and growth (e.g. Ferretti et al. 2014; de Vries et al. 2014) has been reported. Also, the concurrent decline in foliar P, indicative of deterioration of tree nutrient status, may be related to high nitrogen deposition and climate change (Talkner et al., 2015).

Ozone

Ground-level ozone originates from photo-chemical reactions in the atmosphere between volatile organic compounds (VOCs), CO, methane (CH_4) and nitrogen oxides (NOx) under the influence of solar radiation, as well as from transport from the stratosphere. The ozone concentration levels in Europe consist of a hemispherical baseline level with superimposed episodes of elevated or reduced ozone caused mainly by European emissions. NO_x emissions lead to reduced ozone close to the source by a change in the equilibrium ($NO + O_3 \Leftrightarrow NO_2 + O_2$), the so-called titration effect, while further away downwind a gradual build-up of ozone is seen, with the efficiency depending on the solar radiation, atmospheric temperature, humidity etc. Peak ozone concentrations are most important for adverse effects on human health or visible plant damage, while high average concentrations during the growing seasons are threatening crop yields, forests and other ecosystems.

Many recent studies give a fairly consistent picture of the hemispherical baseline ozone level showing a rough doubling from the 1950s in all seasons up to about the year 2000 followed by a decade with a reduced increase, no growth or even reductions at some sites, particularly in the summer (e.g. Parrish et al., 2012; Logan et al., 2012; Derwent et al., 2013). Despite a more than 30% reduction in European emissions of ozone precursors during the last two decades, a decline in mean 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. Therefore, whilst the mean or median ozone concentration has hardly changed, peak concentrations (above >95^{th} percentile) have declined and background concentrations (lower percentiles) have risen in many places (Simpson et al., 2014). EEA (2014b) also reported that measured ground-level ozone concentrations have reduced only marginally or have increased due to long-range transport of pollutants from outside Europe. Reduced precursor emissions might well be masked by large inter-annual variations in ozone, caused by, for example climate, weather or biomass burning events.

Data on ozone concentrations in the last two decades from ICP Vegetation and ICP Forests monitoring sites show similar trends as observed at EMEP monitoring sites (see above) and confirm that the trends are site specific and dependent on the metric used for assessing trends in ozone concentrations. Although a decline in peak ozone concentrations was expected to result in a decline in ozone risk indicators such as AOT40 and SOMO35, where only ozone concentrations above 40 and 35 ppb contribute to the risk of adverse impacts on vegetation and human health respectively, no clear European trends were observed and trends were site-specific, as seen for ozone concentrations. With present day background concentrations of ozone being in the range of ca. 30 – 40 ppb, background concentrations are also starting to contribute to the risk indicators AOT40 and SOMO35, which might explain the lack of a general trend in these risk indicators. In future, the SOMO35 for urban sites could even rise due to a further predicted decline in NO_x emission, which reduces the titration effect of NO_x, resulting in higher urban ozone concentrations. When applying the biologically more relevant flux-based approach (Mills et al., 2011; LRTAP Convention, 2015), there was no trend in the Phytotoxic Ozone Dose (POD) calculated for wheat at ICP Vegetation monitoring sites, which can be explained by the fact that background ozone concentrations contribute considerably to the POD. Although high ozone exposure levels might be a contributing factor in the defoliation of forests, especially in deciduous trees (Klap et al., 2000; Feretti et al., 2007), this is not always the case (Feretti et al., 2012). Hence, the relationship between tree crown condition and ozone effects require more in depth analyses, linking long-term monitoring with mechanistic studies to disentangle the complex interactions between environmental conditions, including ozone, and impacts on tree defoliation.

It remains unclear how emission controls in Europe may be offset by global background ozone changes, 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 (Wild et al., 2012).

Particulate matter

Widespread monitoring of trends in PM, in air, for population exposure and for damage to materials (soiling), did not start until after 2000 (see 3.4). Where EMEP monitoring stations are largely located in background areas with few sources of local air pollution, monitoring stations delivering data to TF Health are located with relevance for assessing population exposure, and are placed in urban background, residential, commercial and mixed areas. Assessments of change in PM concentrations since the 1980s have been based on monitoring of other chemical components, and indicate strong reductions (Tørseth et al., 2014).

For monitoring of population exposure in cities to PMs, there are no indications that population exposure to PM2.5 in the European Region has changed substantially since 2003-2005. However, an overall decrease of emissions of PM2.5 and PM10 (EEA, 2014) and PM10 concentrations (see 3.4) across the European Region has been reported. Soiling is the darkening of building materials as a result of particulate deposition. Dirty buildings are in many cases considered unacceptable even if the structural integrity of the base material is unaffected by corrosion or other types of degradation and costs associated with cleaning can be substantial. Soiling of modern glass has been evaluated since 2005 and the measured response (haze) shows no overall sign of improvement. Values of haze corresponding to visual nuisance detected by the human eye is obtained between a few months to half a year for polluted industrial and traffic areas (Verney-Carron et al., 2012)

Ground-level monitoring of PMs 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 trends in population exposure and assist local authorities in establishing plans for improving air quality. A landmark resolution on "Health and the environment: addressing the health impact of air pollution" has been adopted at the sixty-eighth World Health Assembly (WHA) in May 2015 (WHO, 2015). The Resolution was developed in consideration that air pollution is the largest single environmental risk and a leading cause of disease and death globally. It is a risk factor for ischemic heart disease, stroke, chronic obstructive pulmonary disease, asthma and cancer. Air pollution's negative effect on health brings an enormous economic burden. A recent WHO report reveals that in the WHO European Region alone a staggering US$ 1.6 trillion is the economic cost of the approximate 600 000 premature deaths and of the diseases caused by air pollution in 2010 (WHO Regional Office for Europe, 2015). Among the actions outlined in the adopted Resolution, Member States are urged to improve the morbidity and mortality surveillance for all illnesses related to air pollution, and optimize the linkage with monitoring systems of air pollutants.

The overall compelling scientific evidence on (and significant burden of disease from) air pollution provide convincing arguments for the need to take further action to reduce emissions and improve air quality, with the aim to improve public health, as set forth in the WHA Resolution.

Heavy metals

In recent decades, the deposition of heavy metals has been reduced significantly, most notably for lead due to the switch to unleaded fuel, but also for cadmium. Of the priority metals considered within the LRTAP Convention (cadmium, lead and mercury), the reduction of air concentrations and deposition of mercury have been smallest. Deposition trends modelled by EMEP are in good agreement with trends in heavy metal concentrations measured in mosses, the latter also showing a decline in concentrations for other than the priority metals.

Despite the general decline in deposition, heavy metals still remain a threat to ecosystems due to their accumulation in soil and other components of ecosystems, including food chains. This has implications for human health, as shown for example in Arctic areas. Although levels of lead in humans have fallen in most Arctic countries, they remain elevated in some parts of Russia and Arctic Canada (AMAP, 2015b). Levels of mercury in human blood in Arctic populations in Norway and Sweden have now fallen to similar levels to those found in non-Arctic populations in these countries. However, mercury levels remain elevated, and in some cases exceed guidelines, in parts of Greenland and Canada (AMAP, 2015b).

Trends of heavy metals in environmental media do not show universal declines. Lead and cadmium concentrations in upper soil layers showed decreases between 1994 and 2011 (3.5.3), consistent with the trends observed in mosses and deposition. However, both metals are being transferred to deeper soil layers as documented by increasing concentrations in those layers, indicating that accumulation is ongoing in soils. Contrary to the trend in mosses and deposition, concentrations of mercury increased in the forest floor and in deeper soil layers. In stream waters, hardly any trends in cadmium, lead and mercury were found in the last two decades, but data material is limited.

Heavy metals continue to leach from soils to surface waters, with no clear relation to trends in deposition. Mercury concentrations in freshwater fish show a mixture of trends, from downward in Sweden since 1965, and rising since the 1990s in Sweden, Norway and Canada, although not for all fish species and not for all investigated regions. Mercury in many fish species remains above limits advised for human consumption. Thus, reducing the concentration of mercury in fish should be given a high priority (WHO, 2007). Mercury is subject to hemispheric transport and therefore requires global abatement interventions,

hence the Minamata Convention on Mercury was establish in 2013, a global treaty to protect human health and the environment from the adverse effects of mercury.

In view of the narrow safety margins for impacts of cadmium and lead on human health, further reductions in their emissions and subsequent accumulation in soil and ecosystems is desirable (WHO, 2007).

Exceedances for critical loads for cadmium and lead have declined since 1990. For cadmium, critical loads were almost no longer exceeded in 2010. For lead, the area at risk declined from 67% to 20% in Europe between 1990 and 2010, and magnitude of the exceedance of critical loads declined drastically. Exceedances for lead still occurred in all countries in Europe in 2010. For mercury, high deposition remains an issue of concern in 2010, with a decline from 69% to 56% of the ecosystem area at risk of exceedance of the critical loads between 1990 and 2010.

POPs

Although there is consensus that POPs constitute some of the most dangerous pollutants released into the environment by anthropogenic activities, comprehensive and systematic monitoring of these contaminants in the environment is still lacking in many areas. Although regular monitoring of POPs related to long-range transported atmospheric pollution is conducted under programs such as AMAP and EMEP, currently none of the groups under the WGE report data on a regular basis. Since the evaluation of risks by POPs cannot by performed by a critical loads approach (Gregor et al. 1998), risk assessment should be done by evaluating trends in concentrations and/or exposure, in relation to effect thresholds.

One literature study on POPs, Fjeld et al. (2005) provide an overview of levels of selected POPs in freshwater fish and sediments from North America, Europe and circumpolar Arctic based on data from some major surveys (excluding sites with elevated local pollution). The very few sites with temporal trend data generally showed decreasing levels of legacy POPs in line with results from other monitoring programmes. Also, levels of some new substances (e.g. PBDE) were possibly rising (Fjeld et al., 2005), a tendency that is also documented for some substances and sites, from recent data from EMEP and AMAP (AMAP 2014, 2015b).

ICP Vegetation included a pilot study on POPs in the 2010/11 European moss monitoring (ICP Vegetation 2013). If POPs in mosses can be approved as an appropriate indicator for atmospheric pollution in upcoming moss monitoring studies, this would provide valuable information for combination with EMEP deposition data and would contribute to better understanding of spatial and time trends of POPs exposure in the CLRTAP region.

Comment [HW13]: From Simon:

New internation programms are now being run using passive samplers ... originally run as a campaign these studies are now being repeated and have been adopted by e.g. teh Stockholm Convention GMP. These might be making the older moss studies a bit redundant (moss was essentially being used as a passive sampler .. just an observation ... not really my field)

Because of the global distilling effect many POPs, in particular the more volatile, are transported to and accumulated in polar (but also in alpine) environments. Biota at the top of food chains such as marine mammals or human in the Arctic region therefore show elevated levels of POPs in blood, tissue or other media (Reference). The current report leans to a large extent on information provided by AMAP, to document whether policy to reduce emissions of POPs has been effective.

Levels of POPs in air, biota and humans in the Arctic show generally decreasing trends for most POPs covered by the Conventions. Levels of POPs in the blood of Arctic residents have also generally declined over the past 20 to 30 years. This includes significant falls in levels of DDT and polychlorinated biphenyls (PCBs). However, levels of some POPs in human blood still remain higher in certain Arctic populations than in most general populations in North America and Europe. Trends in levels of POPs that have

emerged in recent years as chemicals of concern in the Arctic, including some regulated brominated flame retardants show a more mixed pattern, with some time-series still showing increasing trends, whereas in others levels appear to have peaked in the early 2000s (AMAP, 2015b).

Declining levels of many 'legacy' POPs in Arctic air, biota and humans are likely associated mainly with national regulatory actions introduced in many countries in Europe and North America during the 1970s-1990s, supplemented by international agreements including the CLRTAP POPs Protocol. Other factors may also be playing a role in influencing trends in levels of some contaminants in the Arctic. These include several factors related to climate change (UNEP/AMAP, 2011). In humans, changes in diet, in some cases associated with risk communications and advice to limit consumption of certain foods, has also played a role.

After substantial declines in the last decades of the 20th century, the slowing rates of decline in levels of regulated or banned POPs– such as DDTs, aldrin, dieldrin, PCBs and chlordanes – in Arctic air and biota indicates that transport of these POPs into the Arctic is now probably associated mainly with re-emissions of POPs that have accumulated in the environment in soils and waters rather than primary emission sources (AMAP, 2015b). Concern, however, remains about, e.g. stockpiles of pesticides that have been banned but have yet to be destroyed, and the many new chemicals with POP-like characteristics that are being introduced into society, in some cases as replacements for banned substances.

AMAPs 2015 assessment of Arctic Pollution Issues (AMAP, 2015b) concludes that controls on pollutants have proven effective; however, there is a need for more timely controls on chemicals of emerging concern. Recognizing that long-range transport remains the most significant source of Arctic contamination, AMAP recommended that [Arctic] states continue to show leadership on international pollution control, and where necessary ratify existing Conventions as soon as possible. At the same time AMAP identified a need for consideration of additional unilateral, regional and global actions to control pollutants of emerging concern (AMAP, 2015b).

5. Conclusions

The trends in ecosystem responses, health and materials confirm that air quality is improving, and that much has been achieved in terms of effects of air pollution policies in the past decade. However, downward trends in effects of air pollution need to be sustained in order to avoid adverse effects on public health and the environment, including recovery of the latter.

The assessments of trends described in this report document that air pollution policy has been most successful for acidification and heavy metals. The area of acidification critical load exceedance and the exceedance itself has been strongly reduced for terrestrial and aquatic ecosystems in many parts of Europe, while signs of recovery have been documented. Surface water chemistry has recovered strongly in most regions, while biological recover also shows improvements but is lagging behind. Full chemical and biological recovery is not foreseeable for the near future because of delay factors, while critical loads remain exceeded in some regions. Corrosion has decreased strongly. Deposition of heavy metals has been strongly reduced, but large areas still show exceedances for lead and cadmium. Mercury remains a problem, with high critical load exceedances and concentrations in freshwater fish exceeding limits advised for human consumption.

Air pollution policies have led to a reduction of pressures caused by the emission of oxidized and reduced nitrogen. However, indirect and direct risk of nitrogen compounds to public health (e.g. NO_x related ozone formation and ammonia related $PM_{2.5}$ concentrations) and the environment (acidification, eutrophication) remain of significant concern on both an urban (local) and regional scale, the latter

including declining biodiversity. Deposition and critical load exceedances are declining, but the assessment of trends until 2020 indicates that still a major part of European natural areas remains at risk of effects of critical load exceedances. Effects of ambient concentrations of particulate matter and ozon seem to have been least reduced. Health impacts of particulate materials have not been reduced since 2000, while records of soiling also point to a lack of improvement. The risk of ecosystem damage from ozone remains high: there is no decline in average air concentrations/deposition despite declining peak concentrations, due to rising background concentrations With regard to persistent organic pollutants spread by long-range atmospheric transport, monitoring records of effects are scarce, and do not allow for an robust evaluation of effectiveness of air pollution policy. However, trends in POPs in arctic areas suggest that for some of the POPs covered by international policies, concentrations in the environment have been reduced. There is an urgent need to establish systematic long-term monitoring of selected POPs including the new emerging ones in background areas, and coordinated international surveys with harmonized methodology and reporting. Reasons for slight increases of POPs concentrations in recent years should further be investigated.

Finally, it is noted that knowledge gaps also remain with respect to current pressing environmental concerns that include air pollution, but not as an isolated issue. Knowledge on interactions and trade-offs between air pollution and climate change on effects for public health, public wellbeing, biodiversity, ecosystem services (e.g. food and timber production, carbon storage) and materials is becoming increasingly important. Improving scientific policy support in these fields is embedded in the Long Term Strategy of the Convention. This requires the continuation and further strengthening of the modelling and monitoring of effects under the synthesizing guidance of European effect oriented policy requirements in general, and the Working Group on Effects in particular.

New References:

Comment [HW14]: Reference list will be checked by Max.

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Comment [MP6]: Question 2015b [was just 2015] is the more general than 2015a … shouldn't we just drop 2015a and revert keep 2015 (i.e. not change to 2015b)?

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