ANNALI DI BOTANICA
Ann. Bot. (Roma), 2015, 5: 31–43
Journal homepage: http://annalidibotanica.uniroma1.it

TWENTY EIGHT YEARS OF ICP VEGETATION: AN OVERVIEW OF ITS ACTIVITIES

Harmens H.[1*], Mills G.[1], Hayes F.[1], Norris D.A.[1], Sharps K.[1]

[1]*Centre for Ecology & Hydrology, Environment Centre Wales, Deiniol Road, Bangor, Gwynedd LL57 2UW, UK*
[*]*Corresponding Author: Telephone: +44 1248 374512, email: hh@ceh.ac.uk*

(Received 11 March 2015; accepted 27 March 2015)

Abstract – Here we look back at the activities and achievements in the 28 years of the International Cooperative Programme on the Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation). The ICP Vegetation is a subsidiary body of the Working Group on Effects of the UNECE Convention on Long-range Transboundary Air Pollution (LTRAP), established in 1979. An important role of the ICP Vegetation is to provide evidence for air pollution impacts on vegetation in support of policy development and review of the LRTAP Convention and its Protocols. The activities and participation in the ICP Vegetation have grown over the years. The main activities include:

- Collate evidence of ozone impacts on vegetation, assess spatial patterns and temporal trends across Europe;
- Develop dose-response relationships, establish critical levels for vegetation and provide European risk maps of ozone impacts;
- Reviewing the literature on ozone impacts on vegetation and produce thematic scientific reports and policy-relevant brochures;
- Determine spatial patterns and temporal trends of heavy metals, nitrogen and persistent organic pollutants concentrations in mosses as a biomonitoring tool of atmospheric deposition of these compounds.

Keywords: Ozone, critical levels, heavy metals, nitrogen, persistent organic pollutants

Introduction

In 1987, the International Cooperative Programme on Effects of Air pollution on Crops (ICP Crops) was established as a subsidiary body of the Working Group on Effects of the UNECE Convention on Long-range Transboundary Air Pollution (LTRAP), established in 1979. In the early years, the programme focussed on assessing the impacts of ground-level ozone on crops. As ozone also affects other vegetation than crops, the remit of the ICP was soon extended to include impacts on air pollution on (semi-)natural vegetation too. Hence, in 1999, the ICP was renamed as the International Cooperative Programme on the Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation). Although ICP Vegetation also includes some research on the impacts of ozone on trees, a separate ICP was already established in 1985 to monitor impacts of air pollution on forests (ICP Forests). In addition to the ICP Vegetation and ICP Forests, the Working Group on Effects consists of four other ICPs (ICP Integrated Monitoring, ICP Waters, ICP Materials and ICP Modelling and Mapping) and the Task Force on Health (Working Group on Effects, 2004). The LTRAP Convention provides the essential framework for controlling and reducing damage to human health and the environment (ecosystems and materials) caused by transboundary air pollution. Over the years, the LRTAP Convention has been extended by eight protocols that identify specific measures to be taken by Parties to cut their emissions of air pollutants (Working Group on Effects, 2004). Fifty-one UNECE member States are Parties to the Convention. An important remit of the ICP Vegetation is to assess spatial patterns and temporal trends regarding air

doi: 10.4462/annbotrm-13064

pollution impacts on vegetation within the UNECE area and provide evidence to the LRTAP Convention for assessments of the sufficiency and effectiveness of air pollution abatement strategies developed within the Convention.

In 2000, the ICP Vegetation further extended its remit by taking over the coordination of the European moss survey from the Nordic Council of Ministers. Since 1990, mosses have been sampled every five year across Europe to provide an indication of the spatial variation and temporal trends of heavy metal deposition to vegetation at a high spatial resolution (Harmens et al., 2010; 2015). In 2005, the moss survey was extended to include the determination of nitrogen in naturally growing mosses (Harmens et al., 2011; 2015) and in 2010 a pilot study was conducted to use mosses as biomonitors of selected persistent organic pollutants (POPs; Harmens et al., 2013). Hence, the participation in the ICP Vegetation has gradually increased over the years. This is reflected in the growing number of countries (in- and outside the UNECE) participating in the annual Task Force Meeting of the ICP Vegetation, with the number of participants increasing from ca. 20 in the early years to ca. 80 in recent years (Figure 1). Below we will describe in more detail the activities of the ICP Vegetation. Further details can be found on the web site: http://icpvegetation.ceh.ac.uk

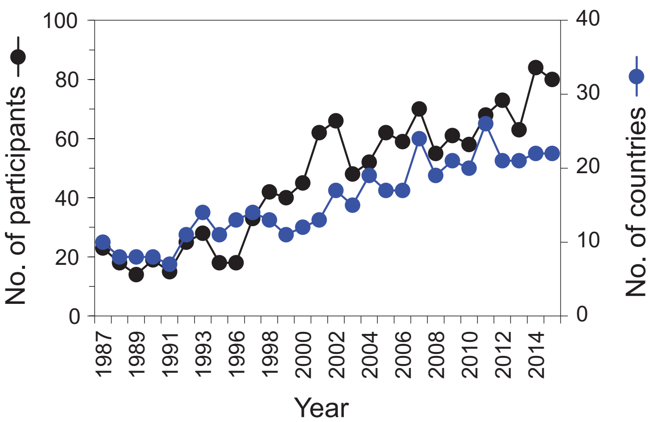

Figure 1. Number of experts and countries participating in ICP Vegetation Task Force meetings between 1987 and 2015.

IMPACTS OF GROUND-LEVEL OZONE ON VEGETATION

Since its establishment in 1987, the ICP Vegetation has studied and collated evidence of impacts of ground-level ozone pollution on vegetation, in the early years mainly on crops but in later years also on (semi-)natural vegetation. In close collaboration with the ICP Forests, information on the impacts of ozone on trees is also evaluated. Activities on ozone can be divided into three categories:

1. Collate evidence of ozone impacts on vegetation, asses spatial patterns and temporal trends;
2. Develop dose-response relationships, establish critical levels for vegetation and provide European risk maps of ozone impacts;
3. Publish thematic scientific reports and policy-relevant brochures on ozone impacts on vegetation by reviewing the literature.

Evidence of ozone impacts on vegetation

The ICP Vegetation has collated a database from ozone exposure experiments to assess the sensitivity of the above-ground biomass of 83 species of semi-natural vegetation. In a meta-analysis, Hayes et al. (2007a) identified some relationships between ozone sensitivity and plant physiological and ecological characteristics, which could be used to predict sensitivity to ozone of untested species. Mills et al. (2007b) conducted further analyse to identify ozone-sensitive communities suitable for mapping exceedances of critical levels (see Section 2.2). Jones et al. (2007) developed a regression-based model for predicting changes in biomass of individual species exposed to ozone, based on their Ellenberg Indicator values and found a relationship with the Ellenberg Indicator for light and salinity. In addition, the ICP Vegetation has populated a database containing 644 records of impacts of ambient ozone on vegetation for 171 species (Hayes et al., 2007b; Mills et al., 2011a). Of the 644 records of visible injury, 39% were for crops (27 species), 38.1% were for (semi)–natural vegetation (95 species) and 22.9% were for shrubs (49 species; Mills et al., 2011a). Effects in ambient air were observed in 18 European countries from Sweden in the north to Greece in the south. This data was collated in various studies that are described in more detail below. Evidence of ozone injury on trees is separately collated by the ICP Forests.

Records of observations of visible-injury symptoms in ambient air from survey and ad-hoc observations

The distribution of records of occurrence of visible injury symptoms shows that ozone injury can occur across the whole of Europe (Hayes et al., 2007b), although it is most commonly recorded in Continental Central Europe and the Western Mediterranean regions (Figure 2). Although records are present as a result of surveys, these surveys only account for approximately one-quarter of the total records of visible injury, with the remaining records from ad-hoc observations. Due to the sporadic nature of surveys, it is not possible investigate temporal trends from survey data.

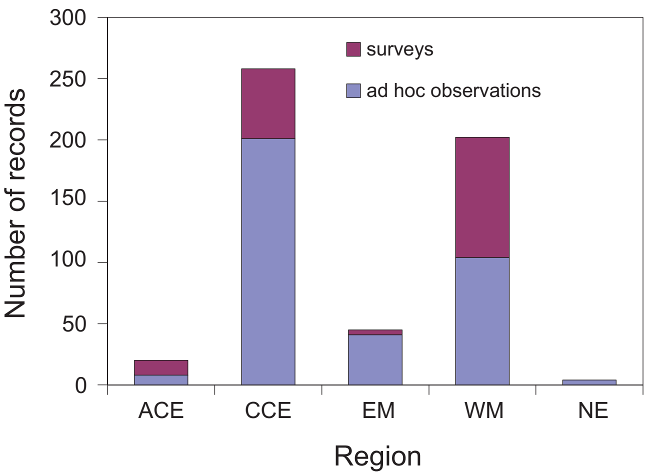

Figure 2. Number of records per region of visible injury symptoms attributed to ambient ozone. ACE = Atlantic Central Europe, CCE = Continental Central Europe, EM = Eastern Mediterranean, WM = Western Mediterranean, NE = Northern Europe (Hayes et al., 2007b).

Biomonitoring studies using a chemical protectant against ozone damage

During the years 1994, 1995 and 1996 participants of the ICP Vegetation conducted studies in ambient air using the ozone-protectant ethylenediurea (EDU) at experimental sites and/or in commercial fields (Ball et al., 1998). For each species used in each study, half of the plants were treated with EDU and half with water. Plants were placed in ambient air and at the end of the exposure period (typically eight weeks), the two sets were compared for the extent of visible injury and/or biomass. Species tested in this way included subterranean clover (*Trifolium subterranean),* bean (*Phaseolus vulgaris),* radish (*Raphanus sativus),* white clover (*Trifolium repens),* red clover (*Trifolium pratense),* tomato (*Lycopersicon esculentum),* soybean (*Glycine max),* watermelon (*Citrullus lanatus)* and tobacco (*Nicotiana tabacum).* In many cases the occurrence of visible leaf injury was reduced for plants treated with EDU. In addition, EDU can also protect plants from decreases in biomass that would otherwise occur in ambient ozone conditions (Hayes et al., 2007b). A comprehensive analysis of the results of exposure of EDU and non-EDU treated *Trifolium repens* (L. cv Menna) at 12 sites from nine countries showed that there was a decrease in the biomass ratio of non-EDU to EDU treated plants with increasing ozone exposure, however there was a lot of scatter in this relationship (Ball et al., 1998).

Biomonitoring with ozone-sensitive and ozone-resistant biotypes of white clover (*Trifolium repens*)

Between 1996 and 2006, the ICP Vegetation biomonitoring programme has involved exposure of an ozone sensitive biotype of white clover (*Trifolium repens* Regal, NC-S) to ambient air (Hayes et al., 2007b; Mills et al., 2011a). Leaf injury was apparent in varying magnitudes ranging from pale cream stipples on the leaf surface to large necrotic patches with leaves severely damaged. At some sites, ozone-resistant plants (NC-R) were also grown; the ratio of the biomass of NC-S to NC-R provides an indication of ambient ozone effects on growth at these sites (Heagle et al., 1995). From 1998 to 2006 there is scored injury data available from a total of 45 sites, representing 16 countries across Europe (Figure 3), including Italy (e.g. Manes et al., 2003). Biomass ratio data is available from 1996 to 2006 for a total of 41 sites from 15 countries. However, each individual site did not necessarily perform the investigation every year meaning there are very few sites with a long time-run of data.

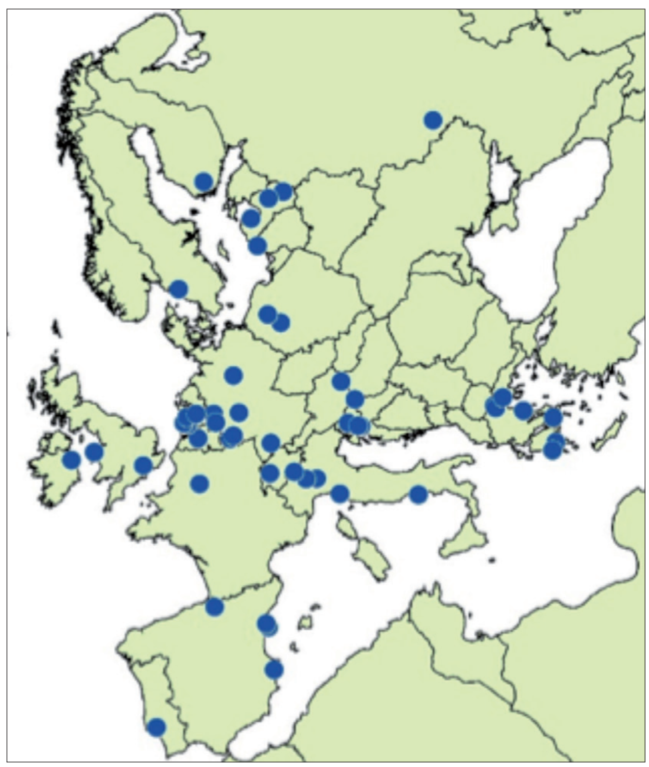

Figure 3. ICP Vegetation clover biomonitoring sites (Hayes et al, 2007b).

Visible injury symptoms on white clover have been recorded in all regions of Europe (Hayes et al., 2007b; Mills et al., 2011a). The highest impacts have generally been found in Switzerland, Italy and Greece, although moderate impacts have also been found in central and northern Europe. The biomass of the sensitive variety of white clover plants was reduced with increasing ozone concentrations. The largest impacts of ozone on the biomass of clover plants were consistently found in southern Europe, particularly in Italy and Greece where biomass reductions of over 30% have been demonstrated in some years. Reductions in biomass of over 10% have been found in the Eastern Mediterranean, Western Mediterranean and Continental Central Europe regions, but such large reductions have not been recorded in either Northern Europe or Atlantic Central Europe (Hayes et al., 2007b; Mills et al., 2011a). For further details of the outcome of the biomonitoring studies with white clover we refer to Hayes et al. (2007b) and Mills et al. (2011a).

Biomonitoring with ozone-sensitive and ozone-resistant biotypes of French bean (*Phaseolus vulgaris*)

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) since 2008. The bean genotypes were develop (Reinert and Eason, 2000) and tested as a bioindicator system (Burkey et al. 2005) in the USA. Although visible leaf injury regularly occurred across the ICP Vegetation network (Figure 4), 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 (e.g. Salvatori et al., 2013). So far, ozone flux-effect relationships have not been developed for bean.

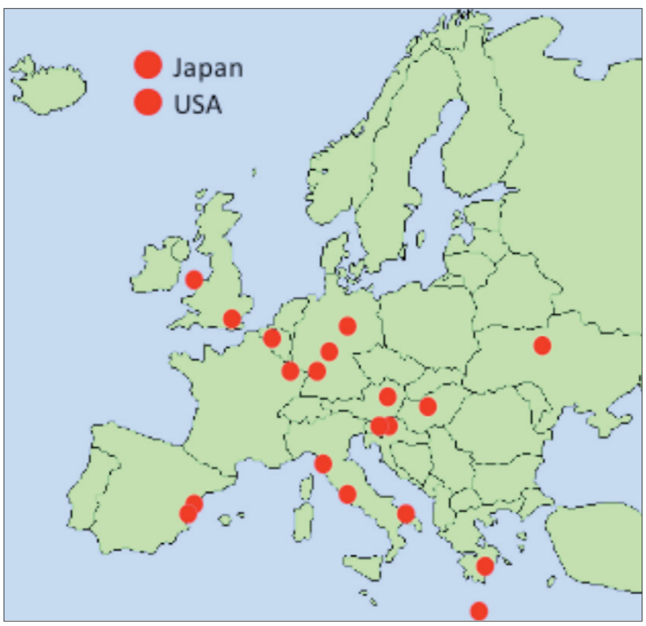

Figure 4. Locations where ozone injury has been detected on bean (*Phaseolus vulgaris*) between 2008 and 2011 in Europe. Injury was also detected in the USA and Japan, and in later years also in China.

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, differences between the sensitive and resistant biotypes do not seem to be strong enough for continued application as a biomonitor for yield effects across all climate regions in Europe.

Evidence from filtered air experiments

Although many research institutes within Europe have the capabilities to investigate the effects of ozone on vegetation experimentally, comparatively few use both charcoal-filtered (CF) and non-filtered (NF) air as treatments. Where these combinations of treatments have been used together, comparisons of responses provide valuable indications of the effect of ambient ozone. Data are available from ICP Vegetation sites in Sweden, Italy, Netherlands, Austria, Switzerland, Belgium, Germany, Spain and Finland (Hayes et al., 2007b). A diverse range of species and response parameters have been investigated in this way, including crop and natural vegetation species. Many studies have investigated growth or yield related parameters. Recently, Pleijel (2011) reviewed the outcome of 30 air filtration experiments for field-grown wheat, representing nine countries in North America, Europe and Asia. Filtration had reduced the average daytime ozone concentration from 35 to 13 nmol mol^{-1}. Twenty six experiments reported improved yield and four experiments reduced yield by filtration, with an average yield improvement of 9% in charcoal-filtered air. Hence, current ambient ozone concentrations adversely affect wheat yield over large parts of the world.

Smart phone application to record incidences of ozone injury in the field

In 2014, the ICP Vegetation has developed a new way of recording incidences of ozone injury in the field, using smart-phone technology for i-phones and android phones, and a web-based recording methodology (http://icpvegetation.ceh.ac.uk). The smart phone App (Figure 5) allows participants to upload photographs of ozone injury direct from the field together with the coordinates for the location where the injury was detected. App users are taken through a short series of questions, with answers selected from drop-down menus. In addition, the App contains an 'Ozone information' section, which includes details of the key symptoms of ozone injury, and other causes of leaf damage that may be mistakenly recorded as ozone injury. There is an 'Examples of ozone injury' page, containing photos of ozone injury on many of the species included in the App species list. Photos of ozone injury were also published in the brochure 'Have you seen these ozone injury symptoms', produced in collaboration with the ICP Forests (http://icpvegetation.ceh.ac.uk/publications/documents/CEHOzoneInjury_webmidres.pdf).

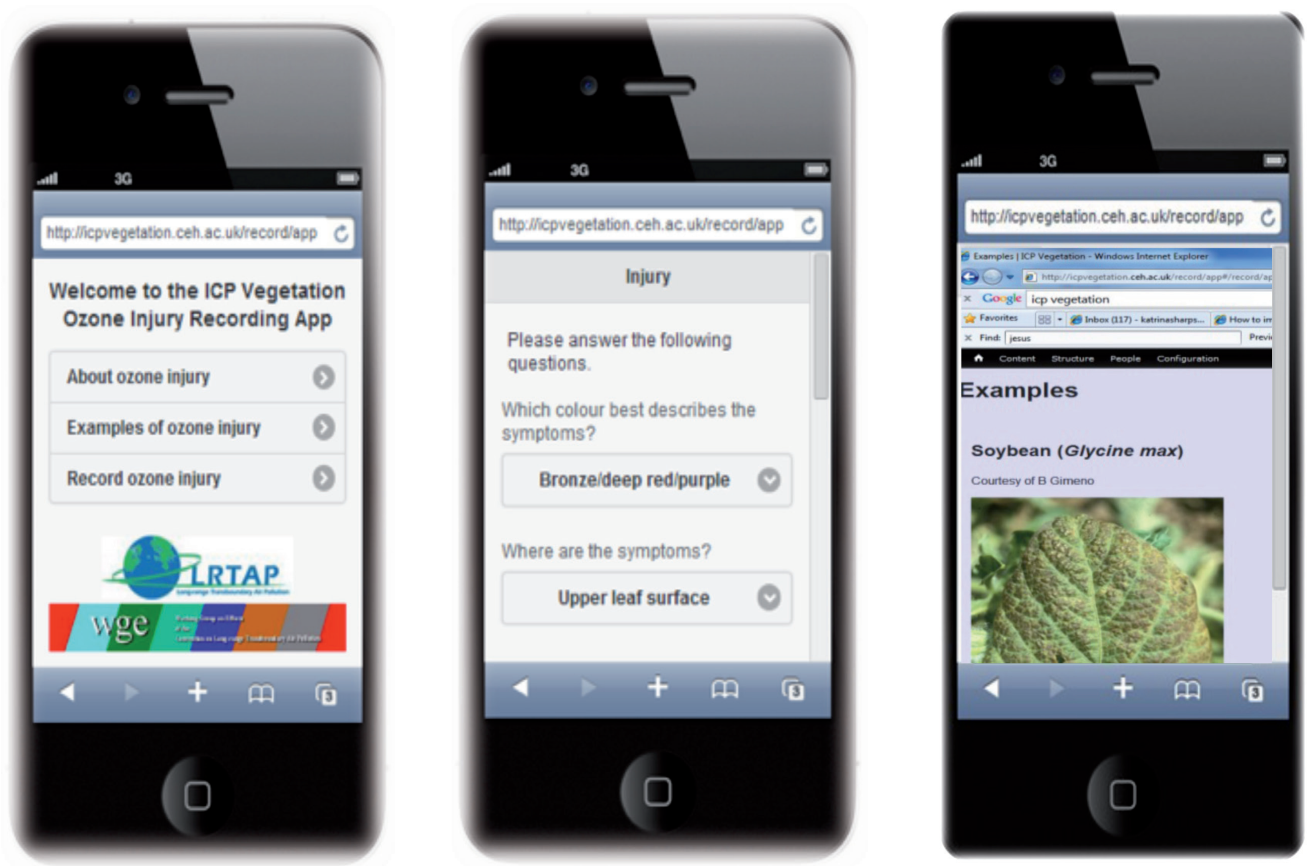

Figure 5. Smart phone App for recording incidences of ozone-induced leaf injury.

Ozone critical levels: from concentration to flux-based approach

History

Over the years, the ICP Vegetation has been responsible for developing ozone critical levels for vegetation and for updating chapter 3 ('Mapping critical levels for vegetation'; LRTAP Convention, 2015) of the 'Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends' of the UNECE LRTAP Convention. The aim of this chapter is to provide information on the critical levels for sensitive vegetation and how to calculate critical level exceedance. Critical levels were defined in an earlier version of this manual (LRTAP Convention, 1996) as "the atmospheric concentrations of pollutants in the atmosphere above which adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge". In the most recent update, the critical levels for vegetation are defined as the "concentration, cumulative exposure or cumulative stomatal flux of atmospheric pollutants above which direct adverse effects on sensitive vegetation may occur according to present knowledge". Critical level exceedance maps show the difference between the critical level and the mapped, monitored or modelled air pollutant concentration, cumulative exposure or cumulative flux.

Critical level values have been set, reviewed and revised for ozone, sulphur dioxide, nitrogen dioxide and ammonia at a series of UNECE Workshops: Bad Harzburg (1988); Bad Harzburg (1989); Egham (1992); Bern (1993); Kuopio (1996), Gerzensee (1999), Gothenburg (2002;), Obergurgl (2005), Edinburgh (2006), Ispra (2009) and associated Task Force meetings of the ICP Vegetation. For sulphur dioxide, nitrogen dioxide and ammonia, concentration-based critical levels have been developed. For ozone, cumulative concentration-based and cumulative stomatal flux-based critical levels have been developed for crops, forest trees and (semi-) natural vegetation.

The concept of ozone critical levels based on an accumulated amount of ozone above a threshold over a fixed period of time was first introduced into the manual in 1996 (LRTAP Convention, 1996), with the inclusion of the ozone parameter AOTX (ozone concentrations accumulated over a threshold of X ppb), as for example described for crops in Mills et al. (2007a). However, several important limitations and uncertainties have since been recognised for using AOTX. In particular, the real impacts of ozone depend on the amount of ozone reaching the sites of damage within the leaf, whereas AOTX-based critical levels only consider the ozone concentration at the top of the canopy. The Gerzensee Workshop in 1999 recognised the importance of developing an alternative critical level approach based on the flux of ozone from the exterior of the leaf through the stomatal pores to the sites of damage (stomatal flux). This approach required the development of mathematical models to estimate stomatal flux, primarily from knowledge of stomatal responses to environmental factors (Emberson et al., 2000). It was agreed at the Gothenburg Workshop in 2002 that ozone flux-effect models were sufficiently robust for the derivation of flux-based critical levels. An additional simplified flux-based risk assessment method for use in large-scale and integrated assessment modelling was discussed at the Obergurgl Workshop (2005) and after further revision (approved at appropriate Task Force meetings) is included in the manual for a generic crop and forest tree. A critical level has been derived for effects on a generic crop, but not for effects on generic trees. As such, the integrated assessment method does not take into account effects of soil moisture in stomatal flux modelling, however, subsequent modelling and mapping within the model developed by EMEP (European Monitoring and Evaluation Programme) uses a simple soil moisture index to take into account effects of soil moisture on the deposition of ozone in a simplistic way (Simpson et al., 2012). At the Ispra Workshop in 2009 and subsequent 23rd Task Force meeting of the ICP Vegetation in 2010, the flux-based critical levels were reviewed, revised where needed, and added for new receptors (Mills et al., 2011b). The critical levels currently included in the 'Modelling and Mapping Manual' are summarised below. Full details of their scientific basis, calculation and applications can be found in Chapter 3 of the manual (LRTAP Convention, 2015).

Stomatal flux-based critical levels for effects of ozone on growth or yield

These take into account the varying influences of air temperature, water vapour pressure deficit (VPD) of the surrounding leaves, light (irradiance), soil water potential (SWP) or plant available water (PAW), ozone concentration and plant development (phenology) on the stomatal flux of ozone (Emberson et al., 2000). They therefore provide an estimate of the critical amount of ozone entering through the stomata and reaching the sites of action inside the plant and are species-specific. The hourly mean instantaneous stomatal flux of ozone based on the projected leaf area (PLA), is accumulated over a stomatal flux threshold of Y nmol m^{-2} s^{-1}. The accumulated Phytotoxic Ozone Dose (i.e. the accumulated stomatal flux) of ozone above a flux threshold of Y (POD_Y), is calculated for the appropriate time-window as the sum over time of the differences between hourly mean values of stomatal flux and Y nmol m^{-2} PLA s^{-1} for the periods when the stomatal flux exceeds Y. The stomatal flux-based critical level of ozone is then the cumulative stomatal flux of ozone, POD_Y, above which direct adverse effects may occur according to present knowledge. Values of stomatal flux-based critical levels (Table 1) have been identified for crops (wheat, potato and tomato), forest trees (represented by birch and beech, and Norway spruce), and (semi-)natural vegetation (represented by *Trifolium* spp. (clover family) and provisionally *Viola* spp. (violet family). The flux-based critical levels and associated response functions are suitable for mapping and quantifying impacts at the local and regional scale, including effects on food security (crops), roundwood supply for the forest sector industry and loss of carbon storage capacity and other beneficial ecosystem services (forest trees), and impacts on the vitality of fodder-pasture and natural grassland species. Where appropriate, they could be used for assessing economic losses (e.g. Mills and Harmens, 2011).

Table 1. Critical levels for ozone. For further details and references see LTRAP Convention (2015).

(a) Flux-based critical levels (see Mills et al., 2011b)			
Receptor	**Effect (per cent reduction)**	**Parameter**	**Critical level (mmol m^{-2} PLA)**
Wheat	Grain yield (5%)	POD_6	1
Wheat	1000 grain weight (5%)	POD_6	2
Wheat	Protein yield (5%)	POD_6	2
Potato	Tuber yield (5%)	POD_6	5
Tomato	Fruit yield (5%)	POD_6	3
Tomato	Fruit quality (5%)	POD_6	4
Norway spruce	Biomass (2%)	POD_1	8
Birch and beech	Biomass (4%)	POD_1	4
Productive grasslands (clover)	Biomass (10%)	POD_1	2
Conservation grasslands (clover)	Biomass (10%)	POD_1	2
Conservation grasslands (*Viola* spp), provisional	Biomass (15%)	POD_1	6
(b) Concentration-based critical levels			
Receptor	**Effect**	**Parameter**	**Critical level (ppm h)**
Agricultural crops	Yield reduction	AOT40	3
Horticultural crops	Yield reduction	AOT40	8
Forest trees	Growth reduction	AOT40	5
(Semi-)natural vegetation communities dominated by annuals	Growth reduction and/or seed production	AOT40	3
(Semi-)natural vegetation communities dominated by perennials	Growth reduction	AOT40	5
(c) VPD-modified concentration-based critical level			
Receptor	**Effect**	**Parameter**	**Critical level (ppm h)**
Vegetation (derived for clover species)	Visible injury on leaves	$AOT30_{VPD}$	0.16

Concentration-based critical levels for effects of ozone on growth or yield

These are based on the concentration at the top of the canopy accumulated over a threshold concentration for the appropriate time-window and thus do not take account of the stomatal influence on the amount of ozone entering the plant. This value is expressed in units of ppm h (μmol mol^{-1} h). The term AOTX (concentration accumulated over a threshold ozone concentration of X ppb) has been adopted for this index; often "X" = 40 ppb (AOT40) is used. Values of concentration-based critical levels are defined for agricultural and horticultural crops, forests and (semi)–natural vegetation. The AOTX-based critical levels have a weaker biological basis than the flux-based critical levels and are suitable for estimating the risk of damage where climatic data or suitable flux models are not available. Economic losses should not be estimated using AOTX-based critical levels and associated response functions.

VPD-modified concentration-based critical level for visible leaf injury

This index is only used to define the short-term critical level for the development of visible injury on crops. The method takes into account the modifying influence of vapour pressure deficit (VPD) on the stomatal flux of ozone by multiplying the hourly mean ozone concentration at the top of the canopy by an f_{VPD} factor to get the VPD-modified ozone concentration. The $[O_3]_{VPD}$ is accumulated over a threshold concentration during daylight hours over the appropriate time-window. This value is expressed in units of ppm h (μmol mol^{-1} h). The term $AOT30_{VPD}$ (VPD-modified concentration accumulated over a threshold ozone concentration of 30 ppb) has been adopted for this index. This critical level can be used to indicate the likelihood of visible ozone injury on vegetation, and is especially useful for estimating effects on leafy vegetable crops where leaf injury reduces the quality and market value.

Literature reviews to inform scientists (full report) and policy makers (short summary brochures) of recent developments

The following synthesis reports and brochures can be downloaded from the ICP Vegetation web site (http://icpvegetation.ceh.ac.uk).

Evidence of widespread ozone damage to vegetation in Europe (1990 – 2006)

The first glossy thematic report produced by the ICP Vegetation was the 'Evidence report' (Hayes et al., 2007b). This report provides field-based evidence for the impacts of ambient ozone on vegetation, collated from material as described above. This report confirmed that the stomatal flux-based risk maps of ozone impacts on vegetation (POD_3) were more coherent with observed impacts of ozone damage than concentration-based (AOT40) risk maps. The outcome of the report was instrumental in the following decision from the Executive Body of the LRTAP Convention: '… ozone effects on vegetation should be incorporated in integrated assessment modelling …'. The Executive Body also noted that '… policies aiming only at health effects would not protect vegetation in large areas of Europe.'

Ozone pollution: A hidden threat to food security

This reports provides an overview of the impacts of ozone pollution on both crop yield and quality, how these impacts are affected by a changing climate, which species are sensitive to ozone, and provides a quantification of economic losses due to ozone impacts on wheat and tomato yield in 2000 and 2020 in EU27 plus Norway and Switzerland (Mills and Harmens, 2011). Mean yield losses for wheat and tomato were estimated to be 13.7% and 9.4% respectively in 2000 and would decline to 9.1% and 5.7% in 2020 under the then current legislation scenario. The report highlighted the urgent need to raise political awareness of the adverse impact of ozone on food production in regions such as South Asia where some of the most important staple foods of wheat, rice maize and bean are sensitive to ozone.

Ozone pollution: Impacts on carbon sequestration in Europe

This reports describes how ozone affects carbon sequestration in the living biomass of trees (Harmens and Mills, 2012). In northern and central Europe, carbon sequestration in the living biomass of trees is reduced by ca. 10%, based on the AOT40 approach, comparing current ambient ozone concentrations with pre-industrial concentration. Slightly higher reductions were estimated using the stomatal flux-based approach. Ozone affects the capacity of the vegetation to act as a sink for greenhouse gases such as carbon dioxide and ozone, and it has been suggested that effects of ozone on vegetation growth and biomass might contribute as much to global warming as the direct effects of ozone as a greenhouse gas due to feedbacks to the climate as a result of ozone impacts on vegetation (Sitch et al., 2007).

Ozone pollution: Impacts on ecosystem services and biodiversity

In this report the ICP Vegetation provided a review of the effects of ozone pollution on ecosystem services and biodiversity (Mills et al., 2013). The review included impacts on ecological processes and supporting services (primary productivity, stomatal functioning, carbon, nitrogen and water cycling), provisioning services (crop and timber production), regulating services (carbon sequestration and global warming, air quality, methane emissions, flowering and pollination), and cultural services (leisure, recreation and amenity). A case study from Italy showed how vegetation can improve air quality, for example through ozone removal (Manes et al., 2012). It was concluded that a comprehensive quantitative assessment of ozone effects on ecosystem services, including an economic valuation, is feasible for some provisioning and regulating services, whilst more research and method development is needed for other ecosystem services.

Air pollution: Deposition to and impacts on vegetation in (South-)East Europe, Caucasus, Central Asia (EECCA/SEE) and South-East Asia

Current knowledge on the deposition of air pollutants to and their impacts on vegetation was reviewed for EECCA (Armenia, Azerbaijan, Belarus, Georgia, Kazakhstan, Kyrgyzstan, Moldova, Russian Federation, Tajikistan, Turkmenistan, Ukraine and Uzbekistan) and SEE countries (Albania, Bosnia and Herzegovina, Bulgaria, Croatia, Cyprus, Greece, Macedonia, Montenegro, Romania, Serbia, Slovenia and Turkey). As an outreach activity, knowledge on this subject was also reviewed for the Malé Declaration countries in South-East Asia (Bangladesh, Bhutan, India, Iran, Maldives, Nepal, Pakistan and Sri Lanka). Air pollution is a main concern in Asia due to enhanced industrialisation, which is directly linked to continued strong economic growth in recent decades (Harmens and Mills, 2014).

Other recent brochures

In 2014, the ICP Vegetation produced a brochure on ozone-induced leaf injury symptoms in collaboration with the Expert Panel on Ambient Air Quality of the ICP Forests. In 2015, the following two brochures were produced: 1) Climate change and reactive nitrogen as *modifiers* of vegetation responses to ozone pollution; 2) Changing ozone profiles in Europe: implications for vegetation.

Mosses as biomonitors of atmospheric pollution

Heavy metals

The survey on heavy metal concentrations in mosses was originally established in 1980 as a Swedish initiative under the leadership of Åke Rühling, Sweden. The idea of using mosses to measure atmospheric heavy metal deposition was developed in the late 1960s by Rühling and Tyler (Rühling and Tyler, 1968). It is based on the fact that mosses, especially the carpet-forming species, obtain most of their nutrients directly from precipitation and dry deposition; there is little uptake of metals from the substrate as these mosses do not have a well-developed root system. The technique of moss analysis therefore provides a surrogate, time-integrated measure of the spatial patterns of heavy metal deposition from the atmosphere to terrestrial systems. 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 deposition analysis. Although the moss concentration data provide no direct quantitative measurement of deposition, this information can be derived by using one of several regression approaches relating the results from moss surveys to precipitation monitoring data (e.g. Berg and Steinnes, 1997; Berg et al., 2003). The first European moss survey on heavy metals was conducted in 1990 and has since then been repeated every five years (Harmens et al., 2004, 2007, 2008, 2010, 2015). In 1990 and 1995, the moss survey was coordinated by the Nordic Working Group on Monitoring and Data, Nordic Council of Ministers. In 2000, the coordination was handed over to the Coordination Centre of the ICP Vegetation at the Centre for Ecology & Hydrology (CEH) Bangor, UK. The next survey is scheduled for 2015 and the coordination of that survey will be conducted by the Joint Institute of Nuclear Research (JINR), Dubna, Russian Federation under auspice of the ICP Vegetation. The coordination was handed over to JINR with the aim to enhance participation of EECCA countries, a priority of the LRTAP Convention in recent years, and reach out to other countries in Asia.

The European moss survey provides data on concentrations of ten heavy metals (arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb), Vanadium (V), zinc (Zn)) in naturally growing mosses. Since 2005, data are also reported for aluminium (Al) and antimony (Sb; Harmens et al., 2010, 2015). The number of sampling sites ranged from ca. 4,500 sites in 2010 to ca. 7,300 sites in 1995. Although spatial patterns are metal-specific, in recent surveys the lowest concentrations of metals in mosses were generally found in Scandinavia, the Baltic States and northern parts of the UK; the highest concentrations were generally found in Belgium and south-eastern Europe, resulting in a more or less declining gradient from south-east to north-west. The recent decline in emission and subsequent deposition of heavy metals across Europe has resulted in a decrease in the heavy metal concentration in mosses for the majority of metals. Since 1990, the median concentration in mosses across Europe has declined the most for lead (77%), followed by vanadium (55%), cadmium (51%), chromium (43%), zinc (34%), nickel (33%), iron (27%), arsenic (21%, since 1995), mercury (14%, since 1995) and copper (11%; Harmens et al., 2015). For cadmium and lead atmospheric deposition has been identified as the primary factor contributing to the spatial variation of concentrations in mosses at the European scale (Holy et al., 2009; Schröder et al., 2010b). For cadmium and lead, significant positive correlations were found between the concentration in mosses and the deposition modelled by the European Monitoring and Evaluation Programme (EMEP) for about two thirds or more of the countries participating in the European moss survey since 1990 (Harmens et al., 2012). The decline in cadmium and lead concentrations in mosses since 1990 (77% and 51% respectively) was in good agreement with the decline in atmospheric deposition modelled by EMEP across Europe (74% and 51% respectively; Travnikov et al., 2012). However, correlations are weaker for mercury (Schröder et al., 2010b), which is probably due to the specific chemistry of elemental mercury (Harmens et al., 2010). The decline (14%) in mercury concentration in mosses since 1995 was less than the decline in atmospheric deposition (27%) modelled by EMEP across Europe (Harmens et al., 2015).

Nitrogen

Determination of the nitrogen concentration in mosses was included for the first time in the 2005 European moss survey (Harmens et al., 2011). The number of moss sampling sites was reduced from 3,070 in 2005 to 2,413 in 2010. Atmospheric nitrogen deposition is the main factor determining the spatial variation of the nitrogen concentration in mosses (Schröder et al., 2010a). The relationship between nitrogen concentration in mosses and EMEP modelled nitrogen deposition (Harmens et al., 2011) or measured nitrogen deposition near the moss sampling sites (Harmens et al., 2014) is asymptotic with saturation of the nitrogen concentration in mosses occurring at a nitrogen deposition rate between 15 – 20 kg ha^{-1} yr^{-1}. Although this makes it difficult to assess the magnitude of exposure in areas with medium to high nitrogen deposition, the moss technique still allows the identification of the areas potentially most exposed. Areas most exposed to high nitrogen deposition are located in western and central Europe. The small decline (5%) in the European average median nitrogen concentration in mosses between 2005 and 2010 is in agreement with the 7% decline reported by EMEP for modelled total nitrogen deposition in the EU27 since 2005 (Fagerli et al., 2012; Harmens et al., 2015).

POPs

A recent review study (Harmens et al., 2013) by the ICP Vegetation has shown that mosses are also suitable as biomonitors of POPs, including polycyclic aromatic hydrocarbons (PAHs), polychlorobiphenyls (PCBs), dioxins and furans (PCDD/Fs), and polybrominated diphenyl ethers (PBDEs; flame retardants). Six countries conducted a pilot study in 2010 on the application of mosses to monitor atmospheric deposition of POPs: France, Norway, Poland, Slovenia, Spain and Switzerland, with only Norway analysing other compounds in addition to PAHs. In Norway, the observed geographical distribution of the concentration of selected POPs (PCB, DDT (dichlorodiphenyl-trichloroethane), HCH (hexachlorohexane) PAHs, PBDEs and PFAS (perflorinated compounds)) in mosses indicated that the concentration in mosses reflect the atmospheric deposition patterns well. For most of the POPs the concentration in mosses decreased with northern latitude (similar to heavy metals), indicating that long-range atmospheric transport contributes to the higher concentrations observed in southern Norway. In Switzerland, high concentrations of PAHs were found in mosses sampled in the region of Basel (chemical industry), whilst low concentrations were observed in the western part of the central plateau where the population density is relatively low. There was a good correlation between the summed PAHs concentration in mosses and the concentration in PM_{10} and soil in Switzerland. The total PAHs concentrations in mosses was significantly lower in Navarra, a rural area in Spain, than in Île-de-France (metropolitan area of Paris) and in Switzerland (Foan et al., 2014). Mosses sampled in Navarra were characterised by a low percentage of heavy PAHs due to the low degree of urbanisation in Navarra. The main PAH emission sources in Switzerland and Navarra were industrial activity and road traffic respectively.

CONCLUSIONS

The ICP Vegetation has expanded since its establishment in 1987 from a group of experts focussing on the impacts of ozone pollution on crops to a group focussing either on the impacts of ozone pollution on crops and (semi-)natural vegetation or the deposition of heavy metals, nitrogen and POPs to mosses. Activities on ozone included collation of evidence of ozone impacts on vegetation, establishing ozone dose-response relationship and critical levels and reviewing literature on the impacts of ozone on various ecosystem services and biodiversity. These activities have been important in supporting the development of Protocols by the LRTAP Convention and assessing the sufficiency and effectiveness of these Protocols. Ozone pollution remains a problem at the global scale, with peak concentrations declining in many parts of Europe but background concentrations rising. There is a need to further assess the modifying influences of other air pollutants (e.g. nitrogen) and climate change (rising carbon dioxide, warming, extreme events such as drought and flooding) on the impacts of ozone on vegetation. Although the deposition of heavy metals has declined in many areas across Europe in recent decades, heavy metal pollution still remains a problem in (south)–eastern Europe. Mercury pollution remains a problem at the global scale.

ACKNOWLEDGEMENTS

We thank the United Kingdom Department for Environment, Food and Rural Affairs (Defra; contract AQ0833 and previous contracts), the UNECE (Trust Fund) and the Natural Environment Research Council (NERC) for funding the ICP Vegetation Programme Coordination Centre at CEH Bangor, UK. We thank all participants of the ICP Vegetation for their invaluable contributions to the programme over the years.

References

Ball G.R., Benton J., Palmer-Brown. D. Fuhrer J., Skarby L., Gimeno B.S., Mills G. (1998). Identifying factors which modify the effects of ambient ozone on white clover (*Trifolium repens*) in Europe. Environmental Pollution 103, 7-16.

Berg T., Hjellbrekke A., Rühling Å., Steinnes E., Kubin E., Larsen M.M., Piispanen J. (2003). Absolute deposition maps of heavy metals for the Nordic countries based on the moss survey. TemaNord 2003:505, Nordic Council of Ministers, Copenhagen, Denmark.

Berg T., Steinnes E. (1997). Use of mosses (*Hylocomium splendens* and *Pleurozium schreberi*) as biomonitors of heavy metal deposition: from relative to absolute values. Environmental Pollution 98, 61-71.

Burkey K.O., Miller J.E., Fiscus E.L. (2005). Assessment of ambient ozone effects on vegetation using snap bean as a bioindicator species. Journal of Environmental Quality 34, 1081-1086.

Emberson L., Simpson D., Tuovinen J.-P., Ashmore M.R., Cambridge H.M. (2000). Towards a model of ozone deposition and stomatal uptake over Europe. EMEP MSC-W Note 6/2000.

Fagerli H., Gauss M., Steensen B.M., Benedictow A.C., Hjellbrekke A.-G., (2012). EMEP/MSC-W model performance for acidifying and eutrophying components and photo-oxidants in 2010. Supplementary material to EMEP Status Report 1/2012.

Foan L., Leblond S., Thöni L., Raynaud C., Santamaria J.M, Sebilo M., Simon V. (2014). Spatial distribution of PAH concentrations and stable isotopic signatures (δ13C, δ15N) in mosses from three European areas - Characterization by multivariate analysis. Environmental Pollution 184, 113-122.

Harmens H., Buse A., Büker P., Norris D., Mills G., Williams B., Reynolds B., Ashenden T.W., Rühling Å., Steinnes E., (2004). Heavy metal concentration in European mosses: 2000/2001 survey. Journal of Atmospheric Chemistry 49, 425-436.

Harmens H., Foan L., Simon V., Mills G. (2013). Terrestrial mosses as biomonitors of atmospheric POPs pollution: A review. Environmental Pollution 173, 245-254.

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., Kvietkus K., Leblond S., Liiv S., Magnússon S.H., Maňkovská 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., Mills G. (Eds) (2012). Ozone pollution: Impacts on carbon sequestration in Europe. ICP Vegetation Programme Coordination Centre, CEH Bangor, UK. ISBN: 978-1-906698-31-7.

Harmens H., Mills G. (Eds.). (2014). Air pollution: Deposition to and impacts on vegetation in (South-)East Europe, Caucasus, Central Asia (EECCA/SEE) and South-East Asia. ICP Vegetation Programme Coordination Centre, CEH Bangor, UK. ISBN: 978-1-906698-48-5.

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., Gonzáles-Miqueo L., Jeran Z., Leblond S., Liiv S., Maňkovská B., Pesch R., Poikolainen J., Rühling Å., Santamaria J. M., Simonèiè 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., Norris D.A., Koerber G.R., Buse A., Steinnes E., Rühling Å. (2007). Temporal trends in the concentration of arsenic, chromium, copper, iron, nickel, vanadium and zinc in mosses across Europe between 1990 and 2000. Atmospheric Environment 41, 6673-6687.

Harmens H., Norris D.A., Koerber G.R., Buse A., Steinnes E., Rühling Å. (2008). Temporal trends (1990 – 2000) in the concentration of cadmium, lead and mercury in mosses across Europe. Environmental Pollution 151, 368-376.

Harmens H., Norris D.A., Sharps K., Mills G., Alber R., Aleksiayenak Y., Blum O., Cucu-Man S.-M., Dam M., De Temmerman L., Ene A, Fernández J.A., Martinez-Abaigar J., Frontasyeva M., Godzik B., Jeran Z., Lazo P., Leblond S., Liiv S., Magnússon S.H., Maňkovská B., Pihl Karlsson G., Piispanen J., Poikolainen J., Santamaria J.M., Skudnik M., Spiric Z., Stafilov T., Steinnes E., Stihi C., Suchara I., Thöni L., Todoran R., Yurukova L, Zechmeister H.G. (2015). Heavy metal and nitrogen concentrations in mosses are declining across Europe whilst some "hotspots" remain in 2010. Environmental Pollution 200, 93-104.

Harmens H., Norris D.A., Steinnes E., Kubin E., Piispanen J., Alber R., Aleksiayenak Y., Blum O., Coşkun M., Dam M., De Temmerman L., Fernández J.A., Frolova M., Frontasyeva M., González-Miqueo L., Grodzińska K., Jeran Z., Korzekwa S., Krmar M., Kvietkus K., Leblond S., Liiv S., Magnússon S.H., Maňkovská B., Pesch R., Rühling Å., Santamaria J.M., Schröder W., Spiric Z., Suchara I., Thöni L., Urumov V., Yurukova L., Zechmeister H.G. (2010). Mosses as biomonitors of atmospheric heavy metal deposition: spatial and temporal trends in Europe. Environmental Pollution 158, 3144-3156.

Harmens H., Schnyder E., Thöni L., Cooper D.M., Mills G., Leblond S., Mohr K., Poikolainen J., Santamaria J., Skudnik M., Zechmeister H.G., Lindroos A.-J., Hanus-Illnar A. (2014). Relationship between site-specific nitrogen concentrations in mosses and measured wet bulk atmospheric nitrogen deposition across Europe. Environmental Pollution 194, 50-59.

Hayes F., Jones M.L.M., Mills G., Ashmore M. (2007a). Meta-analysis of the relative sensitivity of semi-natural vegetation to ozone. Environmental Pollution 146, 754-762.

Hayes F., Mills G., Harmens H., Norris D. (2007b). Evidence of widespread ozone damage to vegetation in Europe (1990 – 2006). ICP Vegetation Programme Coordination Centre, CEH Bangor, UK. ISBN 978-0-9557672-1-0.

Heagle A.S., Miller J.E., Chevone B.I., Dreschel T.W., Manning W.J., Cool P.M.M., Morrison C.L., Neely G.E., Rebbeck J. (1995). Response of a white clover indicator system to tropospheric ozone at eight locations in the United States. Water Air and Soil Pollution 85, 1373-1378.

Holy M., Pesch R., Schröder W., Harmens H., Ilyin I., Alber R., Aleksiayenak Y., Blum O., Coşkun M., Dam M., De Temmerman L., Fedorets N., Figueira R., Frolova M., Frontasyeva M., Goltsova N., González Miqueo L., Grodzińska K., Jeran Z., Korzekwa S., Krmar M., Kubin E., Kvietkus K., Larsen M., Leblond S., Liiv S., Magnússon S., Maňkovská B., Mocanu R., Piispanen J., Rühling Å., Santamaria J., Steinnes E., Suchara I., Thöni L., Turcsányi G., Urumov V., Wolterbeek H.T., Yurukova L., Zechmeister H.G. (2009). First thorough identification of factors associated with Cd, Hg and Pb concentrations in mosses sampled in the European Surveys 1990, 1995, 2000 and 2005. Journal of Atmospheric Chemistry 63, 109-124.

Jones M.L.M., Hayes F., Mills G., Sparks T.H., Fuhrer J. (2007). Predicting community sensitivity to ozone, using Ellenberg Indicator values. Environmental Pollution 146, 744-753.

LRTAP Convention (1996). Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded. Texte 71/96, Umweltbundesamt, Berlin, Germany.

LRTAP Convention (2015). Manual on methodologies and criteria for modelling and mapping Critical Loads and Levels and air pollution effects, risks and trends. Chapter 3: Mapping critical levels for vegetation. http://icpvegetation.ceh.ac.uk

Manes F., De Santis F., Giannini M.A., Vazzana C., Capogna F., Allegrini I. (2003). Integrated ambient ozone evaluation by passive samplers and clover biomonitoring mini-stations. The Science of the Total Environment 308, 133-141.

Manes F., Incerti G., Salvatori E., Vitale M., Ricotta C., Costanza R. (2012). Urban ecosystem services: tree diversity and stability of tropospheric ozone removal. Ecological Applications 22, 349-360.

Mills G., Buse A., Gimeno B., Bermejo V., Holland M., Emberson L., Pleijel H. (2007a). A synthesis of AOT40-based response functions and critical levels of ozone for agricultural and horticultural crops. Atmospheric Environment 41, 2630-2643.

Mills G., Harmens H. (Eds) (2011). Ozone pollution: A hidden threat to food security. ICP Vegetation Programme Coordination Centre, CEH Bangor, UK. ISBN: 978-1-906698-27-0.

Mills G., Hayes F., Jones M.L.M., Cinderby S. (2007b). Identifying ozone-sensitive communities of (semi)–natural vegetation suitable for mapping exceedance of critical levels. Environmental Pollution 146, 736-743.

Mills G., Hayes F., Simpson D., Emberson L., Norris D., Harmens H., Büker P. (2011a). Evidence of widespread effects of ozone on crops and (semi-)natural vegetation in Europe (1990 - 2006) in relation to AOT40 - and flux-based risk maps. Global Change Biology 17, 592-613.

Mills G., Pleijel H., Braun S., Büker P., Bermejo V., Calvo E., Danielsson H., Emberson L., González Fernández I., Grünhage L., Harmens H., Hayes F., Karlsson P.-E., Simpson D. (2011b). New stomatal flux-based critical levels for ozone effects on vegetation. Atmospheric Environment 45, 5064-5068.

Mills G., Wagg S., Harmens H. (Eds) (2013). Ozone pollution: Impacts on ecosystem services and biodiversity. ICP Vegetation Programme Coordination Centre, CEH Bangor, UK. ISBN: 978-1-906698-39-3.

Pleijel H. (2011). Reduced ozone by air filtration consistently improved grain yield in wheat. Environmental Pollution 159, 897-902.

Reinert A., Eason G. (2000). Genetic control of O3 sensitivity in a cross between two cultivars of snap bean. Journal of the American Society of Horticultural Science 125, 222-227.

Rühling Å., Tyler G. (1968). An ecological approach to the lead problem, Botaniska Notiser 122, 248-342.

Salvatori E., Fusaro L., Mereu S., Bernardini A., Puppi G., Manes F. (2013). Different O_3 response of sensitive and resistant snap bean genotypes (*Phaseolus vulgaris* L.). The key role of growth stage, stomatal conductance, and PSI activity. Environmental and Experimental Botany 87, 79-91.

Schröder W., Holy M., Pesch R., Harmens H., Fagerli H., Alber R., Coşkun M., De Temmerman L., Frolova M., González-Miqueo L., Jeran Z., Kubin E., Leblond S., Liiv S., Maňkovská B., Piispanen J., Santamaría J.M., Simonèiè P., Suchara I., Yurukova L., Thöni L., Zechmeister H.G. (2010a). First Europe-wide correlation analysis identifying factors best explaining the total nitrogen concentration in mosses. Atmospheric Environment 44, 3485-3491.

Schröder W., Holy M., Pesch R., Harmens H., Ilyin I., Steinnes E., Alber R., Aleksiayenak Y., Blum O., Coşkun M., Dam M., De Temmerman L., Frolova M., Frontasyeva M., González-Miqueo L., Grodzińska K., Jeran Z., Korzekwa S., Krmar M., Kubin E., Kvietkus K., Leblond S., Liiv S., Magnússon S., Maňkovská B., Piispanen J., Rühling Å., Santamaria J., Spiric Z., Suchara I., Thöni L., Urumov V., Yurukova L., Zechmeister H.G. (2010b). Are cadmium, lead and mercury concentrations in mosses across Europe primarily determined by atmospheric deposition of these metals? Journal of Soils and Sediments 10, 1572-1584.

Sitch S., Cox P.M., Collins W.J., Huntingford C. (2007). Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. Nature 448, 791-794.

Travnikov O., Ilyin I., Rozovskaya O., Varygina M., Aas W., Uggerud H.T., Mareckova K., Wankmueller R. (2012). Long-term changes of heavy metal transboundary pollution of the environment (1990-2010). EMEP Status Report 2/2012.

Working Group on Effects (2004). Review and assessment of air pollution effects and their recorded trends. Working Group on Effects, Convention on Long-range Transboundary Air Pollution. Natural Environment Research Council, UK. ISBN 1 870393 77 5.