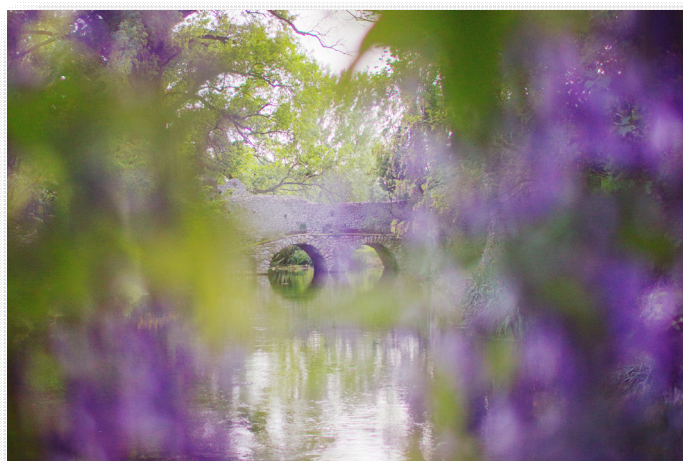


## Effect-based activities on air pollution:

What is the state of the natural and anthropogenic Italian ecosystems?

Edited by Ilaria D'Elia, Alessandra De Marco, Giovanni Vialetto



# Effect-based activities on air pollution: what is the state of the natural and anthropogenic Italian ecosystems?

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## PREFACE

The Division Models and Technologies for Risks Reduction of the ENEA Sustainability Department was established in July 2015 with the specific aim of assemble all the resources, both human and instrumental, that could propose reflections, analyzes and assessments about the interactions among energy strategies, emissions of pollutants and greenhouse gases, air quality, climate change and resulting impacts on health and ecosystems.

The use of numerical models, at different spatial scales, to describe the physical and chemical processes affecting the atmosphere and the ocean is at the hearth of the history and activities of the research groups involved. Nevertheless, great effort and commitment was also dedicated to the development of integrated models to explore links and feedback between the different aspects of the same system. These unique platforms called IAM (Integrated Assessment Modelling) help and support policy-makers to develop and promote sustainable policies.

With this vision we have been developing the national model MINNI (Integrated National Model in support to the International Negotiation on Air Pollution) since 2003. It includes the Integrated Assessment Model GAINS-Italia, which helped our Country: to negotiate revisions of European and International Directives and Protocols with technical data and national scenario simulations; to support technically the actions needed to deal with European infringement procedures; to help the Regional Authorities to fully understand and use the measurements until they have become totally autonomous; to support the early season of the Air Quality Regional Plans together with the assessment of the measures identified in them.

A parallel and equally far-sighted vision has been offered by our colleagues of the actual Division who have been dealing for years with the “pollution effects”. They hold significant roles within the International Convention on Long Range Transboundary Air Pollution (CLRTAP) of the United Nations Economic Commission for Europe (UNECE) both as national representatives in the Executive Body and National Focal Point or Experts within technical Task Forces.

To date a lot of work has been done but we need more and more of each other’s expertise to move forward in the complexity. We cannot have disciplinary “exclusivity” because atmospheric pollution and its effects are no longer (and have never been) a single discipline theme. At the same time we must avoid that integration could generate only simplification.

The main intent of this publication and of this day is to take stock of the situation in Italy, sharing what has been done both on the atmospheric pollution and effects side as well as the many and important experiences of comparison and integration. We would like to start from here in order to deal with what Europe strongly advocates: that the definition of policies to face air pollution considers as a priority the reduction of its effects.

Rome, March 2017.

Gabriele Zanini

*(Head of Division Models and Technologies for the Risks Reduction)*



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## CHAPTER 1 – INTRODUCTION

In the last twenty years, a large effort has been carried out in the Italian community working on the impacts of atmospheric pollution on different ecosystems, both natural or anthropogenic. For long time the work was done without the necessary interconnection between the different sub-networks belonging to the Working Group on Effects. In order to bridge the gaps between scientists and policy makers, the communication between the work done in the different sub-groups has been recently improved and the realization of this monographic text is one of its first tangible result.

The intent of the text is to collate information from the important fields of measurements and modeling to obtain dose-response relationships able to estimate the impacts of air pollutant on the different ecosystems.

Italy is the richest country of the world in terms of biodiversity, agricultural products and number of cultural heritage sites exposed to negative impacts of air pollution and climate change. The Italian country is formed by different peculiar ecosystems, from coastal area to forests, to cities included in the UNESCO list for cultural heritage. It is thus very important to protect our environmental and cultural heritage from environmental pressure and from anthropogenic footprint. A wide choice of measures to control atmospheric pollution and decrease the negative impacts on health and ecosystems is available and possible alternative methodologies, like for example the change in the human behaviours, could have a high importance in a future sustainable society.

More in general, the analysis of the Italian monitoring network shows that the health of the main natural ecosystems has improved, even if there is still some to be done at local level and for specific pollutants. For instance, due to the peculiar conditions of sun exposure and climate, the situation for ozone is still very critical in a Mediterranean environment.

In some cases, the efficacy of the strategies of controlling air pollution can be impaired by climate change, while in some others, strategies to reduce air pollution can be considered as win-win policies, because are beneficial from several points of view.

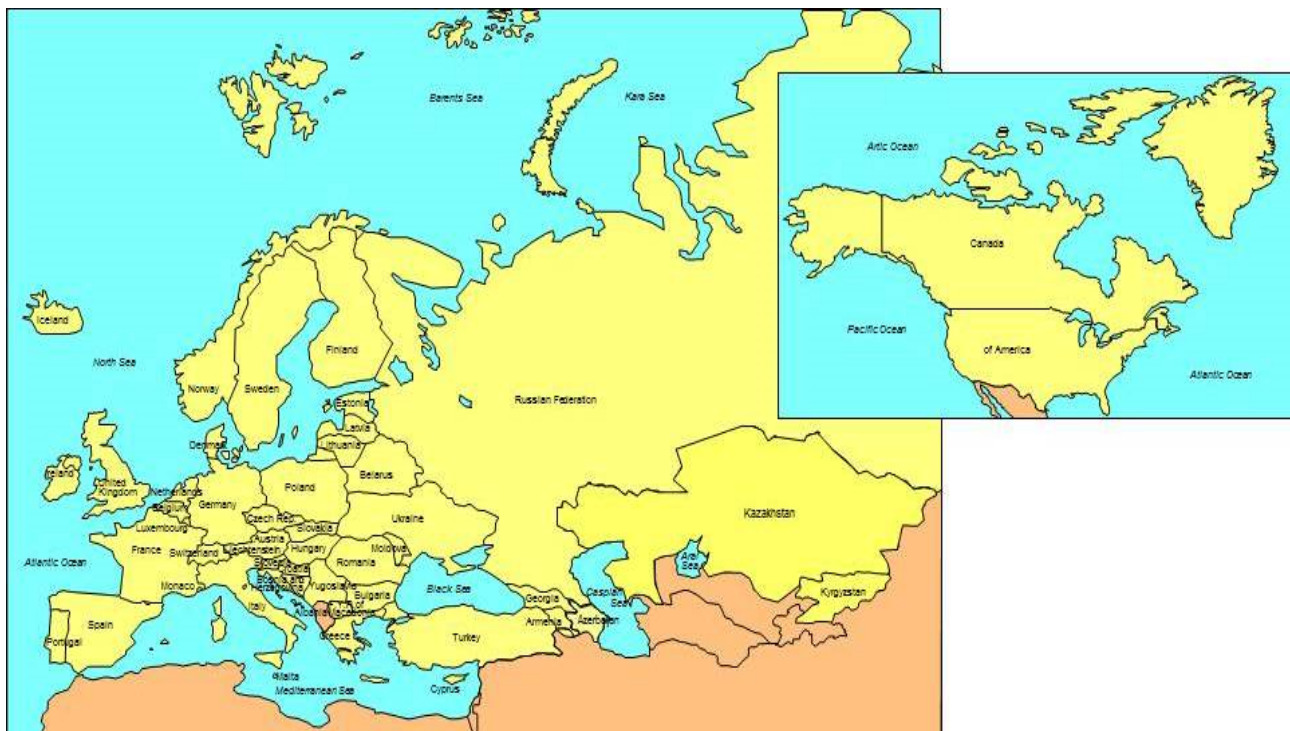
The revision of the National Emission Ceilings Directive (NECD) has established emission reduction targets focusing on the reduction of the impacts of air pollution on ecosystems and human health. This important change captured more attention to the monitoring activities of the impacts of air pollution on natural and anthropogenic ecosystems. This is an emerging issue to promote and to give the due importance to the activities of the Working Group on Effects. In this context, the Italian work has been developed to provide policymakers with a clear picture of what has already been achieved and what can still be done to protect our environment.

The key findings emerged from this work and from the different communities and networks are summarized in the following text.

- **The implementation of the Convention on Long-range Transboundary Air Pollution and its protocols has allowed reaching a significant success in reducing emissions, especially for sulphur, concentrations and deposition trends. Despite this success, exceedances of critical loads and level, of the air quality limit values and high population exposure still exist.**

- Forest condition has improved in Italy since the 1990s, with limited evidence of an impact due to tropospheric ozone. On the other side, there was a distinct effect of nitrogen deposition on forest nutrition, growth and carbon sequestration.
- Biodiversity indices and habitat suitability index in forest sites are recovering consequently to decreasing in pollutant deposition.
- Increasing background O<sub>3</sub> levels in Italy still affect health, productivity and quality of crops and (semi-) natural vegetation. The risk assessment should be based on the effective O<sub>3</sub> dose absorbed through stomata, taking into account the different species/cultivar sensitivity, and the concurrent effect of other stress factors, such as the reactive nitrogen deposition and climatic conditions typical of the Mediterranean area.
- The interactions between air quality and urban vegetation are potentially of great interest, although understanding is still imperfect. In Italy, a few studies have been carried out to estimate potential uptake of pollutant by trees, in particular in two cities: Rome and Florence, in order to estimate and map this Regulating Ecosystem Service.
- Surface water showed a widespread response to decreasing deposition of acidity, sulphate and nitrogen compounds, but the recovery was somewhat delayed, due to the interacting effect of several factors, such as nitrogen saturation of soils in the catchments and climate change.
- Nitrogen deposition will continue to have a prominent role in the acidification processes and in the nitrogen status of surface water. The recovery patterns in the next future will be more and more influenced by climatic factors.
- Atmospheric pollution is a key factor in the deterioration of sensitive materials and materials used in historical and modern cultural heritage are the most vulnerable to air pollution. Even if the decrease of concentrations of air pollutants has led to the decrease of deterioration rates (mainly due to the decrease of SO<sub>2</sub>), current corrosion rates and soiling are still unacceptably high. The cost associated with the damages is enormous but difficult to estimate.
- Health impact assessment of air pollution at national scale in Italy has been carried out in several projects, based on measured data and modelling techniques. Results agreed on showing that PM<sub>10</sub> and NO<sub>2</sub> induce several thousands of premature deaths and hospital admissions per year, due to cardiovascular and respiratory diseases. Confirming conclusions of previous European-scale assessments, air pollution is proved to be a major risk for human health in Italy.
- Many solutions are available and only an integrated approach takes into account the co-benefits of linking air pollution and climate change. The integration of all the different tools, from measures to models, and the coordination among science sectors and different research teams could help in identifying effective environmental policy.

# Effect Oriented Activities in the LRTAP Convention



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## CHAPTER 2 - EFFECT ORIENTED ACTIVITIES IN THE LRTAP CONVENTION

The Convention on Long-range Transboundary Air Pollution (CLRTAP – see web site <http://www.unece.org/env/lrtap/welcome.html>) has achieved successful results in reducing the emissions of a wide range of atmospheric pollutants and consequently in decreasing acidification and eutrophication processes, in soothing ozone peak levels of ozone and photochemical smog, and in ensuring improvements in nitrogen atmospheric levels and deposition rates. The Convention has also proved to be flexible and dynamic in responding to new challenges and problems raised by the transboundary air pollution. The Convention has been also a powerful driver to promote a sound science-for-policy approach to relevant environmental problems not just transferring valuable knowledge and datasets from the scientific community to the decision-makers and involved stakeholders, but also supporting and fostering the whole policy-making process.

However, despite the progress achieved under the Convention, the air quality in the UNECE region is still matter of concern, given its persistent impacts on human health and ecosystems while new environmental problems are emerging (ECE, 2010). A solid and widely recognized scientific basis has been a key success factor for the Convention's outcomes in air pollution abatement. This was enabled by the development of a common and shared knowledge-providers network based on a large system of scientific facilities s managed by authoritative research organizations and aimed at monitoring and modelling a wide set of environmental parameters and ensuring an intense and interdisciplinary exchange of scientists across all the UNECE countries. In addition, the Convention has provided an open-innovation platform for scientists and policymakers to exchange information which has led to a growing and fruitful international cooperation, creating mutual trust, higher policy-harmonization, and the dissemination of good practices in environmental monitoring and assessment. In order to organize its work, the Convention has created an appropriate functional structure, based on expert groups, task forces and research centres. The updated structure of the Convention is showed in Fig. 1.1.

Acidification of soil and water was the main environmental problem in the seventies, caused by the high levels of sulphur and nitrogen atmospheric emissions. Therefore, the reduction of these pollutants was considered as the most urgent need. In the following years, new harmful atmospheric pollutants drew the attention of both the scientific and political community and were thus included into the Convention's protocols. At the same time, new pollutants were being considered in the upcoming policies, such as Non-Methane Volatile Organic Compounds (NMVOC), Heavy Metals (HMs), Persistent Organic Pollutants (POPs), Ozone (O<sub>3</sub>), Particulate Matter (PM) and, more recently Black Carbon (BC).

While the first protocols developed under the Convention focused on technologies capable to reduce emissions, protocols negotiated since the 1990s adopted an effect-oriented approach, aiming at the best cost-effective way to reach the set reduction targets. At the same, it was also recognized that various air pollutants may interact in the atmosphere, thus leading to combined or even synergistic impacts and often depending on the same emission sources. This made a substance-by-substance approach less efficient and was the reason why the so-called multi-pollutant-multi-effect approach was developed. The first protocol based on this new approach was the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (EB, 1999).

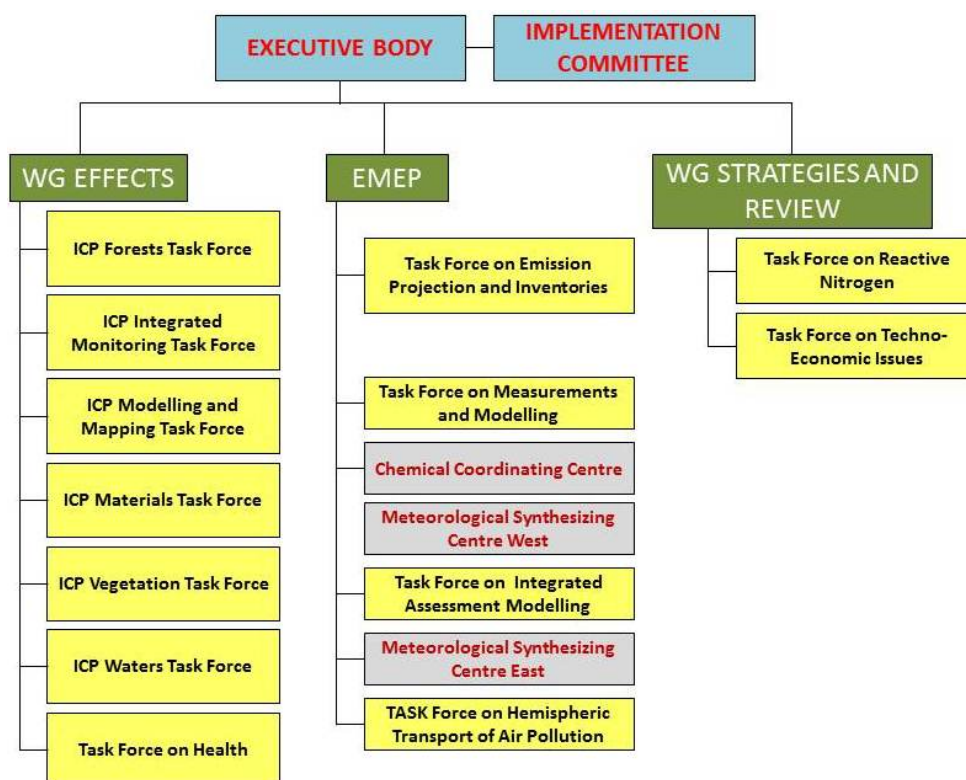


Figure 1.1 – Updated structure of the Convention on Long Range Transboundary Air Pollution.

With the revision of the Gothenburg Protocol, approved in 2012 (ECE, 2012a,b), a further step forward was made, and the particulate matter (PM<sub>2.5</sub> fraction) was also introduced into the Protocol. The focus of the Protocols is targeted to the health protection, which becomes the basis for the optimized emission reduction allocation and commitments for each Party and in doing so, the Convention has achieved considerable success in solving environmental and health problems. These successes are largely due to the scientific grounds of the Convention and the unique way in which science affects any further policy development (look for example Maas et al., 2016). Another key success factor is represented by the Convention's wide geographical coverage. In fact, the Convention embraces most of the northern hemisphere from the West Coast of North America to the Pacific Coast of the Russian Federation (see fig. 1.2).



Figure 1.2 – Map showing the Convention on Long-Range Transboundary Air Pollution signatories.

Italy has a long tradition of activities in the UN-ECE Convention on Long Range Transboundary Air Pollution (UN-ECE LRTAP), since its signature on 14 November 1979, in Geneva and the ratification by the Parliament, occurred on 15 July 1982. The Convention was adopted on 13 November 1979 and left open for signature until 16 November 1979 at the United Nations Office in Geneva. Nowadays, the Convention comprises 32 signatories and 51 Parties.

*Table 1.1 – Status of signature and ratification by Italy of the Protocols of the LRTAP Convention.*

<b>Treaty</b>	<b>Signature</b>	<b>Ratification, Acceptance, Approval, Accession, Succession</b>
LRTAP Convention	14-Nov-79	15 Jul 1982
The 1984 Geneva Protocol on Long-term Financing of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP)	28-Sep-84	12 Jan 1989
The 1985 Helsinki Protocol on the Reduction of Sulphur Emissions or their Transboundary Fluxes by at least 30 per cent	09-Jul-85	5 Feb 1990
The 1988 Sofia Protocol concerning the Control of Emissions of Nitrogen Oxides or their Transboundary Fluxes	01-Nov-88	19 May 1992
The 1991 Geneva Protocol concerning the Control of Emissions of Volatile Organic Compounds or their Transboundary Fluxes	19-Nov-91	30 Jun 1995
The 1994 Oslo Protocol on Further Reduction of Sulphur Emissions	14-Jun-94	14 Sep 1998
The 1998 Aarhus Protocol on Heavy Metals	24-Jun-98	Not yet ratified
The 1998 Aarhus Protocol on Persistent Organic Pollutants (POPs)	24-Jun-98	20 Jun 2006
1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone to the Convention on Long-range Transboundary Air Pollution (Gothenburg Protocol)	01-Dec-99	Not yet ratified

In the following decades 8 Protocols were developed, adopted, signed and ratified by a number of Parties. The table 1.1 summarizes the status of signature and ratification by Italy.

The Protocols on Heavy Metals, on POPs and the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, have been amended and adopted, respectively on 13 December 2012 (Heavy Metals), on 18 December 2009 (POPs), on 4 May 2012 (Gothenburg Protocol). The ratification process of these last three Protocols is still in progress for Italy, although the European Union has accepted both the amended Protocol on Heavy Metals and the amended Protocol on POPs. The last three Protocols, in the amended version mentioned above, are not yet entered into force because the minimum number of ratification by the Parties has not yet been achieved.

The Italian experts participate actively in the works of many technical bodies of the LRTAP Convention. Notably, within the Working Group on Effects, the International Cooperative Programme on Effects of Air Pollution on Materials, including Historic and Cultural Monuments sees the Co-Chair of Italy, since 2005, currently in the person of Pasquale Spezzano. As well, the Task Force on Techno-Economic Issues has the Italian Co-Chair Tiziano Pignatelli, involved since 2006. Moreover, several scientific studies are being conducted under the leadership and /or responsibility of the Italian experts.

Moreover, during the very first discussions on the Convention, it was immediately recognized that a good understanding of the harmful effects of air pollution was a prerequisite for reaching agreement on effective pollution control. To develop the necessary international cooperation in the research on and the monitoring of pollutant effects, the Working Group on Effects (WGE) was established under the Convention in 1980.

The Working Group on Effects provides the Convention's Executive Body with sound and shared scientific information on the degree and geographic extension of the impact of major air pollutants on the human health and some relevant natural and human-heritage targets like water, forests, vegetation and materials.

Such information is the result of monitoring activities on the effects caused by atmospheric pollution across Europe and North America, and are based upon scientific research on dose-response relationships, critical loads and damage evaluation.

WGE is composed by six International Cooperative Programmes (ICPs) and the Task-force on Health. Each ICP includes a Task-Force and a Coordination Centre with the aim of collecting data from monitoring networks. For each ICP, a National Focal Centre (NFC) is established, with the aim to coordinate the national activities, participate in the works of the task-forces, and submit data to the Coordination Centres.

Italy, as Party of the Convention, is therefore committed to appoint the NFCs, to establish and control the monitoring networks, to submit the required data and to participate in the activities of the task-forces. The head of the Italian Delegation to WGE is Alessandra De Marco (ENEA), and the other delegation members are Antonio Ballarin-Denti (Catholic University of Brescia) and Sergio Cinnirella (CNR). All the national Focal Points are reported in table 1.2.

*Table 1.2 – All the National Focal Points to the different International Cooperative Programme under the LRTAP Convention.*

<b>WGE Task Force</b>	<b>Description</b>	<b>National Focal Point</b>	<b>Institution</b>
ICP Forests	ICP on assessment and monitoring of air pollution effects on forests	Angela Farina	State Forestry Authority
		Laura Canini	State Forestry Authority
ICP Integrated Monitoring	ICP on integrated monitoring of air pollution effects on ecosystems	Angela Farina	State Forestry Authority
		Laura Canini	State Forestry Authority
ICP Modelling & Mapping	ICP on modelling and mapping of critical levels and loads and air pollution effects, risks and trends	Patrizia Bonanni	Isprambiente
		Francesca Fornasier	Isprambiente
		Alessandra De Marco	ENEA
		Marcello Vitale	“Sapienza” University of Rome
ICP Materials	ICP on effects of air pollution on materials, including historic and cultural monuments	Pasquale Spezzano (co-chair ICP and NFP)	ENEA
ICP Vegetation	ICP on effects of air pollution on natural vegetation and crops	Fausto Manes	“Sapienza” University of Rome
		Elisabetta Salvatori	“Sapienza” University of Rome
		Alessandra De Marco	ENEA
ICP Waters	ICP on assessment and monitoring of the effects of air pollution on rivers and lakes	Michela Rogora	CNR



# Sources and Emissions of air pollutants



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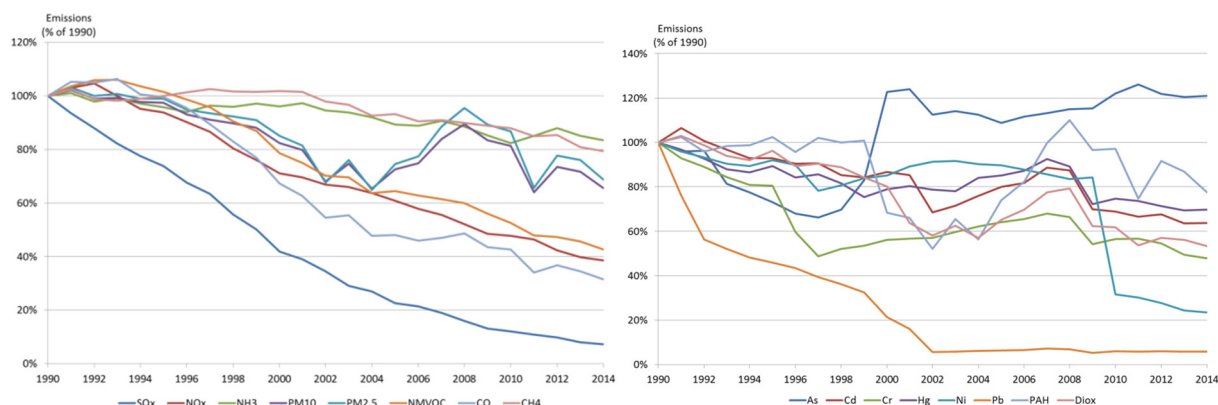
## CHAPTER 3 - SOURCES AND EMISSIONS OF AIR POLLUTANTS

Air pollution is a very important environmental and social issue that poses multiple challenges in terms of management and mitigation (EEA, 2015). Air pollution represents the single largest environmental health risk in Europe and particulate matter has become a major concern for public health whose carcinogenicity has been recognized by the International Agency for Research on Cancer (IARC, 2015). Many measures and policies have been adopted in the past decades at European, national, regional and even local level but a large portion of European population and ecosystems is still exposed to concentrations that exceed the European Union (EC, 2008) and the World Health Organization (WHO, 2005) air quality standards. In this view, the Gothenburg protocol was amended in 2012 and, in 2013, the European Commission adopted the Clean Air Policy Package (COM, 2013) with the aim to further reduce the impacts of harmful emissions on human health and the environment.

Air pollutants are emitted from anthropogenic and natural sources; they may be either emitted directly or formed in the atmosphere and they have a number of impacts on health, ecosystems, the built environment and the climate.

Figure 3.1 shows the trend of the Italian emissions of sulphur oxides (SO<sub>x</sub>), nitrogen oxides (NO<sub>x</sub>), ammonia (NH<sub>3</sub>), primary PM with a diameter of 10 µm or less (PM<sub>10</sub>) and PM with a diameter of 2.5 µm or less (PM<sub>2.5</sub>), non-methane volatile organic compound (NMVOC), carbon monoxide (CO) and methane (CH<sub>4</sub>) between 1990 and 2014. Similarly, figure 3.1 shows the trend in emissions of the toxic metals arsenic (As), cadmium (Cd), nickel (Ni), lead (Pb), mercury (Hg), Polycyclic Aromatic Hydrocarbon (PAH) and dioxins (Diox). In the period 1990-2014, in Italy almost all the primary and precursor emissions have decreased (IIR, 2016). Reductions are especially relevant for the main pollutants, SO<sub>x</sub> (-93%); NO<sub>x</sub> (-61%); CO (- 69%); NMVOC (-57%) and Pb (-94%), while NH<sub>3</sub> shows the smallest reduction (-17%).

Effective action to reduce the impacts of air pollution requires a good understanding of its causes, how pollutants are transported and transformed in the atmosphere, and how they impact on humans, ecosystems and the climate.



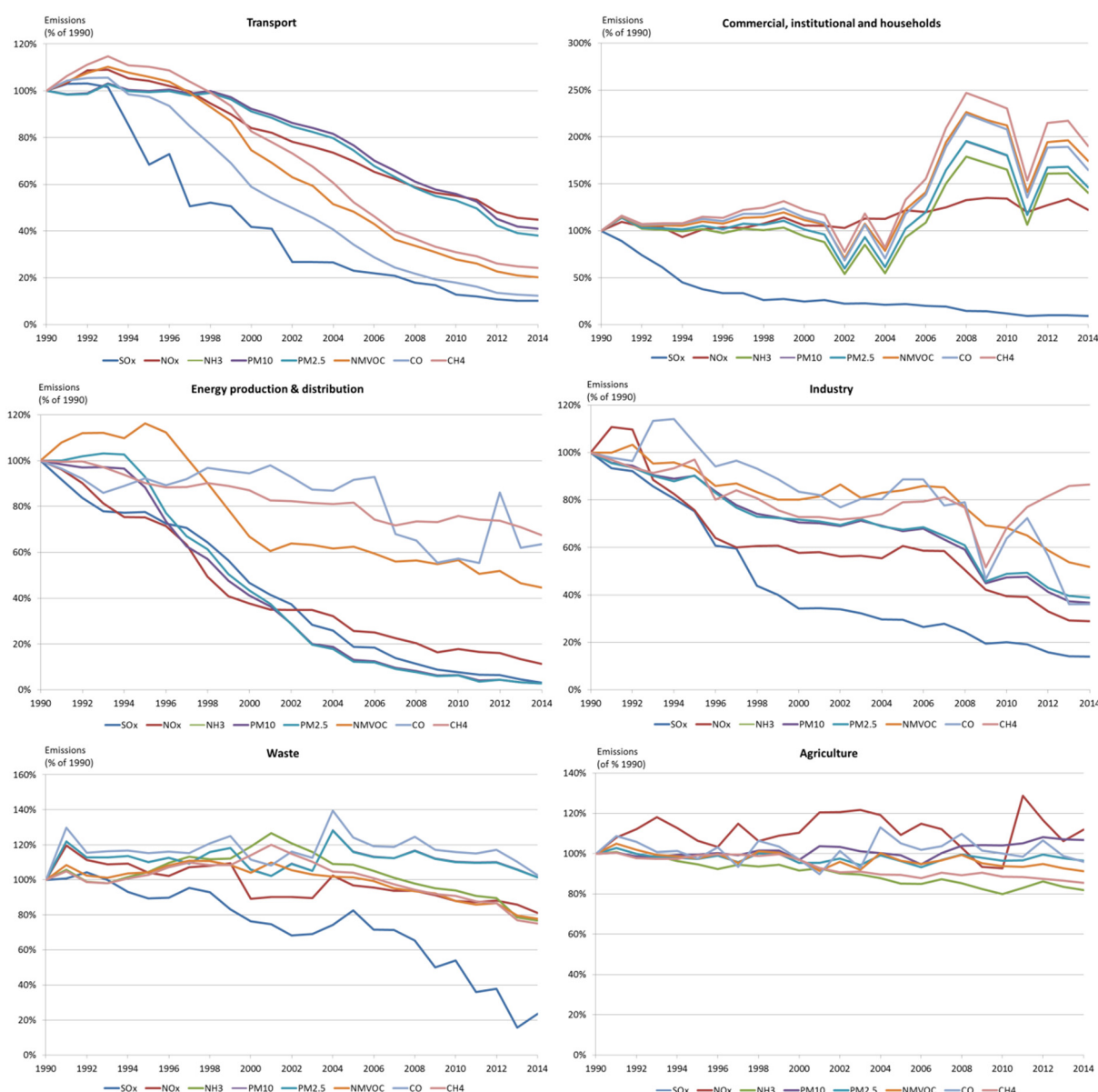
Source: Based on IIR, 2016.

Figure 3.1 – Trend in Italian emissions of SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, NMVOC, CO and CH<sub>4</sub> (top) and of As, Cd, Cr, Hg, Ni, Pb, PAH, Diox (bottom), 1990-2014 (% of 1990 levels).

The major drivers for the trend are reductions in the industrial and road transport sectors, due to the implementation of various European Directives which introduced new technologies, plant emission limits, the limitation of sulphur content in liquid fuels and the shift to cleaner fuels (fig. 3.2). Emissions have also decreased for the improvement of energy efficiency as well as the promotion of renewable energy.

In the following plots, an analysis of the Italian emission trend by sector in the period 1990-2014 is shown. The main source sectors contributing to emissions of air pollutants in Italy are transport, energy, industry, the commercial, institutional and households sector, agriculture and waste.

The transport sector has considerably reduced its emissions of air pollutants in Italy since 1990, as figures 3.2 and 3.3 show, except for PAH emissions, which have increased by 26% from 1990 to 2014. The highest emission reductions from transport between 1990 and 2014 were registered for SO<sub>x</sub> (90%) and for Pb (100%).



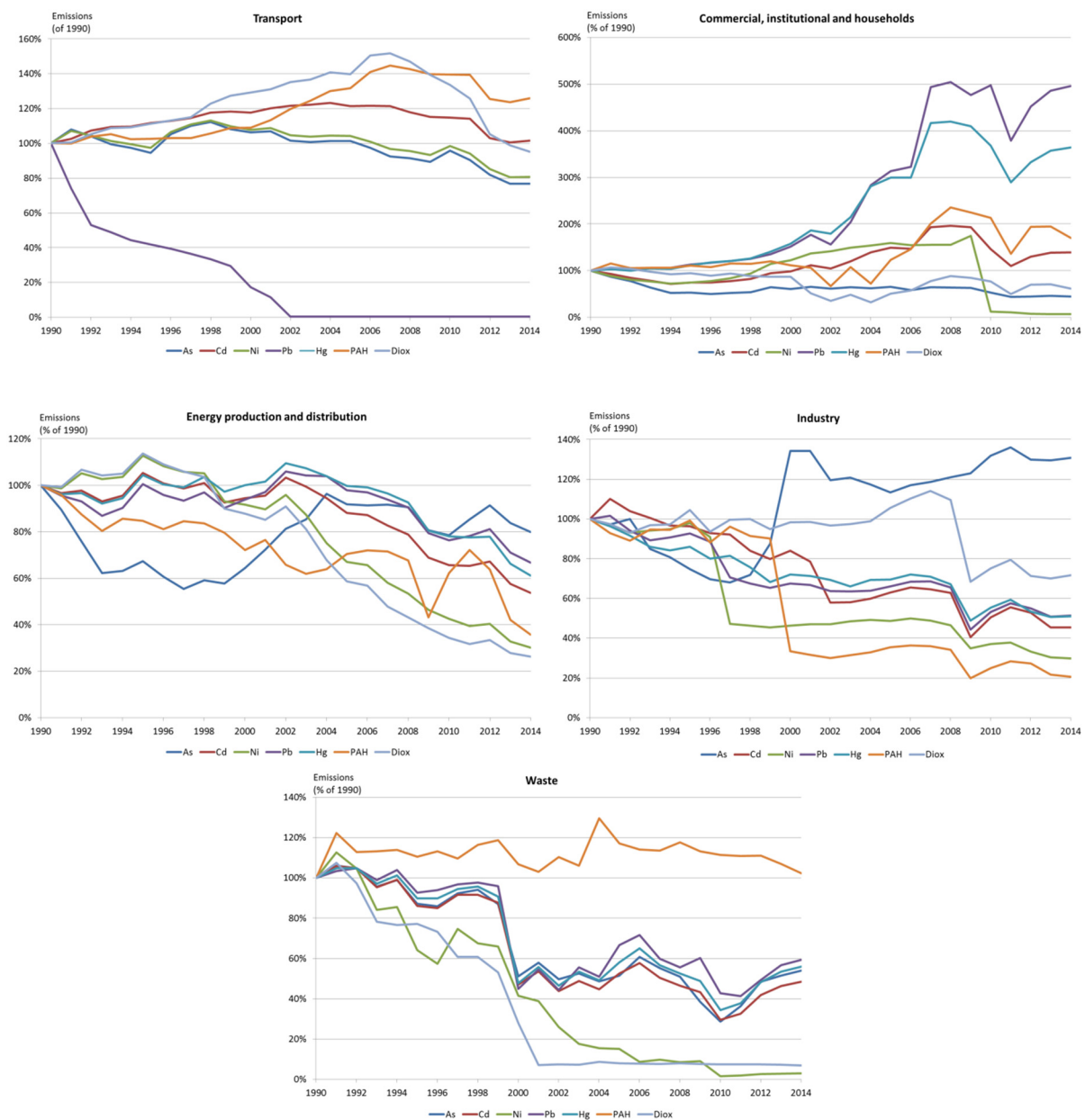
Source: Based on IIR, 2016.

Figure 3.2 – Trend in Italian emissions from main sources of SO<sub>x</sub>, NO<sub>x</sub>, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, NMVOC, CO and CH<sub>4</sub>, 1990-2014 (% of 1990 levels).

With the exception of SO<sub>x</sub> emissions, the commercial, institutional and household sector has significantly increased its emissions, whose trend follows the fuelwood consumption variation. This emission increase is due to the use of household wood and other biomass combustion for heating, owing to government incentives/subsidies, rising costs of other energy sources, or an increased public perception that it is a ‘green’ option (EEA, 2015). Biomass is being promoted as a renewable fuel that can assist with climate change mitigation and contribute to energy security.

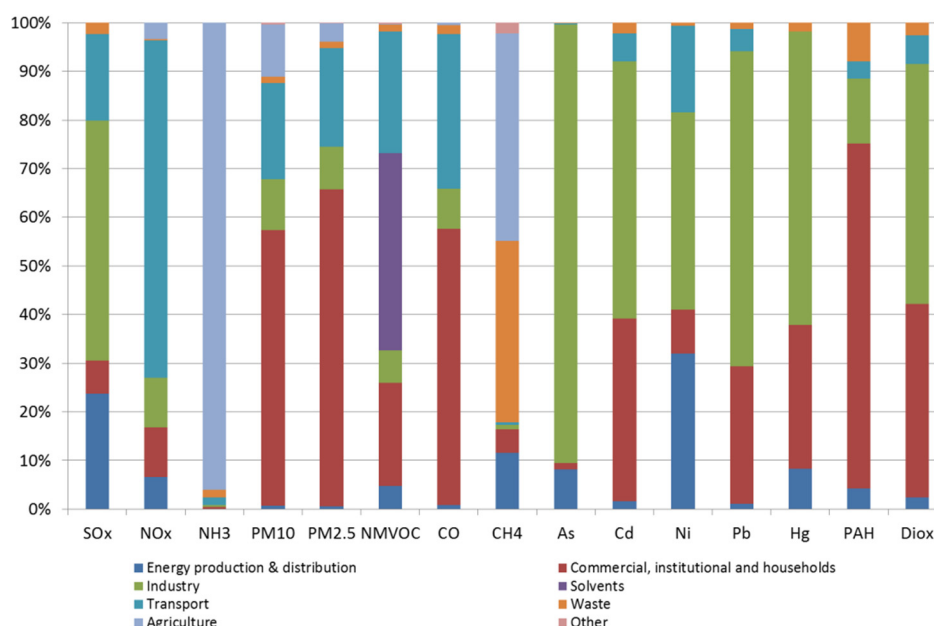
Energy production and industry considerably reduced their air pollutant emissions between 1990 and 2014, with the exception of As emissions from the industrial sector.

Agriculture is the main sector in which emissions of air pollutants have least decreased while waste sector shows a great emission reduction in Diox emissions.



Source: Based on IIR, 2016.

*Figure 3.3 – Trend in Italian emissions from main sources of As, Cd, Ni, Pb, Hg, PAH and Diox, 1990-2014 (% of 1990 levels).*



Source: Based on IIR, 2016.

Figure 3.4 – Main emitting sectors for the main pollutants in the year 2014.

The main emitting sectors in the year 2014 according to the last national emission inventory (IIR, 2016), are shown in fig. 3.4.

Industry is the largest source of SO<sub>x</sub>, As, Cd, Ni, Pb, Hg and Diox, whose emissions respectively represent the 49%, 90%, 53%, 41%, 65%, 60% and 49% of total national emissions.

The transport sector is the biggest contributor to NO<sub>x</sub> emissions, accounting for 69% of total Italian emissions in 2014. However, NO<sub>x</sub> emissions from road transport have not been reduced as much as expected with the introduction of the Euro standards, since emissions in real-life driving conditions for diesel vehicles are often higher than those measured during the approval test.

The commercial, institutional and household sector is by far the most important sector of primary PM<sub>10</sub>, PM<sub>2.5</sub> and PAH emissions contributing respectively to 57%, 65% and 71% of total PM<sub>10</sub>, PM<sub>2.5</sub> and PAH total emissions. Moreover, it is the second most significant emitter of Diox, CO, Cd, Pb and Hg. This sector surely represents a key sector in reducing air pollutant emissions.

The agricultural sector is the greatest emitter of NH<sub>3</sub> and was responsible for 96% of total NH<sub>3</sub> emissions in Italy in 2014. NH<sub>3</sub> emissions have decreased by only 17% from 1990 to 2014 because European policies have cut PM precursor gas emissions, with the exception of NH<sub>3</sub> from agriculture (EEA, 2015).

Emissions have decreased in many sectors except for the commercial, institutional and households sector, whose emissions showed an increase for almost all pollutants and agriculture which showed the lowest reduction. As a consequence, the commercial, institutional and households and agricultural sectors still have a high potentiality to reduce emissions and improve air quality.

In order to reduce the harmful effects of atmospheric pollutants, the National Emission Ceilings Directive (NECD) (EC, 2001) sets EU Member States individual air pollutant emission limits, or 'ceilings', restricting emissions for four important air pollutants: nitrogen oxides (NO<sub>x</sub>), non-methane volatile organic compounds (NMVOC), sulphur dioxide (SO<sub>2</sub>) and ammonia (NH<sub>3</sub>).

*Table 3.1 – Italian progress in meeting ceilings set out in NECD Annexes (EC, 2001).*

Pollutant	NEC Ceilings 2010 (kt)	Emission inventory (kt)				
		2010	2011	2012	2013	2014
SO <sub>2</sub>	475	217	195	176	145	131
NO <sub>x</sub>	990	978	950	867	816	790
NMVOC	1159	1046	954	942	909	849
NH <sub>3</sub>	419	389	402	415	402	393

As of 2010, all Member States are required to meet their emission ceilings. Italy respects its emissions ceilings for all the four pollutants during the period 2010 to 2014 as shown in table 3.1.

Many measures and policies have been adopted in the past decades at European, national, regional and even local level but a large portion of European population and ecosystems is still exposed to concentrations that exceed the World Health Organization (WHO, 2005) and the European Union air quality standards (EC, 2008). In this view, in 2013, the European Commission adopted the Clean Air Policy Package (COM, 2013), with the aim to further reduce the impacts of harmful emissions on human health and the environment, where the proposal for a revised National Emission Ceilings Directive (NECD) plays a major role.

# Actual and potential impact of air pollution on Italian forests: results from the long-term national forest monitoring networks under the ICP Forests



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## CHAPTER 4 - ACTUAL AND POTENTIAL IMPACT OF AIR POLLUTION ON ITALIAN FORESTS: RESULTS FROM THE LONG-TERM NATIONAL FOREST MONITORING NETWORKS UNDER THE ICP FORESTS

This Chapter was prepared for the most part on the basis of evaluations carried out within the LIFE project “Sustainable Monitoring And Reporting To Inform Forest and Environmental Awareness and Protection – SMART4Action” [LIFE13 ENV/IT/000813].

### 4.1 Introduction

Forests in Italy cover ca.  $8.76 \times 10^6$ , stock 1,242 Mt carbon and offer a series of invaluable products and services to the entire society (Gasparini and Tabacchi, 2011; Gasparini et al., 2013). Preserving forest integrity is therefore essential. Although effects are in general less obvious and frequent in comparison with other biotic and abiotic stressors, air pollution may have an impact on several compartments of forest ecosystem, and may endanger forest health, productivity and diversity (Bobbink et al., 2010; Karnosky et al., 2003). In this chapter, we will present status, trends of pressure, impacts and risks placed to forests by the most important air pollution issues in Italy as emerged from data collected by national forest monitoring networks since the mid of the 1990s. To do this, we will concentrate on data originated from harmonized measurements carried out at the national Level I and Level II networks that are parts of the UNECE ICP Forests (Figure 4.1).

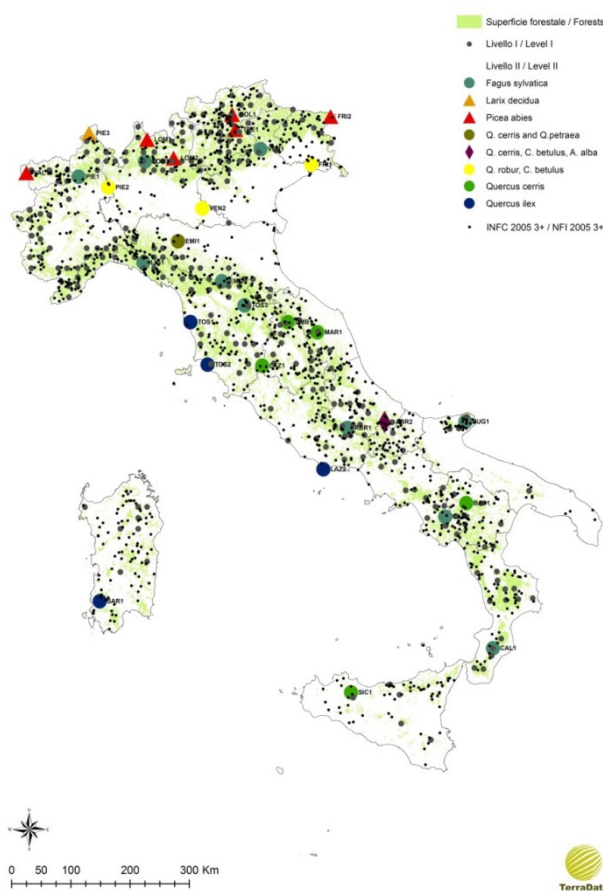


Figure 4.1 – Level I (small black dots) and Level II (large colored symbols) networks in Italy. Colors refer to main tree species in Level II plot (source: LIFE SMART4Action). Sample plots (Phase 3+) of the National Forest Inventory are also reported.

## 4.2 Pressures

Deposition of oxydized ( $\text{NO}_x$ ) and reduced nitrogen ( $\text{NH}_y$ ) and tropospheric ozone ( $\text{O}_3$ ) are the most important air pollution forest-related issues in Italy and elsewhere. Both N deposition and ozone levels have been measured at the sites of the Italian ICP Forests Level II network since the 1990s. Atmospheric deposition is collected in the open field and under forest canopy (throughfall) by different set of funnels (Figure 4.2A). Stemflow collectors (Figure 4.2B) are used in beech plots to collect precipitation running along stems. Ozone concentration is measured using passive samplers (Figure 4.2C) since 1996. A further set of ozone measurements have been carried out over the period 2007 - 2011 on 15 Level I plots in Trentino (Northern Italy) (Gottardini et al., 2010).

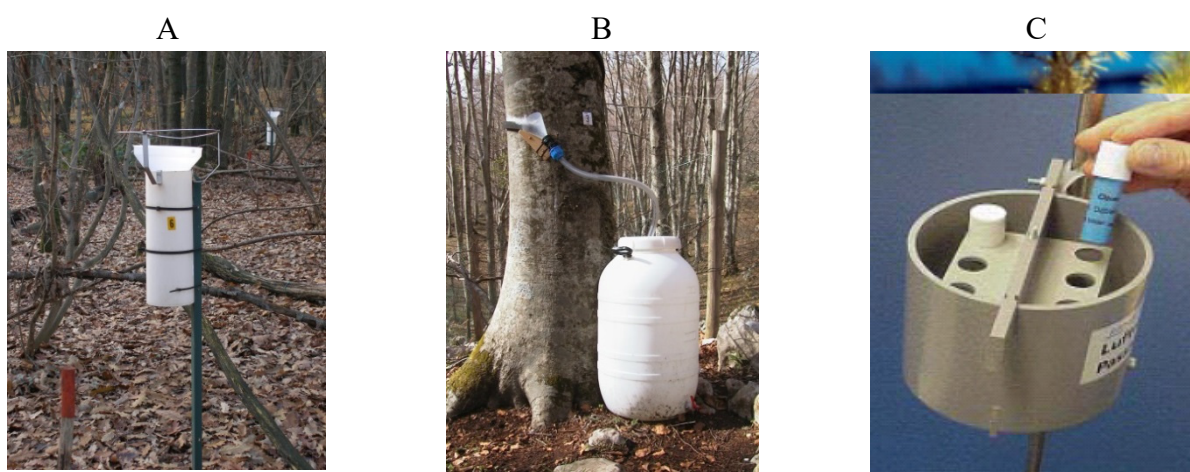


Figure 4.2 – Collectors used for open field and throughfall (A), stemflow (B), and ozone passive samplers (C).

### 4.2.1 Status

Sampling and analysis of atmospheric deposition and of ozone was carried out on a variable number of plots, depending on funding. In particular, in 2009-2011 this activity was carried out and validated on 21 plots. Throughfall deposition represents a good estimate of the deposition reaching the forest soil. The geographic distribution of average throughfall deposition for 2009-2011 of some selected ions and of the pH values obtained from the volume weighted average of hydrogen ion deposition is shown in Figure 4.3.

All the plots show pH values higher than  $> 5$ , mostly around the equilibrium value of 5.6. In the case of sulphate deposition (corrected for the amount deriving by marine aerosol - (CLRTAP, 2004), a geographic pattern is evident, with lower values on the Alps, and higher values in the Southernmost plots. Due to the episodic deposition of Saharan dust, this is leading to episodes with high sulphate and calcium concentration. In the case of nitrate, ammonium and total N (which includes N in organic compounds), high values were measured in the Po plain (where most of industries and farming are located) and in the surrounding hills, while lower values were detected in high elevation Alpine sites.

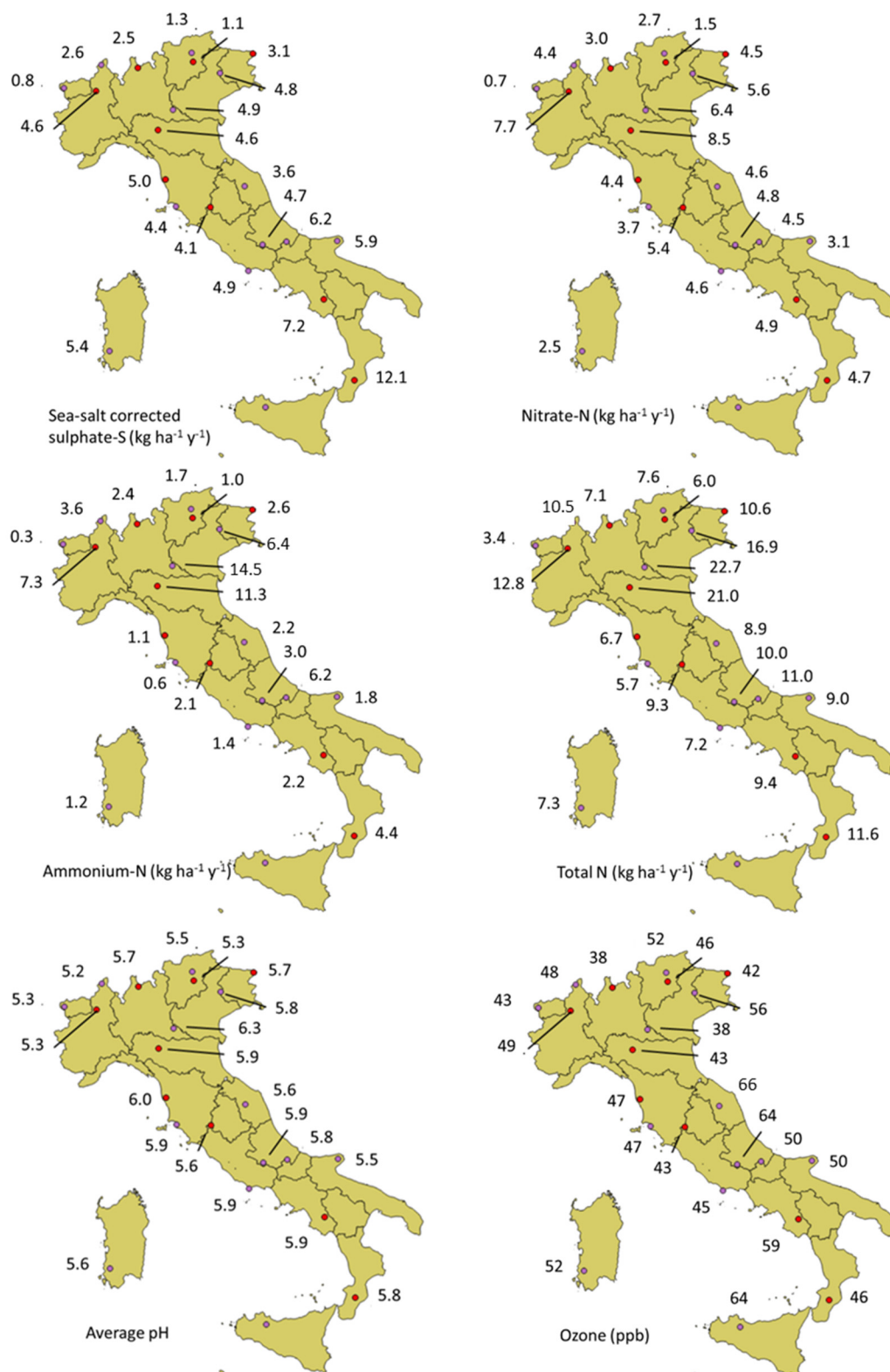


Figure 4.3 – Annual deposition of selected ions and average ozone concentration during the growing season at Level II sites, 2009-2011. Site codes in Figure 4.1. (Source: LIFE SMART4Action).

As for ozone, average concentration during the growing season was in general  $> 40$  ppb in all sites, with slightly higher values in the Southern regions and in the Adriatic part of the peninsula (Figure 4.3).



Figure 4.4 – Mean 2000-2003 AOT40 at 26 Italian Level II sites (dots) and 41 background conventional monitoring stations over the period 2000-2003. After Ferretti et al., 2007.

Table 4.1. Ozone flux estimates available for Level II sites in Italy.

Site	Year	Reporting unit	Flux estimate	Reference
CAL1	2000-2002	seasonal AFst, (mmol O <sub>3</sub> m <sup>-2</sup> )	8-10	Schaub et al., 2007
TOS1	2005	seasonal AFst1.6, (mmol O <sub>3</sub> m <sup>-2</sup> )	53	Bussotti and Ferretti, 2007
TOS2	2005	seasonal AFst1.6, (mmol O <sub>3</sub> m <sup>-2</sup> )	49	Bussotti and Ferretti, 2007
LAZ2	2005	seasonal AFst1.6, (mmol O <sub>3</sub> m <sup>-2</sup> )	47	Bussotti and Ferretti, 2007
TRE1	1999-2011	seasonal AFst1.6, (mmol O <sub>3</sub> m <sup>-2</sup> )	22-35	Gottardini et al., 2012

Accumulated ozone above threshold 40 ppb (AOT40) was estimated to exceed up to 10 times the Critical Level set to protect vegetation (Gerosa et al., 2003, 2007; Ferretti et al., 2007) (Figure 4.4). In terms of ozone flux, estimates were obtained for some Level II sites at individual years (Table 4.1) (Bussotti and Ferretti, 2007).

#### 4.2.2 Trend

Figure 4.5 shows the temporal trend of sulphate, nitrate, and ammonium deposition and ozone concentration in nine forest sites with complete 1997-2013 dataserries. A marked decrease in sulphate deposition is evident in all sites at the end of the 1990s, while the reduction is less evident in the more recent period. Sulphate reduction resulted statistically significant (Seasonal Kendall test on monthly data,  $p < 0.01$ ) in eight out of nine sites. In the case of nitrate and ammonium, a slight decrease was evident only in the last years and it was significant ( $p < 0.05$ ) only in four and one site, respectively.

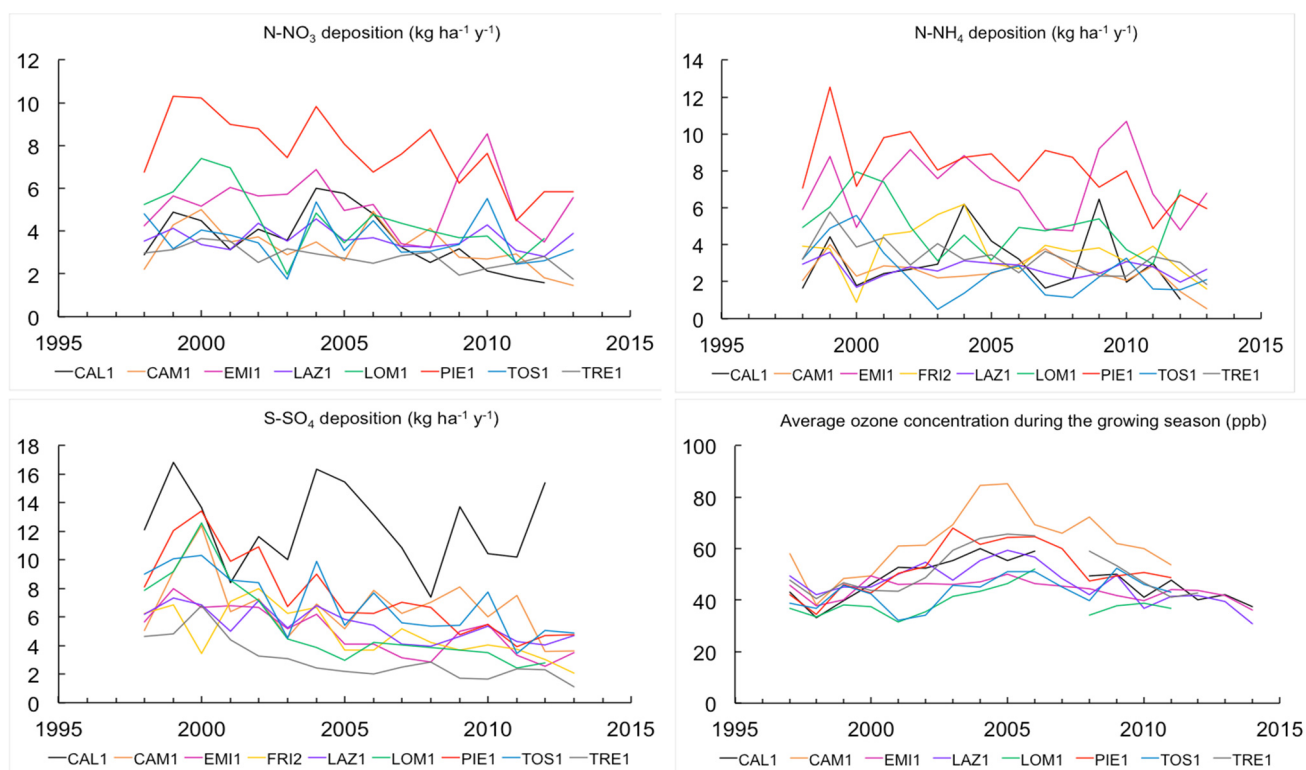


Figure 4.5 – Temporal trend in deposition of selected ions in nine Level II forest plots in Italy.  
(Source: LIFE SMART4Action).

Ozone concentration increased between 1997 and 2005, and decreased between 2005 and 2011 (Figure 4.5). The decreasing pattern is confirmed over the most recent years. As for ozone flux trend, information is available only for one Level II plot, namely TRE1 (Gottardini et al., 2012). At this site, ozone flux in terms of accumulated flux above 1.6 (AFst1.6) follows the same time pattern as AOT40, augmenting from 25 to 35 mmol m<sup>2</sup> PLA between 1999 and 2005, and decreasing to ca. 27 mmol m<sup>2</sup> PLA between 2006 and 2009 (Gottardini et al., 2012).

### 4.3 Impacts

N deposition may affect soil chemistry, nutrient balance, alter species-host interactions, biomass allocation, sensitivity to climate and to ozone. Ozone may reduce photosynthesis, cause premature senescence and direct injury to foliage. All together, this may have direct and indirect impact on forest health, growth, plant species diversity, and nutrition.

#### 4.3.1 Forest health

In Italy, forests health has been measured under quality assured conditions since 1997. The most reliable variable adopted was (and is) defoliation, assessed both on Level I (ca. 250 plots) and Level II (31 plots) networks (see Figure 4.1). Direct injury to foliage due to ozone has been also assessed on some sites.

On Level I, mean defoliation for the 1997-2014 period was higher in broadleaves (39.7% of trees with defoliation higher than 25%) than in conifers (24.1%). Defoliation shows lower level in the South (30.7%) with respect to the North (35.5%) and Central (36.0%) Italy. Broadleaves were more defoliated (43.6%) in Northern Italy and less defoliated (35.8%) in Southern Italy. In conifers, defoliation was higher in Central Italy (36.9%) than in Southern Italy (12.3%).

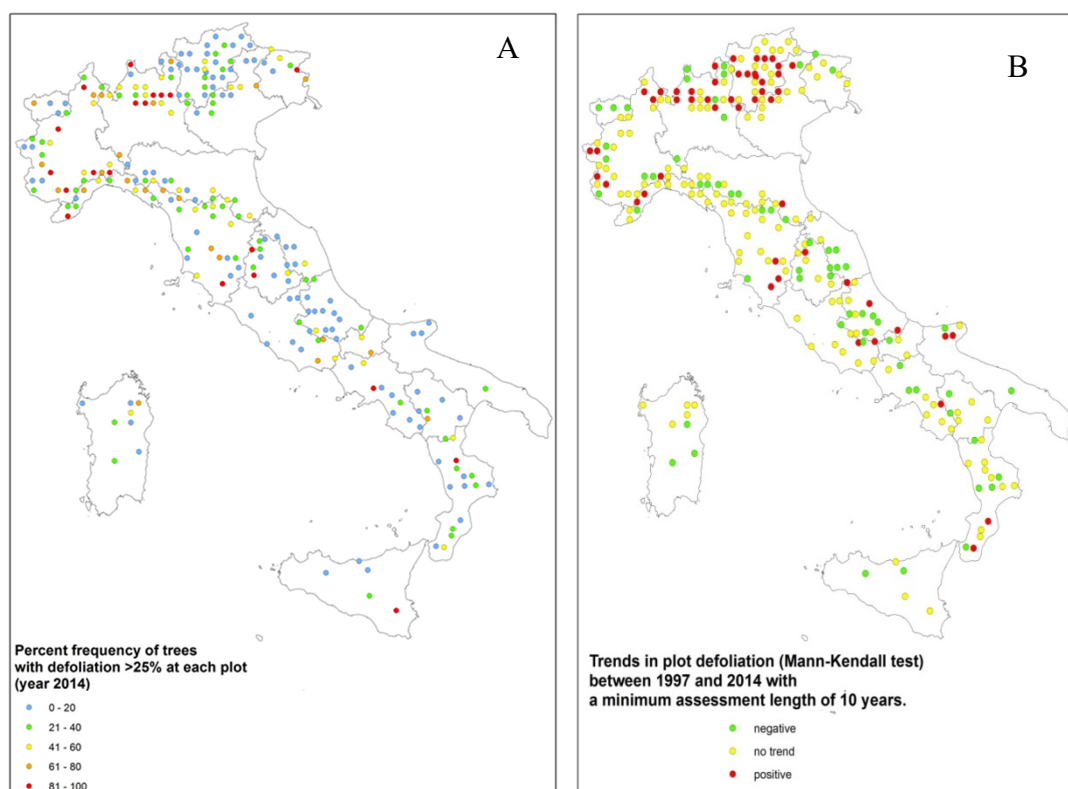


Figure 4.6 – A) Percentage of trees with defoliation higher than 25% in the Level I plots, year -2014. B) 1997-2014 trend of the percentage of trees with defoliation >25%. Plots with at least 10 years of observations were analyzed with Mann-Kendall test. Significant increases (positive, red) and decreases (negative, green) of defoliation are indicated. (Source: LIFE SMART4Action).

The distribution of the defoliated trees (>25%) at each Level I plot (Figure 4.6A) reveals higher frequency in the North-Western regions of Italy (Piedmont, Lombardy, Liguria and partly the Northern areas of Tuscany). The analysis of the 1997-2014 trends (Figure 4.6B) evidenced that plots with significant increase of defoliated trees were concentrated especially in the Northern regions and in the Alpine area. All in all, however, there is a significant ( $P < 0.01$ ) decreasing trend for defoliation, mostly driven by broadleaves.

Level II results confirm substantially the pattern described for the level I. Defoliation is increasing in Northern Italy (with the exception of VAL1) and decreasing in Central and Southern Italy (with the exception of LAZ1).

Foliar symptoms attributable to ozone injury were assessed at 12 Level II sites over the period 2003-2009 (Bussotti and Ferretti, 2007, 2009; Gottardini et al., 2012) and on a subset of Level I plots in Trentino in 2008-2009 (Gottardini et al., 2012). While no symptom was reported for Level I plots in Trentino, 46 *taxa* were found symptomatic at the investigated Level II sites. Off-plot investigation carried out nearby Level I plots in Trentino revealed symptoms on additional species, noticeably on *Viburnum lantana* (Gottardini et al., 2012).

#### 4.3.2. Forest growth

Forest growth has been monitored since 1997 on Level II plots, and 17 plots have full coverage of the monitoring period 1997-2015 (i.e. five subsequent inventories). Current volume increment ( $IcV$ , the difference in volume between two subsequent years) and mean volume increment ( $ImV$ , the ratio between the volume and tree's age) were calculated on the basis by the volume functions provided

by the Italian National Forest Inventory (Tabacchi et al., 2011). Concerning absolute values and expected trends, data are summarized in Table 4.2.

*Table 4.2 – Estimated volume and increments for the 17 CONECOFOR plots (hf=high forests; sc=stored coppices; tc=transitory crops). Standard deviation is reported in italic. (Source: LIFE SMART4Action).*

			Volume (m <sup>3</sup> ha <sup>-1</sup> )					Current volume increment (m <sup>3</sup> ha <sup>-1</sup> yr <sup>-1</sup> )				Mean volume increment (m <sup>3</sup> ha <sup>-1</sup> yr <sup>-1</sup> )				
Plot	Age	Type	1997	2000	2005	2010	2015	2000	2005	2010	2015	1997	2000	2005	2010	2015
ABR1	130	hf	430.3	476.9	524.9	596.0	629.6	15.5	9.6	14.2	6.7	3.8	4.1	4.4	4.8	4.8
CAL1	130	hf	539.2	596.5	682.7	743.9	795.4	19.1	17.2	12.2	10.3	4.8	5.2	5.7	6.0	6.1
CAM1	120	hf	711.3	777.5	835.2	882.4	948.1	22.1	11.5	9.5	13.1	7.0	7.4	7.6	7.7	7.9
FRI2	120	hf	740.3	808.0	892.3	962.6	935.9	22.6	16.9	14.1	-5.3	7.3	7.7	8.1	8.4	7.8
LOM1	100	hf	369.3	416.4	478.9	561.3	676.8	15.7	12.5	16.5	23.1	4.5	4.9	5.3	5.9	6.8
PUG1	95	hf	563.0	605.7	658.5	689.9	754.8	14.2	10.6	6.3	13.0	7.3	7.6	7.7	7.7	7.9
TRE1	210	hf	667.8	742.9	762.6	813.4	866.2	25.0	3.9	10.2	10.6	3.5	3.8	3.8	4.0	4.1
VAL1	160	hf	481.8	503.9	549.5	598.7	648.6	7.4	9.1	9.8	10.0	3.4	3.5	3.7	3.9	4.1
VEN1	140	hf	455.2	502.3	552.2	608.1	658.5	15.7	10.0	11.2	10.1	3.7	4.0	4.2	4.5	4.7
EMI1	65	sc	199.6	209.1	225.2	239.1	238.2	3.2	3.2	2.8	-0.2	4.2	4.2	4.1	4.0	3.7
EMI2	65	sc	215.4	241.4	289.2	320.8	355.6	8.7	9.6	6.3	7.0	4.6	4.8	5.3	5.3	5.5
LAZ1	55	sc	152.6	177.4	190.3	211.7	233.1	8.3	2.6	4.3	4.3	4.1	4.4	4.2	4.2	4.2
MAR1	55	sc	235.0	248.8	266.1	294.2	332.3	4.6	3.5	5.6	7.6	6.4	6.2	5.9	5.9	6.0
SAR1	70	sc	246.9	261.9	261.7	281.9	311.6	5.0	0.0	4.0	5.9	4.7	4.8	4.4	4.3	4.5
TOS1	70	sc	198.6	215.5	238.5	257.7	291.5	5.6	4.6	3.8	6.8	3.8	3.9	4.0	4.0	4.2
PIE1	80	tc	253.0	279.9	307.3	341.1	372.9	9.0	5.5	6.8	6.4	4.1	4.3	4.4	4.5	4.7
SIC1	70	tc	196.3	216.6	211.8	226.9	241.9	6.8	-1.0	3.0	3.0	3.8	3.9	3.5	3.5	3.5
High forests			550.9	603.3	659.6	717.4	768.2	17.5	11.3	11.5	10.2	5.0	5.4	5.6	5.9	6.0
			<i>±130.9</i>	<i>±142.6</i>	<i>±146</i>	<i>±142.8</i>	<i>±125</i>	<i>±5.4</i>	<i>±4.1</i>	<i>±3.1</i>	<i>±7.4</i>	<i>±1.7</i>	<i>±1.7</i>	<i>±1.8</i>	<i>±1.7</i>	<i>±1.6</i>
Stored coppices			208.0	225.7	245.2	267.6	293.7	5.9	3.9	4.5	5.2	4.6	4.7	4.6	4.6	4.7
			<i>±33.2</i>	<i>±31</i>	<i>±34.9</i>	<i>±39.4</i>	<i>±49.8</i>	<i>±2.2</i>	<i>±3.2</i>	<i>±1.3</i>	<i>±2.9</i>	<i>±0.9</i>	<i>±0.8</i>	<i>±0.8</i>	<i>±0.8</i>	<i>±0.9</i>
Transitory crops			224.7	248.2	259.5	284.0	307.4	7.9	2.3	4.9	4.7	3.9	4.1	4.0	4.0	4.1
			<i>±40.1</i>	<i>±44.8</i>	<i>±67.5</i>	<i>±80.8</i>	<i>±92.6</i>	<i>±1.6</i>	<i>±4.6</i>	<i>±2.6</i>	<i>±2.4</i>	<i>±0.2</i>	<i>±0.3</i>	<i>±0.6</i>	<i>±0.7</i>	<i>±0.9</i>

High forests include stands from 95 to 210 years old and are characterized by an average volume of 550.9 m<sup>3</sup> ha<sup>-1</sup> in 1997 and 768.2 m<sup>3</sup> ha<sup>-1</sup> in 2015. Stored coppices include stands from 55 to 70 years with an average volume of 208.0 m<sup>3</sup> ha<sup>-1</sup> in 1997 and 293.7 m<sup>3</sup> ha<sup>-1</sup> in 2015. Transitory crops included just two plots ranging from 70 to 80 years and with an average volume of 224.7 m<sup>3</sup> ha<sup>-1</sup> in 1997 and 307.4 m<sup>3</sup> ha<sup>-1</sup> in 2015.

A positive trend in standing volume has been observed over the concerned period for all the forest types considered (Figure 4.7A). Between 1997 and 2015 high forests had an average increase of 11.84 m<sup>3</sup> ha<sup>-1</sup> per year, more than double if compared to stored coppices and transitory crops (4.62 m<sup>3</sup> ha<sup>-1</sup> per year and 4.34 m<sup>3</sup> ha<sup>-1</sup> per year respectively), which are characterized also by similar trends.

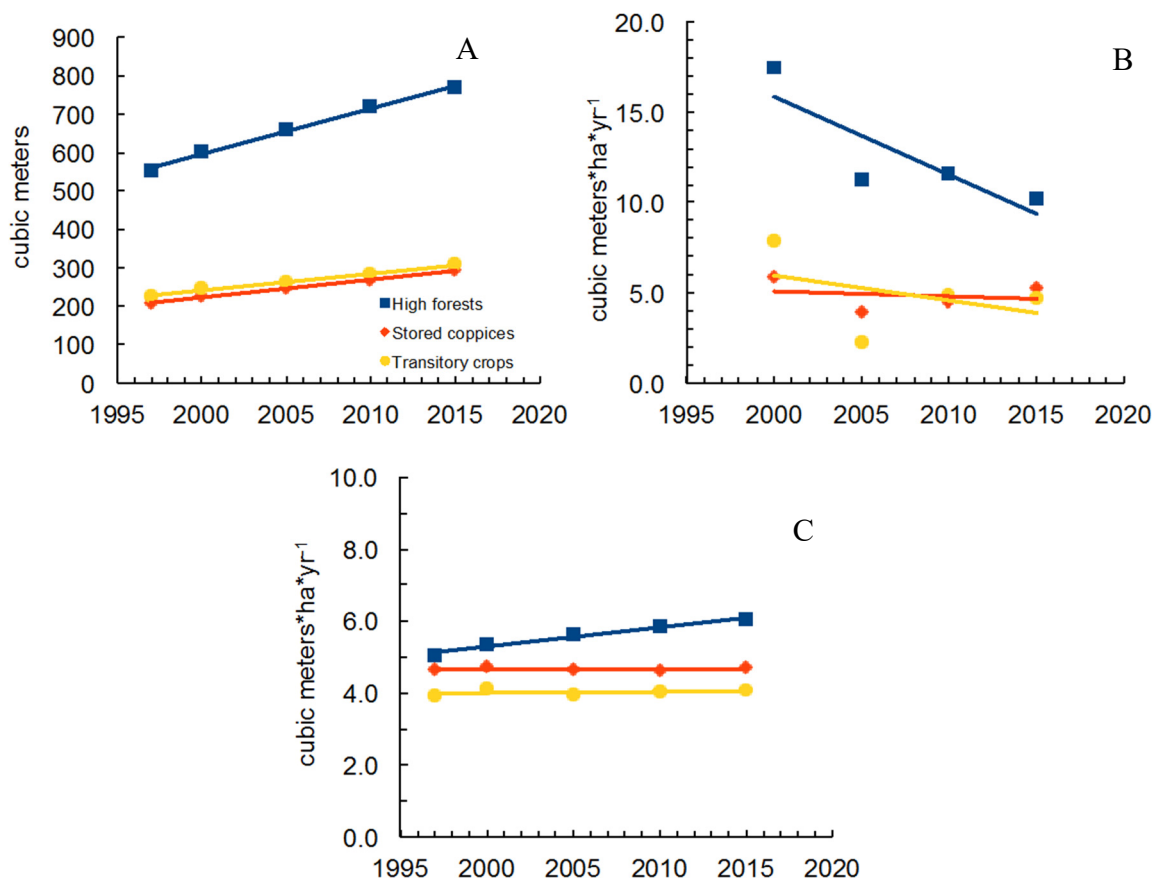


Figure 4.7 – Trends in volume (A), current increment (B) and mean increment (C) at Level II plots in Italy. (Source: LIFE SMART4Action).

The analysis of increments (Figures 4.7 B, C) revealed that high forests are characterized by a decreasing  $IcV$  ( $10.2 \text{ m}^3 \text{ ha}^{-1}$  per year in 2010-2014), but still higher and distant from  $ImV$  ( $6.0 \text{ m}^3 \text{ ha}^{-1}$  per year in 2010-2014). For stored coppices and transitory crops,  $IcV$  is decreasing ( $5.2 \text{ m}^3 \text{ ha}^{-1}$  per year and  $4.7 \text{ m}^3 \text{ ha}^{-1}$  per year in 2010-2014, respectively) and very close to  $ImV$  ( $4.7 \text{ m}^3 \text{ ha}^{-1}$  per year and  $4.1 \text{ m}^3 \text{ ha}^{-1}$  per year in 2010-2014), meaning that the latter is very close to the culmination. The strongest variation of  $IcV$  among subsequent inventory periods (Figure 4.7B) has been recorded for the 2000-2005 as a possible effect of the well-known 2003 heat wave, as reported by various Authors across Europe (Ciais et al., 2005; Leuzinger et al., 2005; Bertini et al., 2011). This variation was much more evident in high forests and less obvious in coppices and transitory crops.

#### 4.3.3 Forest biodiversity

Forest biodiversity has been assessed mostly as diversity of vascular plant species in the Italian forest monitoring networks, Level I and Level II.

Level I was surveyed in 2007 by means of four 10x10 m Sampling Units (SU) on 201 selected plots, according to a harmonised manual (Canullo et al., 2007). It is considered as the representative baseline for the assessment of vascular plant diversity in Italian forests. As grouping factors, the Biogeographical regions and the EFCTs Forest types (EEA 2007, 2012) have been assumed. The spatial distribution of  $\alpha$ -diversity across Level I plots in Italy is given in Figure 4.8, with vascular species density (no. of species  $\times 100 \text{ m}^{-2}$ ) ranging from 2.8 to 54.

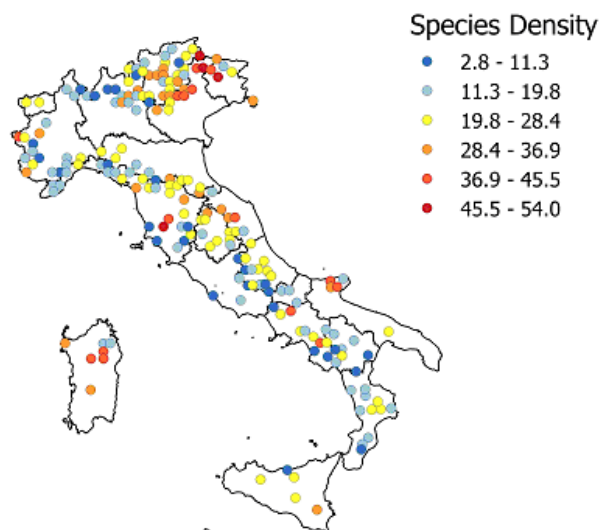


Figure 4.8 – Species density (no. of species  $\times 100 \text{ m}^{-2}$ ) as result of mean species richness between 4 SU on each site.  
(Source: LIFE SMART4Action).

Non Metric Multidimensional Scaling (NMDS) analysis ordinated the sites in terms of similarity of species composition: the most clustered groups by EFTC Forest types are well separated and distributed according both to Latitude and Biogeographical regions (Alpine coniferous forests, Mountainous beech forest, Thermophilous deciduous forests, Broadleaved evergreen forests; Figure 4.9).

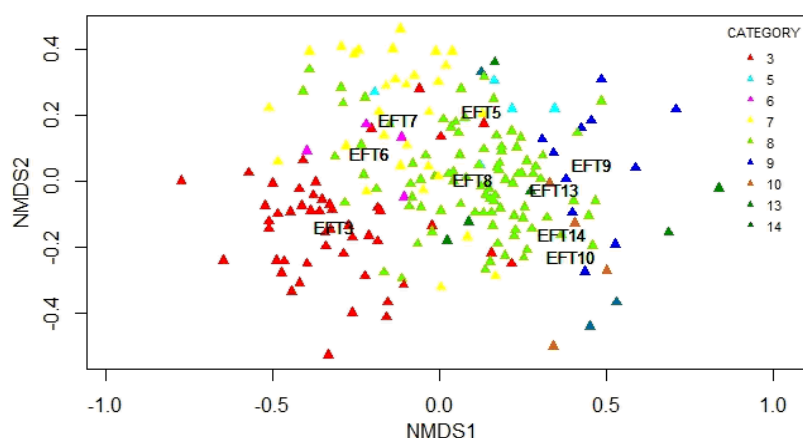


Figure 4.9 – Non Metric Multidimensional Scaling plot shows the relation between species composition (Jaccard dissimilarity index) and the EFTC.  $R = 76.8\%$ ; stress  $\approx 22\%$ . 3 - Alpine coniferous forests; 5 - Oak-hornbeam forests; 6 - Beech forests; 7 - Mountainous beech forest; 8 - Thermophilous deciduous forests; 9 - Broadleaved evergreen forests; 10 - Coniferous forests of the Mediterranean region; 13 - Native plantations; 14 - Exotic plantations and woodlands. (Source: LIFE SMART4Action).

The sampling design in the Level II sites includes 12 10x10 m SUs (Canullo et al., 2013) and proved to capture at least 80% of the expected number of terricolous plant species (including Vascular, Lichens and Bryophytes). For vascular plants, species density was proved to be a good proxy for plot richness (Ferretti et al., 2006). Validated, continuous and comparable datasets cover the period 1999-2012 with 11 plots (Figure 4.10).

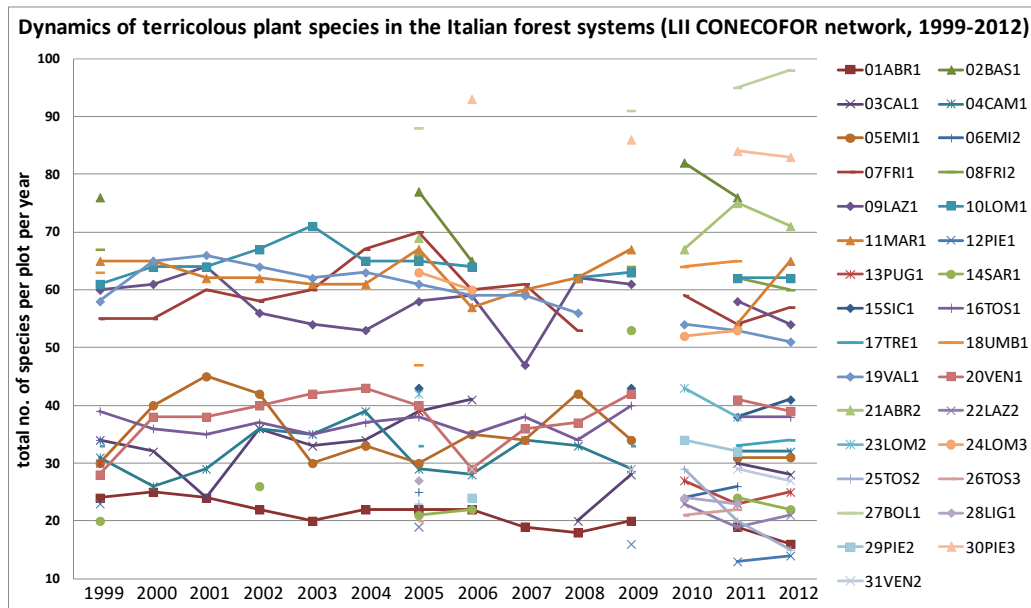


Figure 4.10 –Time pattern of total number of species at individual Level II plots. (Source: LIFE SMART4Action).

Linear model for plant species density (no. of species \* 100 m<sup>-2</sup>) was tested for the selected plots. (within-subject contrasts RMANOVA. Table 4.3). Significant trends ( $P < 0.05$ ) were detected at CAM1 (Campania, South Italy, beech) where the beech forest shows there is an increasing species density, and at LOM1 (Lombardy, North Italy, Norway spruce), with a noticeable reduction in  $\alpha$ -diversity of the transitional Spruce community ( $R^2 = 0.79$ ).

Table 4.3 – Linear model for species density (no. of species \* 100 m<sup>-2</sup>) at the selected Level II plots over the period 1999-2012. Italics:  $p < 0.10$ ; bold:  $p < 0.05$ . (Source: LIFE SMART4Action).

Level II site	$R^2$	$p$	$SE_{regr}$	$b$
ABR1	0,323	<i>0,061</i>	4,815	-0,095
CAL1	0,028	<i>0,099</i>	4,671	-0,111
CAM1	0,118	<b>0,023</b>	9,608	0,171
EMI1	0,046	<i>0,053</i>	3,616	0,067
FRI1	0,055	0,141	4,526	0,111
LAZ1	0,008	0,626	1,562	0,042
LOM1	0,789	<b>0,029</b>	12,153	-0,259
MAR1	0,081	0,149	5,221	-0,110
TOS1	0,104	0,469	2,482	-0,055
VAL1	0,024	0,380	2,204	-0,053
VEN1	0,006	0,371	1,947	0,035

Trends in the species density appear a combination of previous management impact, present changes due to stand dynamics, and external factors. Even if not significant in term of linear trends, is worth to note the visible effect of a strong moth attack (*Lymantria dispar*) in 2002-2003 combined with the 2003 and 2007 heat waves at LAZ1 site (Turkey oak). Beech forest in VEN1 recovered quickly from a severe hailstorm of 1988, leading to still visible effects in 1999. The spruce community in VAL1 has no directional trends, but the annual variability is progressively reducing. The absolute minimum at CAL1 is not apparently linked to known factors.

#### 4.3.4. Forest nutrition

Forest nutrition has been examined in terms of soil and foliar nutritional status.

(i) Soil solid phase. Soil chemistry is measured at Level I and Level II plots. Main results are reported in Figure 4.11 for Level I. In particular:

- carbon content is most frequent between 15-30 g kg<sup>-1</sup> and C:N ratio between 10-15;
- base saturation (BS) is most frequently >75%;
- cation exchange capacity (CEC) is most frequently between 10 and 20 mmolc<sup>+</sup>·kg<sup>-1</sup>, and ca. 50% of the observed plots have a CEC > 20 mmolc<sup>+</sup>·kg<sup>-1</sup>;
- although ca. 50% of plots have a pH >5.5, there is a considerable amount of plots with pH <4.5.

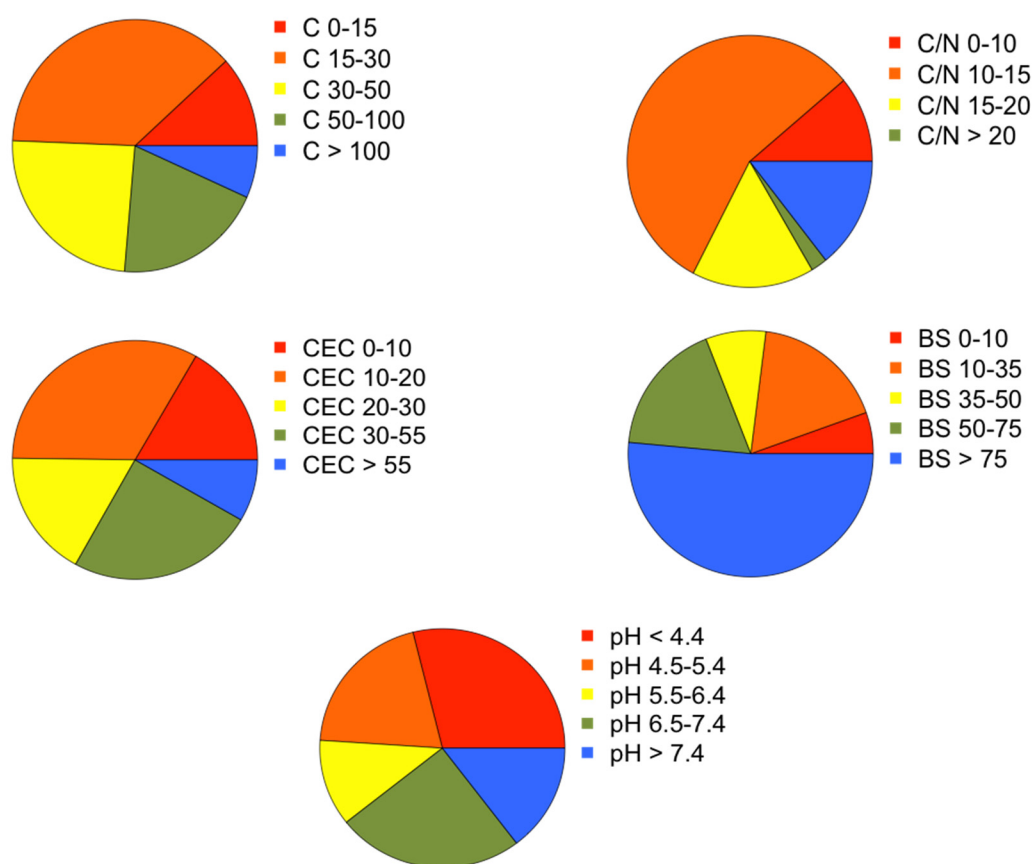


Figure 4.11 – Soil condition on Level I plots in Italy (n: 239). From top left, clock-wise: frequency of plots in different class of carbon (C), C:N, Cation Exchange Capacity (CEC), Base Saturation (BS) and pH.  
(Source: LIFE SMART4Action).

(ii) Soil solution. Soil solution is measured only at Level II sites. It provides important information about changes in the chemistry of soil water that may reflect changes in the atmospheric input and/or changes in ecosystem structure and processes. As such, careful interpretation is always necessary. Main results obtained after evaluating data collected over different time windows (3-15 yrs) and processed by means of Seasonal Mann-Kendall test (SMK) are summarized in Table 4.4.

Table 4.4 – Variable considered, layer examined, no. of plots, range of values among plots and occurrence of significant/non significant trends in soil solution data. (Source: LIFE SMART4Action).

Variable	Layer	No of plot	Range	Significant increasing trend	Significant decreasing trend	Non significant trend
pH	forest floor	6	5.09-6.09	0	3	3
	topsoil	8	4.24-6.59	2	0	6
	subsoil	8	5.04-7.42	4	0	4
NO <sub>3</sub> -N	forest floor	6	0.65-4.03	2	2	2
	topsoil	8	0.09-8.85	1	1	6
	subsoil	8	0.15-2.89	0	2	6
NH <sub>4</sub> -N	forest floor	6	0.08-1.34	0	2	4
	topsoil	3	0.03	1	-	-
	subsoil	8	-	-	-	-
SO <sub>4</sub> -S	forest floor	6	0.5-1.63	0	2	4
	topsoil	8	0.31-2.3	0	5	3
	subsoil	8	0.44-4.71	2	4	2
Base cations	forest floor	6	6.01-15.09	1	3	2
	topsoil	8	0.72-13.88	3	3	2
	subsoil	8	0.85-16.44	2	3	3

(iii) Foliar nutrients. Bi-annual collection and chemical analysis of leaves and needles have been carried out at the Level II plots over the period 1995-2013. Major nutrients have been analyzed: N, S, P, Ca, Mg, K. Main results for the most important tree species are as follows (Figure 4.12A, B):

- in terms of nutritional status, nutrient concentrations are for the most part of species and plots within the acceptable range. Possible exceptions are: high N values reported for some beech sites (Figure 4.12A), and low values reported for holm oak plots; possible low S values at Norway spruce sites; possible low K values at holm oak plots; generalized high values for Ca;
- in terms of time trends, a generalized significant decreasing trend for N (beech, Norway spruce, Turkey oak), S (Norway spruce, Turkey oak). For other nutrients, the time trend is less generalized and more based on species-plot combination (Figure 4.12B).

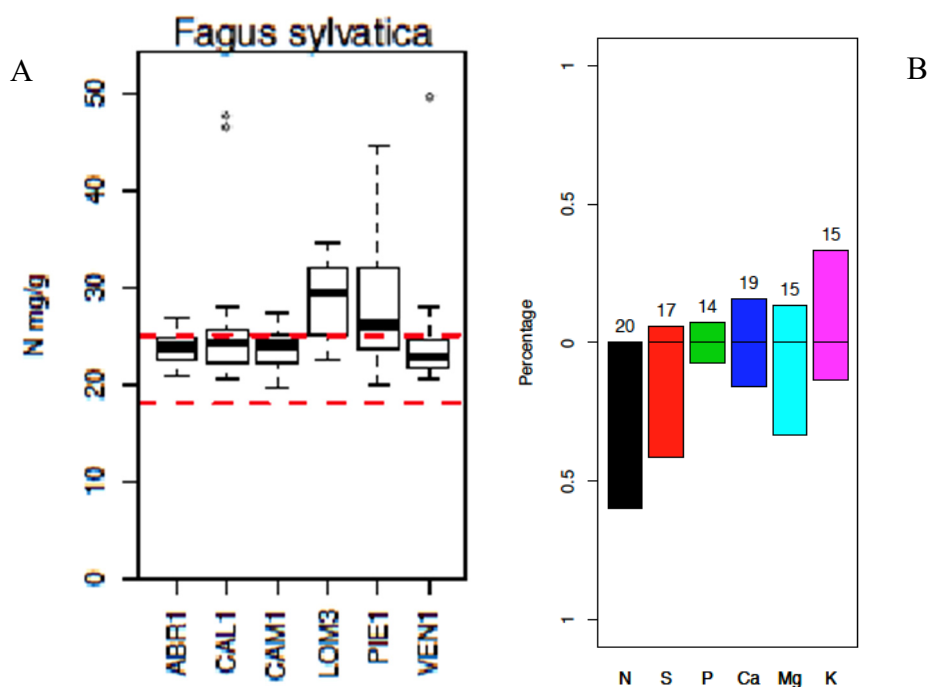


Figure 4.12 – Foliar nutrients. A) mean N values at six beech plots; B) summary of time trends, all sites. Vertical axis shows the % of significant positive trends (above zero) and the % of negative trends (below zero). The % of non significant trend is not explicitly showed. Numbers above bars indicate the number of analyzed sites for the given nutrient. (Source: LIFE SMART4Action).

#### 4.4. Risk assessment

Risk is evaluated here in terms of possible effects of ozone and N deposition on the various impact indicators reported above.

##### 4.4.1 Risk for forest health

No specific study has been undertaken to investigate the effect of N deposition on forest health in Italy. A European-scale study conducted on Level II sites (including Italy), however, revealed that N deposition and related N variables in soil and foliage improve tree defoliation models. The estimated effect is different according to the species being considered (Ferretti et al., 2015). As for ozone, a slight effect has been found for defoliation on beech (Ferretti et al., 2003; Ferretti et al., 2007; Bussotti and Ferretti, 2009). Results from a subset of Level I plots investigated in Trentino (N. Italy) over the period 2007-2011 show that ozone concentration has no significant effect on tree health of assorted species, with Norway spruce and larch being the most frequent ones (Gottardini et al., 2012).

As for direct effect on foliage, symptoms attributed to ozone were assessed at Level II sites and a subset of Level I plots in Trentino. Although several species were found symptomatic (Bussotti and Ferretti, 2007, 2009; Ferretti et al., 2003; Gottardini et al., 2012), statistical relationship with ozone exposure was always weak (Bussotti and Ferretti, 2009). Recent results obtained with single-species approach and *Viburnum lantana* as *in situ* biomonitor were promising, at least at local level (Gottardini et al., 2014; Gottardini et al., submitted).

#### 4.4.2. Risk for forest growth

Distinct effect of N deposition has been detected on growth and C sequestration (Figure 4.13) (Ferretti et al., 2014). In relative terms, the maximal annual response of basal area increment (BAI) was estimated at 0.074–0.085% for every additional kgN. This corresponds to an annual maximal relative increase of 0.13–0.14% of carbon sequestered in the above-ground woody biomass for every additional kgN, i.e. a median value of 159 kgC per kgN ha<sup>-1</sup> per year (range: 50–504 kgC per kgN, depending on the site). The importance of N related variables was further confirmed by a study on 15 Level I plots in Trentino, where N:Mg was the most important predictor of BAI.

No significant effect of ozone has been detected, neither on Level II plots (Ferretti et al., 2003; Ferretti et al., 2014), nor on a subset of Level I plots in Trentino (Gottardini et al., 2012).

All in all, variables related to stand, N deposition, nutrition, soil and climate are by far the most important and significant predictors of growth (Ferretti et al., 2003; 2014).

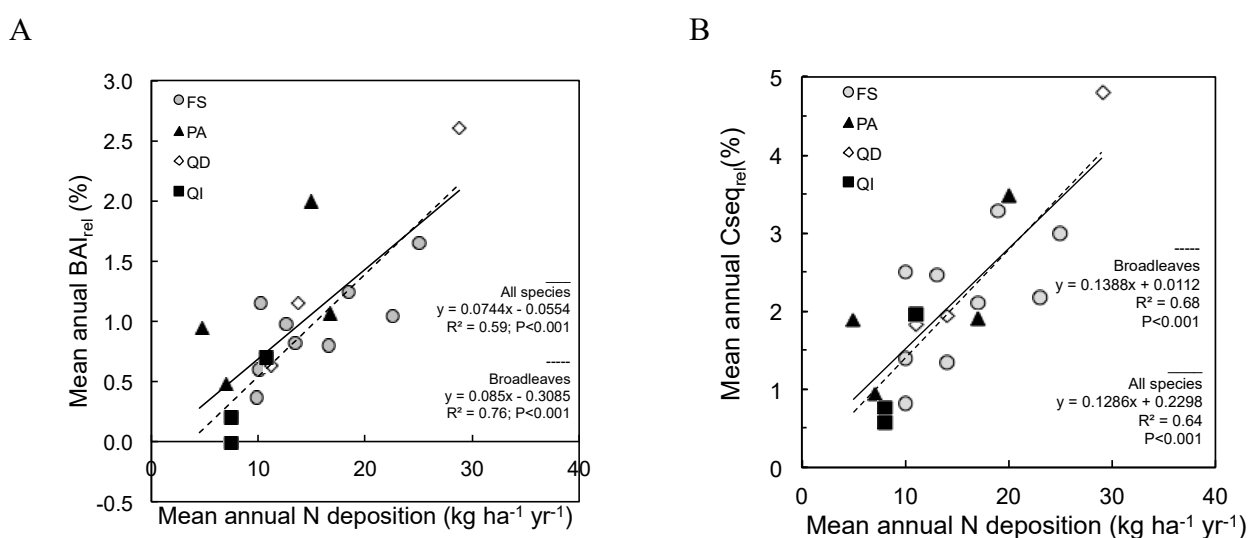


Figure 4.13 – Annual BAI<sub>rel</sub> (A) and annual estimated C sequestered (B) in 2000–2009 plotted against annual N deposition over the same time window at Level II plots in Italy. Solid line: all species; dashed line: broadleaves only. FS: *Fagus sylvatica*; PA: *Picea abies*; QD: deciduous oak (only *Quercus cerris* in this diagram); QI: *Quercus ilex*. (after Ferretti et al., 2014).

#### 4.4.3. Risk for forest biodiversity

Possible air pollution effect on species diversity at Level II plots has been examined in two respects: the expected sensitivity of species composition in relation to ozone and according to the list of sensitive species by the ICP Forests (Ferretti et al., 2003) and the expected effects of stand, soil, meteorology and deposition data on species density (Ferretti et al., 2006).

As for the frequency of ozone sensitive species, an ozone vulnerability index (OVI) that takes into account trees, shrubs, and herbs was calculated. According to this index, beech forests in north and central Italy were potentially the most sensitive to ozone.

As for the set of factors affecting vascular species diversity, multivariate (Generalized Linear Models; Ordinary Least Square Regression) and univariate (Spearman rank order correlation) statistical methods were tested on the set of data collected over the period 1999–2003. All in all, results of the multivariate approach revealed that soil (C, N, C/N, C/P, K, P) and stand (tree species, tree species in the upper layer, leaf litter) are the most important variables: species diversity was found to decrease at increasing level of soil C, N, and C/N.

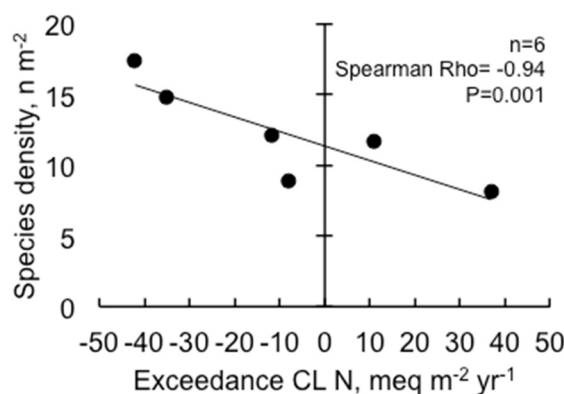


Figure 4.14 – Species density at beech Level II sites vs. exceedance of critical loads (CL) for N for the same sites. CL and exceedances were derived from measurements at the very site. Drawn after Ferretti et al., 2006.

Univariate analysis carried out for beech plots revealed that species diversity decreases as N deposition and exceedance of Critical Load for N increases ( $0.001 < P < 0.05$ ) (Figure 4.14).

#### 4.4.4. Risk for forest nutrition

Distinct effect of N deposition on soil pH and Basic Cation Exchangeable (BCE) was documented, especially for broadleaved forests (Ferretti et al., 2014) (Figure 4.15). Significant effect on foliar nutrient ratios was also reported (Ferretti et al., 2014) (Figure 4.16). With respect to ozone, no study has been undertaken so far to evaluate possible effects on soil biota and foliar nutrients.

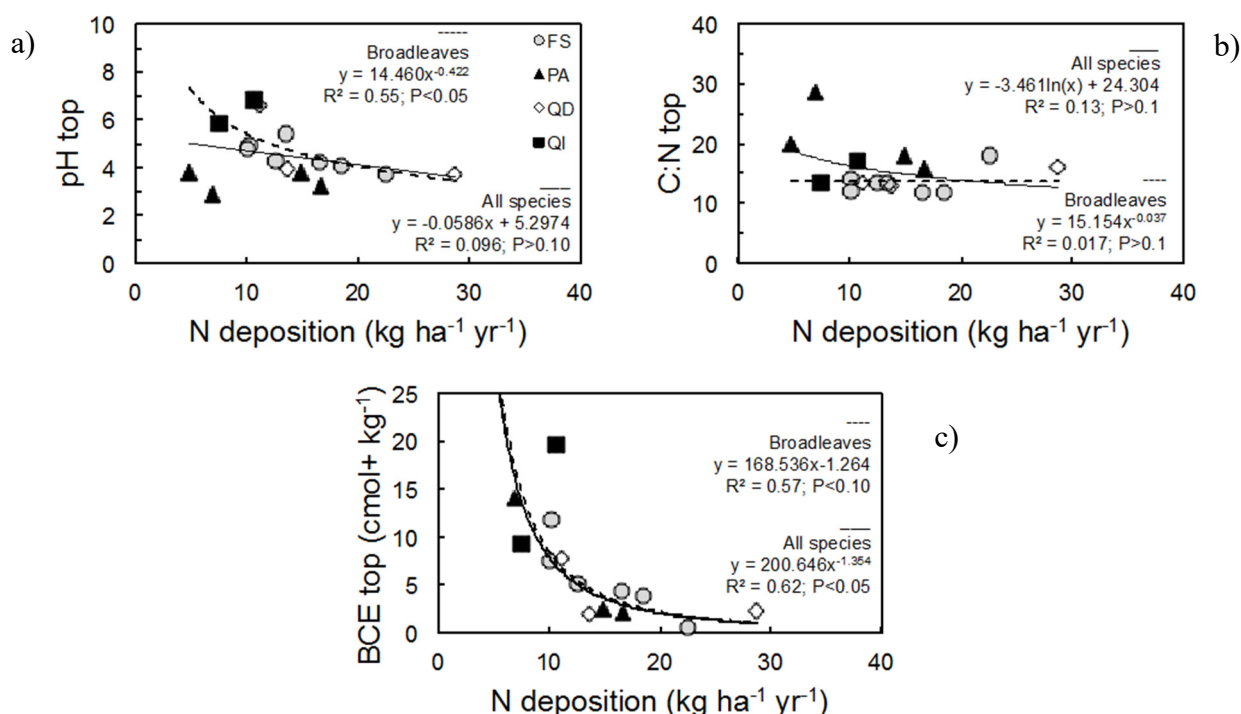


Figure 4.15 – pH (a), C:N (b) and BCE (c) of the mineral topsoil plotted against actual N deposition. Soil data are those obtained after the 1995-1996 survey. Deposition data are mean annual values 2000-2009. Regressions represent always the best fit for the given dataset. Continuous line: all species; dashed line: broadleaves only. FS: *Fagus sylvatica*; PA: *Picea abies*; QD: deciduous oaks; QI: *Quercus ilex*. After Ferretti et al., 2014.

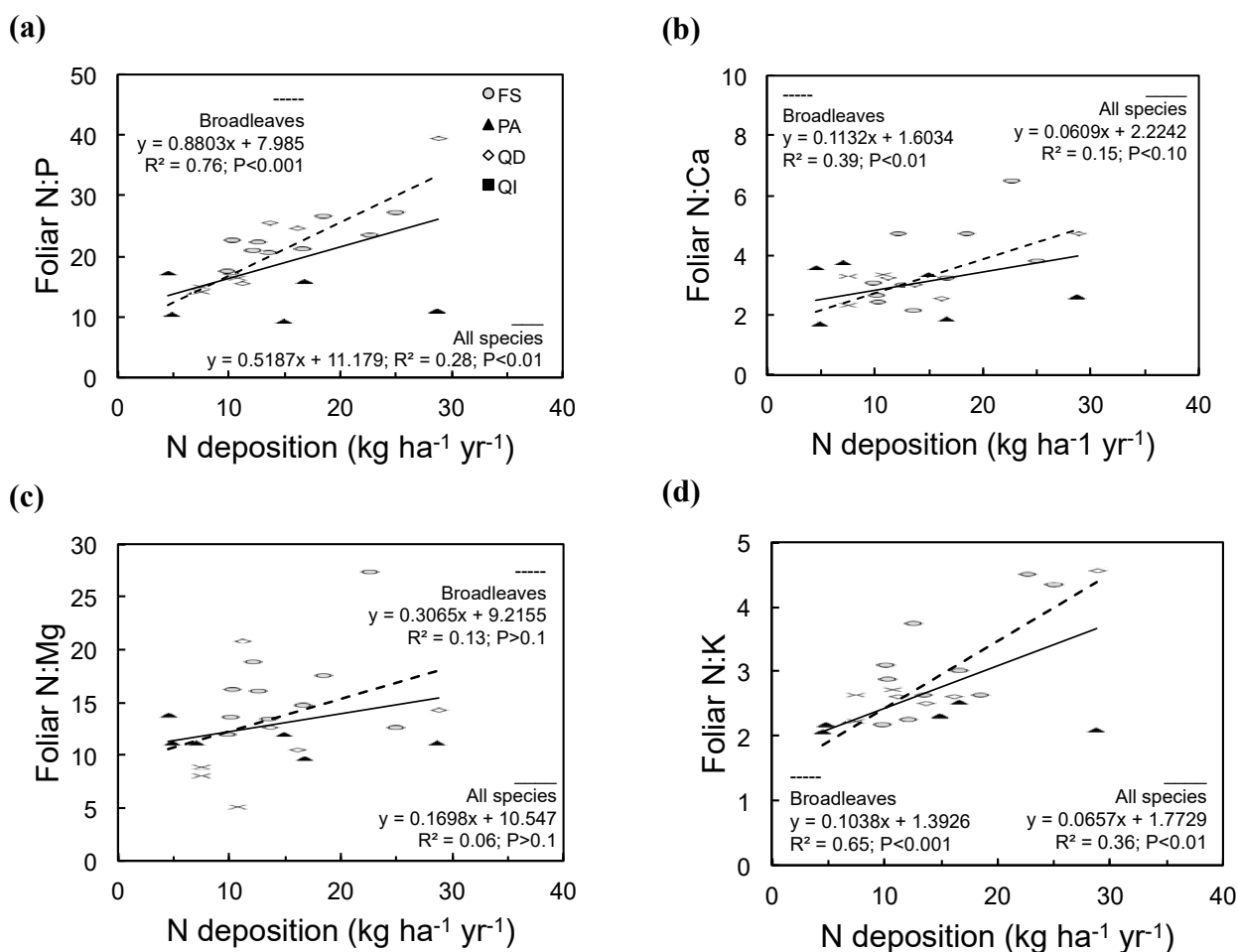


Figure 4.16 – Foliar N:P (a), N:Ca (b), N:Mg (c) and N:K (d) plotted against measured throughfall deposition. Foliar data are mean values after sampling carried out at years 2001, 2003, 2005, 2007, 2009. Deposition data are mean annual values 2000-2009. Continuous line: all species; dashed line: broadleaves only. FS: *Fagus sylvatica*; PA: *Picea abies*; QD: deciduous oak; QI: *Quercus ilex*. After Ferretti et al., 2014.

## 4.5 Conclusions

When using measured data for both pressure and impact, evidence from the Italian forest monitoring networks (UNECE ICP Forests Level I and II) can be summarized as follows:

(i) trends in air pollution issues. There is a significant reduction of sulphate; for nitrate and ammonium the decrease is much slighter, although significant at some sites. After an increase between 1996-2005, mean summer ozone concentration is now decreasing. A decreasing 2000-2013 trend was also reported at the European scale (Sanders et al., 2016).

(ii) Measurable impact. Forest health improved over the 1997-2014 period, with a significant ( $P < 0.01$ ) decreasing trend for defoliation, mostly driven by broadleaves. Forests at Level II sites show positive trends in terms of average increment, and the standing volume is increasing. This latter point is not surprising, as sites are not actively managed and accumulate biomass. Forest biodiversity shows huge variability (Level I) but no clear trend (Level II) over the period 1999-2012. Forest nutrition revealed that acidic soils occur at ca. 50% of Level I plots. In soil solution data collected at level II sites, a decreasing trend for sulphate is evident. There is also significant

evidence of reduced basic cation leaching, which are important in relation to the impacts of acidifying deposition on soil chemistry. Foliar nutrients revealed a composite picture, with mostly decreasing trend for N and S, increasing for K, and much less obvious pattern for P, Ca, and Mg.

(iii) Evidence for risk. Although in terms of reported levels, ozone is potentially a serious risk for Italian forests, measurable effects are quite limited on both Level I and II. Therefore, evidence for actual risk is weak, and limited effect on health and growth were reported by the various study undertaken since early 2000s. On the other end, evidence for N deposition effect on forest nutrition and growth is outstanding. Current level of N deposition was proven to impact soil and foliar chemistry, tree growth and carbon sequestration. Effects on plant diversity were also reported, mainly for beech forests.

# Effects of air pollution on crops and semi-natural vegetation



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## CHAPTER 5 - EFFECTS OF AIR POLLUTION ON CROPS AND SEMI-NATURAL VEGETATION

### 5.1 Introduction

The International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation, formerly ICP Crops) was established in 1987 as a subsidiary body of the Working Group on effects of the UNECE Convention on long-range transboundary air pollution (LTRAP). ICP Vegetation is an international research programme investigating the impacts of air pollutants (particularly tropospheric ozone ( $O_3$ ), heavy metals and nitrogen) on crops and semi-natural vegetation.

Participants meet each year at the Task Force Meeting (TFM) to discuss recent results and the future development of the programme. Recently, Italy hosted the 25<sup>th</sup> and the 28<sup>th</sup> of the TFM, respectively organized by the Math and Physics Department of the Catholic University of Brescia in 2012, and by the National Focal Centre, Department of Environmental Biology, Sapienza University of Rome, in 2014. Some of the works that were presented during the Rome meeting are published in *Annali di Botanica*, Vol. 5, 2015.

### 5.2 Pressures

Tropospheric ozone ( $O_3$ ), nitrogen deposition and heavy metals are currently identified by the ICP Vegetation as the main sources of pressure for crops and (semi)-natural vegetation.

#### 5.2.1 Ozone

$O_3$  is considered as the main oxidizing agents in the near-surface atmosphere of the Mediterranean region (Cristofanelli and Bonasoni, 2009), also acting as a major greenhouse gas (Cooper et al., 2014). Despite the increasing efforts to regulate the precursor emissions this secondary air pollutant (Directive 2008/50/EC, EC, 2008), background  $O_3$  levels are continuously increasing, particularly in Southern Europe (Coll et al., 2009; Cooper et al., 2014). Although the reduction of emissions has decreased the magnitude of  $O_3$  peaks over the last decade, high daily summer  $O_3$  concentrations still occur in the Mediterranean area, favored by high values of irradiance and temperature (Fernández-Fernández et al., 2011; Sicard et al., 2013). Furthermore, nocturnal  $O_3$  pollution episodes are also frequent in the Mediterranean area, particularly along coastal sites located downwind of large urban conurbations, thus posing a concrete risk to vegetation (Fares et al., 2009; Mereu et al., 2009).

Fig. 5.1 describes the temporal trend of the target value for the protection of vegetation, indicated by the EU Directive (EC, 2008) as the AOT40 calculated over a given period using only the one-hour values measured between 08:00 to 20:00 CET in selected monitoring stations with different characteristics. The 5-y average of 9 ppm h (target value for vegetation protection) is systematically exceeded in most of them (see fig. 5.1).

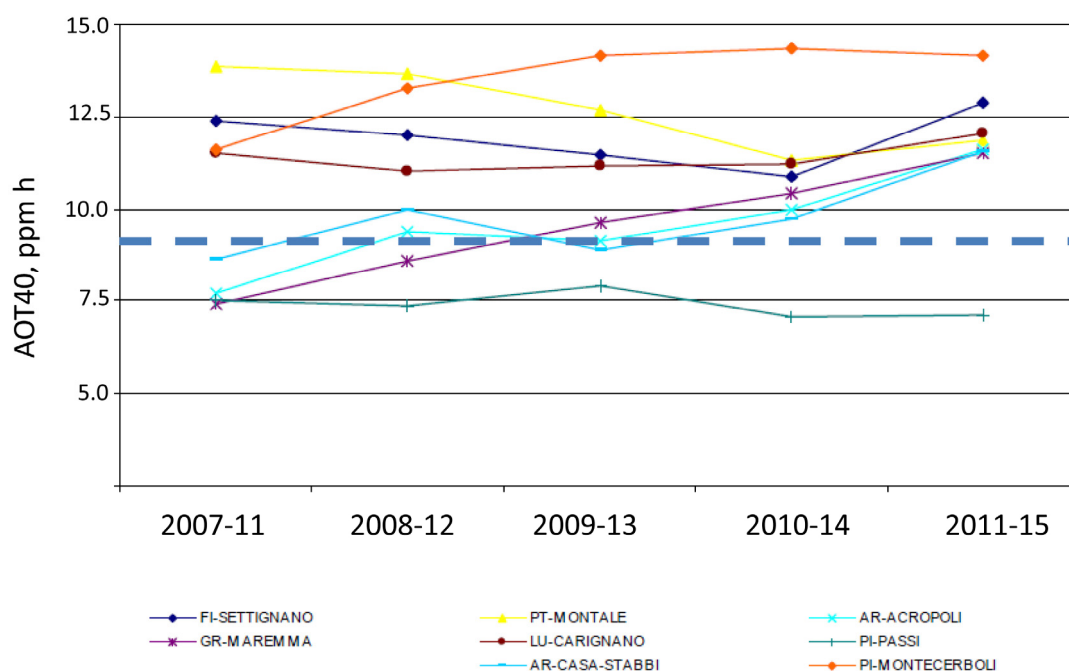


Figure 5.1 – AOT40 (expressed in ppm h) 5-y running averages as observed in 8 Tuscan monitoring stations (Data source: Tuscan Regional Environmental Agency, ARPAT, Florence). The dashed horizontal line shows the target value for the protection of vegetation. GR-Maremma is a rural site; AR-Casa Stabbi is rural/background; AR-Acropoli, FI-Settignano, PT-Montale, LU-Carignano, PI-Passi, PI-Montecerboli are suburban.

It is also interesting to estimate the biological additive effects of O<sub>3</sub> pollution and summer heat waves. This because the meteorological conditions typical of the heat waves (i.e. high temperature, intense solar radiation, dryness) are also responsible of the formation and build-up of photochemically generated ozone (Lorenzini et al., 2014) (Fig. 5.2).

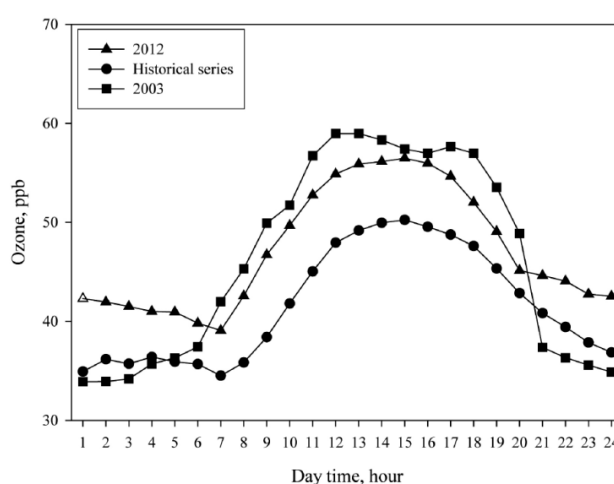


Figure 5.2 – Diurnal variation of ozone hourly concentration in the rural/background monitoring station of Casa Stabbi (Arezzo Province) (1 July-15 August) in 2003 and 2012 (two years with dramatic summer heat waves), in comparison with an historical series (1999-2002 + 2004-2011) (After Lorenzini et al., 2014).

### 5.2.2 Nitrogen

Among the main concerns for the health of Mediterranean vegetation, increasing attention is given to anthropogenic perturbation of the nitrogen (N) cycle (Phoenix et al., 2006; Ochoa-Hueso et al., 2011). Nitrogen is often a limiting factor in many terrestrial ecosystems, but its cycle at global scale has been altered by post-industrial human activities, mainly because of combustion of fossil fuels and fertilization practices (Haber-Bosch reaction) aimed at increasing agricultural production, and resulting in increased N deposition (mainly nitrates and ammonium) (Galloway et al., 2008). NO<sub>x</sub> and NH<sub>3</sub> emitted to the air are the main nitrogen pollutants contributing to the amount of reactive nitrogen rising levels of atmospheric deposition on soils, vegetation surfaces and waters (The European Nitrogen Assessment, 2011).

## 5.3 Impacts

### 5.3.1 Ozone

O<sub>3</sub> is known to affect plant photosynthetic function in different ways, and the effects of the O<sub>3</sub> stress can be observed at biochemical, microscopic and macroscopic level. The first interaction between O<sub>3</sub> and plants occurs at the level of the guard cells of stomata, in which O<sub>3</sub> triggers the production of Reactive Oxygen Species (ROS), thus activating a signalling cascade inducing an overall efflux of anions and K<sup>+</sup> (Vahisalu et al., 2010; Vainonen and Kangasjärvi, 2015). The consequent loss of guard cells turgor, leading to stomatal closure, reduces the gas uptake through stomata; this mechanism restricts the O<sub>3</sub> flux into the leaves (“avoidance mechanism”, Castagna and Ranieri, 2009) but, at the same time, also limits the photosynthetic CO<sub>2</sub> assimilation (Astorino et al., 1995). The O<sub>3</sub> molecules that enter the sub-stomatal chamber can directly peroxidize the membrane lipids of the mesophyll cells (Vitale et al., 2008) or, in the apoplastic space, can generate toxic ROS (like OH<sup>•</sup>, O<sub>2</sub><sup>•-</sup>, H<sub>2</sub>O<sub>2</sub>), which can then move inside the cells (Vainonen and Kangasjärvi, 2015). Furthermore, O<sub>3</sub> triggers the ROS production from different endogenous, enzymatic sources, a process known as “oxidative burst” (Manes et al., 1990a; Scalet et al., 1995). Despite acting as signalling molecules, that leads to the activation of several plant defence responses (Vainonen and Kangasjärvi, 2015), ROS are also responsible of direct oxidative damages to different molecules involved in the photosynthetic process, such as chlorophylls a and b, and Rubisco, whose activity and content have been reported to decline under O<sub>3</sub> stress (Goumenaki et al., 2010). Moreover, detrimental effects of O<sub>3</sub> on the photosynthetic electron transport chain, and in particular on Photosystem II (PSII) function, have been demonstrated (Guidi et al., 2002; Pellegrini, 2014). A reversible, photoprotective down regulation of PSII photochemistry is, however, the most commonly observed PSII response to O<sub>3</sub>, being a consequence of the reduced demand of NADPH and ATP from the Calvin cycle; the latter can be caused by both stomatal closure and biochemical limitations (Bussotti et al., 2011; Mereu et al., 2011; Salvatori et al., 2013).

Also, the role of Photosystem I (PSI) in plant photosynthetic response to O<sub>3</sub> has received increasing attention in the last years. In fact, the measurement of prompt chlorophyll “a” fluorescence (PF) and the JIP test analysis (Strasser et al., 2004; 2010) have shown that the I-P part of the PF transient, which correlates to PSI content and activity (Ceppi et al., 2012; Schansker et al., 2005), is particularly sensitive to O<sub>3</sub> and other oxidative stress factors (Oukkarroum et al., 2009; Bussotti et al., 2011; Pollastrini et al., 2014; Bernardini et al., 2016).

In particular, the amplitude of the I-P phase of the PF ( $\Delta V_{I-P}$ ) was reduced by O<sub>3</sub> stress in many studies, indicating a negative effect of this pollutant on the efficiency of electron transport through PSI, to reduce the end acceptors beyond PSI (Bussotti et al., 2011; Mereu et al., 2011; Pollastrini et al., 2014). Recent studies carried out with the innovative technique of multi signal fluorescence measurement (Fusaro et al., 2016a; Salvatori et al., 2015) have highlighted that, in both crop and natural species, it is possible to distinguish an early O<sub>3</sub> response of the photochemical apparatus, involving PSII only, and a late response, occurring when O<sub>3</sub> cumulative stress becomes more severe, in which PSI activity and content are also modulated.

Ultrastructural alterations and visible leaf injury are reported in sensitive species after ozone stress. The appearance and diffusion of visible leaf injuries due to ozone exposure were reported in several experiments on bean (Gerosa et al., 2009) and durum wheat (Gerosa et al., 2014; Monga et al., 2015). Results showed that microscopical leaf symptoms, assessed as cell death and H<sub>2</sub>O<sub>2</sub> accumulation, preceded by three-four days the appearance of visible symptoms.

Iriti et al. (2006) obtained similar results on currant tomato, suggesting the potential use of this species for ozone biomonitoring protocols, since there was a linear relationship between the intensity and diffusion of visible leaf injuries and the hourly ozone concentration mean.

Basile et al. (2010) have applied Transmission Electron Microscope (TEM) analysis on O<sub>3</sub>-resistant (NC-R) and O<sub>3</sub>-sensitive (NC-S) clones of *Trifolium repens* L. cv. Regal, exposed to ambient O<sub>3</sub> conditions in the Botanical Garden of the Sapienza University of Rome, Italy, from June to October 2005, following the ICP Vegetation Experimental Protocol. This work has highlighted that ultrastructural injuries appeared to be widespread in NC-S leaves, showing dead cells and heavily damaged living cells, while the NC-R clones showed much less damage: the chloroplast maintained an almost intact organization of thylakoids (granal and intergranal regions), but the chloroplast side facing the apoplastic space appeared with no thylakoids. Interestingly, Hsp70 levels, an important stress marker, were only slightly increased in the O<sub>3</sub>-sensitive clone with respect to the O<sub>3</sub>-resistant clone; in contrast, a strong decrease (–60%) in phosphoenolpyruvate carboxylase (PEPCase) protein levels was measured in the ozone-resistant clone.

Pellegrini et al. (2015a) conducted a comparative study on functional leaf traits and the diurnal dynamics of photosynthetic processes on plants of two grape (*Vitis vinifera*) varieties (Aleatico, ALE, and Trebbiano giallo, TRE), exposed under controlled conditions to realistic concentrations of O<sub>3</sub> (80 ppb for 5 h day<sup>-1</sup>, 8:00-13:00 h, + 40 ppb for 5 h day<sup>-1</sup>, 13:00-18:00 h). At the constitutive level, morphological functional traits of TRE improved leaf resistance to gas exchange, suggesting that TRE is characterized by a potential high degree of tolerance to ozone. At the end of the treatment, both varieties showed typical visible injuries on fully expanded leaves and a marked alteration in the diurnal pattern of photosynthetic activity. This was mainly due to a decrease in stomatal conductance (–27 and –29% in ALE and TRE, respectively, in terms of daily values in comparison to controls) and mesophyll functioning (+33 and +16% of the intercellular carbon dioxide concentration). Although the genotypic variability of grape regulates the response to oxidative stress, similar detoxification processes were activated, such as an increased content of total carotenoids (+64 and +30%, in ALE and TRE), enhanced efficiency of thermal energy dissipation within photosystem II (+32 and +20%) closely correlated with the increased depoxidation index (+26 and +22%) and variations in content of some osmolytes.

In summary, it can be concluded that: the daily photosynthetic performance of grapevine leaves was affected by a realistic exposure to ozone. In addition, the gas exchange and chlorophyll *a* fluorescence measurements revealed a different quali-quantitative response in the two varieties. The genotypic variability of *V. vinifera* and the functional leaf traits seem to regulate the acclimation response to oxidative stress and the degree of tolerance to ozone. Similar photoprotective mechanisms were activated in the two varieties, though to a different extent.

Valletta et al. (2016) have investigated the physiological and metabolic effects of O<sub>3</sub> on two wine cultivars of *V. vinifera*, a red grape (San Giuseppe) and a white grape (Maturano) (Fig. 5.3), both autochthonous cultivars of the Latium region and having an economic importance at local scale. The results showed that the white grape cultivar appeared more sensitive to O<sub>3</sub> stress. Moreover, differently from what expected O<sub>3</sub> did not activate stilbene production but, in both cultivars, influenced the content of chlorogenic acid (CGA), an important molecule in the biosynthetic pathway of polyphenols and an antioxidant itself, which decreased in the fumigated samples and recovered after 8 days. The decrease in CGA not necessarily indicates a decrease in its biosynthesis, since it is highly probable that CGA is consumed or as antioxidant, or as precursor to other phenolic compounds.



Figure 5.3 – “Walk-in” Chamber facility of the Department of Environmental Biology, Sapienza University of Rome, with *V. vinifera* plants fumigated with O<sub>3</sub>.

As a result of the above described invisible and visible alterations, and of the increase in metabolic cost for detoxification and repair, a reduction of total biomass, as well as a decreased carbon allocation to heterotrophic tissues (i.e. roots and fruits) (Andersen, 2003), is frequently reported for different species (Salvatori et al., 2013). In crops, such effects translate in a reduction of yield, as well as of food quality, with consequent economic losses to the agricultural sector that are mostly reported for Italy and Spain. In particular, Nali et al. (2002), on the basis of O<sub>3</sub> concentrations recorded by ten monitoring stations in Tuscany, have estimated yield losses due to ozone, which varied in Florence from 8% for corn and alfalfa to 27% for soybean, in Pisa from 5% for corn to 24% for soybean, in Lucca from 3% for corn to 17% for soybean. A preliminary economic estimate for corn, wheat, barley, soybean, tomato and alfalfa, calculated annual damage to be 4.6 MEuro in Florence, 0.5 MEuro in Lucca and MEuro in Pisa.



Figure 5.4 – Open-Top chambers facility of C.R.IN.ES. at Curno (Bergamo).

Several experiments on horticultural yield losses due to ozone were carried out in the Po plain climatic conditions at the CRINES research site (Centro di Ricerca Inquinamento atmosferico ed Ecosistemi, Curno, Bergamo) by the Environmental Physics and Ecophysiology research group of the Catholic University of Brescia (Fig. 5.4).

Results showed mean yield reductions of 40% on bean (cv. “borlotto nano lingua di fuoco” Gerosa et al., 2009), 36% on tomato (cv. “Oxheart”, Gerosa et al., 2008), 18% on lettuce cv. “Romana” and 14% on lettuce cv. “Canasta” (Marzuoli et al., 2016a). In the same experimental site Monga et al. (2015) performed an experiment on durum wheat, highlighting yield losses up to 16% (in cv. “Sculptur”) in plants that were exposed to O<sub>3</sub> enriched air (+50% of the ambient ozone).

Dry biomass reductions up to 25% due to O<sub>3</sub> were observed on alfalfa in the Po plain, and significant differences of forage quality parameters were also found for the same species. However, in all of the above mentioned experiments, a strong intraspecific variability of the response to ozone was observed. For example, ozone caused a slight increase of fruit yield in the tomato cv “San Marzano” (Gerosa et al., 2008), Monga et al. (2015) in a varietal screening experiment found that only 2 of the 5 studied cultivars of durum wheat were ozone sensitive.

The dataset of the experiment on tomato contributed to the definition of a dose-response relationship and critical levels based on ozone flux and fruit yield loss (Gonzalez-Fernandez et al., 2014) that was included in current version of the “Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends” (CLRTAP, 2010). Other experimental datasets have been required for the definition of analogous dose-response relationships for leafy crops species (lettuce, Marzuoli et al., 2016a), bean (Gerosa et al., 2009) and durum wheat (Monga, 2015; Gerosa et al., *in preparation*) in order to define their specific critical levels.

In addition to controlled conditions experiments (with Open-Top Chambers), some open field experiments were conducted in order to characterize the ozone uptake at agricultural ecosystem level, and to quantify the partition of the total ozone flux between the stomatal and non-stomatal components of the deposition pathway. These studies are strictly necessary to properly calculate the

ozone dose absorbed by vegetation for the regional level risk assessment, and for the application of the critical levels.

In this context, it is important to mention the field experiments performed on winter wheat (Gerosa et al., 2003) and soybean (Gerosa, 2002) that highlighted the presence of a significant non-stomatal component of the ozone total flux that could cause serious overestimation of the effects in case it would not be taken into account (Bassin et al., 2004; Tuovinen et al., 2007). These results contributed in the fine calibration of the EMEP model (which contain the DO<sub>3</sub>SE module embedded) for the estimation of the non-stomatal component (Tuovinen et al., 2004).

Finally, a dataset of measurements on onion fields (Gerosa et al., 2007) was used to further improve the ozone flux partition process, identifying the agrometeorological parameters that mostly can influence the quality of the flux estimations.

The ecophysiological and biochemical effects of realistic O<sub>3</sub> exposure under controlled conditions upon medicinal species have been also investigated.

In particular, *Salvia* (Pellegrini et al., 2015b) and the perennial herbaceous lamiacea *Melissa officinalis* (known as lemon balsam) have been considered. Lemon balsam has been selected as a test species to elucidate the variations on biochemical composition of an officinal plant under short- and long-term exposure to O<sub>3</sub>. Whole plants and cell cultures were investigated and biotechnological practical implications envisaged. A consistent fraction of this project (the one focused on rosmarinic acid biosynthesis at the molecular level - Döring et al., 2014a, b) has been performed jointly with a German team. Signaling molecules, membrane integrity, secondary metabolism, programmed cell death, photosynthetic performances are amongst the other key issues taken into account (Pellegrini et al., 2011, 2013b; Tonelli et al., 2015; D'Angiolillo et al., 2015).

The lichen response to O<sub>3</sub> exposure has been evaluated (Pellegrini et al., 2014a), thorough description of the biochemical and physiological mechanisms that are at the basis of the O<sub>3</sub>-tolerance of lichens. Chlorophyll *a* fluorescence emission, histochemical ROS localization in the lichen thallus, and biochemical markers [enzymes and antioxidants involved in the ascorbate/glutathione(AsA/GSH) cycle; H<sub>2</sub>O<sub>2</sub> and O<sub>2</sub> were used to characterize the response of the epiphytic lichen *Flavoparmelia caperata* exposed to O<sub>3</sub> (250 ppb, 5 h d<sup>-1</sup>, 2 weeks) at different watering regimes and air relative humidity in a fumigation chamber. After a two-week exposure Chl*a*F was affected by the watering regime but not by O<sub>3</sub>. The watering regime influenced also the superoxide dismutase activity and the production of ROS. By contrast O<sub>3</sub> strongly influenced the AsA/GSH biochemical pathway, decreasing the AsA content and increasing the enzymatic activity of ascorbate peroxidase, dehydroascorbate reductase and glutathione reductase independently from the watering regime and the relative humidity applied.

### 5.3.2 Nitrogen

Increasing in N deposition is a growing threat to semi-natural grasslands that are listed as a priority habitat for biodiversity conservation in European Union Habitats Directive (92/43/CEE). A study carried out on grassland in Central Italy highlighted that N deposition markedly increases aboveground biomass, maintaining low species diversity (Bonanomi et al., 2006). The strong growth response the authors found out using a relatively low level of nitrogen enrichment, clearly indicated a nitrogen limited community.

Nitrogen eutrophication could exacerbate the deleterious effects of land abandonment on grassland biodiversity (Bonanomi et al., 2006) through enhanced above-ground competition and litter accumulation.

Moreover, since stressors like O<sub>3</sub> or drought can have an impact on roots biomass (Fig. 5.5), puzzling interaction effects might occur. In order to understand the effects of ongoing global environmental change on Mediterranean ecosystems, it is necessary take into account how the multiple stresses affect vegetation since a comprehensive picture is far from being given (Tattini and Loreto, 2014). A recent work by Fusaro et al. (2016b) has investigated how functional and structural traits of two Mediterranean species with different leaf habits (*Fraxinus ornus* and *Quercus ilex*) shift because of nitrogen (N) addition (30 kg ha<sup>-1</sup> y<sup>-1</sup>), also exploring the effect that nitrogen has on the water stress response. Their results have shown that the early successional, deciduous *F. ornus* tends to invest more nitrogen at leaf level enhancing photosynthetic machinery, whereas late successional evergreen *Q. ilex* invests resources on non-photosynthetic biomass, keeping constant N content at leaf level. These differences between species can modify their competitive relations, through influence temperate forest community structure and dynamics. This study also highlights that N can have role in water stress response in both species, but in different way. In *F. ornus* N has ameliorative effect on water stress, determining high gas exchange rates and photosystems functionality. On the contrary, in *Q. ilex* the N addition seems to increase the susceptibility to water stress, possibly because of changes in biomass partitioning due to N. Moreover, studies relative interaction between N deposition and O<sub>3</sub>, highlighted an adverse synergistic effect, since exposure to ambient O<sub>3</sub> concentrations was shown to reduce the Nitrogen Use Efficiency. On the other hand, the growth of plants in response to N (i.e. root development), mitigates impact on biomass and physiology due to O<sub>3</sub> (Marzuoli et al., 2016b). Mechanism of action of N on tree species can contribute to provide a more reliable risk assessment, and accordingly it should be implemented for natural and semi-natural vegetation.

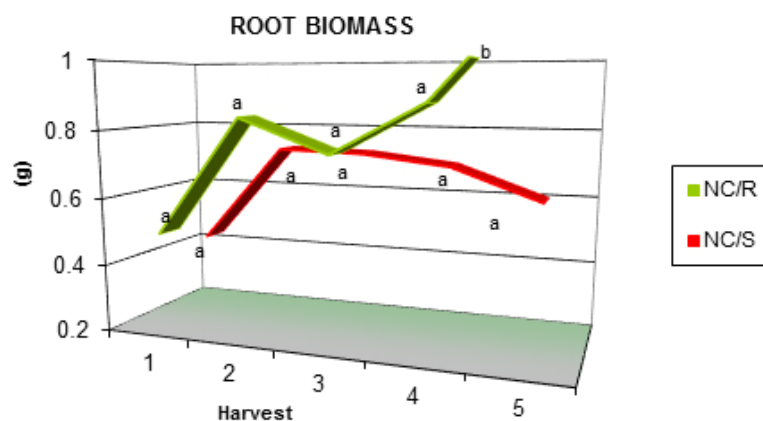


Figure 5.5 – Root biomass of NC/R and NC/S clover clones, harvested after 1, 2, 3, 4 and 5 months of exposure to ambient O<sub>3</sub> in the Botanical Garden of Sapienza University of Rome.

## 5.4 Risk assessment

The methodologies developed within UN/ECE to assess the impacts of air pollution on crops are described in the Chapter 3 of the “Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends.” (UN/ECE, 2010).

### 5.4.1 Ozone

Chapter 3 of the mapping manual considers two main approaches to set critical levels to protect vegetation from tropospheric ozone: the concentration-based critical level ( $O_3$  accumulated over a threshold of  $x$  ppb, AOTx) and the uptake-based critical level (accumulated  $O_3$  dose above a threshold of  $Y$  or phytotoxic ozone dose, PODY). The critical levels are defined as the dose that causes a reduction by 5% of the yield at 95% confidence level (UN/ECE, 2010). The two approaches differ by their data requirements (UN/ECE, 2015a), because PODY is linked to stomatal uptake that is limited by environmental condition such as temperature, light, soil moisture, vapor pressure deficit, phenology and wind (Mills et al., 2011). Since their first establishment in 1996, the  $O_3$  critical levels have been intensively debated among scientists. In particular, many studies carried out in Italy have shown that, despite  $O_3$  exposure at Italian background sites exceeds the AOT40-based critical level, the occurrence of adverse effects of  $O_3$  on forests and crops appears controversial (Manes et al., 2005; Ferretti et al., 2007). At this regard, the use of the stomatal uptake concept (PODY) is highly recommended since, besides pollutant concentration, it takes into account vegetation characteristics and environmental constraints that affect the dose of  $O_3$  effectively absorbed by the plant, an aspect that is of particularly importance under Mediterranean climatic conditions (Ferretti et al., 2007; Manes et al., 2005; Anav et al., 2016; De Marco et al., 2015).

The most common methodology to define the risk assessment is biomonitoring. Biological monitoring can be defined as the measurement of the response of living organisms to changes in the air quality of their environment (Nali et al., 2006), and may provide integrated information on air quality impacts. As plants are more sensitive in terms of physiological reaction to the most prevalent air pollutants than humans and animals (Nali et al., 2006), they are more suitable to be used for biomonitoring. Biomonitoring can be performed through the analysis on the vegetation already present in a given study area (so-called passive biomonitoring) or carried out with selected test plants introduced at the study site (active biomonitoring) (Nali and Lorenzini, 2007). The selection of plants with different sensitivity/resistance to  $O_3$  has been a challenge since 1950's (Karlsson et al., 2003), with the purpose of biomonitoring phytotoxic effects of this pollutant under conditions of ambient exposure. From 1987, these efforts have been coordinated under the ICP Vegetation (Harmens et al., 2015a). Many plant species have been tested as active  $O_3$  biomonitors on pan-European scale, such as radish (*Raphanus sativus* L. cv. Cherry Belle, Manes et al., 1990b; Allegrini et al., 1994), *Phaseolus vulgaris* cv Lit (Astorino et al., 1995), and *Trifolium subterraneum* cv Geraldton. From 1996 to 2004, the ICP Vegetation international biomonitoring programme has involved exposure of ozone sensitive (NC-S) and ozone resistant (NC-R) biotypes of white clover (*T. repens* L. cv. Regal) (Heagle et al., 1994) (Figg. 5.6, 5.7). The clover system has proven to be useful to detect detrimental effects of ambient  $O_3$  (Manes et al., 2003; Nali et al., 2009), however visible injury and biomass reductions proved to be more related to stomatal  $O_3$  uptake than to air  $O_3$  concentration, expressed as AOT40 (Mills et al., 2011).



Figure 5.6 – “Biomonitoring station” with NC-S and NC-R clover clones, in the Botanical Garden of Sapienza University of Rome.

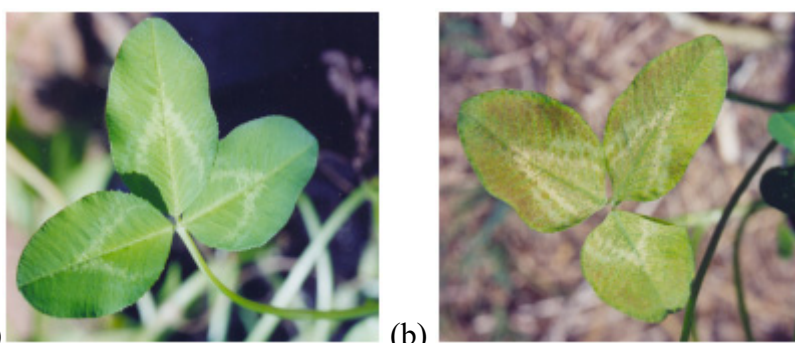


Figure 5.7 – Healthy (a) and O<sub>3</sub>-injured (b) leaves of white clover, NC-S clone.

Moreover, studies carried out in Mediterranean climatic conditions, have shown the importance to consider the effect of environmental variables, such as high air temperatures and Vapour Pressure Deficit (VPD), which can act as confounding factors on the O<sub>3</sub> dose-response relationship of the clover system, even if plants are grown following standard protocols under non limiting water availability (Ferretti et al., 2007; Manes et al., 2005).

Ozone-hypersensitive tobacco (*Nicotiana tabacum* L.) Bel-W3 has been also used worldwide as a reliable and sensitive bioindicator of ozone, showing a characteristic and specific foliar response. The typical foliar lesions induced by ozone under realistic exposure to ambient air are bi-facial greyish necrotic spots, scattered over the lamina (Manes et al., 1990b; Lorenzini et al., 2000). The tobacco system has been extensively investigated, also from the quantitative point of view. Biomonitoring campaigns have been successfully performed all over the world. Ozone-resistant tobacco plants (cv. Bel-B) are routinely inserted in the plots; their sensitivity threshold, in terms of visible injury, for 2 h exposures is 220 ppb vs. 100 ppb of Bel-W3.

Therefore, the appearance of injury on Bel-W3 but not on Bel-B provides further confirmation that such injury is due to ozone. Conventional biomonitoring is performed with adult tobacco plants (about 2 months old, 60 cm in height). However, some years ago, an innovative miniaturized kit for ozone biomonitoring was developed and patented at the University of Pisa, employing plates with tobacco seedlings (typically (Lorenzini, 1994; Lorenzini et al., 1995 – Fig. 5.8). These plates are exposed to ambient air for 7 days in shaded conditions, and the intensity of the injury of cotyledons and the first leaf is visually assessed and related to the dose of ozone to which the seedlings have been exposed (Nali et al., 2004; 2007). More precisely, at the University of Pisa two are the applicative fields based on the tobacco mini-kit, i.e. (i) educational projects of environmental education with young pupils (and their teachers and families) (e.g. Nali and Lorenzini, 2007; Pellegrini et al., 2014b), and (ii) regional wide season-long campaigns to fulfill commitments of local/national environmental authorities to allow permitting of industrial plants such as thermal power stations. Under the guidance of their teachers, the pupils had several opportunities to practice with many basic and applied study areas and disciplines and were initiated into the scientific method in a simple and absorbing manner. Though primarily an educational exercise, the survey provided sound research elements and the picture of pollution that emerged has increased the knowledge of air quality in the investigated area.



Figure 5.8 – The miniaturized kit for biomonitoring of  $O_3$  with tobacco Bel-W3 seedlings. The external dimensions are 12.5x8.5 cm. Please note necrotic lesions induced by the exposure to ambient air in the presence of  $O_3$ .

Recently, ozone sensitive (S156) and ozone resistant (R123) genotypes of snap bean (*Phaseolus vulgaris* L.), selected through genetic crosses, have been proposed as a new biomonitoring system (Burkey et al., 2005) (Fig. 5.9). Several studies have tested the new system under semi-controlled environmental conditions, showing that pod yield was similar in S156 and R123 under  $O_3$  concentrations lower than 30 ppb (S156/R123 pod yield ratio equal to 1), and consistently reduced by increased  $O_3$  levels in the sensitive genotype only (S156/R123 pod yield ratio  $< 1$ ) (Burkey et al., 2005, 2012; Flowers et al., 2007). Given these promising results, the ICP Vegetation  $O_3$  programme has conducted field trials using the snap bean system, following a standardised experimental protocol at pan-European scale since 2008 (Fig. 5.10), the advantage being also the cost-effectiveness of the seed-propagated bean plants, in respect to the vegetative-propagated clover clones (Burkey et al., 2005).

The results of the field trials, however, have shown that the extent of leaf injury on S156 variety, as well as the S156/R123 pod yield ratio, were often not directly related to the AOT40 at the experimental site. This was particularly true for Mediterranean sites, such as the Castelporziano Estate (Rome, Italy), in which the response of the snap bean system during three consecutive summer periods (years 2008-2010), characterized by different meteorological conditions and O<sub>3</sub> levels, was not related to seasonal ozone concentration (Fusaro et al., 2015). Recently, Salvatori et al. (2013) pointed out that the O<sub>3</sub> response of S156 and R123 bean lines varied within plant growth stage, with the different O<sub>3</sub> sensitivity of the two genotypes being more apparent during vegetative growth and flowering. Such differences can be due to different stomatal conductance of the sensitive and resistant plants, and to different effect of O<sub>3</sub> on the I-P phase of the fluorescence transient (Salvatori et al., 2015). A large-scale application of the snap bean biomonitoring system, under different climatic conditions and O<sub>3</sub> levels found at pan-European scale, appears therefore unsuitable.



Figure 5.9 – Ozone sensitive (S156) and ozone resistant (R123) genotypes of snap bean (*Phaseolus vulgaris* L.), during a fumigation experiment carried out in the “walk-in” chambers of the Department of Plant Biology, Sapienza University of Rome.



Figure 5.10 – O<sub>3</sub>-resistant (R123) and O<sub>3</sub>-sensitive (S156) snap bean plants, after three months of ambient air exposure in the Castelporziano Presidential Estate, following the ICP Vegetation biomonitoring protocol (Fusaro et al., 2015).

#### 5.4.2 Nitrogen

The risk assessment of nitrogen pollution in European Mediterranean natural and semi-natural ecosystems is difficult since only few experiments were carried out in the whole region. In Chapter 5 of the Mapping Manual, a list of critical loads ( $\text{kg N ha}^{-1} \text{ y}^{-1}$ ) is reported for natural and semi-natural ecosystems classified according to the European Nature Information System (EUNIS). For example, over a range of 15 and 25  $\text{kg N ha}^{-1} \text{ y}^{-1}$  Mediterranean xeric grasslands are expected to increase production and dominance of graminoids. Between 20 and 30  $\text{kg N ha}^{-1} \text{ y}^{-1}$ , Mediterranean shrub ecosystem changes species richness and composition; however, critical loads have not yet been set.

# **Biodiversity as an important indicator of soil acidity and eutrophication: the role of the modelling in preserving it**



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## **CHAPTER 6 - BIODIVERSITY AS AN IMPORTANT INDICATOR OF SOIL ACIDITY AND EUTROPHICATION: THE ROLE OF THE MODELLING IN PRESERVING IT**

### **6.1 Introduction**

The activities linked to biodiversity protection in Italy (and Europe) are the main issue of the ICP Modelling and Mapping Task Force. The objective to reach no net loss of biodiversity is the target for 2020, and modelling is the most suitable methodology at regional and national scale in order to investigate biodiversity losses.

Biodiversity provides ecosystem services crucial for human well-being. However, rapid population growth, coupled with socio-cultural changes, climate change and the unpredictable nature of economic change and its transition patterns, create challenges for decision-makers. Air pollution is a serious threat to the diversity of life. In general, it can be said that the effects of air pollutants on biological diversity usually affect more lower life forms than higher forms, and, as a result of air pollution, the most affected species decline, while some increase. In general, sensitivity varies from species to species within each group of organisms, but also due, for example, to the pollution load, the stage of life at which the individual is exposed, and the way competition is altered within a particular ecosystem. The effects found on plants affect mostly lichens, bryophytes, fungi, herbaceous flowering species, and trees.

Since 1970s, some evidences concerning negative effects of acid deposition on natural ecosystems have occurred. At the beginning of 1980s, extensive forest damage in central Europe linked to high levels of acid deposition led European Economic Commission to adopt the “Convention on Long-Range Transboundary Air Pollution” to mitigate the impact of air pollution on human health and natural ecosystems through research efforts and the adoption of air pollution abatement policies. At the same time, Critical Loads were derived as indicators of excessive nitrogen and sulphur atmospheric deposition.

Some studies have suggested that in Europe most of the increase in forest growth can be accounted for N deposition (Sutton et al., 2008) and very little by elevated CO<sub>2</sub> concentrations, but this does not seem to apply in all regions. Rehfuss et al. (1999) reported that the combination of CO<sub>2</sub> rise and elevated N deposition accounted for a 15-20% increase in forest net primary productivity. However, the N deposition effect on growth is expected to saturate or even decline in ecosystems with high N inputs (Brumme and Khanna, 2008). Nitrogen emissions and deposition of nitrogen compounds have decreased since 1990 but relatively little compared to sulphur emissions. Agriculture and transport are the main sources of nitrogen pollution (EEA, 2007). In addition, nitrogen components can lead to eutrophication of ecosystems. When these pollutants exceed certain levels (‘critical load’), there is a negative effect on biodiversity. A critical load is defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt, 1988). Exceedances of critical loads by current or future nitrogen loads indicate risks for adverse effects on biodiversity. The currently used methodologies to derived critical loads are all described in Mapping Manual ([www.icpmapping.org](http://www.icpmapping.org)).

Furthermore, excess nitrogen is one of the major threats to biodiversity. Excessive levels of reactive forms of nitrogen in the biosphere and atmosphere constitute a major threat to biodiversity in

terrestrial, aquatic and coastal ecosystems. On land, it causes loss of sensitive species and hence biodiversity by favouring a few nitrogen tolerant species over less tolerant ones.

## 6.2 Measurements of climate variable for modelling simulation

The monitoring activities of meteorological parameters carried out within the national program for integrated control of forest ecosystems (CONECOFOR) represent the Italian application of ICP-FOREST monitoring activities. Meteorological variables have been collected by CREA (Council for Agricultural Research and Economics) in collaboration with the State Forestry Corps through ground monitoring systems of a wide network of automatic stations (in function since 1997 in numerous forest areas of the monitoring network).

The data collection, integrated with other variables surveyed in the CONECOFOR Programme, support integrated studies on forest ecology and plant physiology and, more generally, contribute to define the health status of the Italian forests.

### 6.2.1 Activities description

Permanent monitoring areas for acquisition of meteorological data, inside the forest (“in the plot”) and in “open field” within the radius of 2 km from the first, have been established in accordance with EU Regulations n. 1091/94 and n. 690/95.

This monitoring activity started in 1997 and involved initially 8 permanent areas, for a total of 13 stations. The stages of the implementation of the survey included (i) automatic stations purchase and installation, (ii) periodical collection of data related to the selected variables, as well as (iii) data integration, control, validation and processing. Currently, the geographical structure of the monitoring network has undergone numerous improvements, totalling 26 working stations, of which 23 “open field” and 16 “in the plot”: the current distribution of the stations is shown in figure 6.1.



Figure 6.1 – Location of the meteorological network on the Italian territory.

The stations are installed and managed directly by CREA-RPS. The data analysis is performed in the laboratory of biometeorology at CREA-under the supervision of Dr Silvano Fares and Dr Luca Salvati. Some technical solutions for reducing sampling and data elaboration costs (such as the

upgrade with remote controls, see figure 6.2) are being tested in a small number of forest plots. Until now, 4 areas (ABR1, VEN1, LAZ1 and EMI1) have been implemented with ftp data transmission (datalogger Campbell CR-1000 and modem GPRS Telit GT863-PY).

The main variables registered with a hourly time resolution are:

Open field (Datalogger CR10x, CR1000, CR200x)

1. Wind speed (10 and 2 m) and direction (10 m);
2. Solar radiation (2 m);
3. Temperature and Relative humidity (air 10, 2 and 0.1 m);
4. Soil Temperature (20 cm);
5. Precipitation;
6. Snow depth;
7. Soil volumetric water content and Temperature profiles (10, 30 and 60 cm).

In the plot (Datalogger CR10x, CR1000)

1. Soil volumetric water content and Temperature profiles (10, 30 and 60 cm);
2. Temperature and Relative humidity (air 2 and 0.1 m);
3. Soil Temperature (20 cm);
4. Precipitation;
5. Snow depth;
6. Soil moisture (10, 25 and 60 cm mod. Delta T).

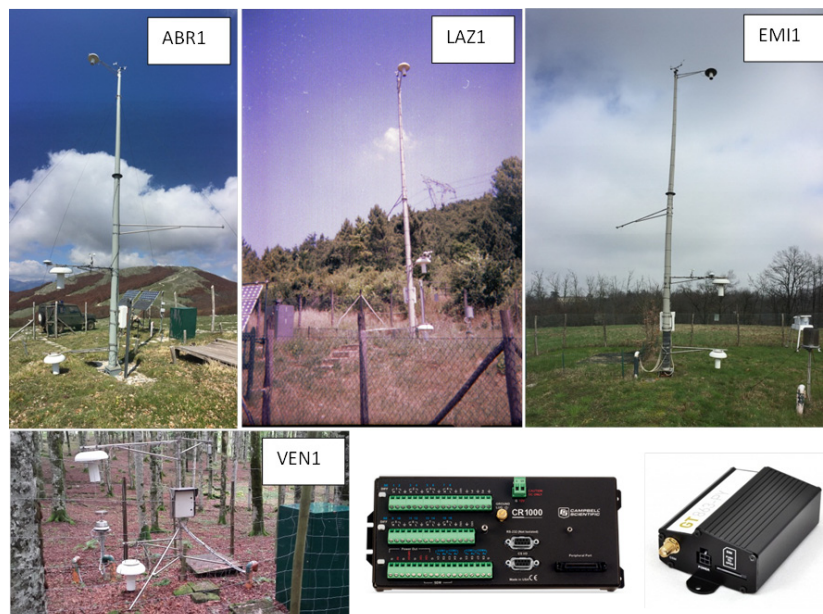


Figure 6.2 – “Open field” and “in the plot” automatic meteorological stations upgraded.

The received meteorological data are processed into a database in two steps: first, descriptive metadata of the permanent site (geographic and physical characteristics), metadata of the stations (acquisition mode, instrumentation installed), metadata of the sensors (specifications), and metadata of the measured parameters and of their processing are registered.

Successively, meteorological data from each weather station are logged and undergo a series of controls and automatic processing for data quality.

The information on the “control” field allow to determine both the type of sensor reporting abnormal data, and the impact on measurement of the error duration. These metadata are useful to assess the completeness of meteorological data in terms of percentage of working time.

Annual activities of the equipment ordinary maintenance are provided in order to ensure the normal operation of the acquisition system, as well as extraordinary interventions for the resolution of specific technical problems.

Finally, the collected data are validated and made available on the website of the ICP forest.

The meteorological data of the 13 selected monitoring sites with the longest time series are reported in table 6.1 as a set of descriptive statistics calculated for five main meteorological variables.

Figures 6.3 and 6.4 show precipitation and temperature trends. The data time lapse is 1st January 1998 – 31st December 2013, with 1-day sampling interval.

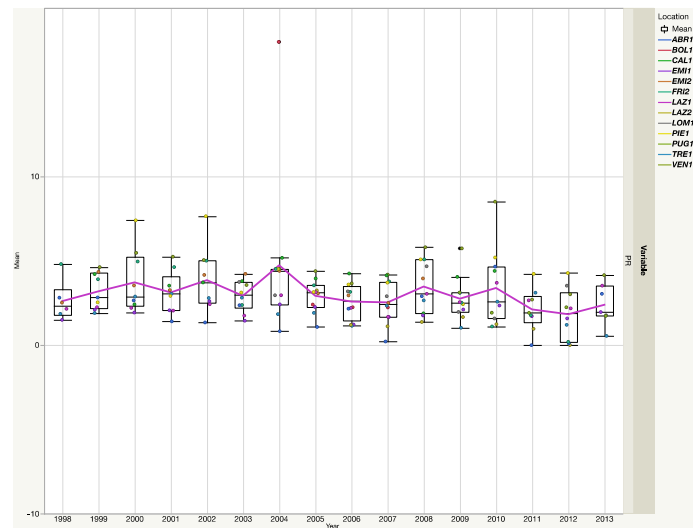


Figure 6.3 – Time trend of average rainfall for the 13 main sites with the longest time series.

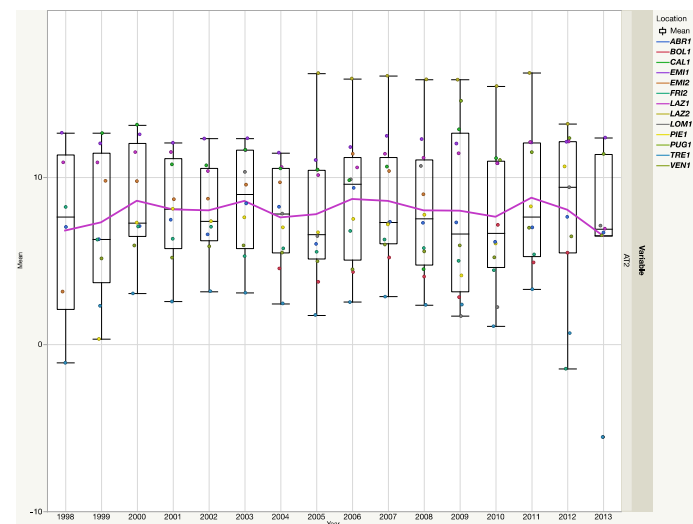


Figure 6.4 – Time trend of average temperature at 2 m for the 13 main sites with the longest time series.

Table 6.1 – Summary statistics, including the mean and median, standard deviation and coefficient of variation values, calculated for each variable, relative to 13 selected sites during the entire analyzed time period (1998-2013).

Note that the variation coefficient (CV) is a dimensionless quantity.

Stations	Statistics	AT01 (°C)	AT2 (°C)	RH01 (%)	RH2 (%)	PR (mm)
01-ABR1	Mean	6.63	7.21	79.62	83.12	2.06
	Median	7.1	7.6	82.1	86.4	0
	Std Dev	6.66	6.74	16.49	15.87	6.61
	CV	100.5	93.48	20.71	19.1	320.65
03-CAL1	Mean	10.83	11	89.17	86.4	3.91
	Median	10.8	11	95.5	93	0
	Std Dev	6.23	6.24	13.87	15.64	10.22
	CV	57.53	56.71	15.56	18.1	261.12
05-EMI1	Mean	12.22	12.02	81.16	80.64	1.92
	Median	12.5	12.3	84.75	85.2	0
	Std Dev	8.17	8.06	17.28	18.37	6.1
	CV	66.89	67.03	21.29	22.78	318.06
09-LAZ1	Mean	11.25	11	79.63	75.62	2.51
	Median	11.1	10.6	82	78.2	0
	Std Dev	7.3	6.92	15.18	17.51	6.99
	CV	64.86	62.93	19.06	23.16	279.01
12-PIE1	Mean	6.09	7.31	85.94	79.27	4.54
	Median	6.1	7.3	93.9	84.9	0
	Std Dev	6.96	6.82	17.23	20.1	15.52
	CV	114.35	93.19	20.05	25.35	341.81
20-VEN1	Media	4.5	5.72	91.34	85.75	4.72
	Median	4.8	6	93.4	90.1	0
	Dev std	7.28	7.2	8.98	14.2	13.85
	CV	161.67	125.89	9.83	16.56	293.21
06-EMI2	Mean	9.8	9.54	82.07	80.06	3.64
	Median	10	9.3	85.9	84.2	0
	Std Dev	7	7.03	16.36	17.36	9.95
	CV	71.45	73.67	19.94	21.69	273.66
08-FRI2	Mean	6.16	5.91	92.63	91.36	3.62
	Median	6.6	6.3	96.3	95.5	0
	Std Dev	7.56	7.64	8.79	10.89	11.05
	CV	122.7	129.15	9.49	11.92	305.05
10-LOM1	Mean	-	7.55	-	71.1	2.96
	Median	-	8.2	-	73	0
	Std Dev	-	6.71	-	17.34	8.13
	CV	-	88.82	-	24.38	274.13
13-PUG1	Mean	-	11.91	-	78.7	2.09
	Median	-	11.7	-	83	0
	Std Dev	-	6.87	-	13.71	6.99
	CV	-	57.71	-	17.41	334.7
17-TRE1	Mean	1.8	2.21	94.8	86.39	2.19
	Median	1.1	2	99.7	91.2	0
	Std Dev	7.24	7.29	9	14.44	5.89
	CV	401.24	330.18	9.49	16.71	268.88
22-LAZ2	Mean	-	15.72	-	77.42	1.3
	Median	-	15.3	-	79	0
	Std Dev	-	5.96	-	12.34	4.43
	CV	-	37.9	-	15.94	340.25
27-BOL1	Mean	-	4.45	-	72.75	3.01
	Median	-	4.7	-	75	0
	Std Dev	-	6.91	-	18.04	8.91
	CV	-	155.35	-	24.79	295.97

### 6.3 Impacts

Adverse effects of excessive deposition, like vegetation changes or forest dieback, does therefore not necessarily lead to immediate damages to ecosystems. To reconstruct and/or predict the temporal development of a soil and vegetation system, dynamic models, that include relevant time-dependent processes, are required. In the last three decades, different dynamic models have been developed, such as FORSAFE (Belyazid et al., 2006), MAGIC (Cosby et al., 1985, 2001), SMART/SMARTml (Bonten et al., 2011) and VSD/VSD+ (Posch and Reinds 2009; Reinds et al., 2001). The last one is used at site level in Italy.

More precisely, a suite of four models has been set to estimate the time required for a new (steady) state to be achieved when chemical and biological parameters response to a change in deposition. Figure 6.5 summarizes the suite of models used to estimate critical loads and biodiversity indices per single site.

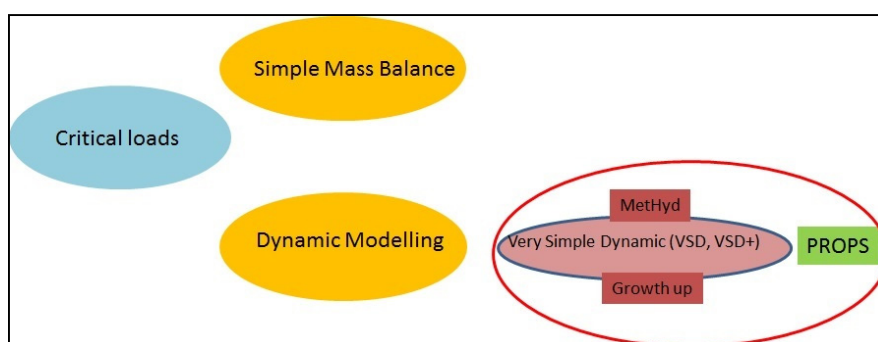


Figure 6.5 – Schematic representation of two methods to estimate Critical load: steady-state and dynamic. The second approach is well represented by the combination of 4 models, able to estimate biodiversity indices.

The **MetHyd model** is a meteo-hydrological pre-processor. It is working elaborating Temperature, Photosynthetic Active Radiation and Precipitation surplus to estimate reduction factor of nitrification rates and of mineralization rates.

The **GrowthUp model** is a pre-processor able to compute N and base cation uptake from user specified tree-growth inputs.

The **Very Simple Dynamic (VSD) model** is the (minimal) extension of the Simple Mass Balance (SMB) steady-state model into a dynamic soil (acidification) model. The **VSD+ model** is an extension of the VSD model with detailed C and N dynamics. The model requires only a minimum set of inputs (compared to more detailed models) and execution time is minimised by reducing the set of model equations to a single non-linear equation. To facilitate the exploration of model behaviour at individual sites, the model is linked to a graphical user interface (GUI). This GUI allows easy (Bayesian) calibration, forward simulation (scenario analyses) and can also be used to compute target loads and delayed times between deposition reductions and ecosystem recovery.

**PROPS module** is the last step of the suite devoted to compute each potential species occurrence probability. The potential biotic is defined using EUNIS class corresponding to Corine land cover class.

### 6.3.1 Biodiversity analysis with dynamic models.

The described dynamic modelling suite has been applied in 5 forest Italian sites. The application of the modelling suite to one Italian site is described in Figure 6.6. The site chosen in this figure is IT10/LOM1 (Val Masino) from the ICP-Integrated Monitoring and/or ICP-Forests networks respectively. This site is characterized by high naturalistic value (protected by Habitat and Bird Directives), high sensitivity (low critical loads) and high pollutant exposure (critical loads were exceeded in the year 2000). The Val Masino site is a secondary *Picea abies* dominated forest with *Abies alba* and *Vaccinium myrtillus*. It belongs to CONECOFOR Programme since 1995 and is included in the ICP Forest European network. This site is in the Central Alps at 900-1190 m a.s.l., and its forest type is classified as EUNIS class G4.6. The VSD+ suite model was applied to simulate soil solution chemistry behaviour in response to net input from atmosphere, net element uptake by vegetation, net nitrogen immobilisation and element weathering.

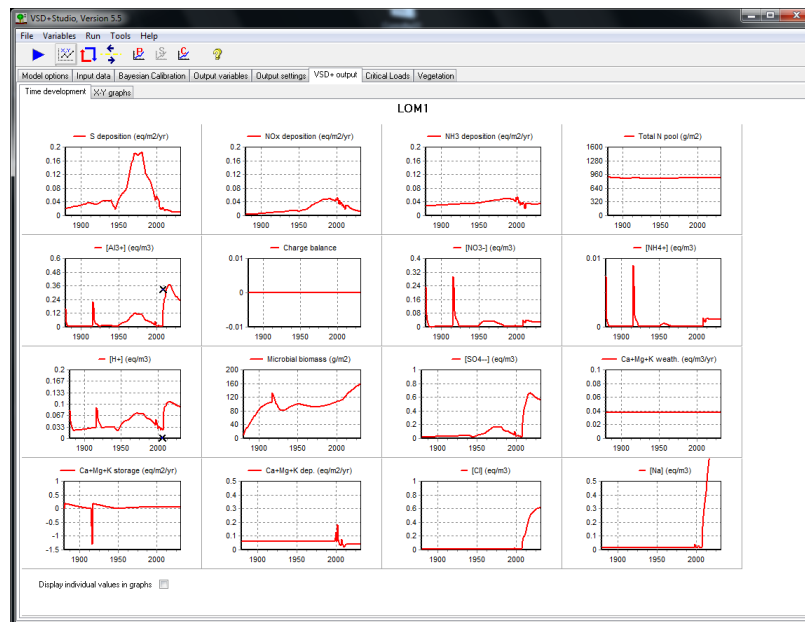


Figure 6.6 – VSD+ run results. Parameter in soil solution and atmospheric deposition are shown.

The outputs obtained from VSD+ were used as inputs for running the PROPS vegetation model. Vegetation types (32 typical species) have been hypothesized from the EUNIS class G4.6. PROPS module estimated the Habitat Suitability Index (HSI), that is defined as the arithmetic mean of the normalized occurrence probabilities of the typical ecosystem species (Figure 6.7A). The PROPS model also computed isolines of normalized occurrence probability as a function of pH and N concentration (Figure 6.7B).

The HSI trend modelled by PROPS in 5 Italian forest sites, characterised by different climatic and environmental conditions is shown in Fig 6.8. The analysis shows an increasing trend for all the sites, even if with different slope, demonstrating a biodiversity recovery after the year 2000, when a decrease in Nitrogen deposition is found all over Italy. The higher rate of increase for the HSI is in the site called IT05, that is in Central Italy, where the dominant tree species is European beech.

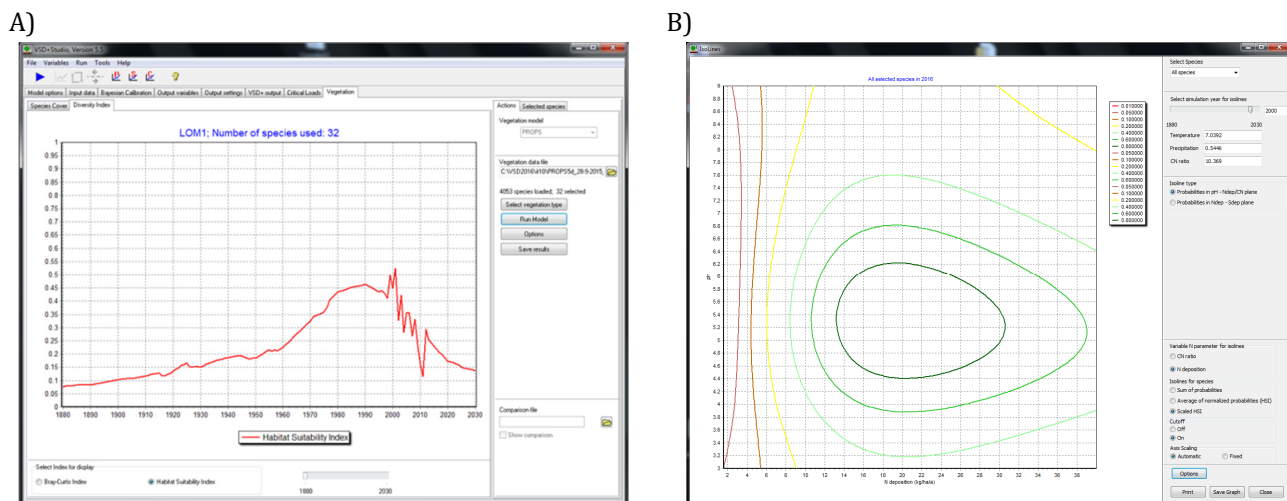


Figure 6.7 – Site IT10 VSD+ results. Habitat Suitability Index during time (A) and occurrence probability isolines as pH and N concentration dependent (B).

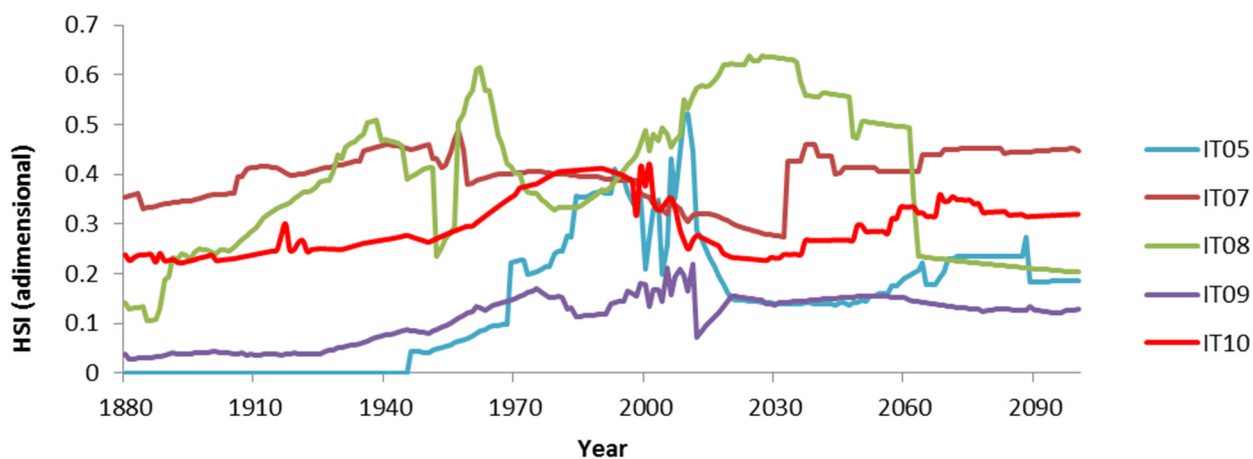


Figure. 6.8 – HSI in five Italian sites from year 1880 to 2100.

In the same way, it is possible to derive N and S range deposition to allow 80% of typical plant species for a specific ecosystem to survive. Those values are N and S biodiversity-based critical loads.

Figure 6.9 shows how derive N and S critical values to ensure the surviving of 80% typical species in three example sites.

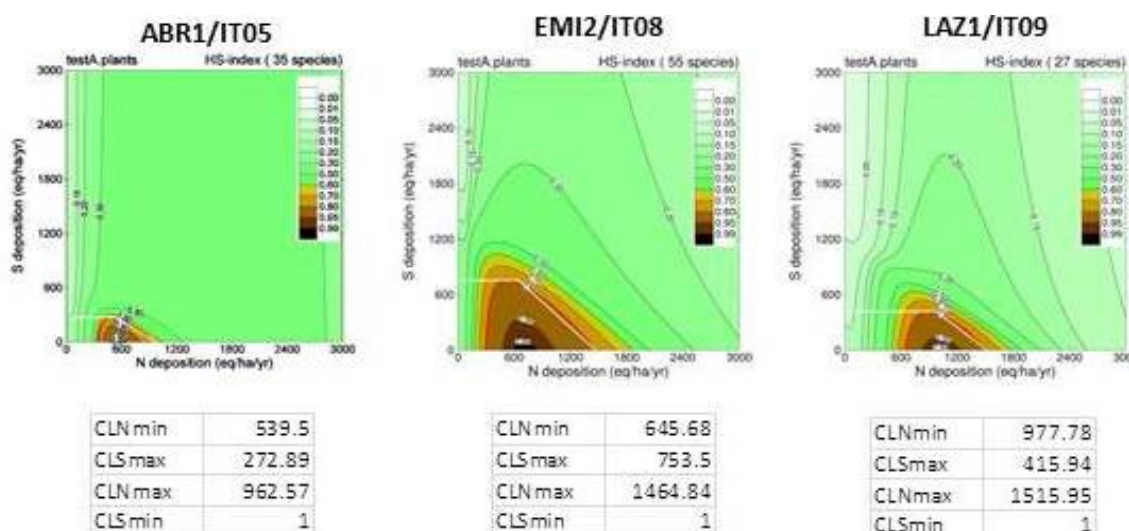


Figure 6.9 – Habitat suitability Index as N and S concentration dependent in three different Italian forest sites. Biodiversity CL (CLNmin, CLSmax, CLNmax, CLSmin) indicates the range of values for S and N indispensable for the maintenance of at least 80% of the typical species for each biocenosis.

### 6.3.2 Biodiversity analysis with statistical models.

The impacts of air pollution on biodiversity are affected by the presence of other factors which themselves have an influence on ecosystems, such as meteorological conditions, forest management practices, soil types, and introduced plant diseases. Sorting out the most important factors is often difficult and sometimes impossible. Some of these factors are interrelated; trees may for instance be so weakened by air pollution as to be especially susceptible to disease. High altitude environments will be among the first to show the effects of acidification.

Although pollution often decreases with altitude, deposition can remain high because precipitation increase. Severe climatic conditions also make plants unable to absorb additional atmospheric nitrogen, which instead leaks by run-off.

We have chosen three differently test sites belonging to the Italian forestry monitoring network (see table 6.2 show sites information):

Table 6.2 – Test sites main information.

Site	IT05/ABR1	IT09/LAZ1	IT10/LOM1
Name	Selvapiana	Monte Rufeno	Val Masino
Latitude	41.8475	42.8306	46.2378
Longitude	13.5975	11.9139	93.5211
Altitude (m slm)	1500	690	1190
No. Of species	24	60	60
Forest type	beech forest	oak forest	spruce (and fir) forest
Age	123	48	93
EMEP50 cod	84036	80035	71039
Protection	Birds and Habitat directives applied	0	Birds and Habitat directives applied
<a href="#">EUNIS</a>	G1.6	G1.7	G4.6

### ***The Pielou's evenness index***

Because the values of the diversity indices are not always comparable among them and depending on the extent to which they can actually vary, we used the evenness as a measure of diversity normalized on a fixed scale (e.g. from 0 to 1), allowing to carry out these comparisons among the three test sites.

$$H' = - \sum p_i \ln(p_i)$$

where  $H'$  is the Shannon-Weaver Diversity Index (Shannon and Weaver 1949),  $p_i$  is the relative abundance of each group of organisms.

$$H'_{max} = \ln S$$

where  $S$  is the species number. The Pielou's evenness (Pielou, 1966) is derived from the Shannon-Weaver index as follows:

$$J = \frac{H'}{H'_{max}}$$

$J$  is constrained between 0 and 1; the less variation in communities among species implies higher values of  $J$ .

In our study, in order to measure the overall plant species diversity of a given transect, a modified version of the Shannon index ( $H'$ ), named  $H_{dunestd}$  (Grunewald & Schubert 2007), was used:

$$H_{dunestd} = - \frac{1}{\ln(k)} \sum p_i \times \ln(p_i)$$

where  $p_i$  = % cover of the  $i^{th}$  species and  $k$  is number of sampled species.

$H_{dunestd}$  has proved to be more useful than the Shannon diversity index in limiting habitats such as the coastal dunes (De Luca et al., 2011), where natural stressful conditions determine the presence of a few species with high dominance (Martínez et al., 2004).

$H_{dunestd}$ , in fact, uses the abundance of species (as cover percentage) in relation to a constant sampling area, and hence, unlike  $H$ , is able to detect changes both in species diversity and total cover.

### ***Characterisation of the ecological niche for plant species: the Ellenberg's indicators.***

Ellenberg defined a set of indicator values for the vascular plants of central Europe (Ellenberg 1979, 1988; Ellenberg et al., 1991). The latest edition of Ellenberg's indicator values applies a 9-point scale for each of six gradients: soil acidity, soil productivity or fertility, soil humidity, soil salinity, climatic continentality and light availability. These have been widely used, both in central Europe and in adjacent parts of western Europe. The basis of indicator values is the realised ecological niche. Plants have a certain range of tolerance of temperature, light, soil pH, and so on. If we wish to make inferences about the ecological conditions pertaining at a site, much useful information can be obtained from the flora. These values are not i.e. mean pH values, but are on an arbitrary scale reflecting soil pH though not directly based on measurements. However, an advantage of indicator values is that they may be more sensitive to the requirements of plants than to a selected physical variable.

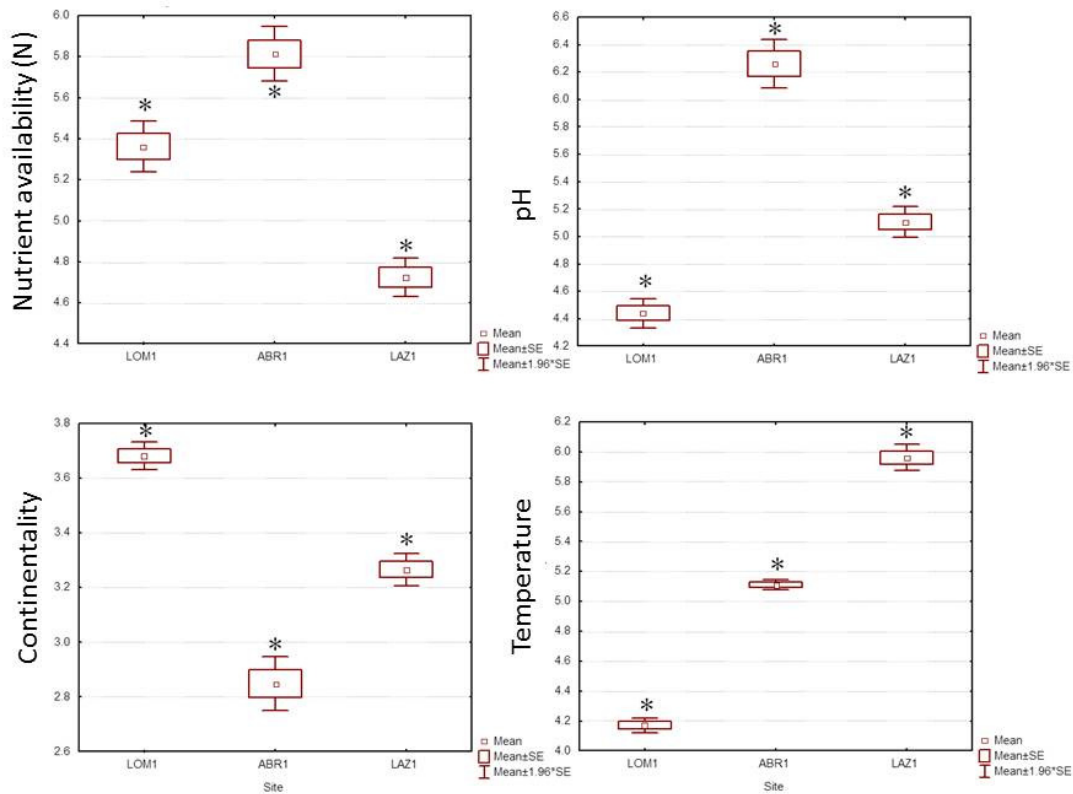


Figure 6.10 – Four Ellenberger's indices (Nutrient availability, pH, Continentality and Temperature) calculated for the three Italian study sites. They well define the sites' characteristics either soil properties or climate.

When the Ellenberg's indices were applied to the three test sites, they showed the ecological ranges that defined peculiar characteristics for each forest site (Figure 6.10). Significant differences of indices were observed among sites (ANOVA test,  $p < 0.05$ ).

The application of two biodiversity indices  $J$  and  $H_{dunestd}$  showed completely different trends among sites (Fig. 6.11). It is useful to remember that  $H_{dunestd}$  is a modified Shannon-Weaver index that was focused on the cover percentage of each species (Attorre et al., 2013). The Pielou's evenness did not shown any difference among test sites (all sites have high equitability i.e. individuals are highly distributed among species), whereas  $H_{dunestd}$  exhibited different biodiversity values, pointing out different patterns of plant species distribution related to the peculiar habitats of forest sites. In fact, a preliminary analysis of species richness at community level revealed that the lowest values occurred in beech forests (ABR1) and the highest in Turkey oak forests (LAZ1), whereas spruce forests (LOM1) were intermediate.

These features highlighted different vegetation dynamics characterised by fluctuations as the commonest on-going process (i.e. LOM1). The regeneration dynamic is also widespread due to the recent abandonment of wood exploitation and coppice management (i.e. ABR1) and low values of  $H_{dunestd}$ , whereas the regression dynamic was predominant in Turkey oak forests as in LAZ1 (and high values of  $H_{dunestd}$ ).

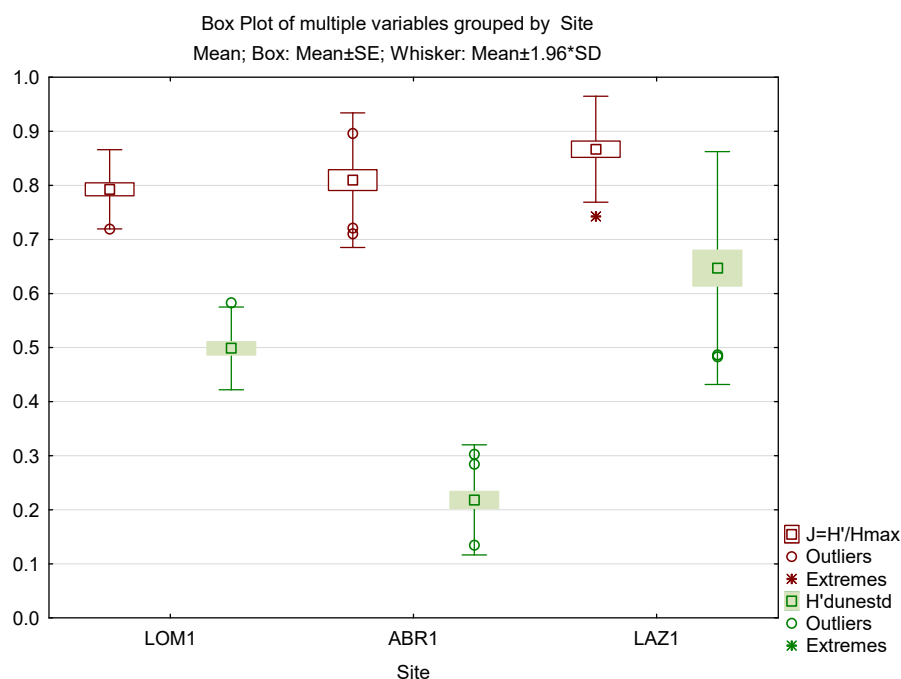


Figure 6.11 – Biodiversity indices ( $J$  and  $H_{dune}$ , light and grey boxes, respectively) calculated for the three study sites. Both indices have been calculated by using the same phytosociological surveys, showing however, different values.

Correlation among variables (biodiversity indices, Ellenberg's indices and exceedances above (BOF N) and under canopy (BSC N) (Both with trend in Figure 6.12) calculated for the three test sites did not highlight a clear aspect for the three test sites. Correlation analysis showed that diversity indices did not correlate with the Ellenberg's indicators except  $J$  in the LAZ1 site (-0.72 Ellen. Temp; -0.88 Ellen. Cont.; 0.75 Ellen. Soil Moist.), but only  $H_{dunestd}$  correlated with BOF N in LOM1.

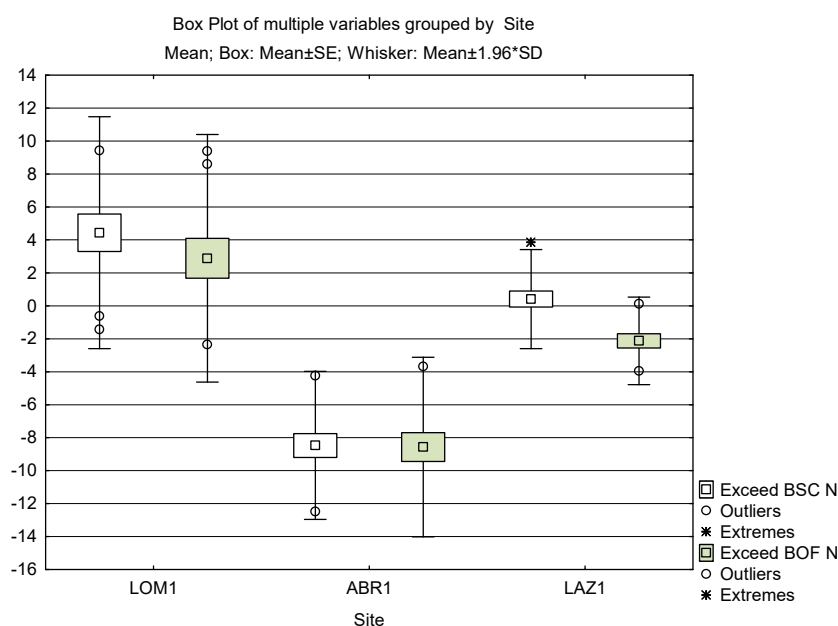


Figure 6.12 – Exceedances above (BOF N) and under canopy (BSC N) for the three study sites.

These simple analyses highlighted that different diversity indices (one based on the species proportion and the other based on proportional coverage area) could suggest different trends, which were not in correlation with climatic data and N depositions for three herbaceous communities growing under different ecological niches. It seems that one decade of data it is not sufficient for assessing a change in herbaceous community's composition. Inferences made on the air pollutant-induced effects in affecting plant community's composition should be carried out with extreme caution.

Our results are highlighting that the application of different biodiversity indices and ecological indicators should be applied in extensive way to other European herbaceous plant communities, to assess if air pollutants, and/or climate, and or anthropogenic activities are causal effects for a changing plant communities. Furthermore, it is important to have a long historical series of Ellenberg indices to assess the spatio-temporal dynamics for the European and Italian forest sites. Some trends are noticeable such as an increase of the Ellenberg Soil pH for ABR1 site or a reduction of Ellenberg Soil N for LOM1 site, although significances are questionable.

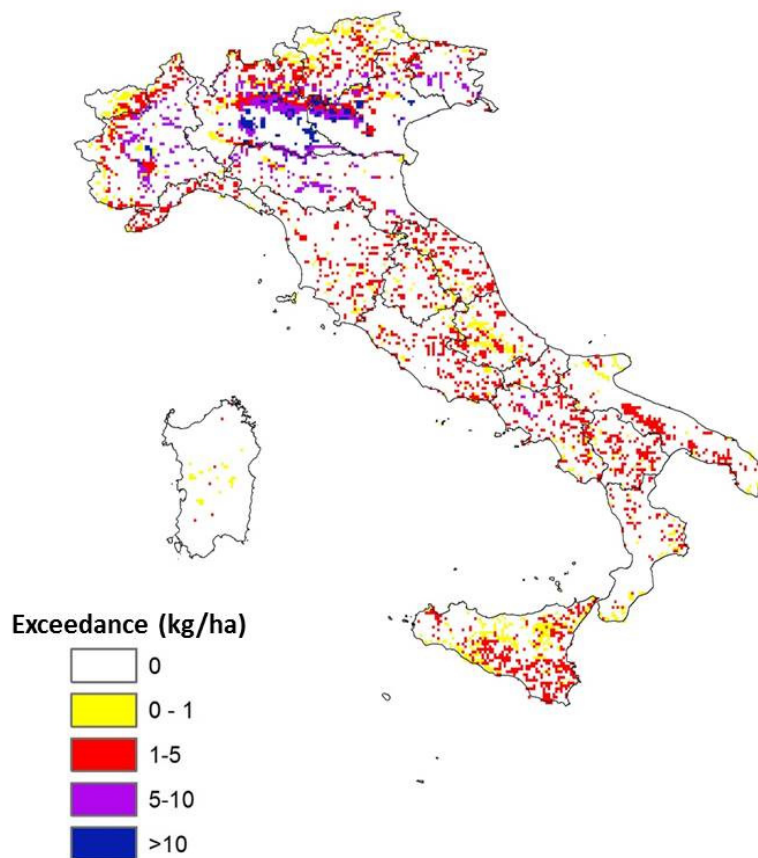
Cause-effect relationships between pollutants and primary production can be difficult to find in the field because a multitude of interactions concurrently could act on the response variable. For this reason, non-linear statistical techniques are becoming of increasing interest. The synergic or antagonistic roles of some limiting factors (such as high temperature, high ozone concentration) in affecting physiological processes (gas exchange, stomatal conductance) are very evident. Further, local variations of ox-N and red-N affected NPP in a complementary manner, acting as positive or negative drivers.

The impacts of air pollution and climate change on crown defoliation were different for each tree species, suggesting species-dependent effects on forests health and vitality. The vulnerability of forest tree species not only depends on exposure to climate change and air pollution but also on adaptive ability of the tree species (Lindner et al., 2010). Changes in climate will be associated to biotic (frequency and consequence of pest and disease outbreak) and abiotic disturbances (changes in fire occurrence and wind storm), causing strong implications for forest ecosystem (Lindner et al., 2010). In this frame nitrogen deposition, affects tree physiology, carbon allocation and plant interactions, resulting in complex relations with other environmental limiting factors such as drought (Matyssek et al., 2006).

## **6.4 Risk assessment**

The risk assessment for critical loads for nutrient nitrogen in Italy for the year 2015 is shown in Figure 6.13.

The deposition over Italy are obtained by GAIN-Italy model with a grid resolution of 20 km and the exceedance map is obtained by difference between nitrogen deposition and critical loads exceedances. Exceedances are present almost at all the latitude in Italy, even if higher levels of exceedances are in the Alps in the Northern Region ( $> 10$  kg/ha/y), due to higher emission levels of nitrogen compounds and consequently higher nitrogen deposition in this area.



*Figure 6.13 – Nitrogen nutrient exceedance in Italy in the year 2015, based on GAINS-Italy model total nitrogen deposition exceedance.*

## 6.5 Conclusions

To estimate the impacts of nitrogen pollution on biodiversity two kind of approaches are suitable, the first one based on dynamic modeling and the second one based on statistical models. Dynamic models estimate the biodiversity indices on a large time frame from 1980 till 2100, while the statistical ones analyse the non-linear relationship between environmental parameters and biodiversity. All the results show that there is a decreasing trend in nitrogen deposition, due to the policies developed to control nitrogen emissions. The decrease is coupled with a recovery trend in biodiversity indices. Despite such recovery trend in Italy, especially in Northern region, there are still nitrogen nutrient exceedance areas, where the ecosystem is exposed to the pressure of nitrogen pollution over the critical loads limits. More effort is needed to further reduce nitrogen pollution in Northern area, and this effort should be addressed to the agricultural sector that is the main constraint to completely recover biodiversity.



# The contribution of Italy to the ICP WATERS Programme



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## CHAPTER 7 - THE CONTRIBUTION OF ITALY TO THE ICP WATERS PROGRAMME

### 7.1 Introduction

The international cooperative programme on assessment and monitoring of air pollution on rivers and lakes (ICP WATERS) was established under the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) in July 1985. The ICP WATERS Programme Centre is hosted by the Norwegian Institute for Water Research (NIVA), while the Norwegian Climate and Pollution Agency leads the programme.

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate in the programme and provide data on a regular basis.

ICP Waters is based on existing surface water monitoring programmes in the participating countries, implemented by voluntary contributions. The ICP site network is geographically extensive and includes long-term data series (more than 20 years) for many sites. At present, the network includes about 200 sites in Europe and North America (Fig. 7.1). The programme yearly conducts chemical and biological intercalibrations (e.g. Escudero, 2015; Fjellheim et al., 2015).

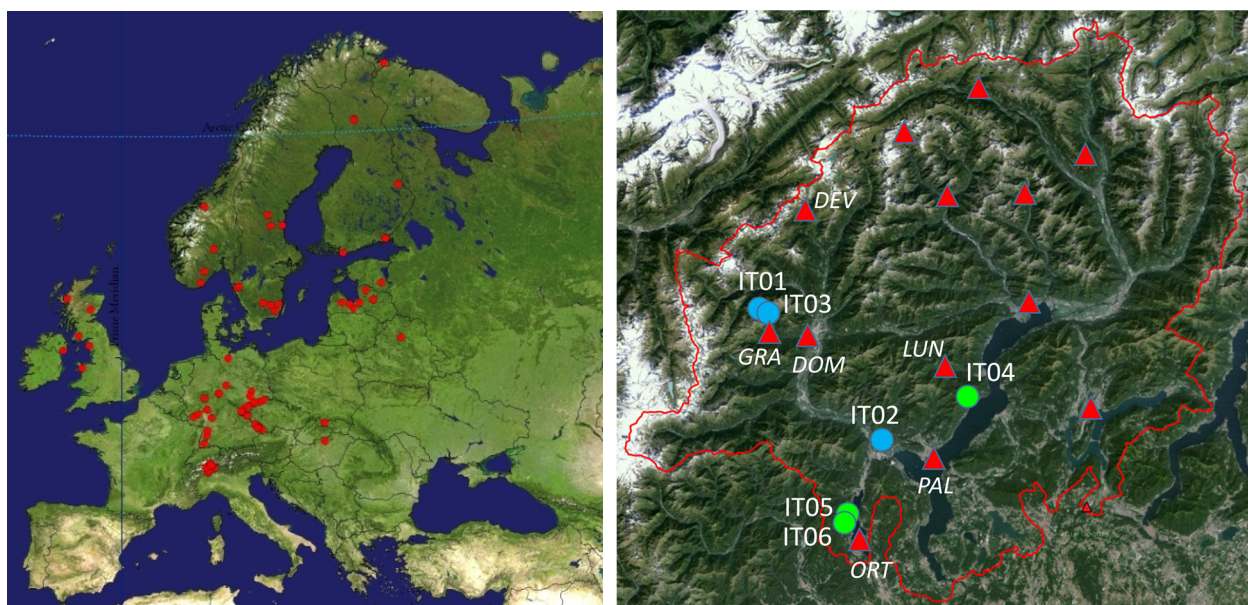


Figure 7.1 – The ICP WATERS sites in Europe (left) and the Italian network in the area of Lake Maggiore watershed, Northern Italy (right). Blue circles: lake sites; green circles: river sites; red triangles: atmospheric deposition sampling stations (including the Swiss network run by “Ufficio aria, clima e energie rinnovabili, SPAAS, DT Canton Ticino”). Site codes as in Tab. 7.1.

Since its establishment, ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution on surface water have been published over the years. Reports, scientific papers, Task Force proceedings and other publications are made available through the ICP WATERS website: <http://www.icp-waters.no>. Beside periodic reports describing the long-term chemical trends at the ICP WATERS sites, reports or papers on specific issues have also been produced: for instance, in depth analysis of biological data providing

evidence of recovery from acidification, or investigation on the so-called “confounding factors”, i.e. factors, other than deposition, which may affect long-term change in water chemistry and biology (Wright et al., 2005). As an example, several studies were devoted to the role of climate change on the reversibility of water acidification (e.g. Wright et al., 2006; Wright and Jenkins, 2001). A further confounding factor which has been addressed using ICP WATERS data was nitrate leaching, which proved to be important in delaying acidification recovery at sites subject to high N deposition (de Wit and Lindholm, 2010).

The Institute of Ecosystem Study of the National Research Council of Italy (CNR ISE; formerly “Istituto Italiano di Idrobiologia”) in Verbania Pallanza has been the National Focal Point for the ICP WATERS since 1995, under the direction and coordination of the Italian Ministry for the Environment, Land and Sea. The regular participation of Italy to the ICP WATERS has led to:

- the development of an Italian network of sites, consisting of three subalpine rivers, one subalpine lake and two high altitude alpine lakes in the area of Lake Maggiore watershed, Piedmont region (Fig. 7.1, Tab. 7.1), to follow the long-term evolution of acidification and recovery, and more generally the response of surface waters in sensitive areas to changing deposition;
- the harmonisation and standardisation of monitoring practices, both for sampling and analysis, following the provision of the Programme Centre (NIVA); this was also allowed by the regular participation of the CNR ISE hydrochemical laboratory to chemical intercomparisons;
- specific research performed on selected sites or areas, to better understand site-specific processes and dynamics at the catchment scale (e.g. nitrogen leaching and response to changing nitrogen deposition; acidification recovery under climate change scenarios).

*Table 7.1 – Main characteristics of the Italian sites included in the ICP WATERS network.*

Site name	Code	Altitude (m a.s.l.)	Catchment area (km <sup>2</sup> )	Lake area (km <sup>2</sup> )	Yearly average flow (m <sup>3</sup> s <sup>-1</sup> )	Data since	Data frequency
Lake Paione Inferiore	IT01	2002	1.26	0.0068		1984	1-2 per year
Lake Mergozzo	IT02	194	10.43	1.83		1978	2-4 per year
Lake Paione Superiore	IT03	2269	0.50	0.0086		1984	1-2 per year
River Cannobino	IT04	193	110.4	-	5.04	1978	Monthly
River Pellino	IT05	290	17.5	-	0.92	1984	Monthly
River Pellesino	IT06	290	3.4	-	0.19	1986	Monthly

The Italian network and the monitoring activities are described in details in Mosello et al. (2000). The CNR ISE has been regularly sending data to the Programme Centre since the establishment of the network. Collected data included base chemical variables (pH, alkalinity, conductivity), nutrients (phosphorus and nitrogen compounds), major ions (Ca, Mg, Na, K, SO<sub>4</sub>, Cl) and selected trace metals (Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn). Besides monitoring surface water bodies, the CNR ISE also runs a network for the assessment of long-term change in atmospheric deposition (Rogora et al., 2016). Analytical methods and QA/QC procedure adopted in the laboratory are described in details at the website <http://www.idrolab.ise.cnr.it>

Results have been presented at national and international conferences and published both in ICP WATERS reports and in scientific papers (e.g. Stoddard et al., 1999; Skjelkvåle et al., 2005; Garmo et al., 2014). In particular, a fruitful cooperation exists with the Swiss colleagues of the “Ufficio aria, clima e energie rinnovabili, SPAAS, DT Canton Ticino” and of the University of Applied Sciences and Arts of Southern Switzerland, who act as ICP WATERS National Focal Point for Switzerland (Steingruber 2015).

## 7.2 Atmospheric pollution pressures on surface waters

Within the ICP Waters aims the main pressures identified as important at the Italian sites of the network are acidification and nitrogen deposition. Furthermore, especially in recent times, attention has been paid to the effects of climate change, in interaction with the other drivers.

**Acidification** of surface waters due to rain acidity has been a problem in the 1970s and 1980s for some high altitude lakes in the Central Alps, characterised by a low alkalinity pool and a limited buffering capacity due to the geological composition of the catchments (Marchetto et al., 1994). Beside the two lakes included in the ICP WATERS network (Lake Paione Superiore and Inferiore; Tab. 7.1), the CNR ISE has been regularly monitoring from the chemical point of view about 30 high altitude lakes since the early 1980s. Most of these lakes, which underwent acidification in the 1980s, partially recovered from the mid-1990s as a response to the decreasing deposition of acidifying compounds, showing an increase of alkalinity and pH (mainly as sulphate ( $\text{SO}_4$ )) (Rogora et al., 2001; 2013). However, few sites remain acidic or still show a high sensitivity to acidification. This is particularly evident at the snowmelt, when alkalinity may be fully depleted by the incoming waters rich in acidifying compounds. At present, nitrate is the dominant acidifying agent in the high altitude lakes, due to the high input of nitrogen compounds from atmospheric deposition (Rogora et al., 2013).

One of the main risk related to water acidification is the dissolution of **trace metals**, which may be harmful for the aquatic biota (especially Al, Cd, Pb, Cu, Ni). For this reason, in addition to standard chemical variables also trace metals have been regularly analysed at the ICP WATERS sites in Italy. Levels proved to be low at all the sites, including high altitude lakes (Tornimbeni and Rogora, 2012). Within the ICP WATERS network, Italian sites proved to be among the most affected by nitrogen (N) inputs from the atmosphere. They were indeed threatened by **N enrichment** due to the N saturation of soils in the catchment and following nitrate ( $\text{NO}_3$ ) leaching to surface waters (Rogora and Mosello, 2007; Rogora, 2007). This is mainly due to the high levels of N deposition which affected the study sites, located north of the Po Plain, one of the most densely inhabited and most industrialised and urbanised areas of Europe. Depositions in the alpine and subalpine areas are particularly high as a combined effect of high pollutant concentrations and high precipitation amounts for orographic effects (Rogora et al., 2006).

Long-term studies at the ICP WATERS sites in Italy also revealed the important role that **climate** drivers may have on the response of surface waters to changing deposition (Rogora and Mosello, 2007; Rogora et al., 2003a). Climatic factors interact with atmospheric deposition affecting the long-term changes in lake water chemistry and biology, with an overall effect which may both favour or contrast recovery patterns according to the specific processes at each site. Studies at the Italian sites also demonstrated that temporal variations of N compounds in surface water may be affected by climatic factors: both increasing temperature and change in precipitation regime proved to be important in the  $\text{NO}_3$  long- and short- term dynamics in rivers and lakes (Rogora, 2007;

Rogora et al., 2013). The assessment of the atmospheric inputs of acidity and S and N compounds in the area of Lake Maggiore watershed have been performed since the beginning of the monitoring through a cooperation between Italy and Switzerland. This area is indeed shared almost equally between the two countries, and ICP WATERS sites, both Italian and Swiss sites, are located here (Fig. 7.1). An extensive network for the study of atmospheric deposition chemistry exists in this area, consisting of 14 stations in total (Rogora et al., 2016). In Italy, emissions of sulphur dioxide ( $\text{SO}_2$ ) reached its maximum between 1965 and 1980, while nitrogen oxides ( $\text{NO}_x$ ) peaked around 1985. Successively, emissions decreased by 90%, 58% and 14%, respectively for  $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{NH}_3$  compared to the values in 1990 (Romano et al., 2014). This resulted in a sharp decrease in the deposition of sulphate, acidity and, at a lesser extent, of N compounds.

The long-term trends of reduced ( $\text{NH}_4$ ) and oxidised ( $\text{NO}_3$ ) N deposition at sites in the Italian part of Lake Maggiore showed a slight tendency to decrease starting in 2006 (Fig. 7.2). The decrease of nitrogen loads was widespread and affected both the northern alpine stations and the southern ones (Rogora et al., 2016). The decrease was more evident for oxidized N than for reduced N. Therefore, deposition of ammonium acquired an increasing importance in time, especially at the southern, more polluted sites: the relative contribution of reduced N to wet N deposition passed from about 50% in the early 1990s to 56-57% in recent years (Rogora et al., 2016).

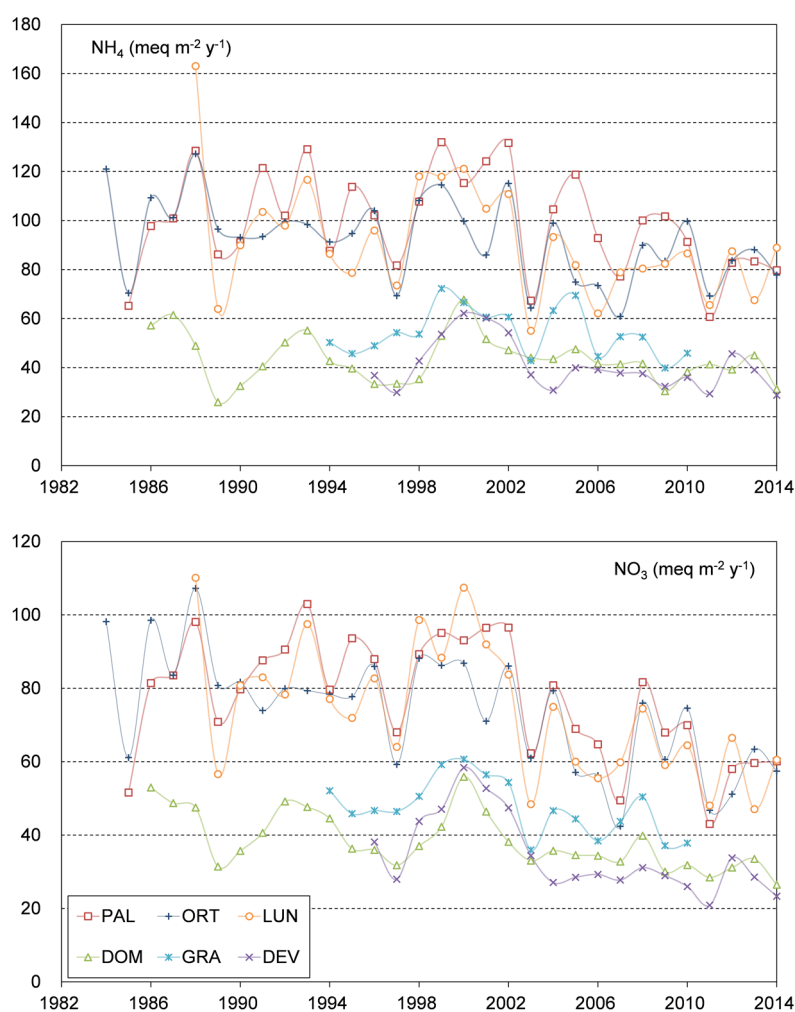


Figure 7.2 – Trends of  $\text{NO}_3$  and  $\text{NH}_4$  annual deposition at the atmospheric deposition sampling sites in Lake Maggiore watershed, North-Western Italy. For site location, see Fig. 7.1.

To assess both temporal change and spatial gradients in the **atmospheric deposition of S and N compounds** in the area of Lake Maggiore, maps of average deposition for 5-year periods were produced (Fig. 7.3). A spatial gradient in the deposition of sulphate and nitrogen compounds was evident both in the 1990s and in recent times (2008-2012), with highest values in the south-eastern part of the area, close to the major emission sources, and decreasing values towards the Alps. However, gradients became less and less evident in time: because of the declining concentrations of acidifying compounds observed in the last two decades, which affected in particular the more polluted southern sites. Despite these recent changes, deposition of inorganic N in North Western Italy, as the sum of ammonium and nitrate, is still between 110 and 140 meq m<sup>-2</sup> y<sup>-1</sup> (15-20 kg N ha<sup>-1</sup> y<sup>-1</sup>) and it has been estimated that total N deposition, including the contribution of dry and organic N deposition, may be 30-40% higher (Rogora et al., 2016).

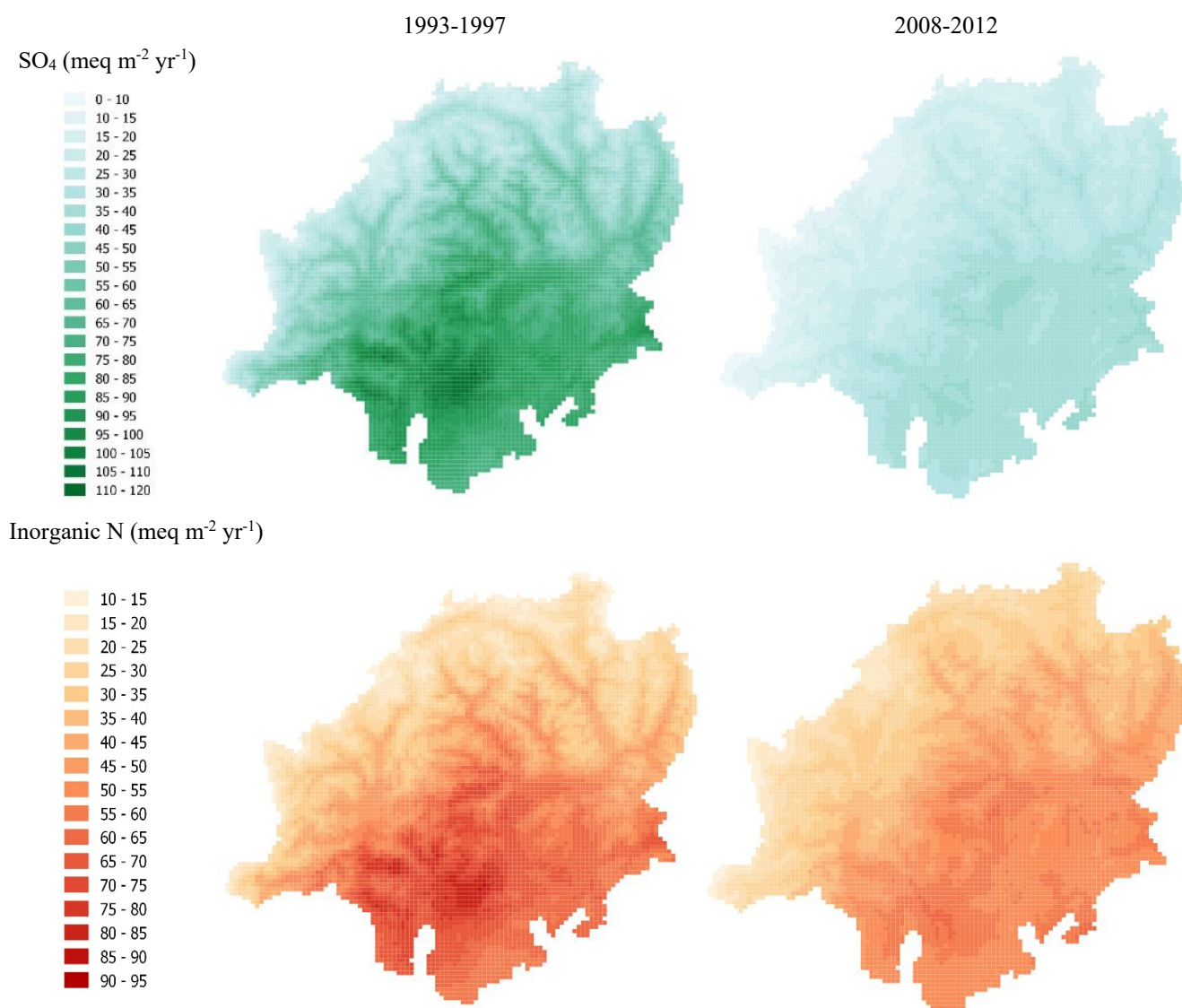


Figure 7.3 – Maps of  $\text{SO}_4$  and  $\text{NO}_3$  deposition over the area of Lake Maggiore watershed in the 1990s compared with the recent period (2008-12). For the location of the sites see Fig. 7.1.

### 7.3 Main impacts of atmospheric pollution on rivers and lakes

The effects of the main pressures on water quality and biota have been assessed at the Italian ICP WATERS sites both through the analysis of long-term data and in-depth studies at specific sites. In the first period since the establishment of the network (1995-2000), the focus has been on acidification and related problems (pH and alkalinity drop, heavy metals dissolution; Mosello et al., 2000; Rogora et al., 2001). Extensive studies were performed by the Programme centre, in cooperation with the National Focal Points, to assess the regional extension of the acidification problem and its relevance at European scale (Stoddard et al., 1999). In Italy, major effects of acid rain were detected at a limited number of sites in the Central Alps, showing significant pH decrease (below 5.6), full depletion of the alkalinity pool and increase of aluminium concentration. Although only few lakes were effectively acidified, most of the lakes in this part of the Alps proved to be sensitive to acidic inputs and potentially threatened, if deposition had remained at the same level (Marchetto et al., 1994). Effects of acidification were evident on both the lake flora and fauna: benthic diatoms assemblage was shifted towards acidophilus species, and zooplankton lost the dominant species, *Arctodiaptomus alpinus*. Palaeolimnological studies outlined that lake acidification paralleled the increasing input of long-range transported industrial pollutants, traced by spherical carbonaceous particles (Guilizzoni et al., 1996; Marchetto et al., 2004).

The Convention on Long Range Transboundary Air Pollution (CLRTAP) entered into force in 1983 and was subsequently extended by eight specific protocols. The Gothenburg Protocol, for instance, for the abatement of acidification, eutrophication and ground-level ozone, was adopted in 1999 and sets emission ceilings for 2010. As an effect of emission reduction, in particular of SO<sub>2</sub>, rain acidity substantially decreased and a chemical recovery of alpine lakes immediately started: for instance, the ICP WATERS site Lakes Paione Inferiore and Superiore showed positive trends of pH and alkalinity since the mid-1990s (Fig. 7.2; Mosello et al., 2000; Marchetto et al., 2004). First signs of biological recovery were identified, such as change in diatom flora and appearance of sensitive species among benthic insects (Marchetto et al., 2004).

Despite this positive response to changing deposition, several critical issues still affect high altitude lakes and sensitive sites in general: a recent study performed on about 40 lakes in Italy (Piedmont) and Switzerland (Canton Ticino) showed that some lakes are still acidic or show a high sensitivity to acidification. This sensitivity is particularly evident at the snowmelt, when alkalinity is still fully depleted in some lakes (Rogora et al., 2013). The study also highlighted the prominent role of N deposition in this area: now, nitrate is the dominant acidifying agent in the studied lakes, due to the high input of nitrogen compounds from atmospheric deposition. As an example, Fig. 7.4 shows the SO<sub>4</sub> to NO<sub>3</sub> ratio in Lake Paione Superiore: as an effect of the decreasing concentration of SO<sub>4</sub> in lake water, NO<sub>3</sub> has become more and more important in time and presently contribute for 40-45% of the sum of acid anions in this lake. Wet deposition of inorganic N (sum of ammonium and nitrate) affecting the ICP WATERS sites has remained fairly constant over a 30-year period and close to 25 kg N ha<sup>-1</sup> y<sup>-1</sup>. These levels of N deposition proved to be among the highest in Europe (Evans et al., 2001), because of the location downwind of the major emission sources (Rogora et al., 2006a). This huge flux of N from the atmosphere caused N saturation of terrestrial catchments and increasing levels of NO<sub>3</sub> in rivers and lakes (Rogora and Mosello 2007; Rogora, 2007). According to pan-European studies, among the few sites in Europe, the Italian sites located in the Lake Maggiore area were showing a significant increase of NO<sub>3</sub> concentrations in the 1980s and 1990s (Skjelkvåle et al., 2005).

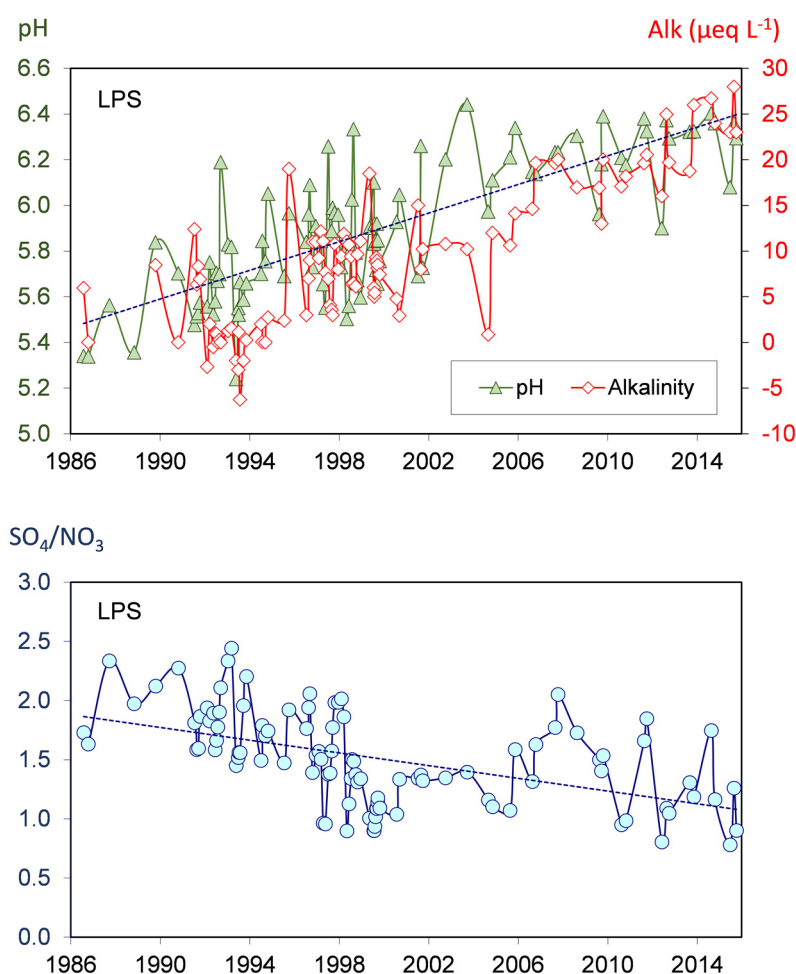


Figure 7.4 - Trends of pH and alkalinity (left panel) and of the  $\text{SO}_4$  to  $\text{NO}_3$  ratio (right panel) in the ICP WATERS site Lake Paione Superiore (IT03).

Furthermore, detailed N budgets performed for the single river catchments showed that the atmospheric input of N cannot be fully retained by soil and vegetation, with N retention varying from 60-70% of the input in the northern part of the area, to 20-30% or even lower in the southern catchments (Rogora et al., 2006b). In a previous analysis of long-term trends in stream water chemistry, the increasing  $\text{NO}_3$  levels and the limited seasonal pattern of  $\text{NO}_3$  were identified as signals of an aggrading level of N saturation in time; according to monthly  $\text{NO}_3$  data, most of the river sites could be classed as being at a medium or high stage of N saturation (Rogora, 2007).

Recently, monitoring data for both rivers and lakes showed a reversal in  $\text{NO}_3$  trends (Rogora et al., 2012), as a response to the decreasing N inputs from the atmosphere observed since 2005-2006. A study performed considering both the ICP WATERS sites and other monitoring sites in the same area, showed how this change was widespread, affecting both high-altitude lakes in the Alps and subalpine lakes and rivers, and occurred at almost the same time at all sites (Fig. 7.5). High altitude lakes show a wide range of concentrations; however, values in the 1980s reached 30-40  $\mu\text{eq L}^{-1}$  in some lakes, while they are mostly below 20  $\mu\text{eq L}^{-1}$  in recent years (Fig. 7.5, right panel). The data for sites subject to a continuous monitoring highlight a coherent pattern, with a dominant positive trend till 2000-2005, followed by a decreasing one (Fig. 7.5, left panel).

The decrease in most recent period was particularly evident for stream sites: for instance, at the ICP WATERS sites Rivers Pellino and Pellesino  $\text{NO}_3$  decreased from 120-140  $\mu\text{eq L}^{-1}$  around 2000 to the present values of about 90  $\mu\text{eq L}^{-1}$  (Fig. 7.3)

Among ICP WATERS lake sites, the watershed of Lake Mergozzo, located in the subalpine part of the area (Fig. 7.1), was probably affected by a state of N saturation:  $\text{NO}_3$  sharply increased in the 1980s and 1990s (Fig. 7.5), then stabilized at around 50  $\mu\text{eq L}^{-1}$  since 2000. The N enrichment of the lake water was ascribed entirely to atmospheric inputs (Rogora et al., 2012). Due to their remote location, lakes Paione Inferiore and Superiore are subject to lower levels of N deposition, and are characterised by distinctly lower  $\text{NO}_3$  concentrations compared to the other sites (Fig. 7.5). Nevertheless, the mean level of  $\text{NO}_3$  in the water of these lakes (20-25  $\mu\text{eq L}^{-1}$ ) is higher than is recorded at other mountain sites in remote areas of the world (Rogora et al., 2008a), so that the Paione lakes and the other mountain lakes in the same area can be considered as possibly impacted by atmospheric N transported from downwind emission sources (Rogora et al., 2012).

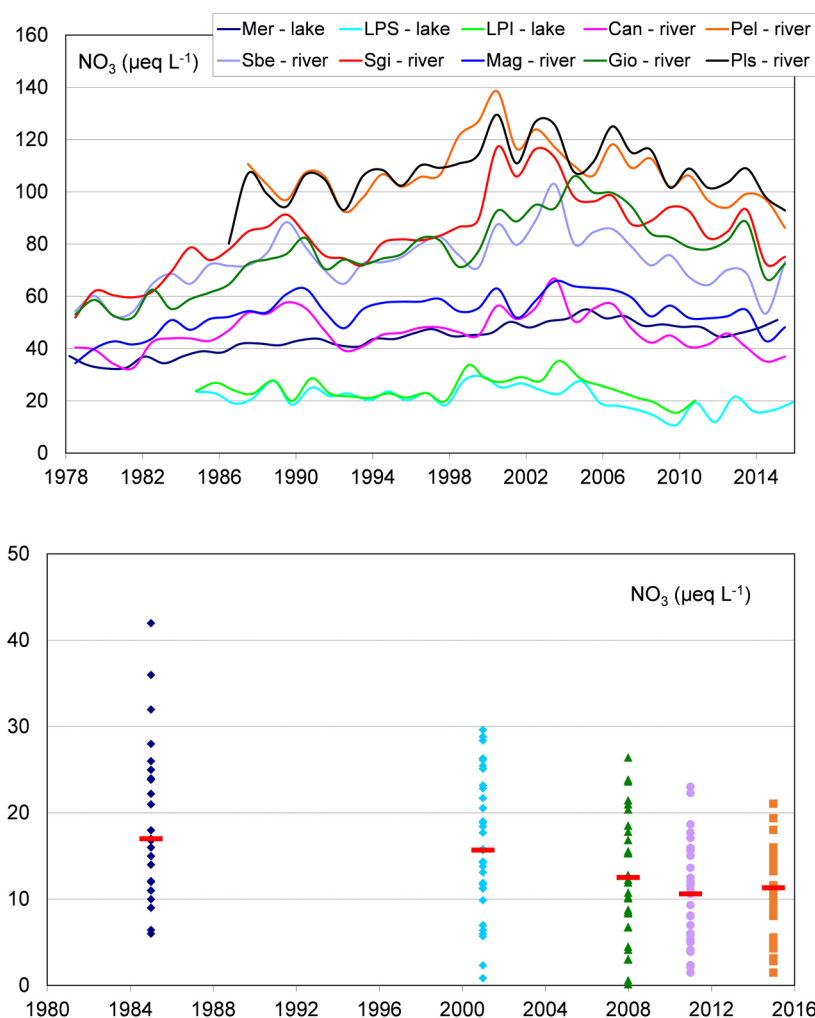


Figure 7.5 – Long -term trends of yearly  $\text{NO}_3$  concentrations (top) at the surface water monitoring sites in the area of Lake Maggiore watershed, including the ICP WATERS sites, and at high altitude lakes in the Central Alps ( $n=30$ ) measured in different periods (bottom). The thick line shows the median value for each survey.

Mer: Lake Mergozzo (IT02); LPS: Lake Paione Superiore (IT03); LPI: Lake Paione Inferiore (IT01); Can: River Cannobino (IT04); Pel: River Pellino (IT05); Pls: River Pellesino (IT06); Sbe: River S. Bernardino; Sgi: River S. Giovanni; Mag: River Maggia; Gio: River Giona.

Besides the similarity of trends, a further feature common to most of the data series was the presence of a high interannual variability in the years between 1999 and 2004; 2002 and 2003 in particular were characterized by peak values of NO<sub>3</sub> at several sites (Fig. 7.5). To better investigate the possible drivers of this short-term variability in water chemistry, climate factors were considered: in particular, the role of temperature and precipitation regime and trends in changing NO<sub>3</sub> concentrations in surface waters was assessed (Rogora, 2007; Rogora and Mosello, 2007). Temperature, especially extreme values, proved to be important in NO<sub>3</sub> export from river catchments: high air temperature and prolonged dry conditions, such as those of the summer of 2003, affected NO<sub>3</sub> dynamics in some of the subalpine rivers in the Lake Maggiore area (Rogora, 2007). These results were explained by the effect of warm periods on temperature-dependent processes such as mineralization and nitrification (Rogora et al., 2008b).

Also, the analysis of other chemical trends (e.g. sulphate, base cations) demonstrated that climatic factors interact with atmospheric deposition affecting the long-term changes in lake water. Some high altitude lakes, for instance, showed an increasing trend of sulphate concentrations, despite the huge decrease of sulphate input from the atmosphere (Rogora et al., 2013). Besides sulphate, also base cations increased in these lakes, with an overall effect on conductivity. An analysis of climate data for the study area puts in evidence how the last two decades (1990-2010) have been characterised by the highest temperature rise and this change mainly affected spring and summer months. Data collected also confirmed a decrease in snowfall as well in the length of the snow cover period (Meteo Svizzera, 2012; Rogora et al., 2003a). A climate related effect can be suggested to explain trends in water chemistry contrasting with those in atmospheric deposition. First of all the presence of less and less snow on the ground and a more exposed portion of the catchments lead to increasing export of weathering products to lake water (Rogora et al., 2003a). In addition, as suggested by some recent studies, the cryosphere, and particularly permafrost degradation processes, may drive lake chemical changes (Thies et al., 2007). For this reason, besides the discrete monitoring of lake water, high frequency measurements of some basic parameters (lake water temperature, conductivity) have been started in the last few years at selected study sites.

#### **7.4 Deposition scenarios and risk assessment**

To assess the impacts of different deposition scenarios on lake and river water chemistry, dynamic modelling has been performed at Italian ICP WATERS sites (Rogora, 2004; Rogora et al., 2003b). Some cooperative studies were also performed at a European level to assess and predict the extent of acidification and recovery, also in relation to confounding factors (Wright et al., 2006; Helliwell et al., 2014). The model used, named MAGIC (Model of Acidification of Groundwater In Catchments), is a process-oriented, intermediate-complexity dynamic model which has been in use for more than 20 years and extensively tested on catchments in Europe and North America for the long term reconstruction and future prediction of soil and surface water acidification at the catchment scale (Cosby et al., 2001). The outputs clearly demonstrated the benefits of achieving the emission reductions in both S and N compounds agreed under the Gothenburg Protocol. It was also clear that, besides the substantial reduction of SO<sub>4</sub> deposition from the peak levels of the 80s, N deposition too had to be reduced to protect freshwaters from further acidification (Rogora et al., 2003b). The results also showed how including other factors specific to the Mediterranean area,

such as dust deposition and climate change, may improve the fit of experimental data and the reliability of the model forecast.

For this reason, a further modelling exercise was performed, in cooperation with researchers from several European countries, adopting a common protocol at 14 intensively-studied sites in Europe and Eastern North America, including Italian sites (Wright et al., 2006). The results suggested that modelling of recovery from acidification should take into account possible concurrent climate changes and, especially in the case of the Italian sites, on the climate-induced changes in nitrogen retention and solute export from the catchments.

A further assessment of the efficacy of deposition reduction on water quality at sensitive sites was performed within the ICP WATERS in the so-called “ex-post analysis” (Wright et al., 2011). The already available MAGIC calibration was used to simulate the evolution of lake chemistry at the site Lake Paione Superiore in response to the deposition scenarios issued by the Coordination Centre for Effects (CCE) in 2010. In particular, projections were made under the scenario of national emission estimates (NAT) and emissions with maximum feasible reductions (MFR) (Fig. 7.6). Under both scenarios, a sharp increase of pH and of the acid neutralising capacity (ANC) of lake water was predicted and an evident improvement of lake water chemical status should be expected. This can be mainly ascribed to the decreasing concentrations of  $\text{SO}_4$ , which will probably decrease further in the next few years and then stabilise (Fig. 7.6).

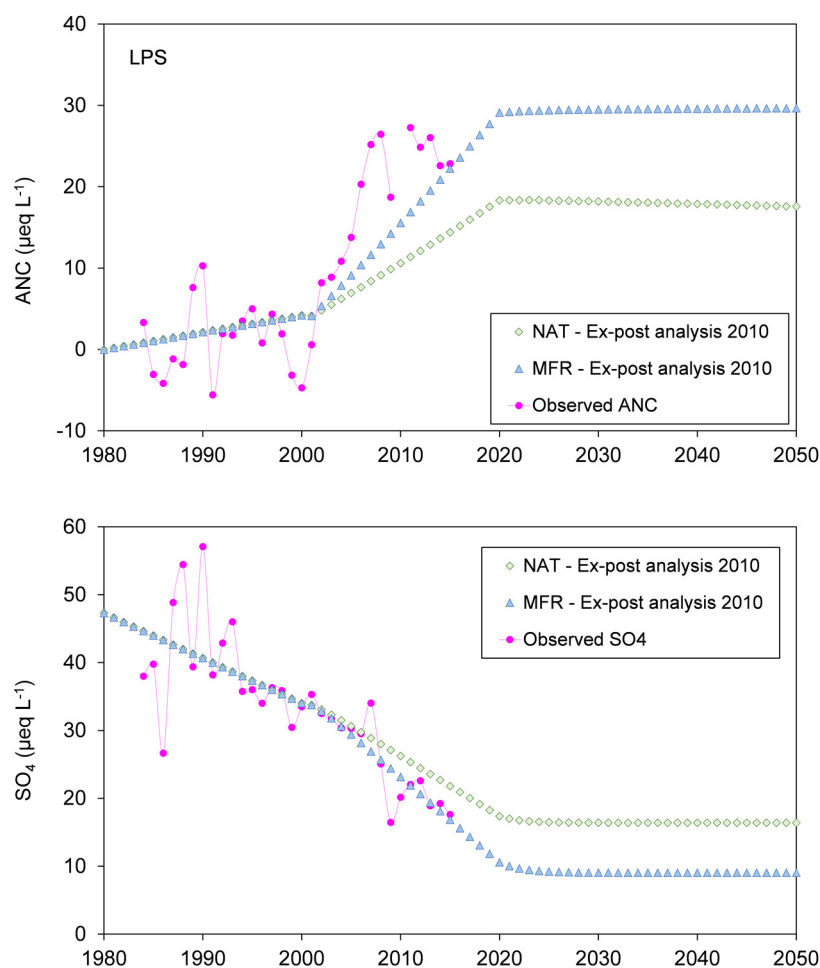


Figure 7.6 – Resulting projections for ANC (top) and  $\text{SO}_4$  (bottom) in Lake Paione Superiore (ICP WATERS site IT03) using the ex-post scenarios provided by CCE. The observed values measured in lake water (autumn samplings) are also shown. NAT: national emission estimates (NAT); MFR: maximum feasible reductions.

However, reduction of N emission in source regions, located in the lowlands, will be crucial in the next few years for the recovery of these types of lakes from acidification and for their nitrogen status as a whole.

Alkalinity, or alternatively ANC (Acid Neutralising Capacity), has been widely used as an index of the acidification status, and of acid sensitivity of surface waters. In Europe, the value of  $20 \mu\text{eq L}^{-1}$  has been identified as the minimum level required for ecosystem protection under UNECE protocols. However, proposed critical limits vary depending on the target group of organisms. Furthermore, even when considering the same target (e.g. macroinvertebrates), critical limits may vary, depending on the typical fauna of sensitive species and their adaptations to native water chemistry. In the high Alps for instance an ANC limit of  $30 \mu\text{eq L}^{-1}$  has been suggested (Raddum and Skjelkvåle, 2001). Some of the monitored lakes in the Alps still show values of ANC below this limit, especially at snowmelt (Rogora et al., 2013). However, it must be also pointed out that some of these lakes are characterised by a very limited “natural” alkalinity pool, due to the lithological composition of their catchments. As a consequence, the alkalinity or ANC critical limits identified at European and international level will be hardly achieved in these sensitive lakes, even under the most optimistic deposition scenarios. Furthermore, when thinking to biological recovery, it should be considered that benthic species in high altitude lakes, which are frequently used as biological indicators, in the future will be probably more affected by changing physical conditions, due to the effects of meteorology and climate, than by the chemistry of lake waters.

In the framework of the UNECE CLRTAP, empirical critical loads of nitrogen (CLN) have been defined for natural and semi-natural ecosystems (Bobbink and Hettelingh, 2011). For instance, for permanent oligotrophic lakes, ponds and pools, a CLN of  $3\text{--}10 \text{ kg N ha}^{-1} \text{ y}^{-1}$  has been identified. At present, although deposition of acidity at ICP WATERS sites in Italy mostly falls below the critical limits, the atmospheric deposition of nitrogen remains still too high with respect to critical levels and further reductions are needed to prevent nitrogen saturation of terrestrial and aquatic ecosystems.

## 7.5 Conclusive remarks

Overall the results of long-term studies at ICP WATERS sites in Italy emphasise the benefits of achieving emission reduction targets. Deposition clearly responded to emission decrease, which has been particularly evident for  $\text{SO}_2$ . Surface water response to changing deposition was widespread but somewhat delayed, due to the interacting effect of several factors, such as N saturation of soils in the catchments and climate change.

Despite the current tendency toward recovery, atmospheric deposition and other global changes will probably keep affecting freshwater quality in the future, especially in the alpine area. The recovery patterns, both from acidification and from N saturation, will be more and more influenced by climatic factors, such as temperature and precipitation, also through indirect effects (snow cover change, retrieving glaciers, permafrost degradation).

Nitrogen deposition, both as oxidised and reduced nitrogen, will continue to have a prominent role in the acidification processes and in the nitrogen status of surface water. From this perspective, further reductions in the emissions of N compounds should be the target of national and international policy.

# Are technical materials and cultural heritage exposed to air pollution risk?

## The contribution of Italy to ICP Materials



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## CHAPTER 8 - ARE TECHNICAL MATERIALS AND CULTURAL HERITAGE EXPOSED TO AIR POLLUTION RISK? THE CONTRIBUTION OF ITALY TO ICP MATERIALS

### 8.1 Introduction

Air pollutants in combination with climatic parameters are key factors in the corrosion and deterioration of several metallic and non-metallic materials. This reduces the operating life of technical materials and threatens objects of cultural heritage, an important component of our individual and collective identity. The impact of pollutants emitted into the atmosphere on materials is cumulative and irreversible because, unlike natural ecosystems, materials have no possibility of self-regeneration. This impact causes massive economic losses for protective measures, substitution of degraded materials, cleaning, maintenance, and restoration work on buildings and historical and cultural monuments exposed outdoors.

The damage to the materials exposed to the atmosphere is the result of complex interactions between chemical, physical and biological parameters. The main forms of degradation caused by atmospheric pollutants on materials and in particular on built cultural heritage are corrosion (loss of material due to chemical attack) and soiling (due to the deposition of particulates on the surfaces). Since materials degradation also occurs in the absence of pollutants, it is important to quantify how much air pollution due to human activities influences and accelerates the background (largely unaffected by human activities) degradation of the materials.

Sulphur dioxide ( $\text{SO}_2$ ) is the main pollutant responsible for the corrosion of materials, but also nitrogen oxides ( $\text{NO}_x$ ), ozone ( $\text{O}_3$ ), particulate matter, carbon dioxide ( $\text{CO}_2$ ), and sea salt from sea spray play an important role. Wet and dry deposition both contribute to the deterioration of materials. Generally, sulphur and nitrogen oxides are oxidised in the atmosphere to sulphuric acid ( $\text{H}_2\text{SO}_4$ ) and nitric acid ( $\text{HNO}_3$ ) that exert their corrosive action on materials by lowering the pH of the precipitation (acid rain).  $\text{SO}_2$  is also the main cause of the sulphation process of stone and bronze surfaces. The deposition of airborne particles on surfaces of buildings and historic monuments causes soiling and accelerates chemical degradation of the materials, thus impacting on both the aesthetic appeal and the decay of such structures.  $\text{O}_3$  has an indirect role as it oxidise sulphur oxides and nitrogen oxides to  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$  and exerts a direct role in the degradation of metals such as copper and copper alloys and in the oxidation of polymeric materials. Chloride content in sea salt is an effective corrosive agent.

$\text{CO}_2$  is generally not considered a pollutant itself, but a climate-altering gas. However,  $\text{CO}_2$  from the air dissolves in rainwater making it slightly acidic (carbonic acid,  $\text{H}_2\text{CO}_3$ ). The rainwater may react with materials that are largely made from calcium carbonates ( $\text{CaCO}_3$ ) – limestone being a common example - transforming the calcium carbonate, slightly soluble, into the more soluble calcium bicarbonate ( $\text{Ca}(\text{HCO}_3)_2$ ). This is washed away causing the material to be weathered (karst effect).

Calcareous stones such as limestone and marble, which are used in most heritage buildings, are the most vulnerable to degradation. Natural degradation factors such as changes in climate and micro-climate around the surfaces, freeze-thaw phenomena, salt crystallization, etc., add to human factors, mainly represented by air pollution, leading to several forms of deterioration, including the build-up of the so-called “black crusts”, heterogeneous deposits consisting of gypsum (calcium sulphate,  $\text{CaSO}_4$ ), calcite resulting from dissolution and subsequent re-precipitation of calcium carbonate, carbonaceous particles and other components.

In addition, air pollutants deposited on materials could enrich it with nutrients, thus favouring the biological colonization (bacteria, fungi, algae, lichens and plants), a further damage factor of the surfaces of architectural works.

## 8.2 Effects on materials

The effects of air pollutants as well as climate parameters on the atmospheric corrosion and soiling of various materials, including materials used in objects of cultural heritage, are investigated by the International Co-operative Programme on Effects on Materials, including Historic and Cultural Monuments (ICP Materials <http://www.corr-institute.se/icp-materials>).

ICP Materials is lead by Sweden, which provides the programme with the Main Research Centre, Swerea KIMAB AB. Since 2005 the chairmanship is shared by Sweden and Italy (ENEA), which together are responsible for the co-ordination and organisation of the programme. A Task Force consisting of representatives from all countries participating in ICP Materials is responsible for the implementation of the programme.

Italy through ENEA is responsible for the sub-centre for stock of materials at risk and cultural heritage which includes use of results including mapping, stock at risk and economic evaluations aimed especially at objects of cultural heritage. Czech Republic, France, Spain, Sweden, Switzerland, and United Kingdom are providing the programme with materials sub-centres. Each sub-centre is responsible for a material or group of materials and prepare, distribute and evaluate corrosion effects after exposure on samples of materials, regardless of where they were exposed. Norway is providing the programme with the environmental sub-centre, which maintains the environmental database and evaluates trends of the environmental data.

The exposure of materials is performed in a network of test sites, located in countries that are Parties of the Convention, covering different climatic conditions and different levels of air pollution. The measurements include a wide range of pollutants, precipitation and climate parameters. A wide range of materials has been selected and exposed that are representative both for technical materials and materials used in objects of cultural heritage. Currently, the ICP Materials network consists of 26 monitoring and exposure stations located in 18 different countries (Figure 8.1).



Figure 8.1 – Map of test sites of the monitoring programme of ICP Materials (left) and test site in Venice with material samples on the rack (right).

Italy participates in the programme since its inception with four sites: Rome (Istituto Superiore di Sanità), Rome-Casaccia (ENEA), Milan (ARPA Lombardia), and Venice (ARPA Veneto).

Figure 8.2 shows the change in the yearly corrosion rates of carbon steel, weathering steel, traditional zinc, blasted zinc, copper, and limestone and the change in soiling of modern glass samples observed at the four Italian stations. Overall, the effects observed in Italy resemble the trend observed by the ICP Materials network (Tidblad et al, 2014; Tidblad et al., 2016) and reflect the decline in emissions of SO<sub>2</sub> and NO<sub>x</sub>. Although there are considerable fluctuations between the measurement campaigns, also attributable to fluctuations in the climatic parameters, corrosion rates of carbon steel declined considerably over the investigated period. Based on only two exposures (1987 and 2011) also the corrosion rate of weathering steel (a low-alloyed steel with increased resistance to atmospheric corrosion) shows similar results after one year of exposure. Corrosion rate of traditionally grinded zinc also declined significantly until the mid-90s, but the trend has diminished in the following years (up to 2000). Glass blasted zinc, investigated from 1997 onwards, shows no clear downward trend (the corrosion of blasted zinc is generally higher because of the rougher surface given by this treatment). Copper samples were exposed during five exposure periods. In general, a significant decrease in the corrosion rate between 1987 and 1997 can be observed while in the following years the improvement is less evident. For limestone, declining trends could be observed initially, particularly in urban areas where the reduction of SO<sub>2</sub> emissions was greater, after that no recognisable downward trend could be observed. Evaluation of soiling of modern glass has been investigated by ICP Materials since 2005. In general, and in analogy with all the sites of the ICP Materials network, haze does not show a clear trend.

Although the degradation of materials in Europe is today significantly reduced, mainly due to reduction of sulphur pollution, the current rates of corrosion and soiling of materials are overall still unacceptably high. Air pollution is still a problem, but more complicated by the superimposition of the effects of a multitude of harmful air pollutants with those of climate parameters such as temperature, relative humidity and amount of rainfall. Effective strategies for the reduction of the effects of air pollutants on materials need to go hand in hand with strategies to cope with climate change.

The research programmes undertaken by ICP Materials lead to the derivation of statistically reliable dose-response functions linking the corrosion or deterioration rate of several materials to the levels of pollutants in combination with climatic parameters. Two sets of dose-response functions have been derived: functions for the SO<sub>2</sub> dominating scenario (Tidblad et al., 1998) and functions for the multi-pollutant scenario (UNECE, 2015). These dose-response functions can be used to calculate the degree of material corrosion or soiling starting from ambient data, and then identify those areas where the risk of damage is greater.

Threshold values of degradation rates, below which impacts are “acceptable” (for materials used in technical constructions) or “tolerable” (for materials used in objects of cultural heritage), have been defined, to which correspond acceptable/tolerable levels, i.e. those concentrations or loads of pollutants that do not lead to unacceptable/intolerable increase in the rate of corrosion or deterioration. Acceptable/tolerable corrosion rates are expressed as a multiple of the background corrosion rate. Such calculations can be used to produce maps showing increased risk of corrosion and soiling at many different scales, from a continent to individual cities.

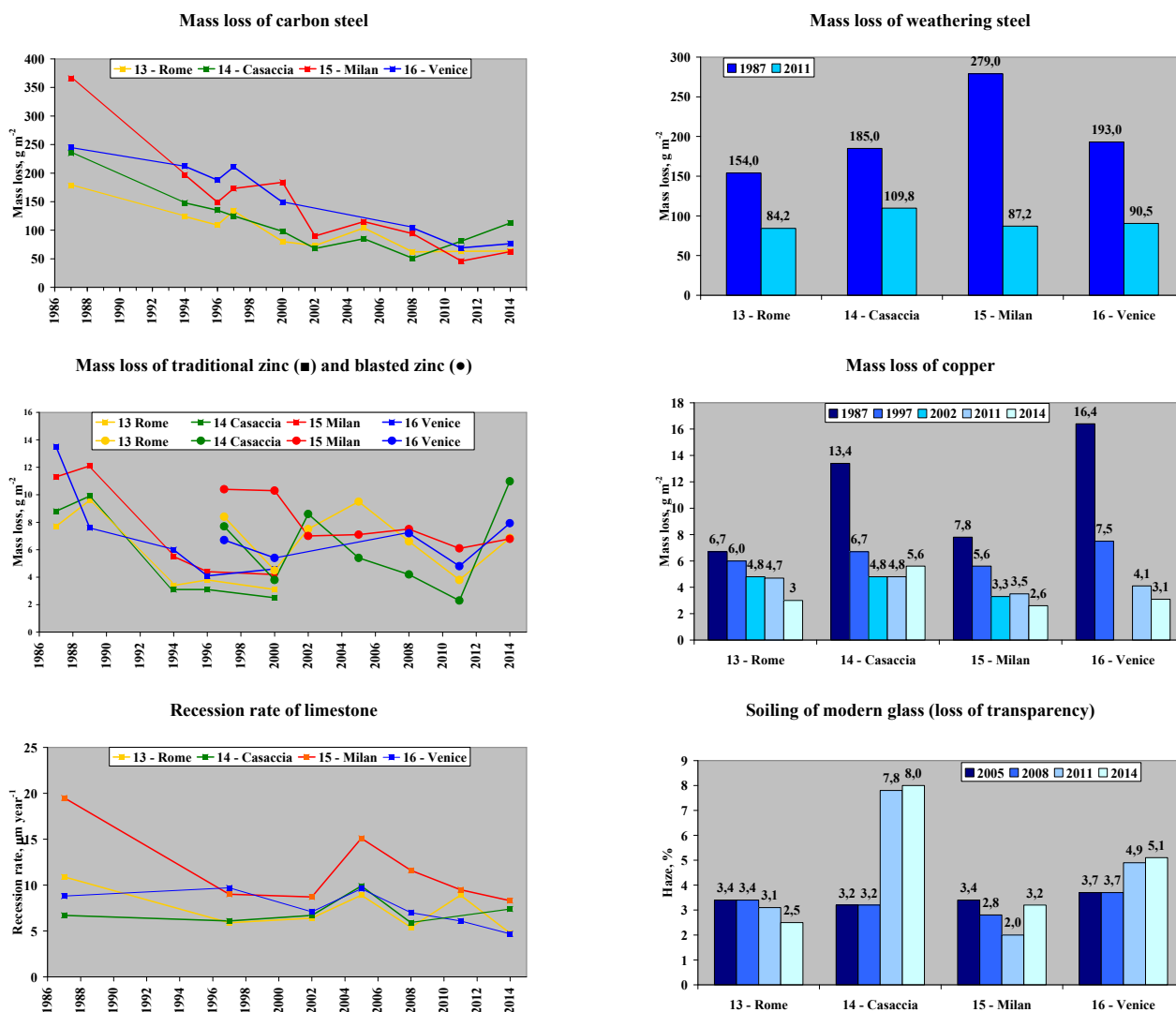


Figure 8.2 – Change in the yearly corrosion rates of carbon steel, weathering steel, traditional zinc, blasted zinc, copper, and limestone and the change in soiling of modern glass samples observed at the four Italian stations.

### 8.3 Effects on historic and cultural monuments and buildings.

The ultimate goal of the ICP Materials program is the calculation of cost of damage caused by deterioration of materials and attributable to atmospheric pollution. It is possible to estimate the difference in cost between two alternative scenarios, one representing the current (or future) air pollution situation and another that quantifies the damage in a “background” scenario. The difference in the deterioration rate can be then used to estimate the cost associated with the air pollution. Performing inventories of stock of materials at risk is one important pieces of information that is required for a cost calculation.

The main results from the Italian sub-centre in this area are the case studies in Italy (Doytchinov et al., 2009; Doytchinov et al., 2010) and the participation in a review of available data on stock of materials at risk (Tidblad et al., 2010a) and economic evaluations (Tidblad et al., 2010b). The Italian sub-centre has also co-authored the chapter on stock at risk studies (Watt et al., 2009) in a book on effects of air pollution on cultural heritage, which was the final product of the European CULT-STRAT project and included significant parts from the ICP Materials programme.

In recent years, the Italian sub-centre has conducted a “Pilot study on inventory and conditions of stock of materials at risk at five UNESCO cultural heritage sites”. The study is presented in four individual ICP Materials reports during the period 2011–2015: Part I: Methodology (Doytchinov et al., 2011); Part II: Determination of stock of materials at risk for individual monuments (Doytchinov et al., 2012); Part III: Economic evaluation (Doytchinov et al., 2014); and Part IV: The relationship between the environment and the artefact (Spezzano et al., 2015).

Five important UNESCO World Cultural Heritage Site in Europe were studied: Greece, Athens, Acropolis, (The Parthenon); France, Paris, The Facades in the Centre of City; Czech Republic, Prague, The National Library; Germany, Berlin, The New Museum; and UK, Bath, Royal Crescent. The study included the evaluation of the dimensions of the monuments and the nature and amount of any material used for its realization by means of field inspection and examination of images, photos and other documents available in literature and on the internet. As the dominating material of the studied monuments is limestone/marble, the multi-pollutant dose-response function for limestone was applied to determine the corrosion and soiling of the materials used in the construction of the monuments.

The main conclusions from the study are as follows: being located in the heart of European capitals, the studied UNESCO sites are impacted by air pollution, mainly due to  $\text{HNO}_3$  (a product of  $\text{NO}_2$  oxidation) and  $\text{PM}_{10}$ , two pollutants that currently seem to play a prominent role in determining damage of limestone. The improvement of air quality between 2000 and 2010, mainly attributable to a significant reduction of air concentration of  $\text{SO}_2$ , produced a small decrease in the recession rate for limestone, first year exposure, which for the studied sites was quantified in about 5-8 per cent. Calculated recession rates after one year of exposure are above the background corrosion rate ( $3.2 \mu\text{m year}^{-1}$ ) and generally close to the target for the year 2050 ( $6.4 \mu\text{m year}^{-1}$ ) or even at one case close to the target for 2020 ( $8.0 \mu\text{m year}^{-1}$ ). Corrosion due to air pollution would result in material deterioration costs ranging from €9.2 per square metre per year ( $\text{m}^{-2} \text{year}^{-1}$ ) to €43.8  $\text{m}^{-2} \text{year}^{-1}$ , depending on the pollution level and the climatic conditions. These costs add to the cost in background areas, estimated from €14  $\text{m}^{-2} \text{year}^{-1}$  to €28  $\text{m}^{-2} \text{year}^{-1}$ .

The predicted loss of reflectance after five years of exposure to the surrounding environment was still unacceptably high for the studied monuments. Predicted soiling rate of limestone indicate that a “tolerable soiling before action”, which represents the threshold triggering significant adverse public reaction of what constitutes acceptable soiling and generally set at 35%, will be reached within 4-7 years after any restoration work. For cultural heritage objects a period of 10-15 years is considered to be appropriate.

In continuation of the pilot study, ICP Materials has launched a Call for Data on “Inventory and condition of stock of materials at UNESCO World Cultural Heritage Site”. The official letter of the Call for Data, a template for submission of data, an explanatory note with instructions on the use of the reporting template, and a brochure exemplifying the step by step approach for the previously assessed UNESCO sites were provided by the Call. These documents have been also made available for downloading on the ICP Materials website.

Main objective of the Call for Data is to invite Parties to participate in studies evaluating material deterioration due to air pollution at UNESCO World Cultural Heritage Site. The ultimate objective is to provide policy makers the evidence of the effects of air pollution not on a generic material or a generic artefact but on easily recognizable symbols of our culture and history.

This Call for Data requires qualitative and quantitative data on both the historic/cultural monument and on the environment. In view of the complexity of the call, the deadline for submission of the data is set to July 2017. Six Parties to the Convention have announced their intention to participate in the Call: Croatia, Germany, Italy, Norway, Sweden, and Switzerland.

#### **8.4 Concluding remarks**

Air pollution is a major environmental problem and has an impact not only on human health but also on the surface of materials and particularly on the surfaces of buildings and cultural monuments exposed outdoors. Interactions between air pollutants and materials lead to an early deterioration and soiling of buildings and monuments. This prompts more frequent maintenance and restoration activities, which is a cost, and which adversely affect the artefact. Most of the cultural heritage in Europe, and Italy in particular, are located in the heart of cities, where higher levels of pollutants are usually found.

Although decreasing trends in corrosion were observed for all materials in connection to the decrease of concentrations of air pollutants (mainly SO<sub>2</sub>), current corrosion rates and soiling are still unacceptably high. The effects observed in Italy, which resemble the trend observed by the ICP Materials network, show that for some material (i.e. carbon steel and copper) a decreasing trend is still evident while for zinc and limestone no decrease can be detected in recent years. A trend can hardly be identified for the soiling of modern glass.

Currently, HNO<sub>3</sub> (a product of NO<sub>2</sub> oxidation) and PM<sub>10</sub> seem to play a prominent role in determining damage of limestone. In developing future policies on air quality, it is important to specifically address impacts on cultural heritage and the built environment. Air pollutants and climate act together in determining corrosion and degradation of materials. Further reduction of air pollutants is one way of compensating for increased risk of corrosion due to climate change.

In general, the damage caused by air pollution to historic and cultural monuments and buildings should be better studied and understood, including the assessment of the cost associated with the damage and the interaction of pollution and global climate change.

#### **8.5 Acknowledgments**

We would like to express our appreciation to Elena Dell'Andrea (Agenzia Regionale per la Prevenzione e Protezione Ambientale del Veneto), Marcello Ferdinandi (Istituto Superiore di Sanità) and Matteo Lazzarini (Agenzia Regionale per la Protezione dell'Ambiente della Lombardia) for their continuous support and assistance in the management of test sites for exposure of materials and measurement of environmental parameters.

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# Vegetation and urban air quality: recent findings



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## CHAPTER 9 - VEGETATION AND URBAN AIR QUALITY: RECENT FINDINGS

As urbanization increases, also the significance of urban forests (UFs; including individual trees, parks and forests) for improving the environmental quality of life in the cities is increasing (Roy et al., 2012). Among the many benefits that UF provide to people, a top relevance is up to air quality improvement, expected to improve human health by removing gaseous air pollutants and particulate matter (PM) from the air (Weber, 2013). Urban air pollution is a major threat to citizens' health (Pascal et al., 2013). A plethora of primary pollutants, e.g. nitrogen oxides (NO<sub>x</sub>), sulphur dioxide (SO<sub>2</sub>), PM, are directly emitted by combustion in industrial processes and vehicles. Secondary pollutants, e.g. ozone (O<sub>3</sub>) and secondary organic aerosol (SOA), are formed by reactions in the atmosphere among precursors like NO<sub>x</sub> and VOC. Among the major air pollutants, PM and O<sub>3</sub> have the largest impact on human health at present (Pascal et al., 2013). UFs act as sink for pollutants and have been argued to phytoremediate the air (Nowak, 2006; Manes et al., 2012). Areas with high urban forest density, in fact, have lower PM than other sites (Irga et al., 2015). Such filtering capacity usually translates into little percent improvements of air quality, but such little values translate into significant savings in terms of human health (Nowak et al., 2013; 2015).

Pollution removal by plants occurs through a combination of two pathways, including deposition to plant surfaces and/or stomatal uptake (Grote et al., 2016). Dry and wet deposition includes scavenging of pollutants by the foliage or bark and, in the cases of reactive air pollutants such as ozone, also in the gas-phase due to emitted reactive substances. Removal depends on air pollution concentrations, meteorological conditions, and resistance in the crown space, through the boundary layer adjacent to surfaces, and uptake of gases through stomata, i.e. the tiny pores on leaf surfaces. These resistances are controlled by vegetation properties at different scales: community (e.g. single trees, green corridors, parks, and forests), canopy (e.g. crown size, shape and density) and foliage (e.g. leaf shape, surface properties and physiology). Larger tree crowns have a higher potential of ameliorating air quality by maximising pollutant deposition (Paoletti et al., 2004), and thus the characteristics of the tree cover - in particular density and continuity of crowns - and of individual tree crowns - size, architecture, and the leaf area per unit ground surface area (*LAI*) - are important drivers for air quality improvement. Also leaf surface characteristics, e.g. cuticular morphology, leaf wettability and hairiness, affect particulate deposition (Kardel et al., 2012). Another important leaf trait affecting the air quality is the persistence of foliage throughout the year (evergreen species) or only during the growing season (deciduous species). As particulate pollution is typically higher in winter (Sieghardt et al., 2005), evergreen species should be recommended for maximizing the deposition of particles (Manes et al., 2014; Fares et al., 2016). In contrast, gaseous pollutants and in particular O<sub>3</sub> are higher during the growing season (Paoletti, 2006; 2009), thus deciduous species are better suited for filtering gaseous pollution.

The capacity to remove pollutants may largely diverge under stress conditions. In a study carried out in the city of Rome, Fusaro et al. (2015) have shown that urban and peri-urban forests were affected by different environmental stressors and forest management practices. Such factors exert a strong effect on the functionality of the two forest sites, thus affecting their ozone removal capacity and the resulting air quality improvement. Understanding these effects is therefore an essential step for a reliable quantification of the air quality amelioration provided by vegetation in metropolitan areas, and for a better management of the Green Infrastructure (*sensu* Tzoulas et al., 2007) of the

city. At this regard, also species selection for air pollution mitigation should consider the ability of tree species to adapt to local conditions.

Non-stomatal removal processes also include chemical deposition resulting from gas-phase reactions between pollutants (mostly O<sub>3</sub> and NO<sub>x</sub>) and biogenic VOC (BVOC) emitted from the ecosystem (e.g. plants or soil) (Fares et al., 2010). Deciduous plants are typically high BVOC emitters, with highest emissions occurring during spring and summer at midday (Holzinger et al., 2006). BVOCs contribute to the formation of important air pollutants e.g. ozone, secondary organic aerosols and PM (Calfapietra et al., 2013). Adopting low BVOC-emitting species in UF is crucial for controlling air quality (Benjamin and Winer, 1998; Ren et al., 2014). In addition to the species-specific BVOC emission factor, however, also the amount of emitting leaves affects the total production of BVOC by trees. Therefore, larger crowns constitute a negative indicator of air quality in BVOC-emitting species. The main BVOCs are isoprene and monoterpenes.

Evidence is emerging that the presence of roadside trees in street-canyons reduces the upwards transport of air pollutant emissions, increases their storage in the canyon and reduces the penetration of clean air from aloft. As a result, higher pollutant concentrations can be observed at pedestrian level within vegetated canyons (Amorim et al., 2013; Salmond et al., 2013; Vos et al., 2013; Gromke and Blocken, 2015). Further investigation of the complex inter-relations between plant characteristics, microclimate, street configuration and pollutant emission, however, is still needed.

Importantly, trees may also affect air quality by emitting primary particles (pollen) and BVOCs (Churkina et al., 2015). Pollens may act as allergens and are possibly more potent in combination with other urban pollutants (Beck et al., 2013). In Europe, 113 million citizens suffer from allergic rhinitis and 68 million from allergic asthma (EFA, 2011), a number that will likely increase due to climate change (Forsberg et al., 2012). Pollen affects human health by triggering those allergic reactions (Bartra et al., 2007; Traidl-Hoffman et al., 2003). Pollen deposition on leaf surfaces, however, helps abating pollen concentration in the air (Dzierzanowski et al., 2011; Terzaghi et al., 2013), likely by mechanisms similar to those regulating particle deposition.

Based on physiological as well as anatomical species-specific traits, Nowak and Crane (1998) developed the iTree model (Formerly, UFORE) to calculate deposition rates of SO<sub>2</sub>, NO<sub>x</sub>, CO, O<sub>3</sub> and PM per leaf area and per tree from climatic and air pollution boundary conditions. This model has been used for several European case studies (e.g. Paoletti, 2009; Paoletti et al., 2011).

By focusing on the species-specific tree properties (i.e. traits) that determine canopy and foliage interaction with major air pollutants, Grote et al. (2016) classified the major tree species of European cities according to their potential of ameliorating air quality (Figure 9.1).

In addition to allergenicity, calculated as pollination duration x intensity x toxicity (Cariñanos et al., 2016), and BVOC emission potential, calculated as lumped isoprene and monoterpene emission potentials under standard conditions (based Karl et al., 2009), Grote et al. (2016) focused also on PM removal efficiency (based on Yang et al., 2015), water use efficiency (WUE, from Wang et al., 2013), and shading capacity calculated as leaf area index x relative leaf abundance throughout the year (based on Tiwary et al., 2016) x crown width/tree height (based on <https://www.horticopia.com/hortpip/index.shtml>). The efficiency of water use makes a plant well adapted to face water stress, which is a common issue for UFs.

Shading is highly appreciated as UF environmental service, because it reduces the urban heat island, i.e. the typical increase of air and surface temperatures that occur in the cities.

The suitability of a tree species for a particular combination of demands is highly case specific. The relative trade-off or synergistic benefit of different traits also depends on the trees immediate surrounding and the importance of the respective ecosystem service. *Aesculus*, for example might be favored for its shading ability but abundance is restricted to sites with good water supply because its WUE is low. *Pinus* species can be favored in Southern Europe, because they not only efficiently remove pollution but are also relatively stress tolerant. In particular, they are drought resistant, a trait less relevant in Northern regions. Stress tolerance may be the first selection criteria in polluted areas even if the gain in ecosystem services is small.

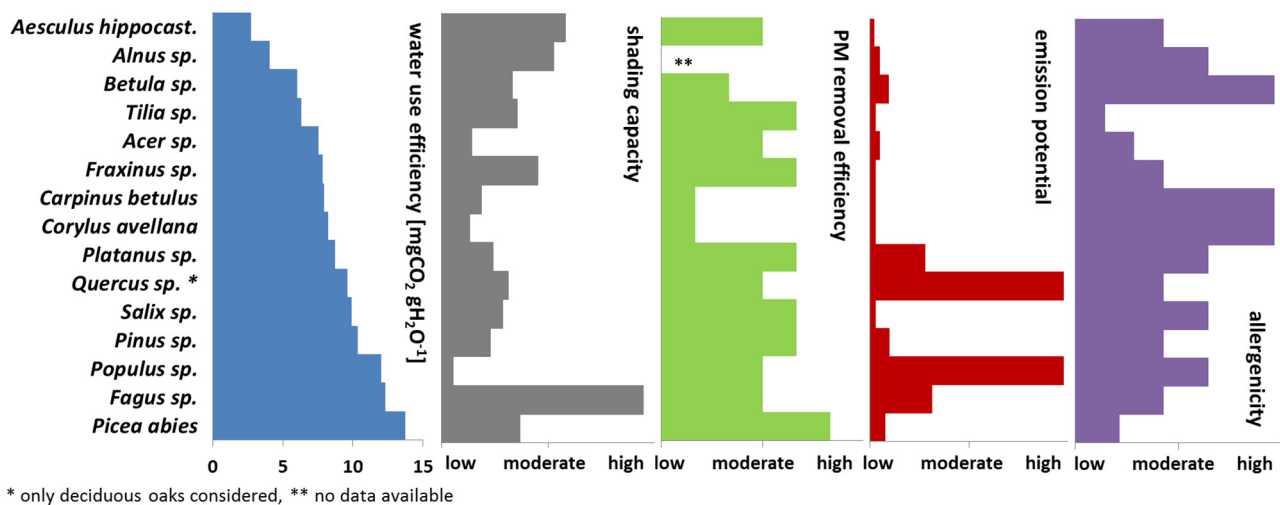


Figure 9.1 – Classification of major tree species in European cities according to their water use efficiency, shading capacity, particulate removal efficiency, isoprene and monoterpene emission potential and allergenicity, following Grote et al. (2016).

In Italy, several studies have recently investigated the ability of urban tree species to remove O<sub>3</sub> and PM from the air. Removal in the city of Florence was estimated per forest type and mapped by Paoletti et al. (2011) and Bottalico et al. (2016).

In the Municipality of Rome, Manes et al. (2012) have quantified the effects of tree diversity on the removal of tropospheric ozone (O<sub>3</sub>) by means of a spatial analysis, integrating system dynamic modeling and GIS. Two years were considered (2003 and 2004), differing in climatic conditions and ozone levels. The results showed that different tree functional groups (evergreen broadleaves, deciduous broadleaves and conifers), have complementary O<sub>3</sub> uptake patterns, related to tree physiology and phenology, thus maintaining a stable community function across different climatic conditions. Based on published unitary costs of externalities and of mortality associated with O<sub>3</sub>, the ecosystem service of O<sub>3</sub> removal of the Rome urban forest has been valued to roughly US\$2 and \$3 million/year, respectively. The removal of PM<sub>10</sub> has been also quantified in the same area and for the same years (Manes et al., 2014), and the Ecosystem Service of PM<sub>10</sub> removal by the three functional groups in the five Sanitary Districts of the Municipality has been mapped. Given the spatial uniformity of PM<sub>10</sub> levels in the urban area, the highest amount of PM<sub>10</sub> deposition rates, during the whole period, were those of the Sanitary District with the largest vegetation cover.

The highest PM<sub>10</sub> depositions for the three functional groups were estimated for the 2004 summer period, in concurrence with the highest mean values of Leaf Area Index. Large urban parks can also significantly contribute to improve air quality locally, i.e at neighborhood level. At this regard, Silli et al. (2015) have estimated the potential PM<sub>10</sub> deposition to vegetation in Villa Ada, a historical park located in the Rome downtown, surrounded by densely built areas and by high-traffic density roads. The results showed that trees may effectively abate suspended particles, with evergreen broadleaved trees being most effective during summer, reducing the average air concentration of PM<sub>10</sub>. During the year 2012, the woody vegetation of Villa Ada removed in total 4417.2 kg of PM<sub>10</sub>.

In a recent study (Marando et al., 2016), the quantification of seasonal PM<sub>10</sub> removal capacity of urban and peri-urban forests has been carried out for the year 2015, considering the whole Metropolitan City (MC) of Rome. This is an administrative unit, which corresponds to the former Province of Rome, and covers an area of 5352 km<sup>2</sup>. Around 22% of this surface is covered by forests, and the overall monetary value of the PM<sub>10</sub> removal service can be estimated to 161.78 million Euros for the year 2015.

At national scale, the PM<sub>10</sub> and O<sub>3</sub> removal from urban and periurban forests has been estimated in ten Metropolitan Cities (Turin, Venice, Milan, Genoa, Bologna, Florence, Rome, Naples, Bari and Reggio Calabria), taking into account the main Physiognomic-Structural Vegetation Categories. The findings remark the importance of the Leaf Area Index in PM<sub>10</sub> removal, and of functional diversity, which is related to stomatal conductance, in the O<sub>3</sub> sequestration process. The overall monetary value of this Ecosystem Service was estimated to be equal to 47 and 297 million USD for O<sub>3</sub> and PM<sub>10</sub>, respectively, in the year 2003 (Manes et al., 2016). This represents, however, a gross estimate, which do not take into account the cost of management and maintenance of the GI.

In a recent study performed in the periurban forest of Castelporziano (Rome), Fares et al. (2016) have measured fluxes of PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> with fast optical sensors and eddy covariance technique, observing that PM<sub>1</sub> is mainly deposited during the central hours of the day, while fluxes of PM<sub>2.5</sub> and PM<sub>10</sub> were negligible. Furthermore, a Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT v4) was applied to simulate the PM emission from traffic in the city of Rome, showing that a significant portion of PM is removed by vegetation in the days when the plume trajectory meets the peri-urban forest.

Guidolotti et al. (2016) implemented the EMEP MSC-W model to scale-down to tree-level O<sub>3</sub>, NO<sub>2</sub> and PM<sub>10</sub> removal in a urban area in Northern Italy, compared the results with the outputs of UFORE (nowadays i-Tree) and found a good agreement with the estimates by this new methodology. In the case of O<sub>3</sub>, pros and cons of laboratory, field, and modeling approaches have been recently discussed and a combination of the three levels of investigation has been recommended as essential for estimating O<sub>3</sub> removal by urban trees (Calfapietra et al., 2016).



# Effects of air pollution on health



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## CHAPTER 10 – EFFECTS OF AIR POLLUTION ON HEALTH

Part of this Chapter was presented at the 8<sup>th</sup> International Congress on Environmental Modelling and Software of the International Environmental Modelling and Software Society (iEMSs) – 10-14 July, 2016, in Toulouse (France).

### 10.1 Introduction

According to the World Health Organization (WHO, 2015), air pollution is the largest environmental risk for human health, with estimates of 600000 premature deaths and 1.6 billion US\$ of economic cost due to mortality and diseases in Europe in 2010 (WHO Regional Office for Europe, 2015). The assessment of the impacts of air quality on health is the endpoint of European and national cost-effective policies, which can be translated into emission reductions and control strategies.

Modelling tools connecting atmospheric dynamics, human response to air pollution and policy options, allow to optimize the use of dedicated resources and data, such as high performance computing and epidemiological cohort studies. In Italy, two recent projects (VIAS and EU LIFE+ MED HISS) estimated the health effects of air pollution on the national scale, following the Health Impact Assessment (HIA) scheme (Figure 10.1), based on existing estimates of the relative risk from previous epidemiological studies, which are applied to the specific exposure of the studied population. The use of a reference national air quality model (MINNI – National Integrated Model to support the international negotiations on atmospheric pollution), combined with monitoring data, allowed the coverage of the whole territory and thus the use of national population data for a comprehensive calculation of exposure and health outcomes.

VIAS (Integrated Assessment of the Impact of Air Pollution on the Environment and Health, 2013-2015; Ancona, 2015) project was funded by the Centre for Disease Control of the Italian Ministry of Health and coordinated by the Department of Epidemiology of the Lazio Region Health Service. VIAS estimated the mortality (from respiratory disease, cardiovascular disease, lung cancer and total) and the months of life lost due to exposure to air pollution. Estimations were carried out both on Italy as a whole and on each of the 20 Italian Administrative Regions. Health effects of PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub> were quantified.

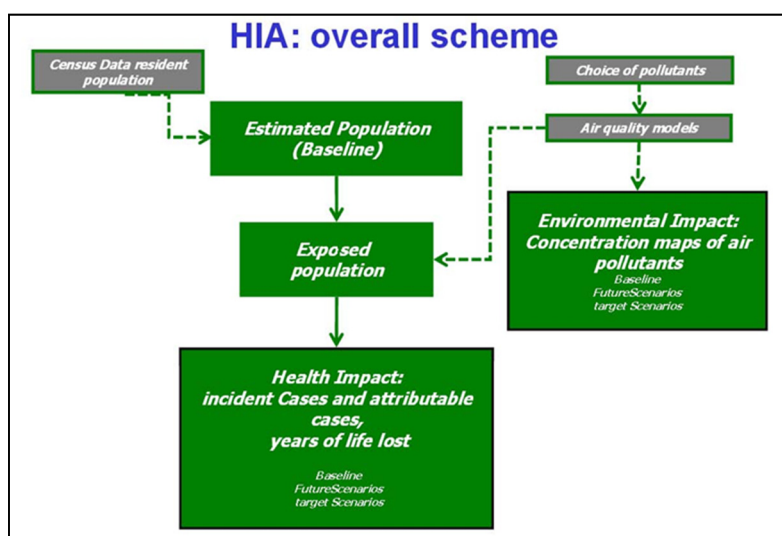


Figure 10.1. Air quality Health Impact Assessment scheme.

## 10.2 Results

To quantify the health gains of Italian population following changes in targets and policies, VIIAS carried out a baseline assessment on years 2005 and 2010. Using model projections on different scenarios (current legislation scenario - CLE, compliance with EU and Italian air quality standards – CLE + Target 1, CLE concentrations reduced by 20% - CLE + Target 2) an estimate of health population for 2020 was produced for future evaluation.

Both the national concentration maps of the analyzed pollutants and the future emission scenarios were provided by the MINNI model system ([www.minni.org](http://www.minni.org)), funded by the Italian Ministry of Environment, which runs an atmospheric modeling system (AMS, Mircea et al., 2014) and an integrated assessment model (GAINS-Italy, D'Elia et al., 2009). The AMS performances have been assessed against measured concentrations in Italy for several simulation years (Mircea et al., 2014, 2016; Ciancarella et al., 2016).

The population data (people aged 30 and over) used for the exposure assessment are related to the year 2005 and were obtained by interpolation of the official national censuses of 2001 and 2011. The concentration of pollutants was provided by the model on a 4x4 km<sup>2</sup> grid, while population data were available on sub-municipal census zones. Therefore, for the calculation of exposure, population data were aggregated on the model grid.

The population exposure was associated with health outcomes by using concentration-response functions (CRFs), which put in relation air pollutants concentrations and health damages. A CRF provides an estimate of the relative risk, i.e. the increment of a health effect associated with a single-unit increment of the ambient concentration of the pollutant. CRFs are site specific, being obtained from observational epidemiological studies. In VIIAS, the WHO guidelines have been applied, using CRFs for mortality and coronary events taken from the WHO HRAPIE review conducted in 2013 (WHO, 2013). Pollutant-specific CRFs on single health outcomes were used. Thresholds of 10 µg/m<sup>3</sup> for PM<sub>2.5</sub>, 20 µg/m<sup>3</sup> for NO<sub>2</sub> and 70 µg/m<sup>3</sup> for O<sub>3</sub> were applied in the assessment, following WHO recommendations on the HIA procedure. The adult population (age 30 years and more) was taken into account following HRAPIE recommendations, as most of the evidence on PM<sub>2.5</sub> long term effects a mortality comes from studies that focused on populations around 30 years of age and above.

VIIAS provided both national aggregated figures and results on 3 geographic areas (North, Center, South and Islands), 20 Member Regions, 2 macro areas (urban, non-urban). A summary of the results at the national level is presented in the following table 10.1.

Total figures in 2005 evidence the relevant impact of PM<sub>2.5</sub> and NO<sub>2</sub> on mortality, ranging from 23000 to 34000 attributable deaths. The average exposure of the population to PM<sub>2.5</sub> is 20.1 µg/m<sup>3</sup>, very close to the EU limit value of 25 µg/m<sup>3</sup> (EC, 2008). Remarkable differences were found among the 20 Member Regions, with Northern Italy showing a higher number of deaths from PM<sub>2.5</sub> (22485) and NO<sub>2</sub> (14008), and between urban and non-urban areas, with deaths attributable to NO<sub>2</sub> being more than doubled (16736 versus 6651) and PM<sub>2.5</sub> deaths only slightly higher in urban areas (19358 versus 15194), confirming the different dispersion pattern and main sources (road traffic and residential heating for NO<sub>2</sub>, secondary aerosol formation for PM<sub>2.5</sub>). The different 2020 scenarios provide a forecast of prevented deaths according to different targets. As an example, attaining the EU limits could save 11000 lives on PM<sub>2.5</sub> and 14000 on NO<sub>2</sub>. The health gains are dependent on the geographical area, as emission reductions are different between Member Regions.

Table 10.1. Average population exposure, attributable deaths for PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub>, and months of life lost for PM<sub>2.5</sub>, in Italy. Results of the VIAS project.

			2005	2010	2020 CLE	2020 CLE - Target1	2020 CLE - Target2
PM <sub>2.5</sub>	general mortality	population exposure (µg/m <sup>3</sup> )	20.1	15.8	18.1	16.2	14.5
		attributable deaths (95% confidence interval)	34552 (20608-43215)	21524	28595	23170	18511
		months of life lost	9.7	5.5	7.7	5.9	4.2
NO <sub>2</sub>	general mortality	population exposure (µg/m <sup>3</sup> )	24.7	17.9	16.6	16.1	13.3
		attributable deaths (95% confidence interval)	23387 (21514-50283)	11993	10117	9021	5247

It is worth noting that the 2020 CLE scenario shows higher exposure and effects for PM<sub>2.5</sub> in 2020 than in 2010, due to an increase in PM<sub>2.5</sub> emissions. This growth is caused by the large spread of wood combustion for residential heating, due both to climate change mitigation policies (encouraging carbon-neutral fuels such as biomasses) and to persistent effects of the economic crisis (leading people to use self-provided wood for heating). This growth overcomes scenario mitigation measures, resulting in an increase of PM<sub>2.5</sub> concentrations and population exposure.

Results were obtained on the MINNI geographical reference, providing national maps of health outcomes, e.g. death rates per 100000 inhabitants from PM<sub>2.5</sub> (Figure 10.2). Large zones in Northern Italy, Tuscany, Rome and Naples show significant death rate values in 2005, with peaks above 250. A decreasing trend is observed between 2005 and 2020, which can be attributed to the growing efficacy of policy mitigation options. Although compliance with the current EU legislation would have a large impact on the health of Italian residents, there is a large scope for further improvement.

Other uncertainties are present in the health outcomes results (synergies and antagonisms between pollutants were not quantified), in the concentration-response functions (conclusions of epidemiological studies conducted in different conditions and populations are extrapolated to Italy) and in the model outputs (coarse resolution for NO<sub>2</sub>, scenario assumptions are realistic but subjected to periodical revisions).

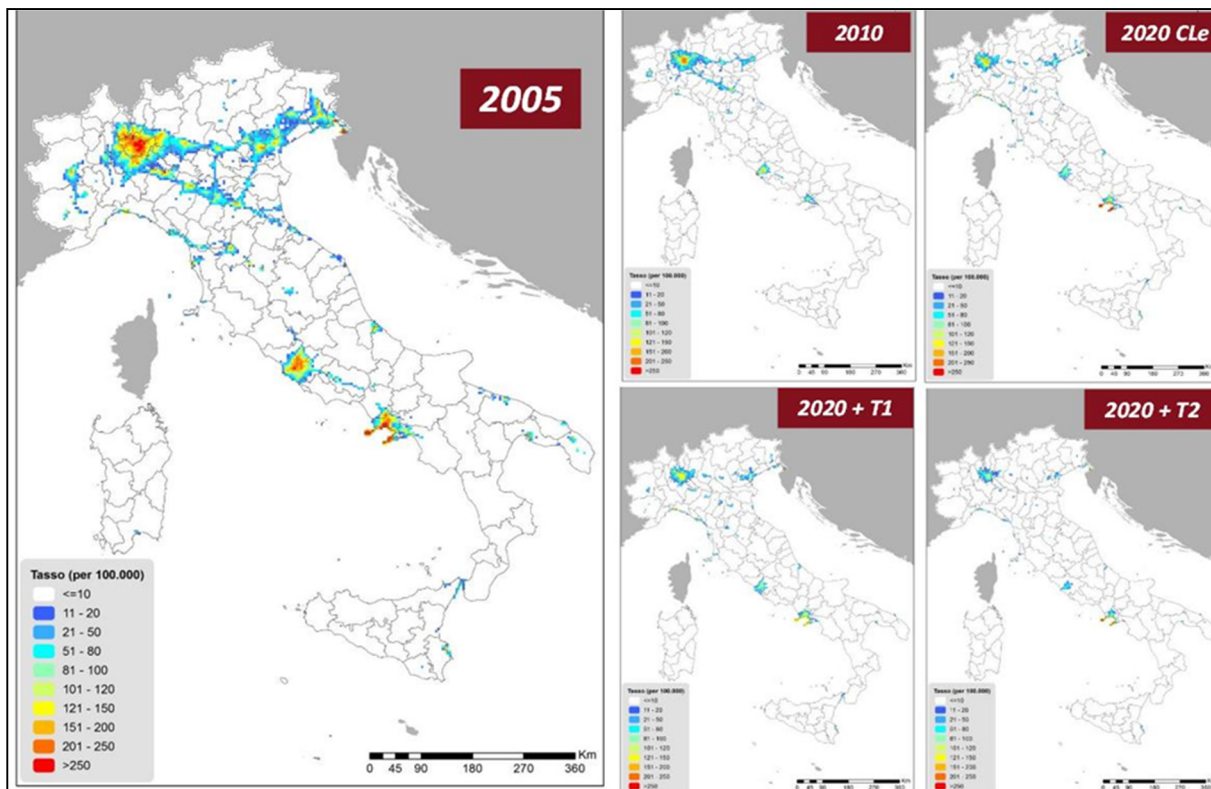


Figure 10.2. Death rates attributable to  $PM_{2.5}$  each 100000 inhabitants in the 5 case studies.  
Results of the VIAS project.

MED HISS (Mediterranean Health Interview Surveys Studies: long term exposure to air pollution and health surveillance) is a recently completed EU LIFE+ Pilot project (2013-2016) involving four countries (Italy, France, Slovenia and Spain). MED HISS is aimed to set up a surveillance system of long term effects of air pollution, based on the common availability of routine air quality and health data. Three main information sources were used: 1) the National Health Interview Surveys (NHIS, Hupkens et al., 1999), available in all countries, representative of the total population and covering both urban and rural areas; 2) mortality and hospital admissions registries; 3) air pollution models.

The use of NHISs data already available, which include individual information on main potential confounders (smoking, BMI, occupation, education, etc.), allows the recruiting of retrospective cohorts, with a remarkable saving of resources, that are normally allocated on building the information on the observed population. Furthermore, NHISs data on entire national populations try to overcome the existing limitations on the health effect of air pollution, historically based on cohort studies from outside EU, or conducted in Europe, but with restrictions on age or location.

Cohorts are being followed-up for mortality and morbidity, and each subject will be assigned a level of exposure to air pollution ( $PM_{10}$ ,  $PM_{2.5}$ ,  $NO_2$ ,  $O_3$ ), derived from national-scale dispersion models. As population and health data are available at municipality level, a dedicated work package up-scaled the model gridded concentrations on municipalities, using a weighted block averaging procedure (Ignaccolo, 2012) that accounts for built-up surface percentages collected from CORINE LAND COVER data (<http://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2006-raster-3>). MED HISS cohorts are representative of all populations and areas of residence (urban, rural, metropolitan) and long term effect will be evaluated for a wide range of diseases.

Long term effects of air pollution on mortality and hospital admissions (the latter rarely assessed in epidemiological studies) were calculated for Italy with the 1999-2000 NHIS, with different follow-up periods and number of cohort members using the Cox proportional hazard model (Cox, 1972), with pollutants and age time-varying variables, adjusting for other variables (age, gender, educational level, activity status, living alone, BMI, smoking, physical activity).

Hazard ratios related to  $10 \mu\text{g}/\text{m}^3$  of  $\text{PM}_{2.5}$  increase are in line with other cohort studies for mortality for all causes (1.04, confidence interval 95% 1.02-1.06) and for circulatory system diseases (1.03, 1.00-1.06) and lung cancer (1.12, 1.04-1.21). Risks of first-ever hospital admissions related to  $\text{PM}_{2.5}$  were found to be significant for circulatory system diseases (1.04, 1.01-1.07), lung cancer (1.18, 1.08-1.29), kidney cancer (1.18, 1.02-1.36) and myocardial infarction (1.15, 1.10-1.21).

The relative risks allowed to assess the health outcomes in Italy for 2010 related to  $\text{PM}_{2.5}$ : 33533 (20429-41368) attributable deaths, 14 years of life lost on average for each death, 9.2 months of reduction of life expectancy for each individual. Northern Italy and urban areas, being more polluted, show higher mortality.

As MED HISS proposed a new methodology, several practical issues emerged during the project.

Data availability is different in the 4 countries, both generated by air quality models (in France-CHIMERE, in Italy-MINNI, in Slovenia-ARSO and in Spain-CALIOPE) and for health data. Model datasets are inhomogeneous because available on different years, due to differences in the national practices and non-mandatory use of models for regulatory purposes (e.g., MINNI model data on Italy required assimilation of measured data, to be comparable to the other countries).

Inhomogeneous anonymization and linkage procedures and schemes of NHIS questionnaires are in force across Europe. The individual linkage between NHIS sampled subjects and mortality/morbidity data was not possible in Spain and Slovenia for preserving privacy and was substituted with an ecological approach based on aggregated data, which allowed to calculate relative risk rates for mortality from  $\text{PM}_{2.5}$ .

National-scale epidemiological studies can be as well based on measured data of pollutant concentrations. A recent epidemiological study has been carried out through a voluntary collaboration between researchers from ENEA, Italian National Institute for Environmental Protection and Research (ISPRA) and Istituto Superiore di Sanità (ISS) (Uccelli et al., 2016). It concerns female lung cancer mortality and long term exposure to particulate matter (PM) in Italy starting from data detected by the official monitoring stations. It is the first Italian epidemiological study on female lung cancer mortality on all the municipalities of province capital cities with available measured mean annual concentrations of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ . PM is one of the main responsible of the impact of air pollution on human health and it has recently been classified in Group 1 by IARC, besides outdoor air pollution (IARC, 2015). Even though in Italy a reduction of the emissions has been observed in the last 25 years, PM concentrations are still high in comparison with both the European targets and the WHO guidelines of air quality (WHO, 2006). The study considered female population only, in order both to reduce the confounding effect of occupational exposures and to focus on an association previously investigated mainly in men. The dose-response relationship between female lung cancer mortality in the 2000-2011 period and  $\text{PM}_{10}$  and/or  $\text{PM}_{2.5}$  available mean annual outdoor concentrations (respectively in 64 and 32 municipalities) and the burden of death due to such exposures were computed.

Standardized mortality rates (SMRate) were calculated by the ENEA's epidemiological database, which includes the Italian mortality data (both general and cause specific) up to 2013, the 3<sup>rd</sup> International Classifications of Diseases (ICD VIII, IX and X), and the Italian decennial census populations from 1961 and their annual interpolations. Mortality data, codified and recorded by the National Institute of Statistics (ISTAT), are the only health data immediately available in Italy for all municipalities. Multiple regression analysis of SMRates, as a function of PM concentrations, considering percentage of smokers and deprivation indexes as additional explanatory variables, was performed for PM<sub>10</sub> only due to the relatively low number of PM<sub>2.5</sub> monitoring stations' measures. An SMRate increase of 0.325 for 1 µg/m<sup>3</sup> increment of PM<sub>10</sub> concentration was calculated.

On the basis of such an increase and of the attributable risk evaluated from the overall difference of SMRates between the 2 subgroups of municipalities equal/below and above the WHO guideline of 20 µg/m<sup>3</sup>, a proportion ranging between 13-16% female lung cancer mortality could be attributed to PM<sub>10</sub> levels exceeding the WHO guideline. Therefore, about 300 female lung cancer deaths could be prevented each year in the investigated municipalities with an overall annual population of 8, 146, 520.



# What remains to be done to reduce air pollution?



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## CHAPTER 11 - WHAT REMAINS TO BE DONE TO REDUCE AIR POLLUTION?

The previous chapters have shown how air pollution and its environmental effects have considerably improved over the past 20 years. This important result has been achieved also as a consequence of the implementation of the different protocols under the Convention on Long Range Transboundary Air pollution (CLRTAP), and in particular of the national emission ceilings under the Gothenburg Protocol of 1999 and the NEC Directive (EC, 2001). However, critical issues remain, such as the outstanding effect of N deposition on forest nutrition and growth and on water quality. Current levels of N deposition are still too high to prevent impact on soil, foliar and water chemistry, tree growth and carbon sequestration.

The implementation of the current protocols and directives contributes to reduce, but not eliminate, the negative impacts on human health, materials and environment and further measures are needed in order to eliminate as much as possible negative effects. The impact assessment of the Clean Air Policy Package (COM, 2013) shows, for example, that by 2030 the clean air policy package, compared to the business as usual scenario, is estimated to avoid 58000 premature deaths, save 123000 km<sup>2</sup> of ecosystems from nitrogen pollution, save 56000 km<sup>2</sup> protected Natura 2000 areas from nitrogen pollution and save 19000 km<sup>2</sup> of forest ecosystems from acidification. Moreover, health benefits alone will save €40-140 billion in external costs and provide about €3 billion in direct benefits (Amann et al., 2014).

The last two most important negotiation processes, the Gothenburg Protocol at the international level and the NEC Directive at the European level, have introduced for the first time a cost-effectiveness and effect-based principle as the rationale to derive quantitative and differentiated national reduction obligations based on the reduction of health impact (Amann et al., 2011; COM, 2013; Amann et al., 2015). This new approach highlighted how the reductions of the effects on human health and ecosystems are leading all the air pollution policies.

To support the methodological aspects of the policy design through this new approach and make it applicable at a regional level within a country, the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) has developed MINNI (*National Integrated Model to support the International Negotiation on atmospheric pollution*) ([www.minni.org](http://www.minni.org)), funded by the Italian Ministry of Environment, Land and Sea (see box 11.1).

MINNI is an Integrated Modelling System that links atmospheric science with the economics of emission abatement measures and policy analysis and consists of several interdependent and interconnected components: the national AMS (*Atmospheric Modeling System*, Mircea et al., 2014) and the national GAINS-Italy (Ciucci et al., 2016; D'Elia et al., 2009). They interact in a feedback system through ATMs (*Atmospheric Transfer Matrices*) and RAIL (*RAINS-Atmospheric Inventory link*).

## BOX 11.1

### MINNI (National Integrated Model to support the International Negotiation on atmospheric pollution)

The MINNI project started as an agreement between the Italian Ministry of Environment and ENEA, in collaboration with AriaNet and IIASA to support the Italian emission reduction policies. MINNI has then evolved into a scientific project, coordinated by ENEA, to investigate thematic concerns concerning the air pollution field, including the development and maintaining of a state-of-the-art National Integrated Model and its validation using experimental data.

The project focuses on natural and anthropogenic causes of atmospheric pollution, both from national and trans-boundary origin, and on its impact on human health, ecosystems and country's cultural heritage. In particular air concentrations of ozone, heavy metals, Persistent Organic Pollutants (POPs), primary and secondary particulate matter are studied, along with acid depositions and depositions leading to eutrophication.

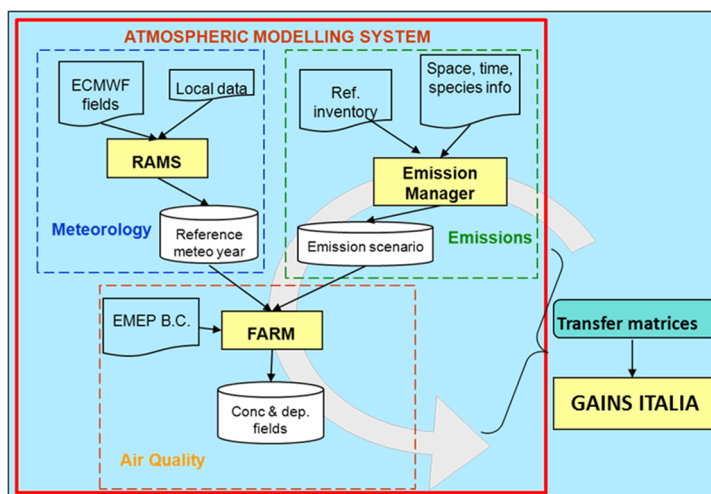
MINNI consists of two main modeling systems:

- **Atmospheric Model System (AMS)**
- **GAINS-Italy (Greenhouse Gas - Air Pollution Interactions and Synergies)**

The Atmospheric Model System (AMS) belongs to the family of chemical transport models (CTM), which describe physico-chemical processes in the atmosphere. The outputs of such models are four dimensions pollutant concentrations (considering the time), starting from known meteorological and emissions conditions with a spatial resolution of 4 km and temporal of one hour. Such models allow to link emissions to concentrations, a crucial point for management of recovery actions.

The integrated assessment model system GAINS-Italy allows evaluation of impacts and costs. Starting from information on emission abatement technologies and economic scenarios of the energy and productive sectors, GAINS-Italy produces alternative and/or future emission scenarios and provides support for the evaluation of the cost-efficiency of abatement options. The efficiency is expressed in terms of concentration reductions.

The two components are linked in a feedback system, by ATM Atmospheric Transfer Matrices (ATM) and RAINS-Atmospheric Inventory Link (RAIL).



In accordance with the most updated emission scenario elaborated by ENEA and ISPRA (D'Elia and Peschi, 2016), the baseline scenario, which assumes full implementation of current legislation, both at a European and national level, will make it possible for Italy to comply with the emission reduction commitments of the Gothenburg Protocol for all pollutants, while the new NEC targets from the year 2030 will require additional measure for all pollutants (see fig. 11.1).

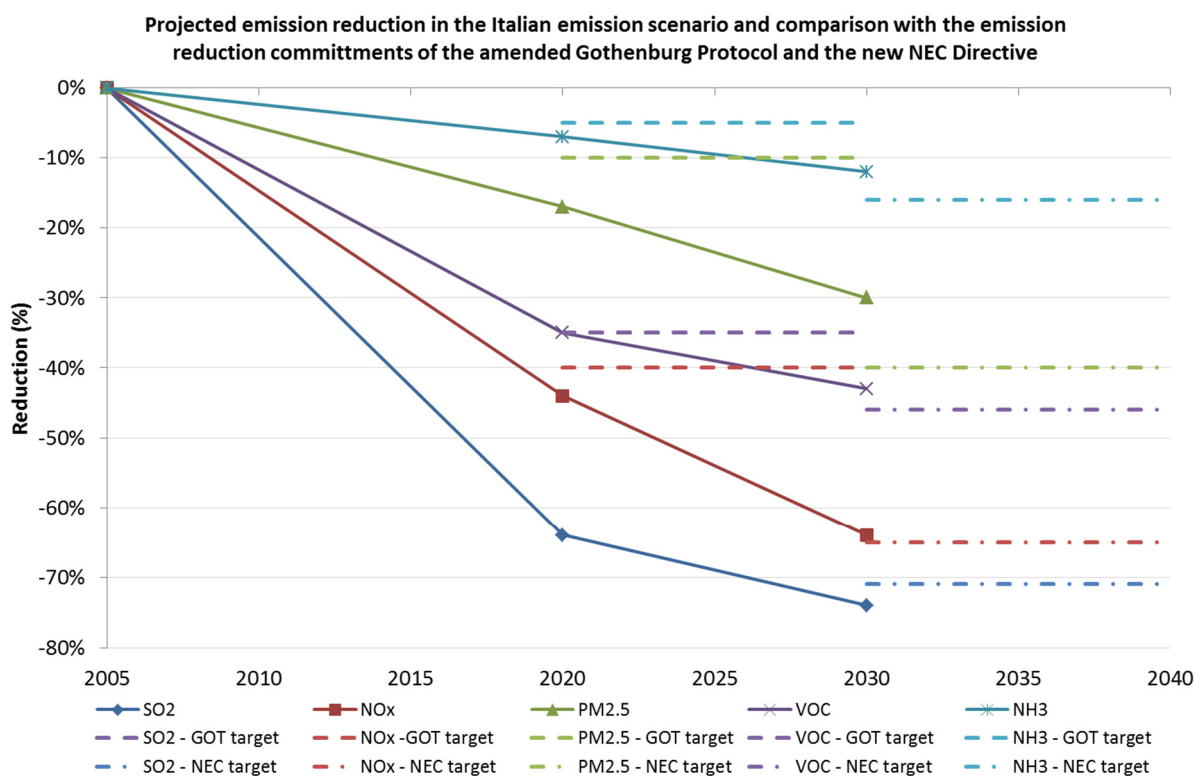


Figure 11.1 – Projected emission reduction in the last national emission scenario and comparison with the NEC targets.

Different and cost-efficient measures are available to reduce emissions, for example the Ecodesign Directive in the residential sector could reduce PM<sub>2.5</sub> emissions in a range between 35% and 55% in the year 2030 with respect to the base year 2005 depending on the implementation hypotheses; while measures on urea-based fertilizers and of the low N feeding for livestock could lead to an ammonia reduction of 22% (D'Elia and Peschi, 2016). Scenarios are not forecasts and they merely describe future developments based on major driving forces and on their impacts. In order to define the additional reduction needed and the most efficient measures that should be put in place to reach the emission reduction commitments, sensitivity analyses are unavoidable, especially on Euro 6 standards on diesel cars whose complete failure would let the NOx target unreachable.

The amended Gothenburg protocol (ECE, 2012a; 2012b) and the other last new protocols and the new NEC Directive (EC, 2016) also address newly recognized challenges. Emission reduction commitments have been introduced for a long list of new pollutants, from fine particulate matter (PM<sub>2.5</sub>) to some heavy metals (lead, cadmium and mercury) and a list of persistent organic pollutants (including inter alia dioxins, PAHs, DDT, PCB etc.). In particular, measures for fine particulate matter also include black carbon, that not only may have harmful health effects, but it is also a powerful greenhouse gas. It is very important to consider climatic policies in combination with air quality policies to have a win-win policy.

Ozone, mercury and POPs also show the importance of the global transport of atmospheric pollutants. From this point of view, cooperation between CLRTAP and other northern hemispheric and worldwide organizations is becoming increasingly important and is highly encouraged.

Another possibility to reduce the air pollution is linked to the so called unconventional measures. One of the most representative examples of unconventional measure is the reduction of meat consumption, strictly linked to human diet and personal choices. An example of the change in pollutants emissions linked to change in the human consumption was reported by Westhoek et al. (2014) that demonstrated a reduction in the emission of nitrogen compounds around 40% halving meat consumption all over Europe.

Changes toward a more plant-based diet could help substantially in mitigating emissions of GHGs, because the 24% of GHGs emissions (an average for all the selected countries) is due to food consumption. Unfortunately, this is a largely unexplored area of climate policy. Few authors have proposed changes that lower meat consumption.

Indeed, anthropogenic emissions of greenhouse gases (GHGs) related to food production accounts for about 15% at a world level. Carlsson-Kanyama (1998 and 2003) have shown that food choices and diet can influence the energy requirements for the provision of human nutrition and the associated GHG emissions. Meals similar in caloric content may differ by a factor lasting from 2 to 9 in GHG emissions (Engstrom et al., 2007). An analysis of the energy inputs showed that meals with similar nutritional value had a difference in GHG emissions of up to a factor of 4, depending on the items chosen (Carlsson-Kanyama et al., 2003). All of these studies identified certain foods as more resource demanding/polluting, including animal products and certain vegetable-intensive ways produced.

Many improvements have been reached through the Convention on Long-range Transboundary Air Pollution and its protocols but long-term risks due to ozone, nitrogen, heavy metals and persistent organic pollutant continue to exist and a significant proportion of the Italian population is exposed to high concentrations of fine particles and ozone. Lots solutions are available and an integrated approach to air pollution and climate change could lead to significant co-benefits. Effective environmental policy could only be developed implementing monitoring programmes, developing models to predict concentrations and deposition levels, and assessing their effects and measures. The integration of all the different tools, from measures to models, and the coordination among science sectors and different research teams could help in identifying cost-effective solutions.



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## **CHAPTER 6 - BIODIVERSITY AS AN IMPORTANT INDICATOR OF SOIL ACIDITY AND EUTROPHICATION: THE ROLE OF THE MODELLING IN PRESERVING IT.**

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## CHAPTER 7 - The contribution of Italy to the ICP WATERS Programme

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