



Centre for
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NATURAL ENVIRONMENT RESEARCH COUNCIL

Air Pollution and Vegetation



ICP Vegetation

**Annual Report
2011/2012**

wge

Working Group on Effects
of the
Convention on Long-range Transboundary Air Pollution

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Air Pollution and Vegetation

ICP Vegetation¹ Annual Report 2011/2012

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Finally, we wish to thank all of the ICP Vegetation participants for their continued contributions to the programme and other bodies within the LRTAP Convention.

Front cover photo: Dr Laurence Jones (CEH, Bangor)

Executive Summary

Background

The International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) was established in 1987. It is led by the UK and has its Programme Coordination Centre at the Centre for Ecology and Hydrology (CEH) in Bangor. It is one of seven ICPs and Task Forces that report to the Working Group on Effects (WGE) of the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) on the effects of atmospheric pollutants on different components of the environment (e.g. forests, fresh waters, materials) and health in Europe and North-America. Today, the ICP Vegetation comprises an enthusiastic group of over 200 scientists from 35 countries in the UNECE region with outreach activities to other regions such as Asia, Central America and Africa. An overview of contributions to the WGE workplan and other research activities in the year 2011/12 is provided in this report.

25th ICP Vegetation Task Force meeting

The Programme Coordination Centre organised the 25th ICP Vegetation Task Force meeting, 31 January - 2 February 2012 in Brescia, Italy, in collaboration with the local hosts at the Ecophysiology and Environmental Physics Laboratory - Mathematics and Physics Department, Università Cattolica del Sacro Cuore. The meeting was attended by 73 experts from 21 countries, including Egypt and South Africa. Participation at the annual Task Force meeting has been rising steadily over the 25 years from ca. 20 experts from 10 countries to the current level of over ca. 70 participants from more than 20 countries. The Task Force discussed the progress with the workplan items for 2012 and the medium-term workplan for 2013 - 2015 for the air pollutants ozone, heavy metals, nutrient nitrogen and persistent organic pollutants (POPs). The meeting was preceded by a one-day ozone workshop where the following two themes were discussed:

- Quantifying ozone impacts on Mediterranean forests;
- Mapping vegetation at risk from ozone at the national scale.

New scientific developments presented at the workshop support the use of the stomatal flux-based method rather than the concentration-based method for ozone impact assessments on vegetation. The workshop recommended to further develop the ozone flux-based method and provide further field-based validation of ozone flux-effect relationships and critical levels via epidemiological studies. A book of abstracts, details of presentations and the minutes of the 25th Task Force meeting and ozone workshop are available from the ICP Vegetation web site (<http://icpvegetation.ceh.ac.uk>).

Reporting to the Convention and other publications

In addition to this report, the ICP Vegetation Programme Coordination Centre has provided a technical report on 'Effects of air pollution on natural vegetation and crops' (ECE/EB.AIR/WG.1/2012/8) and contributed to the joint report (ECE/EB.AIR/WG.1/2012/3) and impact analysis report (ECE/EB.AIR/WG.1/2012/13) of the WGE. It contributed to the Guidance Document VII on health and environmental improvements for the revision of the Gothenburg Protocol. The ICP Vegetation also published the glossy report and summary brochure for policy makers on the threat of ozone to food security, and published a glossy report on the impact of ozone on carbon sequestration in Europe. The ICP Vegetation contributed to and facilitated printing of the colour brochure of the WGE on 'Impacts of air pollution on human health, ecosystems and cultural heritage' in English, French and Russian. In addition, a colour leaflet was produced on 'Mosses as biomonitors of atmospheric heavy metal pollution in Europe' in English and Russian. Three scientific papers have been published or in press and the ICP Vegetation web site was updated regularly with new information.

Contributions to the WGE common workplan

Further implementation of Guidelines on Reporting of Air Pollution Effects

The ICP Vegetation continued to monitor and model deposition to and impacts on vegetation for the air pollutants ozone, heavy metals, nitrogen and POPs.

Final version of the impact analysis by the WGE

To support the revision of the Gothenburg Protocol, the WGE has conducted an analysis on the impacts of air pollution on ecosystems, human health and materials under different emission scenarios, including the application of recently developed effects indicators such as the phytotoxic ozone dose (POD; flux-based approach). Results from the ICP Vegetation show that despite predicted reductions in both ozone concentrations and stomatal fluxes in 2020, large areas in Europe will remain at risk from adverse impacts of ozone on vegetation, even after implementation of maximum technically feasible reductions, with areas at highest risk being predicted in parts of western, central and southern Europe.

Ideas and actions to enhance the involvement of EECCA/SEE countries in Eastern Europe, the Caucasus and Central Asia and on cooperation with activities outside the Air Convention

Working with the lead participant of the European moss survey in the Russian Federation, the ICP Vegetation is actively encouraging the participation of more EECCA/SEE countries. For example, Albania took part for the first time in the moss survey in 2010/11 and attended the ICP Vegetation Task Force meeting for the first time in 2012. Together with the Stockholm Environment Institute (SEI) in York (UK) the ICP Vegetation has produced a position paper on outreach activities to Malé Declaration countries in South Asia. Suggestions were provided and discussed for further collaboration in the near future at the third meeting of the Task Force of the Malé Declaration, 9-10 August 2012, Chonburi, Thailand. However, implementation of further collaboration is severely hindered by the lack of available funds. The ICP Vegetation also has developed collaboration with experts in Egypt, South Africa, Cuba and Japan, who have attended recent Task Force meetings of the ICP Vegetation.

Progress with ICP Vegetation-specific workplan items in 2011/12

Supporting evidence for ozone impacts on vegetation

Since 2008, participants of the ICP Vegetation have been conducting biomonitoring campaigns using ozone-sensitive (S156) and ozone-resistant (R123) genotypes of *Phaseolus vulgaris* (Bush bean, French Dwarf bean). In 2011, the biomonitoring of ozone effects using bean was scaled down compared to the previous two years, reflecting less interest from the participants. Nevertheless, experiments were conducted with ozone-sensitive and ozone-resistant bean (*Phaseolus vulgaris*) at nine sites across Europe and one in the USA. The data from the 2011 and previous biomonitoring and ozone exposure experiments were combined in a database for dose-response analysis. The database contains data from Belgium, France, Germany (3 sites), Greece (2 sites), Hungary (2 sites), Italy (3 sites), Japan, Slovenia (2 sites), Spain (3 sites), South Africa, UK (2 sites), Ukraine and the USA. Visible leaf injury regularly occurred across the network, but there was no clear dose-response relationship with ozone parameters. Similarly, there was no clear relationship between concentration-based parameters and the ratio of the pod weight for the sensitive to that of the resistant bean. Flux-effect relationships will be explored in the coming year.

Ozone impacts on carbon sequestration in Europe

Terrestrial vegetation, particularly forests, is an important sink for the greenhouse gases carbon dioxide (CO₂) and ozone. However, the air pollutant ozone has a negative impact on cell metabolism and growth of ozone-sensitive plant species. Hence, this will result in a positive feedback to global warming as less CO₂ and ozone will be sequestered by vegetation, resulting in a further rise of their concentrations in the atmosphere. The future impacts of ozone on carbon (C) sequestration in European terrestrial ecosystems will depend on the interaction with and magnitude of the change of the physical and pollution climate, represented by rising temperatures, increased drought frequency, enhanced atmospheric CO₂ concentration and reduced nitrogen deposition. For the first time we applied the DO₃SE (Deposition of Ozone for Stomatal Exchange) model to estimate the magnitude of the impact of ambient ozone on C storage in the living biomass of trees. The Phytotoxic Ozone Dose above a threshold value of $Y \text{ nmol m}^{-2} \text{ s}^{-1}$ (POD_Y) was calculated applying known flux-effect relationships for various tree species. When applying a standard parameterisation for deciduous and conifer trees, current ambient ground-level ozone was estimated to reduce C sequestration in the living biomass of trees by 12.0 to 16.2% (depending on ozone, meteorological and climate input data)

in the EU27 + Norway + Switzerland in 2000. The flux-based approach indicated the highest ozone impact on forests in central Europe, where moderate ozone concentrations coincide with a climate conducive to high stomatal ozone fluxes and with high forest carbon stocks. A considerable reduction was also calculated for parts of northern Europe, especially when applying climate region-specific parameterisations. Under drought-free conditions (i.e. no limitation of soil water availability for tree growth), the predicted reduction in C sequestration in the living biomass of trees increased from 12.0 to 17.3% in the year 2000, with the highest reductions predicted for the warmer and drier climates in the southern half of Europe, particularly in the Mediterranean. Although a decline in stomatal ozone flux was predicted in 2040, C sequestration in the living biomass of trees will still be reduced by 12.6% (compared to 16.2% in 2000).

The above results only describe ozone effects on the living biomass of trees and do not take into account any effect of ozone on soil carbon cycling, impacts of potential changes of forest management in the future or feedbacks to the climate system. Although the flux-response functions used were derived for young trees (up to 10 years of age), there is scientific evidence from some epidemiological studies that the functions are applicable to mature trees as assumed in this study. There is a clear need to include the impacts of ozone on vegetation in global climate change modelling.

Progress with European heavy metals and nitrogen in mosses survey 2010/11

Mosses have been collected for element analysis every five years since 1990 and the most recent survey was conducted in 2010/11. A total of 26 countries will submit or have already submitted data on heavy metals, of which 14 countries will also submit (or have submitted) data on nitrogen concentrations in mosses. Nitrogen concentrations were reported for the first time in the 2005/6 European moss survey. As a pilot study, six countries have agreed to submit data on POPs concentrations in mosses to further assess the suitability of mosses as biomonitors of atmospheric POPs pollution. The final report on the 2010/11 European moss survey will be published in 2013.

Relationship between (i) heavy metal and (ii) nitrogen concentrations in mosses and their impacts on ecosystems

A review of the scientific literature showed that little is known about the relationship between heavy metal concentrations in mosses and the impacts of heavy metals on terrestrial ecosystems. Toxicity effects of heavy metals are usually limited to areas close to pollution sources, with impacts often declining exponentially with distance from the pollution source. For example, in agreement with an observed gradient of reducing heavy metal concentrations in mosses away from a heavy metal pollution source, an increase was observed in the abundance of soil mesofauna with distance from the pollution source. However, in the European survey, mosses are not sampled close to pollution sources and hence concentrations are often too low to be associated with an impact on terrestrial ecosystems in the sampling areas. This does not mean that we should not be concerned about heavy metal deposition in remote areas as metals will accumulate in the soil and might become a problem in the future if bio-available concentrations reach critical limits.

A review of the scientific literature showed that little is also known about the relationship between nitrogen concentrations in terrestrial mosses and impacts of nitrogen on terrestrial ecosystems. The nitrogen concentration in the moss species used in the European moss survey tends to be a good indicator of total atmospheric nitrogen deposition up to a deposition flux of ca. $15 \text{ kg ha}^{-1} \text{ y}^{-1}$. Above this level, the nitrogen concentration in mosses tends to saturate, although the level at which saturation occurs varies geographically. Empirical critical loads have been defined for various habitats (ECE/EB.AIR/WG.1/2010/14), however, the effects indicators for exceedance have not been related so far to nitrogen concentrations in mosses per se. For many terrestrial ecosystems with an empirical critical load below $15 \text{ kg ha}^{-1} \text{ y}^{-1}$ nitrogen effects have been reported on moss species (e.g. changes in moss species composition or abundance). Recent studies have shown that vegetation responses to nitrogen deposition might depend more on the nitrogen form (ammonia or nitrate) than dose. Vegetation tends to be more sensitive to ammonia (ECE/EB.AIR/WG.5/2007/3) than nitrate exposure.

Future activities of the ICP Vegetation

The ICP Vegetation Task Force has agreed to conduct a review and publish a glossy state of knowledge report on 'Ozone impacts on biodiversity and ecosystem services' in 2013. Highlights from this study will be submitted for inclusion in the WGE's report on impacts of air pollution on biodiversity and ecosystem services. As one of its core activities the ICP Vegetation will continue ozone stomatal flux model development and flux map validation. Hence, we will continue to collate supporting evidence for ozone impacts on vegetation and review the robustness of flux-effect relationships for the establishment of new flux-based ozone critical levels for additional plant species. In 2013, the ICP Vegetation will report on the outcome of the 2010/11 European moss survey for heavy metals, nitrogen and POPs. The ICP Vegetation will also continue to explore opportunities for outreach activities to other regions of the globe.

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1 Introduction

1.1 Background

The International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) was established in 1987, initially with the aim to assess the impacts of air pollutants on crops, but in later years also on (semi-)natural vegetation. The ICP Vegetation is led by the UK and has its Programme Coordination Centre at the Centre for Ecology and Hydrology (CEH) in Bangor. The ICP Vegetation is one of seven ICPs and Task Forces that report to the Working Group on Effects (WGE) of the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) on the effects of atmospheric pollutants on different components of the environment (e.g. forests, fresh waters, materials) and health in Europe and North-America. The Convention provides the essential framework for controlling and reducing damage to human health and the environment caused by transboundary air pollution. So far, eight international Protocols have been drafted by the Convention to deal with major long-range air pollution problems. ICP Vegetation focuses on the following air pollution problems: quantifying the risks to vegetation posed by ozone pollution and the atmospheric deposition of heavy metals, nitrogen and persistent organic pollutants (POPs) to vegetation. The work of the ICP Vegetation contributed significantly to the recent revision of the Gothenburg Protocol (finalised in May 2012), aiming to abate acidification, eutrophication and ground-level ozone.

The ICP Vegetation comprises an enthusiastic group of over 200 scientists from 35 countries in the UNECE region (**Table 1.1**). In addition, scientists from China, Cuba, Egypt, India, Japan, Pakistan and South Africa participate as the ICP Vegetation stimulates outreach activities to other regions in the world and invites scientists in those regions to collaborate with and participate in the programme activities. The contact details for lead scientists for each group are included in Annex 1. In many countries, several other scientists (too numerous to mention individually) also contribute to the biomonitoring programmes, analysis, modelling and data synthesis procedures of the ICP Vegetation.

Table 1.1 Countries participating in the ICP Vegetation; in italics: not a Party to the LRTAP Convention.

Albania	FYR of Macedonia	Romania
Austria	Germany	Russian Federation
Belarus	Greece	Serbia
Belgium	Iceland	Slovakia
Bulgaria	<i>India</i>	Slovenia
<i>China</i>	Italy	<i>South Africa</i>
Croatia	<i>Japan</i>	Spain
<i>Cuba</i>	Latvia	Sweden
Czech Republic	Lithuania	Switzerland
Denmark	Montenegro	Turkey
<i>Egypt</i>	Netherlands	Ukraine
Estonia	Norway	United Kingdom
Finland	<i>Pakistan</i>	USA
France	Poland	Uzbekistan

1.2 Air pollution problems addressed by the ICP Vegetation

1.2.1 Ozone

Ozone is a naturally occurring chemical present in both the stratosphere (in the 'ozone layer', 10 – 40 km above the earth) and the troposphere (0 – 10 km above the earth). Additional photochemical

reactions involving NO_x, carbon monoxide and non-methane volatile organic compounds (NMVOCs) released due to anthropogenic emissions (especially from vehicle sources) increase the concentration of ozone in the troposphere. These emissions have caused a steady rise in the background ozone concentrations in Europe and the USA since the 1950s (Royal Society, 2008). Superimposed on the background tropospheric ozone are ozone episodes where elevated ozone concentrations in excess of 50-60 ppb can last for several days. Ozone episodes can cause short-term responses in plants such as the development of visible leaf injury (fine bronze or pale yellow specks on the upper surface of leaves) or reductions in photosynthesis. If episodes are frequent, longer-term responses such as reductions in growth and yield and early die-back can occur.

The negotiations concerning ozone for the Gothenburg Protocol (1999) were based on exceedance of a concentration-based critical level of ozone for crops and (semi-)natural vegetation. However, since then the biologically more relevant stomatal flux-based was developed, estimating the flux of ozone from the exterior of the leaf through the stomatal pores to the sites of damage (Emberson *et al.*, 2000; Pleijel *et al.*, 2007). During 2009/10, flux-based critical levels of ozone for vegetation were reviewed at an LRTAP Convention workshop in Ispra, November 2009 and new/revised flux-based critical levels were agreed at follow-on discussions at the 23rd ICP Vegetation Task Force meeting, February 2010 (Harmens *et al.*, 2010a; LRTAP Convention, 2010; Mills *et al.*, 2011b). They include policy-relevant indicators for i) agricultural crops to protect security of food supplies; ii) forest trees to protect against loss of carbon storage in living trees and loss of other ecosystem services such as soil erosion, avalanche protection and flood prevention; iii) grassland (productive grasslands and grassland of high conservation value) to protect against for example loss of vitality and fodder quality. The flux-based approach is now the preferred method for assessing the risk of ozone impacts on vegetation, as described in Annex 1 of the revised Gothenburg Protocol. Particulate matter (PM_{2.5}), and thereby also black carbon as a component of PM_{2.5}, has now been included in the Gothenburg Protocol. The recently revised Gothenburg Protocol requires that EU member states jointly cut their emissions of sulphur dioxide by 59%, nitrogen oxides (a precursor of ozone) by 42%, ammonia by 6%, volatile organic compounds (a precursor of ozone) by 28% and particles by 22% between 2005 and 2020. Once the national emission reduction obligations have been implemented in 2020, the revised Protocol is expected to result in significant reductions in human health impacts and adverse impacts on the environment from air pollution. Despite these emission reductions, air pollution will still pose significant risk to human health and the environment after 2020.

The ozone sub-group of the ICP Vegetation contributes models, state of knowledge reports and information to the LRTAP Convention on the impacts of ambient ozone on vegetation; dose-response relationships for species and vegetation types; ozone fluxes, vegetation characteristics and stomatal conductance; flux modelling methods and the derivation of critical levels and risk assessment for policy application.

1.2.2 Heavy metals

Concern over the accumulation of heavy metals in ecosystems and their impacts on the environment and human health, increased during the 1980s and 1990s. Currently some of the most significant sources include:

- Metals industry (Al, As, Cr, Cu, Fe, Zn);
- Other manufacturing industries and construction (As, Cd, Cr, Hg, Ni, Pb);
- Electricity and heat production (Cd, Hg, Ni);
- Road transportation (Cu and Sb from brake wear, Pb and V from petrol, Zn from tires);
- Petroleum refining (Ni, V);
- Phosphate fertilisers in agricultural areas (Cd).

The heavy metals cadmium, lead and mercury were targeted in the 1998 Aarhus Protocol as the environment and human health were expected to be most at risk from adverse effects of these

metals. This Protocol is currently under review. Atmospheric deposition of metals has a direct effect on the contamination of crops used for animal and human consumption (Harmens *et al.*, 2005).

The ICP Vegetation is addressing a short-fall of data on heavy metal deposition to vegetation by coordinating a well-established programme that monitors the deposition of heavy metals to mosses. The programme, originally established in 1980 as a Swedish initiative, involves the collection of naturally-occurring mosses and determination of their heavy metal concentration at five-year intervals. European surveys have taken place every five years since 1990, with the latest survey having been conducted in 2010/11. The results of this recent survey will be published in 2013 and will also include data on nitrogen and POPs concentrations in mosses. Spatial and temporal trends (1990 – 2005) in the concentrations of heavy metals in mosses across Europe have been described by Harmens *et al.* (2008; 2010b). Detailed statistical analysis showed that spatial variation in the cadmium and lead concentrations in mosses is primarily determined by the atmospheric deposition of these metals, whereas it is less clear which factor primarily determines the mercury concentration in mosses (Harmens *et al.*, 2012; Holy *et al.*, 2010; Schröder *et al.*, 2010b).

1.2.3 Nitrogen

In recent decades, concern over the impact of nitrogen on low nutrient ecosystems such as heathlands, moorlands, blanket bogs and (semi-)natural grassland has increased. The empirical critical loads for nitrogen were reviewed and revised recently (Bobbink and Hettelingh, 2011; ECE/EB.AIR/WG.1/2010/14). In 2009, the WGE gathered evidence on the impacts of airborne nitrogen on the environment and human health with the aim of drawing attention to the current threat of atmospheric nitrogen deposition to the environment and human health (ECE/EB.AIR/WG.1/2009/15). Details on the contribution of the ICP Vegetation can be found in Harmens *et al.* (2009). Previously, plant communities most likely to be at risk from both enhanced nitrogen and ozone pollution across Europe were identified (Harmens *et al.*, 2006). In 2005/6, the total nitrogen concentration in mosses was determined for the first time at almost 3,000 sites to assess the application of mosses as biomonitors of nitrogen deposition at the European scale (Harmens *et al.*, 2011b; Schröder *et al.*, 2010a). The European nitrogen in moss survey was repeated in 2010/11. There are many groups within Europe studying atmospheric nitrogen fluxes and their impact on vegetation (e.g. Nitrogen in Europe (NinE), ECLAIRE, COST 729). The ICP Vegetation maintains close links with these groups to provide up-to-date information on the impacts of nitrogen on vegetation to the WGE of the LRTAP Convention.

1.2.4 Persistent organic pollutants (POPs)

POPs are organic substances that possess toxic and/or carcinogenic characteristics, are degrading very slowly, bioaccumulate in the food chain and are prone to long-range transboundary atmospheric transport and deposition. In 1998, the Aarhus Protocol on POPs was adopted and a list of 16 substances was targeted to eliminate any discharges, emissions and losses in the long term. In 2009, seven new substances were included. In 2001, the Stockholm Convention on POPs was established as a global treaty under the United Nations Environment Programme (UNEP), and new substances were added in 2009. Mosses are known to accumulate POPs (Harmens *et al.*, in press) and in the 2010/11 ICP vegetation European moss survey six countries have determined the concentration of selected POPs (polycyclic aromatic hydrocarbons (PAHs) in particular) in mosses to assess spatial patterns of POPs deposition to vegetation.

1.3 Workplan items for the ICP Vegetation in 2012

For the first time the Executive Body of the LRTAP Convention agreed on a biannual workplan at its 29th meeting in December 2011 (see ECE/EB.AIR/109/Add.2). Here we will report on the following workplan items for the ICP Vegetation in 2012:

- Supporting evidence for ozone impacts on vegetation;
- Impacts of ozone on carbon sequestration, including linkages between ozone and climate change;
- Progress with European heavy metals and nitrogen in mosses survey 2010/11;
- Relationship between (i) heavy metal and (ii) nitrogen concentrations in mosses and their impacts on terrestrial ecosystems.

In addition, the ICP Vegetation was requested to report on the following common workplan items of the WGE:

- Further implementation of the Guidelines on Reporting of Monitoring and Modelling of Air Pollution Effects;
- Final version of the impact analysis by the WGE;
- Ideas and actions to enhance the involvement of EECCA/SEE countries in the Eastern Europe, the Caucasus and Central Asia and on cooperation with activities outside the Air Convention.

The remaining items agreed in the biannual workplan for 2012-2013 will be reported in 2013 (see Section 6.2).

Progress with most of the above workplan activities is described in Chapter 3. In Chapter 4, the impacts of ozone on carbon sequestration are described and Chapter 5 provides a review on available knowledge on the relationship between i) heavy metal and ii) nitrogen concentrations in mosses and impacts on terrestrial ecosystems. Finally, new activities of the ICP Vegetation are described in Chapter 6, including the medium-term workplan for 2013 – 2015 (up-dated at the 25th ICP Vegetation Task Force Meeting, 31 January – 2 February 2012, Brescia, Italy).

2 Coordination activities

2.1 Annual Task Force meeting

The Programme Coordination Centre organised the 25th ICP Vegetation Task Force meeting, 31 January – 2 February 2012 in Brescia, Italy, in collaboration with the local host at the Ecophysiology and Environmental Physics Laboratory - Mathematics and Physics Department, Università Cattolica del Sacro Cuore. The meeting was attended by 73 experts from 21 countries, including 19 Parties to the LTRAP Convention and guests from Egypt and South Africa. A book of abstracts, details of presentations and the minutes of the 25th Task Force meeting are available from the ICP Vegetation web site (<http://icpvegetation.ceh.ac.uk>).

The Task Force discussed the progress with the workplan items for 2012 (see Section 1.3) and updated the medium-term workplan for 2013 - 2015 (see Section 6.2) for the air pollutants ozone, heavy metals, nutrient nitrogen and POPs. In addition, the ozone expert groups established in 2011 (Harmens *et al.*, 2011a) reported on progress and activities conducted since the 24th ICP Vegetation Task Force meeting in 2011. To support the reporting on impacts of ozone on biodiversity and ecosystems services in 2013, an additional temporary expert group was established to review this theme. The Task Force took note of the conclusions and recommendations from the one-day ozone workshop on 31st January 2012 in Brescia, Italy.

At the workshop presentations were given and discussions were held on the following two themes:

- Quantifying ozone impacts on Mediterranean Forests;
- Mapping vegetation at risk from ozone at the national scale.

The following conclusions were drawn at the workshop:

- New scientific developments support the current text and conclusions in the Modelling and Mapping Manual, i.e. the flux-based method provides better indicators than the concentration-based method for ozone impact assessments on vegetation;
- Current flux-based critical levels for beech/birch were validated by epidemiological studies on mature trees in Switzerland.

The following recommendations were made at the workshop:

- Further field-based validation of ozone flux-effect relationships and critical levels for vegetation is required via epidemiological studies;
- Validation of DO₃SE with eddy covariance flux data would make a valuable contribution;
- The ozone flux-based method should be further developed by:
 - Expanding the number of species with flux-effect relationships;
 - Standardising the method of up-scaling for mature trees;
 - Further qualifying and quantifying uncertainties;
 - Developing a standard protocol for estimating the maximal stomatal conductance (g_{max});
 - Including effects of biogenic volatile organic compounds (BVOC);
 - Further developing the Ball–Berry photosynthesis model in DO₃SE;
 - Further stimulating the cooperation with ICP Forests and make use of their available data;
 - Adding a new technical annex to chapter 3 of the Modelling and Mapping Manual with flux parameterisations for additional species.

Some of these recommendations are already being addressed in the European Framework 7 project 'ECLAIRE' (Effects of Climate Change on Air Pollution and Response Strategies for European Ecosystems; <http://www.eclair-fp7.eu/>) which includes contributions from several ICP Vegetation participants and other LRTAP Convention bodies.

At the Task Force meeting, participants of the European moss survey reported on progress with data analysis and submission of the 2010/11 survey on heavy metals, nitrogen and POPs. They are keen

to conduct the next European moss survey in 2015 (depending on available national funds). The chair reiterated that continued and additional participation from countries in Southern-Eastern Europe (SEE), Eastern Europe, Caucasus and Central Asia (EECCA), with potential outreach to other parts of Asia, is highly desirable.

The Task Force acknowledged and encouraged further fruitful collaborations with the bodies and centres under the Steering Body to EMEP, in particular EMEP/MSC-West, EMEP/MSC-East, the Task Force on Integrated Assessment Modelling and the Task Force on the Hemispheric Transport of Air Pollution, and bodies under the Working Group of Strategies and Review, in particular the Task Force on Reactive Nitrogen. In addition, the Task Force encouraged further development of outreach activities to other regions in the world (see Section 3.1.3).

Over the years participation in the ICP Vegetation and attendance of the Task Force meetings has been rising (**Figure 2.1**). Originally named as the ICP Crops, focussing on the impacts of ozone on crops, the programme started to incorporate impacts on (semi-)natural vegetation later on and therefore gained its current name in the mid-1990s. In 2001, the ICP Vegetation took over the coordination of the European moss survey on heavy metals from the Nordic Council of Ministers and therefore widened its scope and further enhanced participation in its activities.

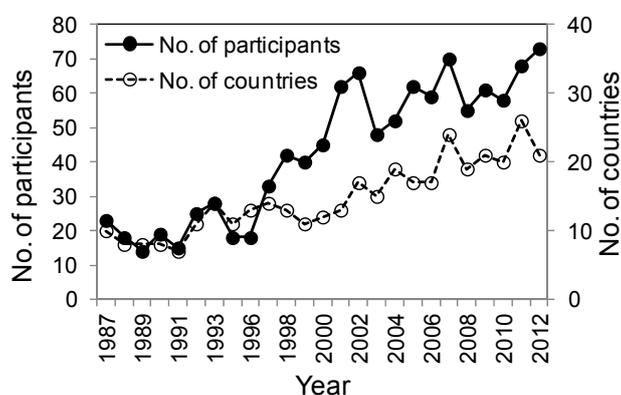


Figure 2.1 Participation in ICP Vegetation Task Force meetings since 1987.

The **26th Task Force meeting** will be hosted by IVL - Swedish Environmental Research Institute, and will be held in Halmstad, Sweden, from 28 - 31 January 2013.

2.2 Reports to the LRTAP Convention

The ICP Vegetation Programme Coordination Centre has reported progress with the 2012 workplan items in the following documents for the 31st session of the WGE (<http://www.unece.org/index.php?id=24661>):

- ECE/EB.AIR/WG.1/2012/3: Joint report of the ICPs, Task Force on Health and Joint Expert Group on Dynamic Modelling;
- ECE/EB.AIR/WG.1/2012/8: Effects of air pollution on natural vegetation and crops (technical report from the ICP Vegetation);
- ECE/EB.AIR/WG.1/2012/13: 2012 impact assessment: effects indicators as tools to evaluate air pollution abatement policies (see Section 3.1.2);

The ICP Vegetation also contributed to Guidance Document VII on health and environmental improvements from the WGE, submitted to the 30th meeting of the Executive Body, 30 April – 4 May 2012 (informal document 3) for the revision of the Gothenburg Protocol.

In addition, the Programme Coordination Centre for the ICP Vegetation has:

- published the final glossy report and a summary brochure on 'Ozone pollution: A hidden threat to food security' (Mills and Harmens, eds, 2011);
- published a glossy report on 'Ozone pollution: Impacts on carbon sequestration in Europe' (Harmens and Mills, eds, 2012), see Chapter 4;
- published the current annual report on line;
- contributed to the colour brochure of the WGE on 'Impacts of air pollution on human health, ecosystems and cultural heritage' and facilitated printing of the brochure in three languages (English, French and Russian) with a contribution in kind from the Swiss Federal Office for the Environment (FOEN);
- contributed to the final report on 'Impacts of air pollution on ecosystems, human health and materials under different Gothenburg Protocol scenarios to support the revision of the Gothenburg Protocol' (see also ECE/EB.AIR/WG.1/2012/13);
- published a leaflet on 'Mosses as biomonitors of atmospheric heavy metal pollution in Europe' (also available in Russian).

2.3 Scientific papers

The following papers have been published or are in press:

Grünhage, L., Pleijel, H., Mills, G., Bender, J., Danielsson, H., Lehmann, Y., Castell, J.F., Bethenod, O. (2012). Updated stomatal flux and flux-effect models for wheat for quantifying effects of ozone on grain yield, grain mass and protein yield. *Environmental Pollution* 165: 147-157.

Harmens, H., Norris, D. A., Cooper, D.M., Mills, G., Steinnes E., Kubin, E., Thöni, L., Aboal, J.R., Alber, R., Carballeira, A., Coşkun, M., De Temmerman, L., Frolova, M., Frontasyeva, M., González-Miqueo, L., Jeran, Z., Leblond, S., Liiv, S., Maňková, B., Pesch, R., Poikolainen, J., Rühling, Å., Santamaria, J. M., Simonè, P., Schröder, W., Suchara, I., Yurukova, L., Zechmeister, H. G. (2011). Nitrogen concentrations in mosses indicate the spatial distribution of atmospheric nitrogen deposition in Europe. *Environmental Pollution* 159: 2852-2860.

Harmens, H., Ilyin, I., Mills, G., Aboal, J.R., Alber, R., Blum, O., Coşkun, M., De Temmerman, L., Fernández, J.A., Figueira, R., Frontasyeva, M., Godzik, B., Goltsova, N., Jeran, Z., Korzekwa, S., Kubin, E., Kviatkus, K., Leblond, S., Liiv, S., Magnússon, S.H., Maňková, B., Nikodemus, O., Pesch, R., Poikolainen, J., Radnović, D., Rühling, Å., Santamaria, J.M., Schröder, W., Spiric, Z., Stafilov, T., Steinnes, E., Suchara, I., Tabor, G., Thöni, L., Turcsányi, G., Yurukova, L., Zechmeister, H.G. (2012). Country-specific correlations across Europe between modelled atmospheric cadmium and lead deposition and concentrations in mosses. *Environmental Pollution* 166: 1-9.

Harmens, H., Foan, L., Simon, V., Mills, G. (in press). Terrestrial mosses as biomonitors of atmospheric POPs pollution: A review. *Environmental Pollution*.

3 Ongoing research activities in 2011/12

In this chapter, progress made with the common WGE and ICP Vegetation workplan for 2012 is summarised. New ICP Vegetation workplan items in 2012 are described in detail in Chapter 4 and 5.

3.1 Contributions to WGE common workplan items

3.1.1 Further implementation of the Guidelines on Reporting of Air Pollution Effects

Table 3.1 provides an overview of the monitoring and modelling effects reported by the ICP Vegetation according to the Guidelines (ECE/EB.AIR/2008/11).

Table 3.1 Monitoring and modelling effects reported by the ICP Vegetation.

Parameter	Ozone	Heavy metals	Nitrogen	POPs
Growth and yield reduction	X			
Leaf and foliar damage	X			
Exceedance critical levels	X			
Climatic factors	X			
Concentrations in mosses		X	X	X

3.1.2 Contributions to the completed version of the impact analysis by the WGE

Background

To support the revision of the Gothenburg Protocol (finalised in May 2012), the WGE has conducted an analysis on the impacts of air pollution on ecosystems, human health and materials under different emission scenarios. The objectives of this analysis were to:

- Provide information on effects of air pollution on ecosystems, human health and materials to support decisions for the revision of the Gothenburg Protocol;
- Demonstrate application of new science and effects indicators, developed since 1999, to illustrate the potential impact of policy/decisions on the environment, human health and materials;
- Illustrate effectiveness of emission scenarios to improve the environment and human health.

Here we report on the analysis conducted by the ICP Vegetation regarding the impacts of ozone on vegetation. This is an update of the interim analysis reported last year (Harmens *et al.*, 2011a). The update reflects the discussions during the various phases of the negotiations of the Gothenburg Protocol revision. The updated analysis on the risk of ozone impacts on forests is based on scenarios published by IIASA in August 2011 (described in CIAM report 4/2011, Amann *et al.*, 2011). The analysis for the risk of ozone impacts on crops were not updated and are therefore based on scenarios published by IIASA in October 2010 (described in CIAM report 1/2010, Amann *et al.*, 2010).

The updated scenarios and projections applied for forests were:

- NAT2000: historical data for the year 2000 based mainly on national information;
- COB2020: Cost Optimised Baseline for the year 2020. This dataset is generated assuming that only current (2011) legislation still applies in 2020 (comparable to NAT2020 described previously and applied for crops);
- MID2020: assuming a higher ambition level for environmental targets than COB2020;
- MTFR2020: data based on a scenario assuming that all technically feasible technologies are implemented by 2020.

The baseline activity data on energy use, transport, and agricultural activities were issued from different sources, including national submissions to IIASA and from specialized sectorial energy, transport and agricultural models (e.g., PRIMES, TREMOVE and CAPRI). They were then used as input data for the GAINS model with which scenarios were optimised so that emissions control scenarios would achieve environmental targets for human health and environmental impacts (acidification, eutrophication, effect of ground-level ozone) as discussed in the 48th session of the WGSR. MTFR represents the reduction that would be obtained if the most stringent regulations were implemented. Any decision leading to some emission reduction will lead to a situation between the baseline and the MTFR scenario.

For the development of the 1999 Gothenburg Protocol, AOT40² was used to indicate the risk to vegetation of adverse impacts of ozone. Since then, a biologically more relevant impact indicator has been developed, the Phytotoxic Ozone Dose above a threshold Y (POD_Y), which gives a better correlation between the locations where ozone damage was reported in Europe between 1990 and 2006 and maps of ozone flux (POD_Y) than maps of AOT40 (Hayes *et al.*, 2007; Mills *et al.*, 2011a). Recently, new or revised flux-based critical levels were developed for crops (potato, tomato, wheat), trees (beech/birch, Norway spruce) and white clover as a representative species of grasslands and (semi-)natural vegetation (Harmens *et al.*, 2010a; LRTAP Convention, 2010; Mills *et al.*, 2011b).

Crop yield and economic losses based on ozone flux-effects indicators

Using the flux-based approach and NAT scenarios, economic losses due to ozone for wheat were estimated to be 3.2 billion euros in EU27+Switzerland+Norway in 2000 reducing to 1.96 billion euros in 2020 (**Table 3.2**). Although the percentage wheat yield reduction is predicted to decline in 2020, only a very small reduction in the proportion of EMEP grid squares exceeding the critical level is predicted. Proportional reductions in yield and economic value for tomato, an important crop for southern areas, were similar to those for wheat for NAT2020 compared to NAT2000. Further details can be found in a report describing the impacts of ozone on food security (Mills and Harmens, 2011).

Table 3.2 Predicted impacts of ozone pollution on wheat and tomato yield and economic value, together with critical level exceedance in EU27+Switzerland+Norway in 2000 and 2020 under the current legislation scenario (NAT scenario). Analysis was conducted on a 50 x 50 km EMEP grid square using crop values in 2000 and an ozone stomatal flux-based risk assessment.

Crop	Wheat		Tomato	
	NAT2000	NAT2020	NAT2000	NAT2020
Emission scenario				
Economic losses (billion Euro)	3.20	1.96	1.02	0.63
Percentage of EMEP grid squares exceeding critical level*	84.8	82.2	77.8	51.3
Mean yield loss (%)*	13.7	9.1	9.4	5.7

* Calculated for the grid squares where the crop is grown.

Mapping risk of ozone impacts on forest growth: application of flux-based methodology

Comparison of ozone risk maps for forests applying the different scenarios and projections shows that despite the predicted reductions in stomatal fluxes in the future, large areas in Europe will remain at risk from adverse impacts of ozone on forest growth, with areas at highest risk being predicted in parts of western, central and southern Europe (**Figure 3.1**). In **Figure 3.2**, the proportion of grid squares in each category illustrated on the maps is shown for the four scenarios. Although for the 2020 scenarios there is a decrease in the proportion of grid squares in the highest categories, there

² The sum of the differences between the hourly mean ozone concentration (in ppb) and 40 ppb for each hour when the concentration exceeds 40 ppb, accumulated during daylight hours.

remains a high proportion of grid squares in the middle to high categories (25 – 26% and 11 – 16% for a POD_1 of 24 – 28 $mmol\ m^{-2}$ and 28 – 32 $mmol\ m^{-2}$ respectively), indicating a continuing risk of damage. Hence, additional measures to reduce the emissions of ozone precursors will be required to protect large areas in Europe from adverse impacts of ozone on forests in 2020.

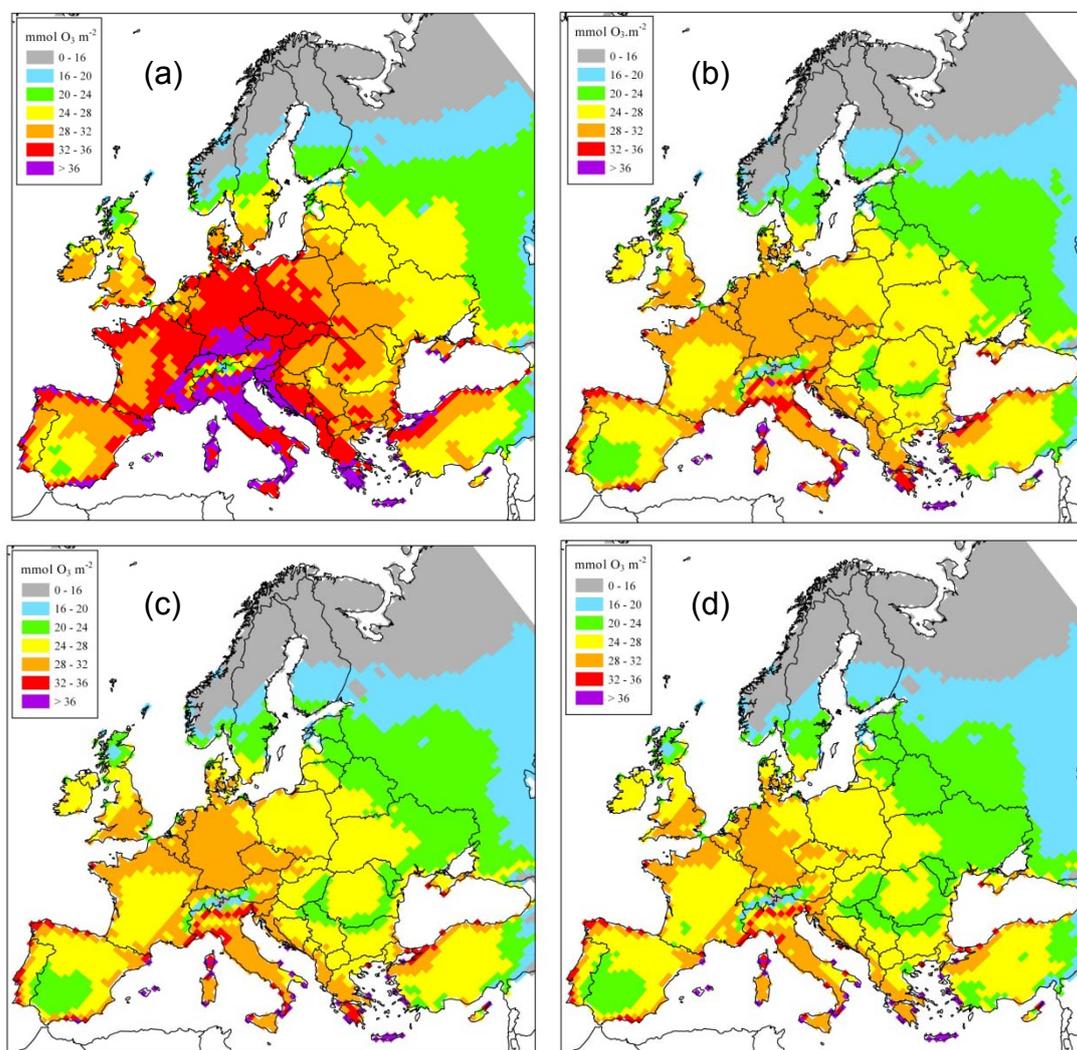


Figure 3.1. The risk of adverse ozone impacts in on biomass production in forest using the generic deciduous tree flux model (POD_1) for a) 2000 (NAT2000) and 2020: b) COB2020, c) MID2020, and d) MTFR2020.

For further details we refer to the full impacts report produced by the WGE (see http://www.unece.org/fileadmin/DAM/env/documents/2012/EB/n_14_Report_WGE.pdf), with a summary of the results being reported in ECE/EB.AIR/WG.1/2012/13. In collaboration with the other ICPs and Task Force on Health, the ICP Vegetation Programme Coordination Centre also produced a glossy brochure on 'Impacts of air pollution on human health, ecosystems and cultural heritage', summarising the results, conclusions and recommendations (see <http://icpvegetation.ceh.ac.uk>). The brochure is also available in French and Russian.

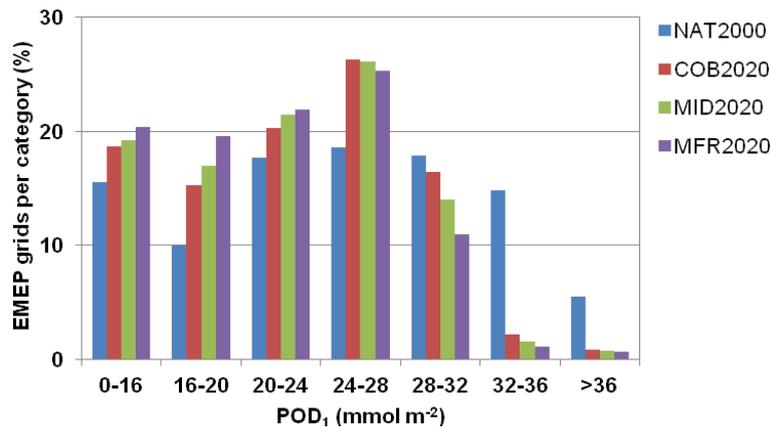


Figure 3.2 Proportion of grid squares within specified categories of POD₁ calculated using the generic deciduous tree flux model as calculated with the different datasets and scenarios.

3.1.3 Ideas and actions to enhance the involvement of EECCA/SEE countries in the Eastern Europe, the Caucasus and Central Asia and on cooperation with activities outside the Air Convention

Working with the lead participant of the European moss survey in the Russian Federation, the ICP Vegetation is actively encouraging the participation of more EECCA/SEE countries. For example, Albania took part for the first time in the moss survey in 2010/11 and attended the ICP Vegetation Task Force meeting for the first time in 2012. Every year we try to find funds for experts in EECCA/SEE countries to participate in our annual Task Force meeting. In 2012, a short leaflet was produced on the results of the 2005/6 European moss survey, which was translated into Russian for distribution in EECCA countries. The glossy brochure on 'Impacts of air pollution on human health, ecosystems and cultural heritage' (see Section 3.1.2) was translated into Russian and widely distributed within the Convention and by contacts in EECCA countries.

Via the Stockholm Environment Institute (SEI) in York (UK), which hosts the secretariat of the Global Atmospheric Pollution Forum, the ICP Vegetation has continued to encourage collaboration with South Asia, particularly with Malé Declaration countries. For example, the ICP Vegetation has produced a position paper on outreach activities with the Malé Declaration, which was presented at the Third Meeting of the Task Force on Future Development of Malé Declaration on Control and Prevention of Air Pollution and its Likely Transboundary Effects for South Asia (Malé Declaration), held on 9-10 August 2012 in Chonburi, Thailand. Suggestions were provided and discussed for the near-term collaboration (next three years) between the two regional atmospheric pollution programmes. The Malé Declaration showed a general interest in intensifying the collaboration with the LRTAP Convention as outlined in the position paper submitted by the ICP Vegetation and SEI, but expressed the need for funding to make this collaboration happen. Furthermore, the ICP Vegetation has developed collaboration with experts in Egypt, South Africa, Cuba and Japan, who have attended recent Task Force meetings of the ICP Vegetation. Further collaboration is often hindered by the lack of available funds.

3.2 Progress with common ICP Vegetation workplan items

3.2.1 Supporting evidence for ozone impacts on vegetation

Since 2008, participants of the ICP Vegetation have been conducting biomonitoring campaigns using ozone-sensitive (S156) and ozone-resistant (R123) genotypes of *Phaseolus vulgaris* (Bush bean, French Dwarf bean) that had been selected at the USDA-ARS Plant Science Unit field site near Raleigh, North Carolina, USA. The bean lines were developed from a genetic cross reported by Dick

Reinert (described in Reinert and Eason (2000)). Individual sensitive (S) and tolerant (R) lines were identified, the S156 and R123 lines were selected, and then tested in a bioindicator experiment reported in Burkey *et al.* (2005). A trial of this system occurred in central and southern parts of Europe during the summer of 2008. This was extended in 2009 and included again in the ozone biomonitoring programme for 2010 and 2011.

In 2011, the biomonitoring of ozone effects using bean was scaled down compared to the previous two years, reflecting less interest from the participants. Nevertheless, experiments were conducted with ozone-sensitive and ozone-resistant bean (*Phaseolus vulgaris*) at nine sites across Europe and one in the USA. As in previous years, bean seeds of the strains S156 and R123 were kindly provided by Kent Burkey (USA). Seeds of both varieties and an updated experimental protocol (ICP Vegetation, 2011) were sent out to participants who recorded the occurrence of visible injury to leaves and quantified the reduction in pod yield of the sensitive compared to the resistant variety for plants exposed to ambient ozone. Some participants carried out stomatal conductance measurements to contribute to the development of a flux-effect model.

The data from the 2011 and previous biomonitoring and ozone exposure experiments conducted in 2008, 2009 and 2010 were combined in a database for dose-response analysis. The database contains data from Belgium, France, Germany (3 sites), Greece (2 sites), Hungary (2 sites), Italy (3 sites), Japan, Slovenia (2 sites), Spain (3 sites), South Africa, UK (2 sites), Ukraine and the USA. Over 3000 stomatal conductance measurements have been made on the bean plants and used to generate an ozone flux model using the Emberson *et al.* (2000) approach. Over the course of the bean biomonitoring experiment, hourly accumulated ozone fluxes ranged from 4.4 (Bangor, UK) to 18.9 (Seibersdorf, Austria) mmol m^{-2} . Visible leaf injury regularly occurred across the network (**Figure 3.3**), but there was no clear dose-response relationship with concentration-based ozone parameters. Similarly, there was no clear relationship between concentration-based parameters and the ratio of the pod weight for the sensitive to that of the resistant bean. Flux-effect relationships will be explored in the coming year.



Figure 3.3 Locations where ozone injury has been detected on bean between 2008 and 2011.

Overall, the bean biomonitoring system does seem to provide a good indication of the occurrence of ozone concentrations that are high enough to visibly damage plants. As such it is very valuable for use in countries as proof or otherwise that ozone levels are causing damage. However, we are concerned that differences between the sensitive and resistant biotypes are not strong enough for continued application as a biomonitor for yield effects across all climate regions in Europe.

3.2.2 Progress with European heavy metals and nitrogen in mosses survey 2010/11

The European moss biomonitoring network was originally established in 1990 to estimate atmospheric heavy metal deposition at the European scale. The network provides a time-integrated measure of heavy metal and potentially nitrogen deposition from the atmosphere to terrestrial ecosystems (Harmens *et al.*, 2010b; 2011b). It is easier and cheaper than conventional precipitation analysis as it avoids the need for deploying large numbers of precipitation collectors with an associated long-term programme of routine sample collection and analysis. Therefore, a much higher sampling density can be achieved than with conventional precipitation analysis.

Mosses have been collected for element analysis every five years since 1990 and the most recent survey was conducted in 2010/11. A total of 26 countries will submit or have already submitted data on heavy metals, of which 14 countries will also submit (or have submitted) data on nitrogen concentrations in mosses (**Table 3.3**). As a pilot study, six countries have agreed to submit data on POPs concentrations in mosses. A recent review has shown that POPs concentrations in mosses can also be a useful indicator for the atmospheric deposition of selected organic compounds such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Harmens *et al.*, 2011a, in press). In addition, some countries will determine the sulphur concentration in mosses.

Table 3.3 Countries (regions) participating in the European moss survey 2010/11. All twenty six countries will report on heavy metals, 14 countries will report on nitrogen and six countries will report on selected POPs concentration in mosses.

Country	N	POPs
Albania		
Austria	✓	
Belarus		
Belgium	✓	
Bulgaria	✓	
Croatia	✓	
Czech Republic	✓	
Denmark (Faroe Islands)		
Estonia	✓	
Finland	✓	
France	✓	✓
FYR of Macedonia		
Iceland		
Italy (Bolzano)	✓	
Kosovo*		
Norway		✓
Poland	✓	✓
Romania		
Russian Federation		
Serbia		
Slovakia	✓	
Slovenia	✓	✓
Spain (Galicia, Navarra, Rioja)	✓ (Navarra, Rioja)	✓ (Navarra)
Sweden		
Switzerland	✓	✓
Ukraine (Donetsk)		

* For this study considered as a separate region.

4 Impacts of ozone on carbon sequestration and linkages between ozone and climate change

4.1 Introduction

4.1.1 Background

Since the industrial revolution, concentrations of carbon dioxide (CO₂) in the atmosphere have been rising, initially slowly but in recent decades more rapidly (IPPC, 2007). This is primarily due to an increase in fossil fuel burning associated with population growth and enhanced social and economic development. In recent decades deforestation, especially in the tropics, has also contributed considerably to the rise in atmospheric CO₂ as tropical forests are a major sink for CO₂. The rise in atmospheric CO₂ concentrations has resulted in a rise in the surface temperature of the earth (global warming). In addition to CO₂ increasing, the atmospheric concentrations of other gases contributing to global warming (greenhouse gases) such as nitrous oxide, methane, halocarbons and ozone have risen too. Depending on future scenarios, the earth's surface temperature is predicted to rise by a further ca. 2 – 4°C by the end of the 21st century. Currently, ozone is considered to be the third most important greenhouse gas, after CO₂ and methane (IPPC, 2007). In contrast to CO₂ and halocarbons, ozone is a short-lived greenhouse gas, so any reductions in ground-level ozone production will reduce atmospheric ozone concentrations within months and hence reduce its contribution to global warming. Long-lived greenhouse gases will stay in the atmosphere for a long time, so even when emissions are kept constant at the 2000 level, a further rise in surface temperature of 0.5°C is predicted by the end of the 21st century. We have investigated how ozone pollution is currently, and likely in the future to continue to be, reducing carbon (C) sequestration in the living biomass of trees (and other vegetation), thereby potentially exacerbating global warming. Here we provide a summary of this study, further details can be found in Harmens and Mills (2012).

4.1.2 Ozone pollution

As well as being a greenhouse gas, ozone is also an important atmospheric pollutant and has adverse effects on human health and the environment. Ozone is a naturally occurring chemical that can be found in both the stratosphere (as the so-called "ozone layer", 10 - 40 km above the Earth) and the troposphere (at "ground level", 0 - 10 km above the Earth). At ground level there is always a background concentration of ozone resulting from natural sources of the precursors and stratospheric incursions. Of concern for human health and vegetation (including C sequestration and food production) is the additional tropospheric ozone which is formed from complex photochemical reactions from fossil fuel burning in industrial and transport activities. As a result of these emissions, there has been a steady rise in the background ozone concentration in Europe since the 1950s to the current 30 – 40 ppb (Royal Society, 2008). Background ozone concentrations in Europe are still rising and predicted to rise until at least 2030, in part due to hemispheric transport of the precursors of ozone from other parts (developed and developing areas) of the world. Background concentrations in Europe have now reached levels where they have adverse impacts on vegetation. During periods of hot dry weather and stable air pressure, ozone episodes occur where concentrations rise above 60 ppb for several days at a time.

4.1.3 Vegetation as a sink for atmospheric CO₂ and ozone

Atmospheric gases such as CO₂, ozone and water vapour are exchanged through microscopic stomatal pores on leaves. This for instance enables plants to fix CO₂ for photosynthesis and hence growth, and to transpire for the adjustment of the internal water balance. The more open the stomata are, the more CO₂ and ozone will enter the plant and the more water will transpire. Ozone entering the plant has the potential to damage plant cells by forming reactive oxygen species, which can lead to detrimental effects on photosynthesis and growth and/or ultimately to cell death. The magnitude of these damaging effects depends on the plant species and genotype, concentration of ozone, duration

of exposure, climate and soil conditions. Plants are able to detoxify a certain amount of ozone, but above this amount damage to vegetation is likely to occur, either as acute damage due to exposure to 'high' ozone concentrations that usually occur during ozone episodes or as chronic damage due to prolonged exposure to elevated background ozone concentrations. Hence, terrestrial vegetation is considered an important sink for the greenhouse gases CO₂ and ozone. However, if ozone concentrations are high enough to reduce photosynthesis (i.e. CO₂ fixation) and/or above-ground plant growth, then less CO₂ and ozone will be taken up by the vegetation, leading to a positive feedback to atmospheric CO₂ and ozone concentrations and therefore global warming (Sitch *et al.*, 2007).

4.1.4 Ozone impacts in a changing climate

The future impacts of ozone on C sequestration in European terrestrial ecosystems will depend on the interaction with and magnitude of the change of the physical and pollution climate, represented by rising temperatures, increased drought frequency, enhanced atmospheric CO₂ concentration and reduced nitrogen deposition. Ecosystems are inherently complex, and for any one aspect of functioning, there are multitudes of driving factors. Exposure studies on the interaction between ozone and other pollutants (nitrogen) and climate change often show the following:

- *Elevated CO₂ concentrations* – Elevated ozone and CO₂ often affect plant physiology and soil processes in opposite directions. Hence, the overall response and resulting impact on C sequestration might well be cancelled out when both gases are enriched in the atmosphere (Harmens and Mills, 2012).
- *Warming* – A rise in temperature stimulates ozone formation and directly affects the stomatal uptake of ozone since this process is temperature dependent. Warming can also indirectly affect the uptake of ozone via impacts on relative humidity, plant development and soil water availability, all of which influence the stomatal gas exchange (Emberson *et al.*, 2000). Some studies have shown that atmospheric ozone concentrations modify the response of plant species and genotypes to warming (Kasurinen *et al.*, 2012).
- *Enhanced drought* – It has often been postulated that drought will protect vegetation from ozone damage as the stomatal pores shut down more during periods of drought to prevent water loss. However, the interactions between ozone and drought (mediated via plant hormones) are more complex than first thought and drought might not protect ozone sensitive species from adverse impacts of ozone (Mills *et al.*, 2009; Wilkinson and Davies, 2009, 2010).
- *Nitrogen deposition* – Relatively few studies have investigated the impacts of both ozone and nitrogen on vegetation. Evidence suggests that ozone and nitrogen can have both synergistic and antagonistic effects on species and ecosystem processes, and that they may interact in unpredictable ways to affect plant communities (Harmens *et al.*, 2006).

Relatively few studies have investigated the interactive impacts of two or more drivers of change. The outcome of such studies often indicates complex interactions and non-linearity in responses. There is an urgent need for more field-based, larger scale experiments where vegetation is exposed to multiple drivers of climate change for several years (at least one decade) to further investigate the overall impact of a combination of drivers of change on terrestrial ecosystems. Modelling studies to predict future impacts of change should also be based on a multifactorial approach. So far, the impacts of ozone on vegetation and feedbacks to the climate have hardly been considered in global climate modelling. Recent modelling studies have shown that the indirect impact of ozone on global warming via its impacts on vegetation might be contributing as much to global warming as its direct effect as a greenhouse gas (Sitch *et al.*, 2007).

4.2 Impacts of ozone on C sequestration in the living biomass of trees

4.2.1 First flux-based assessment for Europe for the current (2000) and future climate (2040)

The DO₃SE (Deposition of Ozone for Stomatal Exchange) model (Emberson *et al.*, 2000) was applied by Büker, Emberson and colleagues (SEI-York) to estimate the magnitude of the impact of ambient ozone on C storage in the living biomass of trees. The Phytotoxic Ozone Dose above a threshold value of $Y \text{ nmol m}^{-2} \text{ s}^{-1}$ (POD_Y) was calculated applying known flux-effect relationships for various tree species (LRTAP Convention, 2010; Mills *et al.*, 2011b).

The following input data were used (Harmens and Mills, 2012):

- i) Ozone and meteorological data provided by EMEP for the year 2000 (Simpson, pers. comm.), and ii) ozone and climate data provided by the Rossby Centre regional Atmospheric climate model (RCA3) for current (2000) and future (2040) years (Kjellström *et al.*, 2005).
- Land cover data to identify the distribution of forest tree species: i) for EMEP data the species-specific JRC land cover data (<http://forest.jrc.ec.europa.eu/distribution>) and for ii) RCA data the UNECE Long-Range Transboundary Air Pollution (LRTAP) Convention harmonised land cover data were used (Cinderby *et al.*, 2007).
- Forest C stock data were derived from the European forests inventory dataset (Forest Europe, 2011).

In addition, for the year 2000 using EMEP ozone and meteorological data, the application of generic parameterisations for trees in DO₃SE (POD₁) were compared with the application of climate region specific parameterisations (Emberson *et al.*, 2007), with a mixture of POD₁ and POD_{1.6}, (Karlsson *et al.*, 2007) and a deactivated soil moisture deficit (SMD; Büker *et al.*, 2011) module (POD₁), i.e. no limitation of soil moisture on stomatal conductance and hence ozone flux (no influence of drought). Reductions in C stock due to ozone were calculated from the potential C stock present in both years should there have been no impact of ozone on the C stock. This calculation assumes that the trees were exposed to the same ozone flux as in 2000 or 2040 during the build-up of the C stock.

The 2040 scenario runs used the GEA-LOW-CLE emissions generated by IIASA for the year 2050 (<http://cityzen-project.eu>), together with RCA meteorology for 2040-2049. Thus, both emissions and meteorology were changed. The GEA-LOW-CLE emission scenario is based on the illustrative scenario of the GEA Efficiency pathway group in terms of energy demand and use, and the implementation of a stringent climate policy corresponding to a maximum of 2 °C rise in global temperature target. In addition, this scenario assumes global implementation of extremely stringent pollution policies (SLE) until 2030. These stringent air quality control strategies are much more ambitious than the currently planned legislations, but are still lower than the so called Maximum Feasible Reduction (MFR) which describes the technological frontier in terms of possible air quality control strategies by 2030.

Table 4.1 Estimated percentage reduction of C storage in the living biomass of trees due to ozone in 2000 and 2040 in the EU27 + Norway + Switzerland. SMD_{off} = soil moisture deficit module switched off; POD_Y = Phytotoxic Ozone Dose above a threshold value of $Y \text{ nmol m}^{-2} \text{ s}^{-1}$. The reduction is calculated from the potential C stock present in both years should there have been no impact of ozone on the C stock.

Modelled input data	Year	Parameterisation DO ₃ SE model	POD _Y	Reduction C storage (%)
EMEP	2000	Generic	POD ₁	12.0
	2000	Climate region-specific	POD _{1/1.6}	13.7
	2000	SMD _{off}	POD ₁	17.3
RCA3	2000	Generic	POD ₁	16.2
	2040	Generic	POD ₁	12.6

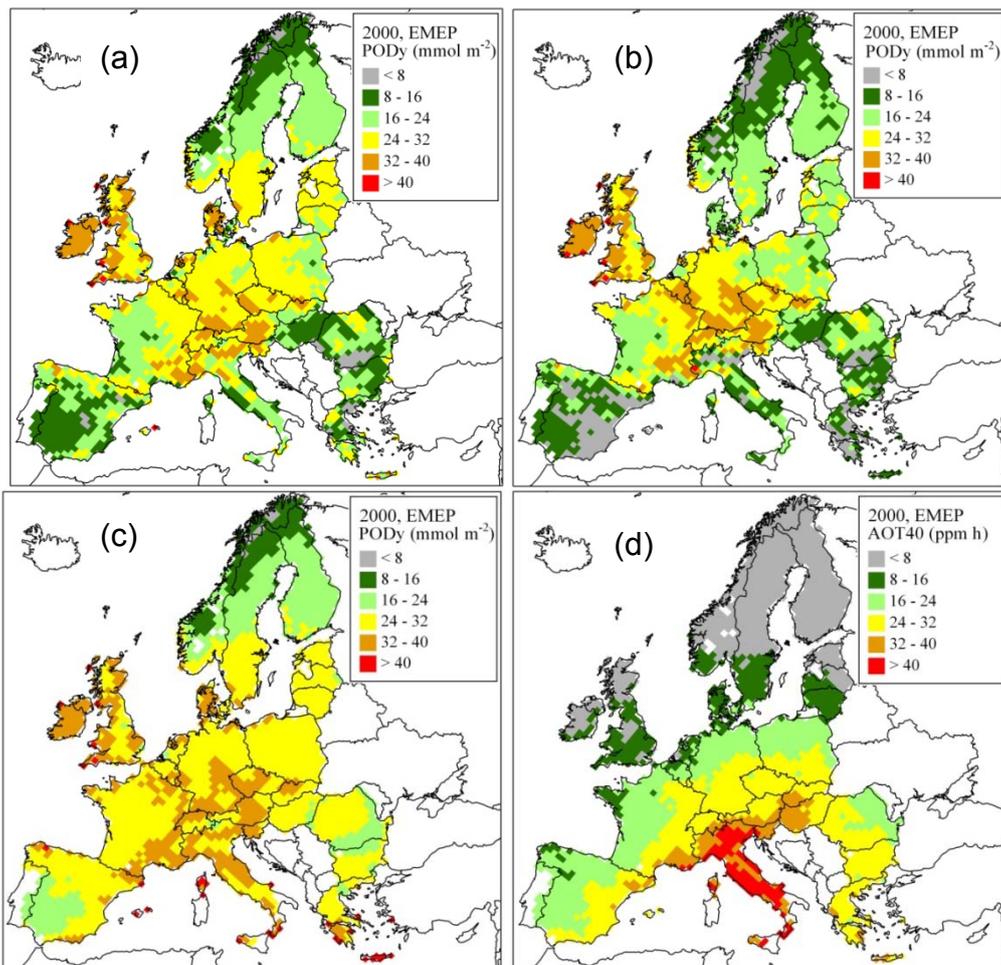


Figure 4.1 POD_Y in 2000 calculated from EMEP input data and applying the following parameterisations in DO_3SE : (a) generic parameterisation ($Y = 1 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$), (b) climate region specific parameterisation (Y is a mixture of 1 and $1.6 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$), and (c) generic parameterisation with soil moisture module switched off (i.e. no soil water limitations). For comparison, (d) AOT40 in 2000 is also shown (Harmens and Mills, 2012).

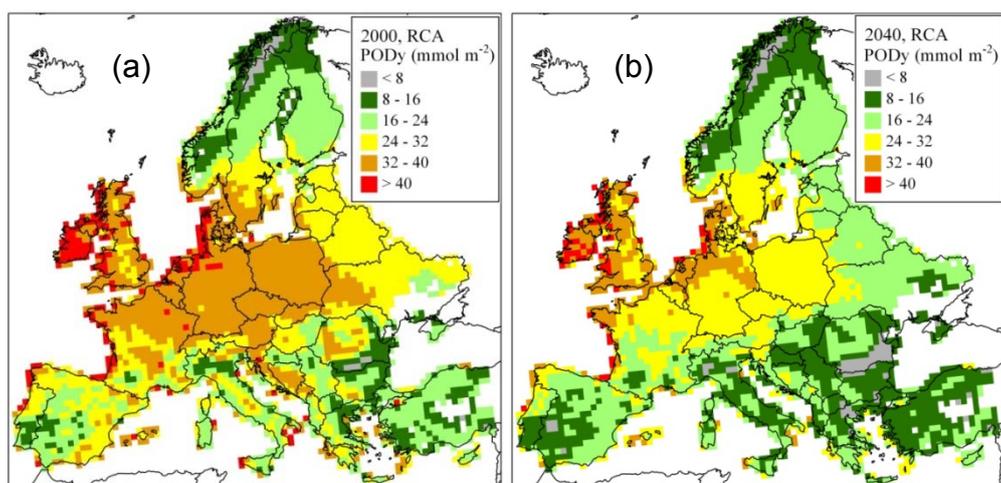


Figure 4.2 POD_Y in (a) 2000 and (b) 2040, calculated from RCA input data and applying the generic parameterisation in DO_3SE ($Y = 1 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$; Harmens and Mills, 2012).

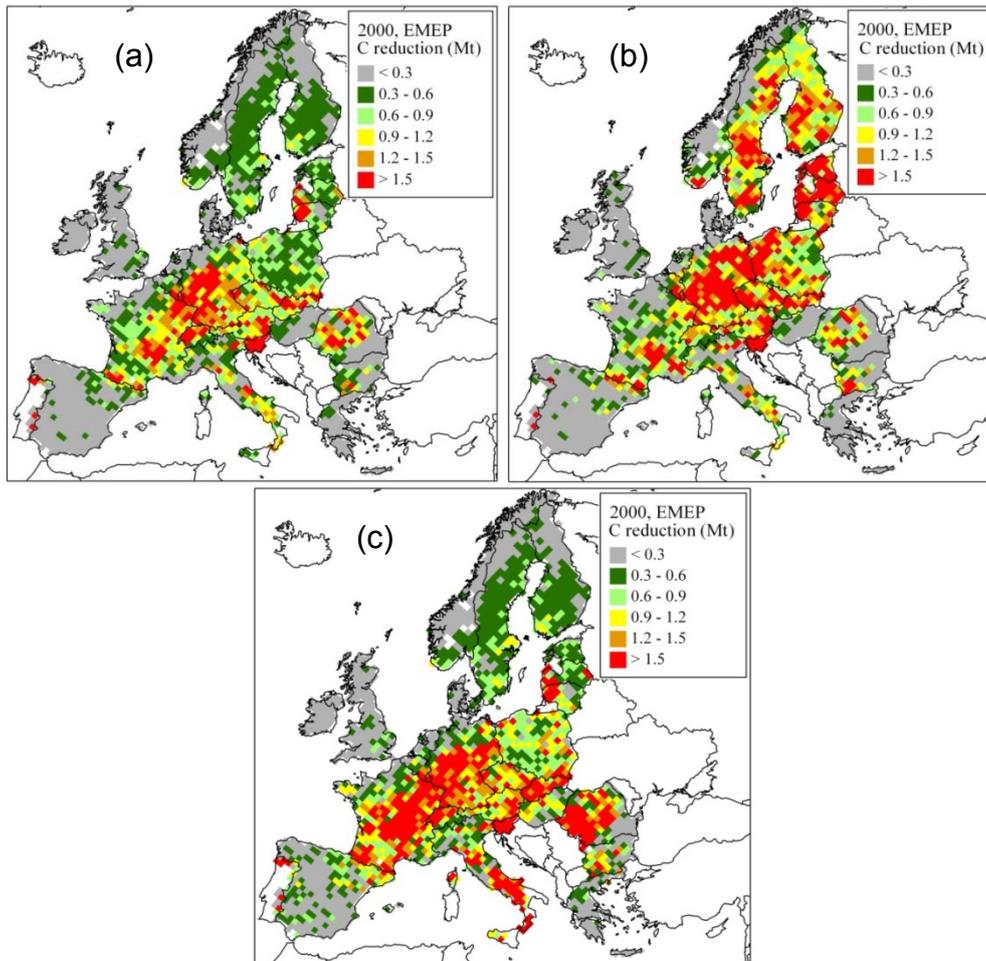


Figure 4.3 Absolute reduction (Mt C) in C storage in the living biomass of trees due to ozone in 2000, applying POD_Y calculated from EMEP input data and applying the following parameterisations in DO_3SE : (a) generic parameterisation ($Y = 1 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$), (b) climate region specific parameterisation (Y is a mixture of 1 and $1.6 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$), and (c) generic parameterisation with soil moisture module switched off (i.e. no soil water limitations). The reduction is calculated from the potential C stock present in both years should there have been no impact of ozone on the C stock (Harmens and Mills, 2012).

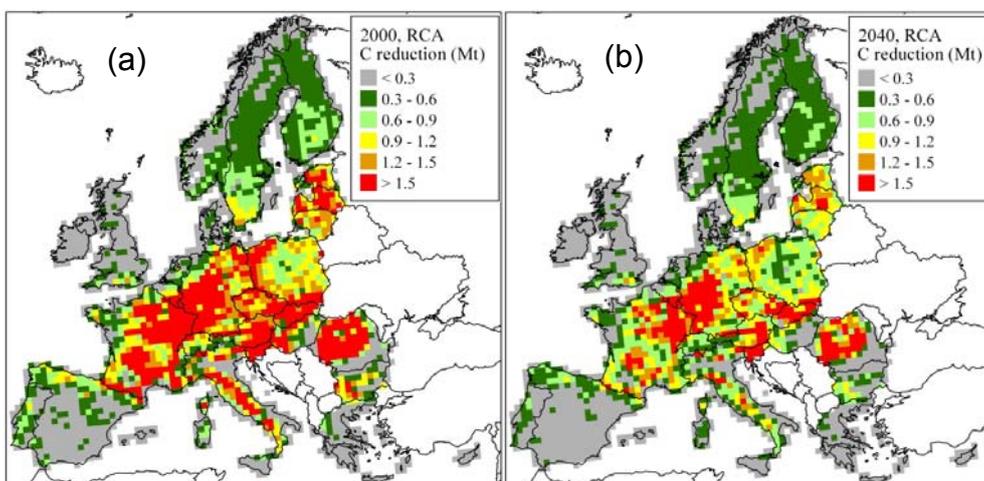


Figure 4.4 Absolute reduction (Mt C) in C storage in the living biomass of trees due to ozone applying POD_Y in (a) 2000 and (b) 2040, calculated from RCA input data and applying the generic parameterisation in DO_3SE ($Y = 1 \text{ nmol m}^{-2} \text{ PLA s}^{-1}$). The reduction is calculated from the potential C stock present in both years should there have been no impact of ozone on the C stock (Harmens and Mills, 2012).

The main results are (**Table 4.1, Figures 4.1 - 4.4**):

- When applying the flux-based methodology and a generic parameterisation for deciduous and conifer trees, a reduction of C sequestration in the living biomass of trees by 12.0 (EMEP input data) to 16.2% (RCA input data) was calculated. The flux-based approach indicates a high risk of ozone impacts on forests in Atlantic and Continental Central Europe, and also a considerable risk in southern parts of northern Europe (in comparison with the concentration based approach).
- The climate-region specific parameterisation for 2000 revealed slightly higher C reductions (13.7%) due to ozone compared to the generic parameterisation (12.0%) for calculating POD_Y .
- The deactivation of the soil moisture deficit module of the DO_3SE model, which simulates drought-free stomatal ozone uptake conditions throughout Europe, led to an increase in C reduction, especially in the warmer and drier climates in Central and Mediterranean Europe.
- Although a decline in stomatal ozone flux was predicted in 2040, C sequestration in the living biomass of trees will still be reduced by 12.6% (compared to 16.2% in 2000). The decline in stomatal ozone flux in 2040 is mainly a result of a predicted reduction in atmospheric ozone concentrations across Europe.

Whilst the spatial patterns and temporal trends indicated above can be postulated with a considerable degree of certainty, the absolute values of C reductions have to be interpreted with care. It should be remembered that these are for effects on annual increment in living tree biomass only, and do not take into account any effect on soil C processes, including any direct or indirect ozone effects on below-ground processes that affect the rate of C turnover in the soil. Furthermore, the response functions used were derived for young trees (up to 10 years of age). However, there is scientific evidence from epidemiological studies that the functions are applicable to mature trees within forests too (Braun *et al.*, 2010).

4.2.2 Case study in northern and central Europe applying the AOT40 method

A more detailed study based on relative growth rates of trees was conducted by Karlsson (IVL, Sweden) to assess the impacts of current ambient atmospheric ozone concentration (in comparison to pre-industrial ozone levels in the range of 10 -15 ppb, i.e. $AOT40 = 0$) on C sequestration in the living biomass of trees in temperate and boreal forests (Harmens and Mills, 2012). As exposure-response relationships based on AOT40 are more commonly reported in the literature, the AOT40 method was applied here. The AOT40 values per country used here were annual means for the growing season of trees for the period 2000-2005 and were provided by EMEP. However, it should be noted that the AOT40 approach might underestimate the risk of ozone impacts on vegetation in northern European countries in particular (e.g. Hayes *et al.*, 2007; Mills *et al.*, 2011a; see also above). Using data from forests inventories on forest types, age classes and structure, growth and harvest rates and combining these with AOT40-based dose response relationships for young trees, calculated yearly growth increment values were converted to C stock changes. The estimated percent reduction in the change of the living biomass C stock across forests in ten countries was 10%. However, for different countries these values ranged between 2 and 32% (**Table 4.2**).

The most important factor that determines the changes in the forest living biomass C stock is the gap between growth and harvest rates. If this gap is small, then a certain growth reduction caused by ozone will have a relatively large impact on the C stock change, and vice versa. By far the most important countries for C sequestration in the living biomass C stocks in northern and central Europe are Sweden, Finland, Poland and Germany. Ozone-induced growth reductions will also result in an economic loss for forest owners.

Table 4.2 Estimated reductions in annual C sequestration due to current ambient ozone exposure as compared to pre-industrial ozone levels in northern and central Europe.

Country	Decline (%)	Country	Decline (%)	Country	Decline (%)
Czech Republic	32.0	Finland	2.2	Lithuania	13.8
Denmark	5.8	Germany	12.3	Norway	1.8
Estonia	4.5	Latvia	8.8	Poland	12.8
All countries	9.8			Sweden	8.6

4.2.3 A global perspective of impacts on C storage in terrestrial ecosystems

The JULES (Joint UK Land Environment Simulator) model has been run with ozone fields and observed climatology over the period 1901-2040 to assess the impacts of ozone on the global C and water cycle (Sitch *et al.*, 2007). In JULES, the plant damage due to ozone directly reduces plant photosynthesis, and thereby indirectly, leaf stomatal conductance. With elevated near surface ozone levels, the model simulates decreased plant productivity, and as less CO₂ is required for photosynthesis, reduced stomatal conductance. Therefore, the plant is able to preserve water supplies. However, some recent studies have shown that ozone impairs stomatal functioning such that ozone might enhance rather than reduce stomatal conductance (Mills *et al.*, 2009; Wilkinson and Davies, 2009; 2010). As no direct effect of ozone on stomatal functioning is currently incorporated into JULES, the indirect effect of ozone on stomata via photosynthesis was switched off ('fixed stomata') in the current study to investigate the consequences for the global C and water cycle. In JULES, the ozone flux-based method was applied (Sitch *et al.*, 2007).

Table 4.3 Simulated future percentages changes (% Δ) in carbon (C) and water cycle (runoff) variables globally for three time periods: 1901-2040, 1901-2000 and 2000-2040. GPP = Gross Primary Productivity, Veg = vegetation, Gs = stomatal conductance (Scenario: SRES A2).

1901-2040	% Δ GPP	% Δ VegC	% Δ SoilC	% Δ TotalC	% Δ Runoff	% Δ Gs
Control	-15.4	-10.9	-9.7	-10.0	12.6	-13.3
Fixed stomata	-17.9	-11.8	-10.5	-10.9	1.4	-1.6
2000-2040						
Control	-6.9	-5.0	-4.1	-4.4	4.5	-5.0
Fixed stomata	-8.1	-5.5	-4.6	-4.8	0.6	-0.5
1901-2000						
Control	-9.2	-6.2	-5.8	-5.9	7.7	-8.7
Fixed stomata	-10.7	-6.7	-6.2	-6.4	0.8	-1.1

Applying ozone stomatal flux response relationships in JULES, the model predicted that the reduction in C stored in vegetation is 6.2% globally and almost 4% in Europe in 2000 compared to 1900, and is predicted to rise to 10.9% globally and ca. 5 to 6% in Europe by 2040 (**Table 4.3**) due to a predicted rise in atmospheric ozone concentrations in the future emission scenario applied. As expected, results from the control run suggest a large indirect effect of ozone (via photosynthesis) on stomatal conductance and runoff. Unsurprisingly, stomatal conductance and river runoff changed little through time in the fixed stomata simulation, where the indirect effect of ozone on stomata via photosynthesis was switched off. However, despite the difference in stomatal conductance response between simulations, the differences in the response of the C cycle are rather modest. It can be concluded that in the absence of a direct effect of ozone on stomatal conductance, ozone-vegetation impacts act to increase river runoff and freshwater availability substantially due to a reduced water loss from soil via transpiration from vegetation. However, such an increase might not occur if ozone has adverse impacts on stomatal functioning, reducing their responsiveness to environmental stimuli.

In addition, Sitch and colleagues analysed the impacts of ozone and drought interactions on plant productivity in Europe by applying the climate of the year 2003, which was a very dry year across the

whole of Europe. Large reductions in plant productivity were simulated under drought conditions. The net impact of ozone is to further reduce plant productivity under drought. In the absence of a direct effect of ozone on stomatal conductance, ozone acts to partially offset drought effects on vegetation (Harmens and Mills, 2012).

4.2.4 Recommendations

Policy More stringent reductions of the emissions of precursors of ozone are required across the globe to further reduce both peak and background concentrations of ozone and hence reduce the threat from ozone pollution to C sequestration. It would be of benefit to better integrate policies and abatement measures aimed at reducing air pollution and climate change as both will affect C sequestration in the future. Improved quantification of impacts of ozone within the context of climate change is urgently required to facilitate improved future predictions of the impacts of ozone on C storage in the living biomass of trees. Stringent abatement policies aimed at short-lived climate forcers such as ozone provide an almost immediate benefit for their contribution to global warming.

Research There is an urgent need for more field-based, larger scale experiments where vegetation is exposed to multiple drivers of climate change for several years (at least one decade) to further investigate the overall impact of a combination of drivers of change on C sequestration in terrestrial ecosystems. Further development of the ozone flux-based method and establishment of robust flux-effect relationships are required for additional tree species, in particular for those species representing the Mediterranean areas. Field-based ozone experiments should also include the assessment of ozone impacts on below-ground processes and soil C content. Further epidemiological studies on mature forest stands are required for the validation of existing and new ozone flux-effect relationships. Experiments are needed on the interacting effects of climate change and ozone, including quantifying impacts of reduced soil moisture availability, rising temperature and incidences of heat stress, impacts of rising CO₂ concentrations and declining nitrogen deposition. Impacts of other drivers of change on existing flux-effect relationships should be investigated. Further development of climate region-specific parameterisations for flux models is needed to improve the accuracy of predictions. Existing flux models (e.g. DO₃SE) will have to be further developed to include more mechanistic approaches for the accurate prediction of combined effects of ozone, other pollutants and climate change, on various plant physiological processes and hence C sequestration.

There is an urgent need to further include ozone as a driver of change in global climate change modelling to quantify its impact (either directly or indirectly via impacts on vegetation) on global warming. Such modelling should further investigate the mechanisms of interactions between ozone and other drivers of global warming. Finally, there is a need to quantify the economic impacts of ozone on forest growth in order to establish the economic consequences for the wood industry. For enhanced C storage in the living biomass in the future, the ozone-sensitivity of tree species and varieties should be considered as a factor in future breeding and forests management programmes.

5 Relationship between (i) heavy metal and (ii) nitrogen concentrations in mosses and their impacts on ecosystems

5.1 Heavy metals

Some heavy metals (Co, Cu, Fe, Mo, Mn, Ni, Zn) play an essential role in cell metabolism, whereas others (e.g. Cd, Hg, Pb) are not known to be essential for life. Essential heavy metals are needed in small quantities only (micronutrients), and both essential and non-essential heavy metals are potentially toxic when they are available in excess for uptake by organisms in the environment (e.g. Woolhouse, 1983; Sánchez, 2008; Boyd, 2010). Most likely, the ability of heavy metal ions to bind strongly to O, N and S atoms is the basis of their toxicity (Borovok, 1990). It has been shown that metals can modify chemical communication between individuals, resulting in 'info-disruption' that can affect animal behaviour and social structure and hence intraspecies and interspecies interactions, however 'info-disruption' by metals in terrestrial habitats is not well studied (Boyd, 2010). The atmospheric deposition of heavy metals increased since the industrial revolution, but in recent decades the deposition of heavy metals in Europe has begun to decline (Travnikov *et al.*, 2012) due to the use of cleaner fuel in combustion processes (e.g. less coal and more gas, unleaded petrol), implementation of air pollution abatement policies (applying the latest technology to filter emissions at the source) and closing down many heavy polluting sources in parts of Europe.

Terrestrial mosses primarily receive heavy metals from atmospheric deposition as they lack a root system. In the last three decades, mosses have been applied successfully as biomonitors of heavy metal deposition across Europe (Harmens *et al.*, 2008; 2010b). Detailed literature reviews on the application of mosses as biomonitors of heavy metals have been conducted by Burton (1990), Tyler (1990), Onianwa (2001) and Zechmeister *et al.* (2003). Although the heavy metal concentration in mosses provides no direct quantitative measurement of deposition, this information can be derived by using regression or correlation approaches relating the measured heavy metal concentrations in mosses to deposition data (e.g. Berg and Steinnes, 1997; Berg *et al.*, 2003; Zechmeister *et al.*, 2004). Recently, Bouquete *et al.* (2011) recommended that the results of moss biomonitoring studies should be regarded as qualitative or semi-qualitative, rather than attempting to provide absolute data, which may not be temporally representative, and may have a high degree of uncertainty associated with them.

Here we discuss whether there is any field-based evidence for a relationship between heavy metal concentrations in terrestrial mosses and impacts of heavy metals on terrestrial ecosystems. Many studies have demonstrated that the highest metal concentrations in mosses are often found within 500–2000 meters of emission sources, showing a significant decreasing gradient over this distance (Türkan *et al.*, 1995; Fernández *et al.*, 2000, 2007; Salemaa *et al.*, 2004; Santamaria *et al.*, 2010), although values higher than background levels have been obtained at distances of over 20 km from the industry (Zechmeister *et al.*, 2004; Schintu *et al.*, 2005). Heavy metal deposition near roads generally declines to background levels within 250 m distance from the road, however elevated deposition can be observed up to 1000 m from very busy roads (Zechmeister *et al.*, 2005).

In the scientific literature there is a lack of a direct comparison between heavy metal concentrations in mosses and impacts on ecosystems. Impacts of heavy metals on terrestrial ecosystems are often most pronounced in areas close to pollution sources (such as heavy metal industry and mines), with impacts declining with distance from the pollution source. Similarly, heavy metal concentrations in mosses tend to decline in a gradient away from pollution sources (Zechmeister *et al.*, 2003), often exponentially (Zechmeister *et al.*, 2004). Some studies have made an indirect comparison, for example, Santamaria *et al.* (2012) reported that the declining gradient of heavy metal concentrations in mosses away from a pollution source (González-Miqueo *et al.*, 2010) coincided with an increase of the abundance of soil mesofauna.

The European moss survey aims to provide an indication of the deposition of heavy metals away from pollution sources, primarily in rural areas, and the contribution of long-range transport to heavy metal deposition to vegetation. In agreement with the decline in the annual deposition of heavy metals in recent decades across Europe (Travnikov *et al.*, 2012), the heavy metal concentration in mosses has also declined (Harmens *et al.*, 2010b). Heavy metal concentrations in mosses tend to reflect the accumulated heavy metal deposition over a growing period of two to three years. Hence, they provide no indication of historical accumulation of heavy metals in soils over a longer period. However, temporal trends can be determined by repeated sampling of mosses in time (Harmens *et al.*, 2010b).

Although deposition of heavy metals to above-ground plant parts can lead to uptake via the leaves (Harmens *et al.*, 2005), the risk of heavy metal toxicity to terrestrial ecosystems is often expressed as a function of the free metal ion concentration in soil solution. The LRTAP Convention has developed the critical loads approach based on established critical limits of heavy metals in soil solution (UBA, 2004). These critical limits are based on no-observed effect concentration (NOEC), often determined for single metals in standardised laboratory conditions for specific indicator species of toxicity. Little is known about the toxicity of metal mixtures in soil solutions and hence the NOEC for metal mixtures. Exceedance of the critical loads provide an indication of the risk of adverse impacts of heavy metals on terrestrial ecosystems. Hettelingh and Sliggers (2006) and the Task Force on Heavy Metals (2006) concluded that available information on the metals chromium, nickel, copper, zinc, arsenic and selenium suggests that none of these metals achieve high enough concentrations as a result of long-range atmospheric transport and deposition to cause adverse effects on terrestrial ecosystems. However, although the area of exceedance of the critical loads for these heavy metals is small, even small exceedances may result in effects in the future due to the accumulative nature of heavy metals in soils. These results support the focus of the 1998 Aarhus Protocol on Heavy Metals on the metals cadmium, lead and mercury.

In 2000, the European ecosystem area at risk of adverse impacts of cadmium, lead and mercury was estimated to be <1, 42 and 77% respectively (Hettelingh and Sliggers, 2006) whereas in 2010 it was estimated to be <1, 15 and 71% (Slootweg *et al.*, 2010). However, hardly any field-based evidence is available to validate the critical load exceedance calculations for terrestrial ecosystems. Tipping *et al.* (2010) suggested that the critical loads calculations for mercury might overestimate the level of critical load exceedance. In the UK, there was almost no exceedance of the critical load for mercury in 2010. This is a vast improvement from the area of the critical load exceedance in 1970, which was 13% for mercury in rural areas of the UK. Although mercury concentrations in mosses were determined at sites across Europe, data from less sites and countries is available for mercury than for cadmium and lead. Schröder *et al.* (2010b) showed that the correlation between metal deposition rates and concentrations in mosses is rather weak for mercury and considerably lower than for cadmium and lead, indicating that mosses might not be that suitable as biomonitors of mercury deposition or air concentrations. This might be related to the specific chemistry of mercury pollution (Harmens *et al.*, 2010b).

In 2000 and 2010, the highest areas of critical load exceedance for cadmium were estimated in Bulgaria and Macedonia (Hettelingh and Sliggers, 2006). Although these countries also have high levels of cadmium concentrations in mosses (Harmens *et al.*, 2010b), high levels in mosses were also observed in other countries such as Belgium and Slovakia, where hardly any critical load exceedance was estimated. For lead the highest areas of critical load exceedance was calculated for the European part of the Russian Federation in 2000 and 2010, however, data on the lead concentration in mosses is scarce for this region.

One should bear in mind that ecosystems are exposed to different stressors and that it is difficult to disentangle impacts of single stressors in the field. De Zwart *et al.* (2010) made a first attempt to estimate the loss of species due to cadmium and lead depositions in Europe. One of the endpoints for the critical loads of cadmium and lead is the ecotoxicological effect of metal ions in soil solution on soil micro-organisms, plant and invertebrates. Depositions will (eventually) result in a concentration in soil solution in equilibrium with each other, depending on ecosystem properties like leaching, uptake

and soil characteristics such as pH, organic matter and clay content. Toxicity data for soil dwelling organisms and terrestrial plants are comparatively scarce. Hence, De Zwart *et al.* (2010) applied publically available data on acute median lethal or effective concentrations (LC50 or EC50) based on aquatic toxicity tests. There is no indication that the sensitivity of organisms living in the soil is intrinsically different from the sensitivity of organisms living in surface waters, provided that the evaluation is based on the truly bioavailable fraction of the metals. Based on this approach, De Zwart *et al.* (2010) concluded that toxicity effects of cadmium and lead are close to zero in the vast majority of ecosystems across Europe. There is also little evidence of adverse effects at current levels of metal deposition on vegetation in the UK (RoTAP, 2012).

In summary, according to current knowledge, the relatively low levels of heavy metals in mosses (compared to previous decades) due to long-range transport are unlikely to indicate any adverse impact of heavy metals on ecosystems. A straightforward relationship between heavy metal concentrations in mosses and calculated critical load exceedances is not to be expected as the heavy metal concentration in mosses reflect atmospheric deposition of heavy metals whereas critical load exceedances for soil solution is not only determined by heavy metal deposition but also affected by soil characteristics. It should be noted that despite a general decline in heavy metal deposition across Europe in recent decades, metals accumulate in soils and might therefore become a problem in the future if bio-available concentrations reach critical limits in soil solution (UBA, 2004). The risk, estimated by the use of critical limits, is more important for assessing current threats from heavy metals to biota than critical loads, which are relevant at 'steady state', and which may not be achieved for centuries. Changes in soil composition as a result of changes in climate, or mechanical disturbance, may release the stored material in a bioavailable form, and this is one of the largest uncertainties when considering the impact of future climate on heavy metals in the environment (RoTAP, 2012).

5.2 Nitrogen

Nitrogen is a macronutrient and essential for the growth of the majority of living organisms. However, species differ in their requirement for nitrogen intake for a healthy growth: some species have adapted to living in a low nitrogen environment, whereas others have adapted to living in a high nitrogen environment. In response to the rising demand for food and energy, increasing anthropogenic nitrogen emissions have resulted in atmospheric nitrogen deposition becoming an important and dominant source of nitrogen for some ecosystems (Erisman *et al.*, 1998; Galloway *et al.*, 2008; Sutton *et al.*, 2011). Global anthropogenic nitrogen depositions are now around the same order of magnitude as nitrogen input from natural sources, leading to a more than doubling of the nitrogen pool available to terrestrial organisms in less than a century (Vitousek *et al.*, 1997). Reactive nitrogen compounds are mainly present in the atmosphere in oxidised or reduced forms. The main anthropogenic sources for oxidised forms of nitrogen are combustion processes in transport, industry and energy production, estimated to contribute up to 70% of oxidised nitrogen emissions (Bragazza *et al.*, 2005). Emission sources of reduced forms of nitrogen are primarily related to agricultural activities such as animal husbandry (manure) and the application and production of fertilizers. Nitrogen emitted into the atmosphere is subject to short and long range atmospheric transport (Galloway *et al.*, 2008). Reactive nitrogen can be redistributed from emission hot-spots (i.e. agricultural and densely populated regions) to remote regions with undisturbed ecosystems naturally adapted to very low nitrogen inputs and availability. Enhanced nitrogen deposition may result in acidification and eutrophication of ecosystems, potentially leading to changes in plant diversity (Bobbink *et al.*, 2010; Stevens *et al.*, 2011). In large parts of Europe the critical loads of eutrophication for ecosystems, including those where mosses play an important role, are exceeded and are predicted to remain exceeded in the near future (Hettelingh *et al.*, 2011).

Several studies have shown that mosses have the potential to be indicators of atmospheric nitrogen deposition (Harmens *et al.*, 2011b, and references therein). However, sometimes the relationship between atmospheric nitrogen deposition and the nitrogen concentration in mosses is weak (e.g. Stevens *et al.*, 2011) or shown to be species-specific (Arroniz-Crespo *et al.*, 2008; Salemaa *et al.*,

2008). In 2005, ectohydric moss species were sampled for the first time at the European scale to indicate spatial patterns of atmospheric nitrogen deposition across Europe (Harmens *et al.*, 2011b). Detailed statistical analysis of the European moss data (Schröder *et al.*, 2010a) revealed that the total nitrogen concentration in mosses is significantly and best correlated with EMEP modelled air concentrations and atmospheric nitrogen deposition rates in comparison to other predictors that might contribute to the spatial variation of nitrogen concentrations in mosses. The variation in the total nitrogen concentration in mosses was best explained by the variation in ammonium (NH_4^+) concentration in air, followed by nitrogen dioxide (NO_2) concentrations in air. An apparent asymptotic relationship was found between EMEP modelled total atmospheric nitrogen deposition and the total nitrogen concentration in mosses (**Figure 5.1**; Harmens *et al.*, 2011b). Factors potentially affecting this relationship were discussed in more detail in the same study. Saturation appears to start at nitrogen deposition rates of ca. $15 \text{ kg ha}^{-1} \text{ y}^{-1}$, which might indicate the threshold of adverse impacts of nitrogen on the moss species sampled. For many habitats in Europe a nitrogen deposition of $15 \text{ kg ha}^{-1} \text{ y}^{-1}$ is within the range or even above the empirical critical load for nitrogen (Bobbink and Hettelingh, 2011).

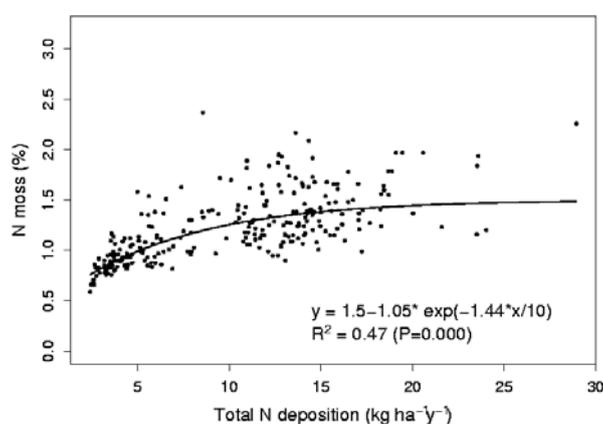


Figure 5.1 Relationship between EMEP modelled average annual total nitrogen deposition for 2003-2005 and averaged nitrogen concentration in mosses in 2005/6 for EMEP grid cells where at least five moss sampling sites were present (Harmens *et al.*, 2011b).

Although many studies have reported separately on the nitrogen concentration in mosses and on the impacts of elevated nitrogen deposition on terrestrial ecosystems, we are not aware of studies reporting on the relationship between both. Based on a survey of 153 acid grasslands from Atlantic Europe, Stevens *et al.* (2011) reported on a positive albeit weak relationship between the nitrogen concentration in the moss species *Rhytidiadelphus squarrosus* and atmospheric nitrogen deposition. Such a relationship was not observed for two vascular plant species. Nevertheless, Stevens *et al.* (2011) concluded that *R. squarrosus* was not a good indicator of atmospheric nitrogen deposition in acid grasslands. In the same study, grass species richness as a proportion of total species richness increased whereas forb species richness decreased with increasing nitrogen deposition, indicating a change in species composition. Over a period of 14 years, Zechmeister *et al.* (2007) observed that a few moss species (*Hypnum cupressiforme*, *Leucodon sciuroides*) responded to ambient nitrogen deposition levels by an increment in their population coverage. However, most moss species remained stable in their overall abundance. Although species turnover rates were rapid, observed changes in species composition could only to some extent be attributed to effects of airborne pollution. The moss communities as a whole did not show directional changes attributable to the observed levels of nitrogen deposition and the decrease of sulphur deposition. Thus, the substantial exceedance of critical loads for eutrophication effects did not lead to acute injuries. If at all, such injuries tended to be chronic injuries of individuals within the moss population.

Within the LRTAP Convention, the critical load approach has been developed to identify areas at risk from adverse effects of air pollution (UBA, 2004; Hettelingh *et al.*, 2007). Modelled critical loads of nitrogen are based on the acceptable nitrogen concentration in soil solution, i.e. the critical value at

which nitrogen starts to leach from the soil. Applying the mass balance method, the critical nitrogen load from deposition can then be calculated. In addition, empirical nitrogen critical loads for vegetation have been defined (Bobbink and Hettelingh, 2011), based on the effects of elevated nitrogen deposition on vegetation. Compared to modelled critical loads, empirical critical loads are generally higher for the most sensitive ecosystems (Hettelingh *et al.*, 2007). Nevertheless, mapped exceedances of empirical and modelled critical loads show a good resemblance. Areas in western Europe are particularly at risk from critical load exceedance, as shown for example for modelled critical loads in **Figure 5.2**. Although the same areas also have high concentrations of nitrogen in mosses, in parts of continental and eastern Europe the nitrogen concentrations in mosses are relatively higher than the critical load exceedance. One should bear in mind that whereas nitrogen concentrations in mosses are well correlated with atmospheric deposition of nitrogen (Schröder *et al.*, 2010a), soil characteristics significantly affect the exceedance of nitrogen critical loads.

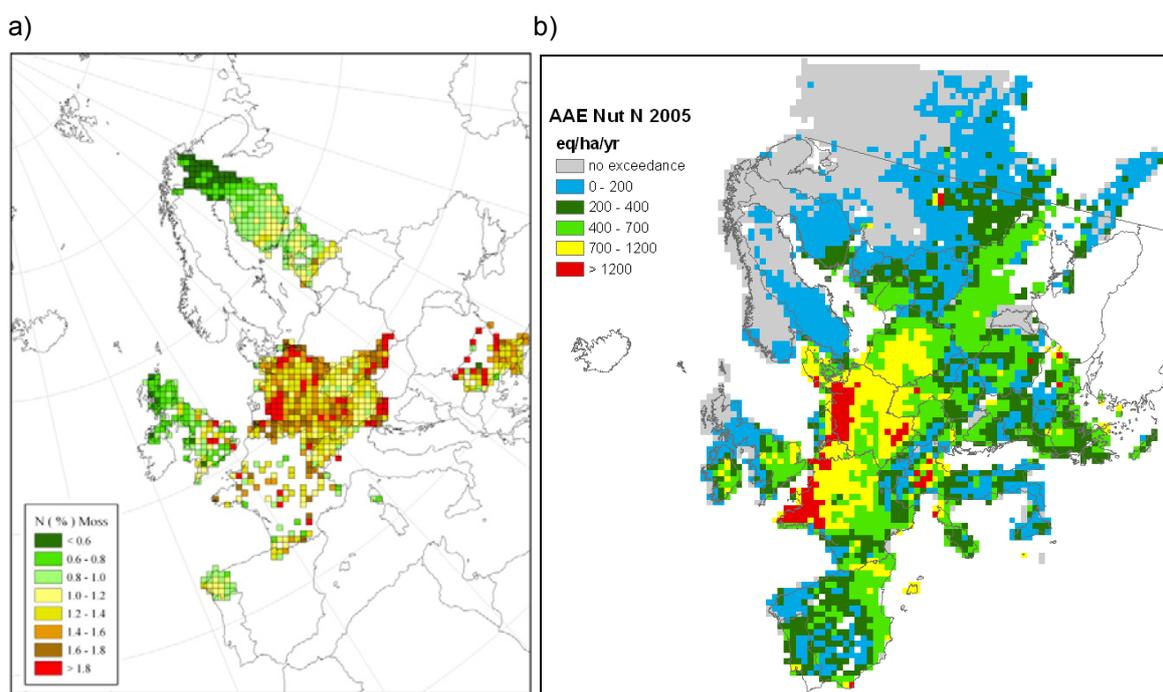


Figure 5.2 a) Mean concentration of nitrogen in mosses per EMEP grid square in 2005/6 and b) the average accumulated exceedance (AAE) of modelled critical loads of nitrogen (Nut N) in 2005. The size of the coloured squares reflects the area exceeded. Source AAE data: ICP Modelling and Mapping, Coordination Centre for Effects.

In the UK, critical loads for effects of nitrogen deposition on major sensitive habitats are exceeded for 58% of their area (RoTAP, 2012). There is strong evidence that nitrogen deposition has significantly reduced the number of plant species per unit area (species richness) in a range of habitats of high conservation value over large areas of the UK. The observed loss of plant species richness is primarily due to a decline in frequency of species adapted to low nutrient habitats. In cases where overall species richness has not changed, species characteristic of low nutrient habitats have been replaced by species adapted to higher nutrient availability, with undesirable implications for habitat conservation. Graminoid cover tends to have increased at the expense of forb cover. Moss (and lichen) species richness and community composition is more dynamic than that of vascular plants, with a replacement of more sensitive species by a more nitrogen pollution-tolerant community. There is no evidence of further declines in species richness over the last 20 years in areas of high nitrogen deposition, where much of the decline may have preceded the 1980s. However, there is evidence that current nitrogen deposition in many parts of the UK is associated with further declines in the frequency of sensitive plant species. Taken together, the data from field surveys and experimental studies provide a strong body of coherent evidence that exceedance of critical loads of nitrogen deposition is associated with adverse effects on terrestrial biodiversity at a UK scale. Whereas field

surveys (either repeated in time or over a gradient of nitrogen deposition) might be able to show a relationship between nitrogen deposition and impacts on terrestrial ecosystems (Maskell *et al.*, 2010), causality of such a relationship can only truly be tested where only atmospheric nitrogen deposition is manipulated as a driver of change. In field surveys it is difficult to disentangle the impact of nitrogen deposition from other changes, such as natural succession, land-use, management history, climate change, recovery from acidification (RoTAP, 2012). One should remember that nitrogen deposition has both eutrophying and acidifying effects and observed impacts of enhanced nitrogen deposition might be through accelerated soil acidification rather than eutrophication as such (RoTAP, 2012; Stevens *et al.*, 2011).

Recent studies have shown that vegetation responses to nitrogen deposition might depend more on the nitrogen form (ammonia or nitrate) than dose. Vegetation tends to be more sensitive to ammonia than nitrate exposure (Leith *et al.*, 2005; Pitcairn *et al.*, 2006; Sheppard *et al.*, 2008; 2009; Verhoeven *et al.*, 2011). Ammonia is more likely than wet deposition (ammonium and nitrate) to cause changes in vegetation for a given rate of nitrogen deposition. To reflect these new findings, the critical levels for ammonia were reduced in 2007, with lower critical levels being set for mosses and lichens than for herbaceous plant species (ECE/EB.AIR/WG.5/2007/3; Cape *et al.*, 2009). However, one should bear in mind that the critical load for total nitrogen deposition makes no distinction between the forms in which nitrogen is deposited. In the UK, the critical level for ammonia for lower plants is exceeded over 69% of the land area, and that for higher plants is exceeded over 19% of the UK (RoTAP, 2012).

In gradient studies in areas with high nitrogen deposition, Pitcairn *et al.* (2006) found that the nitrogen concentration in mosses responds differently to wet and dry deposited nitrogen and appears to respond more to concentrations of nitrate and ammonium in precipitation than to total nitrogen deposition at wet deposition sites. Regional studies in the UK have shown maximum nitrogen concentrations in mosses of 1.6% in areas dominated by wet deposition, despite relatively large inputs of nitrogen, whereas in gradient studies around livestock farms dominated by dry deposition, tissue nitrogen values of up to 4% were measured (Pitcairn *et al.*, 2006). Nordin *et al.* (2006) found that moss species in boreal forests take up predominantly ammonium, whereas biomass production tended to be higher with nitrate fertilization. This resulted in a higher nitrogen concentration in the mosses after ammonium exposure only. However, similar Spearman rank correlation coefficients were found between the total nitrogen concentration in mosses and EMEP modelled air concentrations or atmospheric deposition rates of different nitrogen forms (Schröder *et al.*, 2010a).

In summary, nitrogen concentrations in mosses can provide a good indication of terrestrial ecosystems being at risk from adverse impacts of enhanced atmospheric nitrogen deposition and can serve as an early warning system (Harmens *et al.*, 2011b). This could be true particularly for areas that have traditionally been exposed to low atmospheric nitrogen deposition and are currently being exposed to rising levels of nitrogen pollution. However, in large areas in Europe it might not be possible anymore to establish a relationship between the total nitrogen concentration in mosses and impacts of atmospheric nitrogen deposition on terrestrial ecosystems (if such a relationship could be established at all) due to the historic rise in nitrogen deposition that has changed ecosystem properties already. A combination of bioindicators is likely to be best to establish the current state of terrestrial ecosystems, in particular for areas of high conservation value (Nordin *et al.*, 2009).

6 Future activities of the ICP Vegetation

6.1 Review of ozone impacts on biodiversity and ecosystem services

The ICP Vegetation will review the potential (and where available, quantified) impacts of ozone in Europe on the provisioning, regulating, supporting and cultural services involving vegetation. This will include a review of current knowledge of whether ambient ozone impacts on plant biodiversity. We will incorporate results from scientific publications and national reports to provide an up-to-date synthesis of current knowledge. Highlights from this study will be submitted for inclusion in the WGE's report on impacts of air pollution on biodiversity and ecosystem services.

We plan to include the following chapters in the glossy report from this study:

1. Introduction
2. Sensitivity of European vegetation to ozone and the potential for impacts on biodiversity
3. Impacts on provisioning services, including food (see Mills and Harmens, 2011) and timber production
4. Impacts on regulatory services including pollination, C sequestration (see Harmens and Mills, 2012) and climate, air quality and water resources
5. Impacts on supporting services, including nutrient cycling, water cycling and primary production
6. Impacts on cultural services, including leisure, recreation and amenity
7. Contributions from ICP Vegetation participants on nationally-funded research on this subject
8. Conclusion and research recommendations.

6.2 Medium-term workplan (2013-2015) of the ICP Vegetation

As one of its core activities the ICP Vegetation will continue ozone stomatal flux model developments and flux map validation. Hence, we will continue to collate supporting evidence for ozone impacts on vegetation and review the robustness of flux-effect relationships for the establishment of new flux-based ozone critical levels for additional plant species. In 2013, the ICP Vegetation will report on the outcome of the 2010/11 European moss survey for heavy metals, nitrogen and POPs. The ICP Vegetation will also continue to explore opportunities for outreach activities to other regions of the globe.

The following medium-term workplan was adopted at the 25th Task Force Meeting of the ICP Vegetation (Brescia, Italy, 31 January – 2 February 2012):

2013 (see ECE/EB.AIR.109/Add.2):

- Report on supporting evidence for ozone impacts on vegetation;
- Report on the European heavy metals and nitrogen in mosses survey 2010/11;
- Report on the pilot study of mosses as biomonitors of POPs.

2014:

- Report on supporting evidence for ozone impacts on vegetation;
- Update of chapter 3 of the Modelling and Mapping Manual by inclusion of a new annex describing further technical developments;
- Report on ozone impacts on vegetation in a changing climate;
- Report on heavy metal and nitrogen concentrations in mosses in EECCA/SEE countries;
- Report on preparations for the moss survey 2015/16.

2015:

- Report on supporting evidence for ozone impacts on vegetation;
- Report on air pollution impacts on vegetation in EECCA/SEE countries;
- Report on interacting effects of co-occurring ozone and nitrogen pollutants on vegetation;
- Report on progress with the moss survey 2015/16.

Common workplan items of the WGE for 2013 have been described in the biannual workplan for the LRTAP Convention (see ECE/EB.AIR.109/Add.2) and include:

- i) Report on the further implementation of the Guidelines on Reporting of Monitoring and Modelling of Air Pollution Effects;
- ii) Report on ideas and actions to enhance the involvement of EECCA/SEE countries in the Eastern Europe, the Caucasus and Central Asia and on cooperation with activities outside the Air convention;
- iii) Report on impacts on biodiversity and ecosystems services.

Common workplan items beyond 2013 will be decided at the WGE meeting in September 2013. These and the ICP Vegetation-specific workplan items for 2014 and 2015 are subject to approval by the Executive Body of the LRTAP Convention in December 2013.

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Annex 1. Participation in the ICP Vegetation

In many countries, several other scientists (too numerous to include here) also contribute to the work programme of the ICP Vegetation.

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Air Pollution and Vegetation

ICP Vegetation

Annual Report 2011/2012

This report describes the recent work of the International Cooperative Programme on effects of air pollution on natural vegetation and crops (ICP Vegetation), a research programme conducted in 35 countries in the UNECE region, with outreach activities to other regions. Reporting to the Working Group on Effects of the Convention on Long-range Transboundary Air Pollution, the ICP Vegetation is providing information for the review and revision of international protocols to reduce air pollution problems caused by ground-level ozone, heavy metals, nitrogen and persistent organic pollutants (POPs). Progress and recent results from the following activities are reported:

- Contributions to revision of the Gothenburg Protocol.
- Impacts of ozone on carbon sequestration.
- Ozone biomonitoring programme.
- European heavy metal and nitrogen in mosses survey 2010/2011, including a pilot study on POPs.
- Relationship between i) heavy metals and ii) nitrogen concentrations in mosses and impacts on terrestrial ecosystems.

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