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Impacts of air pollution on human and ecosystem health, and implications for the National Emission Ceilings Directive: Insights from Italy

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ABSTRACT

Across the 28 EU member states there were nearly half a million premature deaths in 2015 as a result of exposure to $PM_{2.5}$, O_3 and NO_2. To set the target for air quality levels and avoid negative impacts for human and ecosystems health, the National Emission Ceilings Directive (NECD, 2016/2284/EU) sets objectives for emission reduction for SO_2, NOx, NMVOCs, NH_3 and $PM_{2.5}$ for each Member State as percentages of reduction to be reached in 2020 and 2030 compared to the emission levels into 2005. One of the innovations of NECD is Article 9, that mentions the issue of "*monitoring air pollution impacts*" on ecosystems. We provide a clear picture of what is available in term of monitoring network for air pollution impacts on Italian ecosystems, summarizing what has been done to control air pollution and its effects on different ecosystems in Italy. We provide an overview of the impacts of air pollution on health of the Italian population and evaluate opportunities and implementation of Article 9 in the Italian context, as a case study beneficial for all Member States. The results showed that SO_4^{2-} deposition strongly decreased in all monitoring sites in Italy over the period 1999–2017, while NO_3^- and NH_4^+ decreased more slightly. As a consequence, most of the acid-sensitive sites which underwent acidification in the 1980s partially recovered. The O_3 concentration at forest sites showed a decreasing trend. Consequently, AOT40 (the metric identified to protect vegetation from ozone pollution) showed a decrease, even if values were still above the limit for forest protection (5000 ppb h^{-1}), while PODy (flux-based metric under discussion as new European legislative standard for forest protection) showed an increase. National scale studies pointed out that PM10 and NO2 induced about 58,000 premature deaths (year 2005), due to cardiovascular and respiratory diseases. The network identified for Italy contains a good number of monitoring sites (6 for terrestrial ecosystem monitoring, 4 for water bodies monitoring and 11 for ozone impact monitoring) distributed over the territory and will produce a high number of monitored parameters for the implementation of the NECD.

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1. Introduction

Air pollution has significant negative impacts on human health (IARC, 2015; Atkinson et al., 2010; Cadelis et al., 2014; Correia et al., 2013; Fang et al., 2013; Meister et al., 2012) as well as on natural and anthropogenic ecosystems (Ochoa-Hueso et al., 2017; Rao et al., 2017; Varotsos et al., 2012; Monks et al., 2015), climate (Bytnerowicz et al., 2007; Sitch et al., 2007; Shindell et al., 2009; Myhre et al., 2013), and consequently society and economy (EEA, 2016a). Numerous research studies showed the associations between exposure to air pollutants and adverse health outcomes, such as increased hospital admissions for respiratory and cardiovascular diseases, congestive heart failure, increases in asthma attacks and acute bronchitis, and altered lung function (Fattore et al., 2011; Al-Hemoud et al., 2018; Goudarzi et al., 2017). Despite international and national air pollution strategies to control air pollutant emissions, a large proportion of European populations and ecosystems are still exposed to concentrations exceeding the limit or target values (EEA, 2018) recommended by Air Quality Directive (Directive 2008/50/EC) and the World Health Organization for the protection of human health (WHO, 2017).

The Geneva Convention on Long-range Transboundary Air Pollution (CLRTAP), signed in 1979, and its following eight protocols, are focused on human and ecosystem protection from the negative impacts of air pollution and recommend the reduction of eight air pollutants: Sulphur Oxides (SOx), Nitrogen Oxides (NOx), Non-Methane Volatile Organic Compounds (NMVOC), Heavy Metals (HMs), Persistent Organic Pollutants (POPs), tropospheric ozone (O_3), Particulate Matters (PM_{10}, $PM_{2.5}$) and Black Carbon (BC). For the protection of human health, the Directive 2008/50/EC has introduced a threshold of 60 ppb for the daily maximum 8-h ozone average. The threshold level should not be exceeded on > 25 days a year, averaged over 3 years (target value) and per calendar year (long-term objective). In 2016, 17% of European Union (EU) stations exceed the target value for human health protection. Besides, 87% of the background stations reported values above the long-term objective (EEA, 2018). The latest European Environment Agency air quality report (EEA, 2018) highlights that the stations with NO_2 concentrations above the annual limit value (40 μg/m^3) are widely distributed all over Europe, as in previous years, and 88% of these values were observed at traffic stations. Even if the decrease of NO_2 concentrations observed over the period 2000–2014 at all types of stations will continue at the same rate until 2020, 7% of stations would still have concentrations exceeding the annual limit value (EEA, 2016a). The concentrations of PM_{10} continue to exceed the daily (50 μg/m^3 not to be exceeded > 35 days a year) and annual mean (40 μg/m^3) EU limit values across Europe. The PM_{10} limit value (20 μg/m^3) adopted by the WHO was exceeded at 48% of EU stations, located in 27 countries (EEA, 2018) while $PM_{2.5}$ limit value (10 μg/m^3) was exceeded at 68% of stations located in 32 out of 37 countries.

To achieve adequate air quality levels and avoid significant negative impacts and risks for human and ecosystems health, the revision of the National Emission Ceilings Directive (2016/2284/EU, hereafter NECD) replaces earlier legislation (Directive 2001/81/EC) and sets objectives for emission reduction for each Member State as percentages of reduction to be reached by 2020 and 2030 compared to 2005 levels. The NECD is currently the main legal instrument to reduce overall emissions of air pollutants in Europe and, given the transboundary nature of an important part of air pollution, its enforcement throughout the entire Europe is of crucial importance. The NECD sets 2020 and 2030 emission reduction commitments for five classes of air pollutants: Sulphur dioxide (SO_2), Nitrogen Oxides (NO_x), Non-Methane Volatile Organic Compounds (NMVOCs), ammonia (NH_3) and Particulate Matter below 2.5 μm of aerodynamic diameter ($PM_{2.5,}$ the new pollutant included in the Directive). Tropospheric O_3, being a secondary air pollutant, it is not specifically included in the NECD as it is not emitted directly by human activities; however, it is controlled indirectly via regulation of its precursors, namely VOCs and NOx (Chameides et al., 1988). Ozone is probably one of the most damaging air pollutants for forests and crops (Lefohn et al., 2018; Mills et al., 2011a; Ochoa-Hueso et al., 2017) and can lead to more frequent hospital admissions and increase the risk of mortality for cardiovascular and respiratory diseases (Ghorani-Azam et al., 2016; WHO, 2013). Ozone may become worse in the future (Sicard et al., 2017) leading to public health concerns (WHO, 2013).

The health impact of air pollution in the EU is expected to be halved by 2030, compared to 2005 levels. However, according to the estimations of EEA 2018, across the 28 EU member states there were nearly half a million premature deaths in 2015 because of exposure to $PM_{2.5}$, O_3 and NO_2. Such impacts correspond to an estimated cost of 330–940 billion euros (3–9% of EU Gross Domestic Product, GDP). Therefore, NECD represents an opportunity for all EU citizens which will benefit from improved air quality and for industry because the measures foreseen for reducing air pollution will boost innovation and enhance European competitiveness in the field of green technologies. The NECD is an opportunity also for public authorities that will save billion euros through lower health care costs by reaching the expected air pollution standards (EC, 2016). Beyond the human health, economic negative impacts are expected by agricultural yield losses (Avnery et al., 2011; Van Dingenen et al., 2009), maintenance and restoration costs for Cultural Heritage (Rabl, 1999) and reduction of ecosystem services provided by aquatic (Beier et al., 2017) and terrestrial (Hein et al., 2018) ecosystems.

One of the innovations of NECD is Article 9, that mentions the issue of "*monitoring air pollution impacts*" on ecosystems. According to this article, "*Member States shall ensure the monitoring of negative impacts of air pollution upon ecosystems through a cost-effective and risk-based approach, based on a network of monitoring sites that should be representative of their freshwater, natural and semi-natural habitats and forest ecosystem types*". Annex V of NECD reports a series of indicators for monitoring air pollution impacts, although the choice among them is not mandatory. These indicators should be monitored based on the methodologies proposed by the CLRTAP and its International Cooperative Programs (e.g. ICP Forests, ICP Waters). Member States that do not use the optional indicators will need to explain how the selected indicators fulfill the objective of Article 9.

In Italy, the Ministry for Environment, Land and Sea - General Directorate for Waste and Pollution (Division on Air, noise and electromagnetic pollution), is responsible for the NECD enforcement and for setting a National Network to monitor air pollution impacts in collaboration with research institutions and local administrations. Italy is an interesting case study for the application of the NECD Article 9, because of the high level of functional biodiversity and the wide variety of environmental, landscape and climatic conditions (Blasi et al., 2008, 2014; Capotorti et al., 2012) that make difficult the selection of representative monitoring sites. Moreover, due to the multi-stress conditions that typically affect Mediterranean ecosystems, the identification of monitoring indicators is a critical issue (Manes et al., 2007; Fusaro et al., 2015; Ochoa-Hueso et al., 2017). In the last 20 years, however, a large effort has been carried out by the Italian scientific community to study the impacts of air pollution on different ecosystems, both natural or anthropogenic (e.g. D'Elia et al., 2017; Paoletti, 2006; Rogora et al., 2012, 2013).

Effective actions to reduce the impacts of air pollution requires a good understanding of the main sources of emission, how the pollutants are transported and transformed in the atmosphere, and how they affect human health and ecosystems. Based on these consideration, the aims of this manuscript are: 1) to provide a clear picture of what is available in term of monitoring network for air pollution impacts on Italian ecosystems; 2) to summarize what has been done to control air pollution and its effects on different ecosystems in Italy; 3) to provide a framework/summary/overview of the expected impacts of air pollution on health in Italian population; and 3) to evaluate opportunities and implementation of Article 9 of the NECD in the Italian context. This limited case study may be beneficial for all Member States.

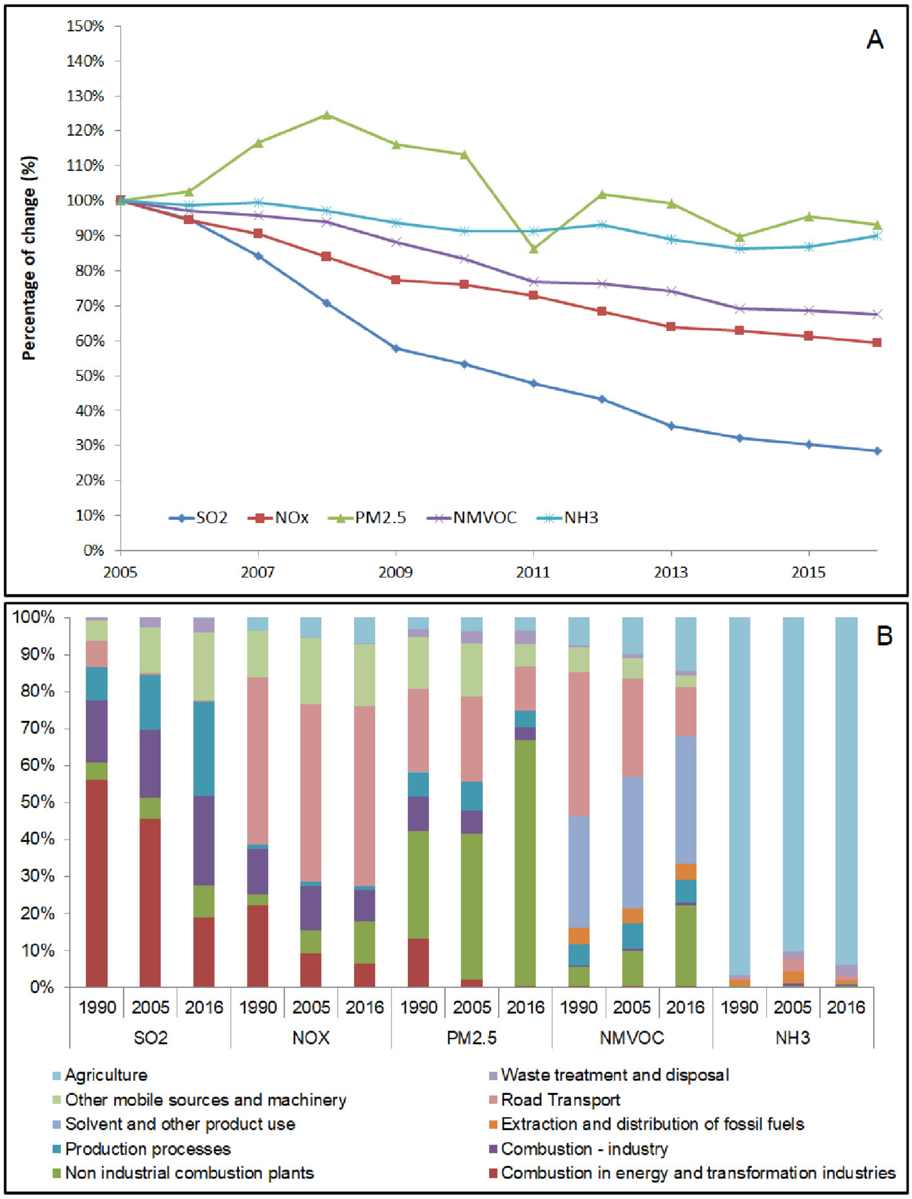


Fig. 1. Total Italian emissions (expressed as percentage of change relative to 2005) from 2005 to 2016 (A) and partitioning by sector for the years 1990, 2005 and 2016 (B) for SO_2, NOx, $PM_{2.5}$, NMVOC and NH_3.

2. Sources and emissions of air pollutants in Italy and relative concentration into the atmosphere

Trends of Italian emissions from 2005 to 2016 (ISPRA, 2018) for the five pollutants covered by NECD (SO_2, NO_x, NH_3, NMVOC and $PM_{2.5}$), are shown in Fig. 1A while a comparison among sector variations in the years 1990, 2005 and 2015 is shown in Fig. 1B. A significant decreasing trend is observed for SO_2 ($p < 0.05$). The national target of SO_2 emissions, set by the previous NECD (Directive 2001/81/EC) at 475 Gg for 2010 (EC, 2001) and the new target established for 2020 (65% emission reduction respect to the base year 2005) were reached. The decreasing trend of SO_2 is mainly determined by the reduction in emissions from energy combustion and transformation in the industrial sector due to the use of natural gas in place of coal in the energy production (Fig. 1B); this result follows the enforcement of the Directive EEC 75/716 (EC, 1975), which introduced more restrictive constraints in the Sulphur content of liquid fuels. The main source of NOx emissions is road transport (about 49% in 2016) and other mobile sources and machineries that contribute to the total emissions for 17% in 2016 (Fig. 1B). Similarly, the NO_x emissions show a decreasing trend from 2005 to 2016 (around 41%), mainly related to a large reduction of NOx emissions by the road transport (−60% and −40% in 2016 relative to 1990 and 2005, respectively) and energy combustion sectors (−89% and −59% in 2016 relative to 1990 and 2005, respectively) (Fig. 1B). Conversely, the non-industrial combustion plants emissions show an increase by +35% in 2016 relative to 1990 and +11% relative to 2005. However, the target value of emissions fixed for 2010 by the NECD (EC, 2001) at 990 Gg has already been reached and continues to be respected. In Western Europe, the introduction of improved vehicle technologies and stringent Euro standards reduce NOx road traffic emissions from 1990, despite increases in fuel consumption (Vestreng et al., 2008; Sicard et al., 2013). In the national emission inventory, NOx emissions from road transport were calculated with the software COPERT 5, version 5.1 (ISPRA, 2018).The atmospheric emissions of ammonia showed a slight decline in 2005–2016 of about 10% (Fig. 1A), reaching the national target of NH_3 for 2010 (Directive 2001/81/EC) and 2020 (new NEC Directive, 2016/2284/EC). In 2016, the main source of emissions was agriculture with a 94% of contribution out of the total NH_3 emissions (Fig. 1B), despite the introduction of abatement technologies and a reduction in the livestock number (EC, 2010). The emissions of NMVOC decreased between 2005 and 2016, with a variation of about 32% (Fig. 1A). Solvent and other product use and non-industrial combustion plants are the main sources of emission for NMVOC (34% and 22% respectively in 2016) (Fig. 1B). The decrease of NMVOC emissions can be attributed to the vehicles equipment into catalytic exhaust pipes and the progress in the storage and distribution of hydrocarbons (Sicard et al., 2013). The $PM_{2.5}$ emissions show a fluctuating non-linear trend from 2005 to 2016 with a slight reduction of about 7%, in 2016, relative to 2005. In 2016, the non-industrial combustion plant sector represented the most emitting sector, contributing to 66% of total $PM_{2.5}$ emissions due to the relevant contribution of residential biomass burning, followed by the road transport sector with 12%.

Overall, all emissions from the industrial and road transport sectors decline over the period 2005–2016. The identification of sources and the quantification of emissions suggest that the implementation of various European Directives introducing new technologies, Euro standards in road transport, plant emission limits, limitation of Sulphur content in liquid fuels and shift to cleaner fuels, in conjunction with the improvement of energy efficiency and the promotion of renewable energy, led to these positive achievements. The promotion of renewable energy adopted for climate change mitigation policies, like the increasing of residential wood combustion, could have a counter-productive effect on air pollutant emissions and air quality, particularly evident on $PM_{2.5}$ emissions whose trend reflects the residential wood consumption.

Moving from emissions to air pollutant concentration into the atmosphere, a critical problem is the number of monitoring sites. Indeed, despite large efforts to improve the quality of collected data, the observations are often not sufficient to cover the whole country and to represent an area with complex topography like Italy (García-Gómez et al., 2014). The spatial distribution of monitoring stations is rarely homogeneous, and gaps can be noted for the quantification of population and ecosystem exposures. To overcome this issue, the additional support of a modeling system to evaluate air quality impacts is necessary. In fact, Air Quality Directive (EC, 2008) clearly stated the importance of using modeling tools in addition to the ground measurements for analyzing emission trends and pollutant concentrations for air quality assessment and planning. In Italy, the Atmospheric Modeling System of MINNI project (MINNI, National Integrated Model to support the international negotiation on atmospheric pollution – extensively described in Mircea et al., 2014, 2016; D'Elia et al., 2009; Ciucci et al., 2016) was developed by ENEA with support from the Italian Ministry of Environment, Land and Sea Protection, assesses air quality at national level every five years according to Legislative Decree D.lgs. 155/2010 as requested by Air Quality Directives, and evaluates national Air Quality Plans in support to the NECD according to D.lgs. 81/2018. Fig. 2 shows the annual mean of hourly concentrations of SO_2, NO_2, O_3, and $PM_{2.5}$ simulated by AMS-MINNI (Mircea et al., 2016) for the years 2005 and 2010. The performances of AMS-MINNI in predicting air pollutant concentrations are described in detail by Mircea et al. (2014) and Ciancarella et al. (2016). By comparing the pattern of the two years, it can be observed a difference in SO_2, NO_2 and $PM_{2.5}$ concentrations in urban and highly industrialized areas such as Po Valley because of the emission control strategies. On the other hand, the O_3 concentrations in 2010 outside Po Valley are higher than those simulated in 2005; this is due to the high sensitivity of O_3 concentrations to meteorological conditions, in particular to air temperature which was higher on average in 2010 than in 2005, and due to the non-linear response of O_3 formation to precursor emission reductions. The same difference observed between 2005 and 2010 by the modeling concentration is confirmed by measurements data. Thus, the modeling system not only compensates for the scarcity of measurements, but also allows to better investigate the response of O_3 concentrations to emission reductions in areas with different VOC/NOx chemical regimes and estimate O_3 impacts on ecosystems.

3. Measurements and impacts of air pollution in Italian ecosystems

3.1. Terrestrial ecosystems

Because of the nature of crops, which usually have an annual growth cycle, the long-term monitoring of pollution impacts has limited meaning for this kind of vegetation. For these reasons, in the following section we focused the discussion on forests, putting aside semi-natural vegetation and crops.

Forest ecosystems play a key role in providing ecosystem services (Manes et al., 2012, 2016) that could be impaired by the impacts that pollutants have on tree functionality (Fusaro et al., 2015; Wittig et al., 2009; Li et al., 2017).

Forests in Italy cover ca. 10.467.533 ha and represent 34.7% of the land area (INF, 2015). Italian forests stock 1242 Mt. carbon and provide tangible and intangible benefits to society (Vizzarri et al., 2015), such as effective visitor and tourism management, or wellbeing. In Italy, two monitoring networks were set-up to comply with the requirements of EU regulations and the CLRTAP: a large-scale monitoring network of around 260 plots on a systematic grid (Level I plots, ICP Forests) and an intensive monitoring network of 31 selected sites (Level II, ICP Forests). Based on the data collected at those sites, many studies have been performed to investigate air pollution impacts on Italian forest health and growth (e.g. Paoletti, 2009; Paoletti et al., 2018; De Marco et al., 2013). Another monitoring network in the frame of the European Strategy Forum on Research Infrastructures (ESFRI) has the potential to provide long-term data on carbon assimilation and therefore better understanding the effect of climate change and pollutant exposure on key agricultural and forest ecosystems. This is the Integrated Carbon Observation System (ICOS) with 9 active sites in Italy (Fares et al., 2018).

Air pollution affecting Italian forest ecosystems is primarily in the form of nitrogen (N) deposition and tropospheric O_3 (Paoletti, 2006; Gentilesca et al., 2018). The deposition of some atmospheric compounds has been monitored at ICP Forests sites in Italy since 1998 (9 Level II ICP Forests sites).

Sulphate (SO_4^{2-}) deposition showed an evident decreasing trend over 1998–2013 in all monitoring sites in Italy (on average by 40%), while nitrate (NO_3^-) and ammonium (NH_4^+) depositions showed a slight decrease only in a few sites (D'Elia et al., 2017). Bulk open-field deposition of SO_4^{2-} showed a decreasing trend especially in the first decade of that period (Fig. 3), as an effect of the decreasing emission of SO_2. A slight decrease in NO_3^- and NH_4^+ deposition was evident only in the most recent years and just at a few sites (Fig. 3). Inorganic N deposition still exceeds 10–12 kg N ha^{-1} $year^{-1}$ at Northern sites, where critical loads exceedance are found (D'Elia et al., 2017). These results agree with those reported by other studies on deposition trends in Northern Italy (Rogora et al., 2016) and over Europe (Waldner et al., 2014). N deposition may affect species composition, plant diversity, nutrient balance and increase the susceptibility to secondary stress (e.g. aridity) (Fusaro et al., 2017, 2018) and disturbance factors e.g. plant pathogens, insect pests (Bobbink et al., 2010; Holmberg et al., 2018; Dirnböck et al., in press). Earlier studies reported weak and not significant correlations between tree crown defoliation and air pollutant depositions at European level (De Marco et al., 2015, 2017; Vitale et al., 2014; Augustaitis et al., 2010).

The annual mean of hourly O_3 concentrations showed an increase between 1997 and 2005 and a decrease between 2005 and 2011 (Seasonal Kendall test on monthly data, $p < 0.01$, D'Elia et al., 2018), even if the slope at forest sites was below 10% (LIFE SMART4*Action*, unpublished data).

Ozone may i) reduce CO_2 assimilation at leaf level leading to a

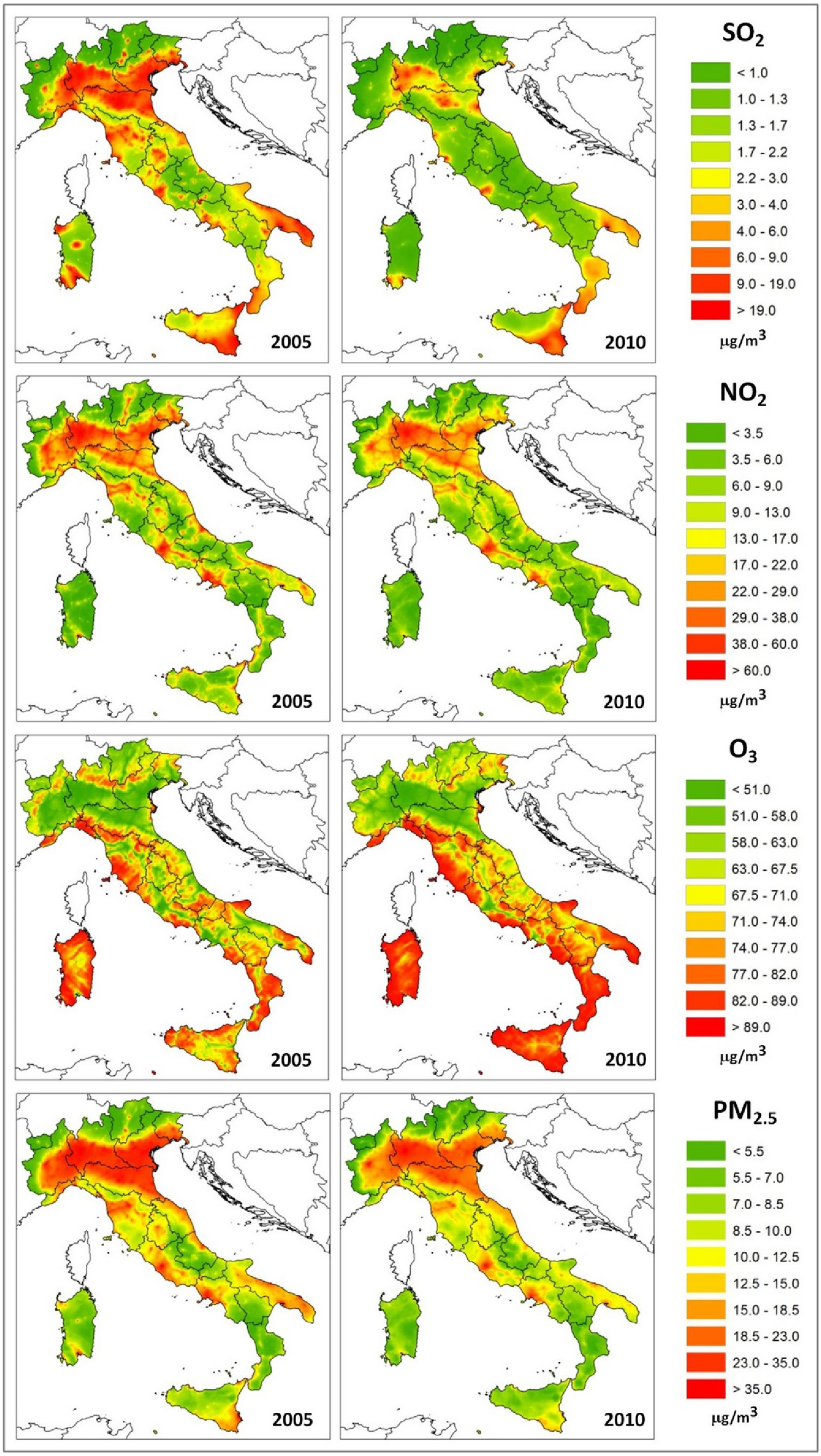


Fig. 2. SO_2, NO_2, O_3, and $PM_{2.5}$ annual mean of hourly concentrations in 2005 and 2010 in Italy by MINNI atmospheric modeling system at a spatial resolution of 4 km.

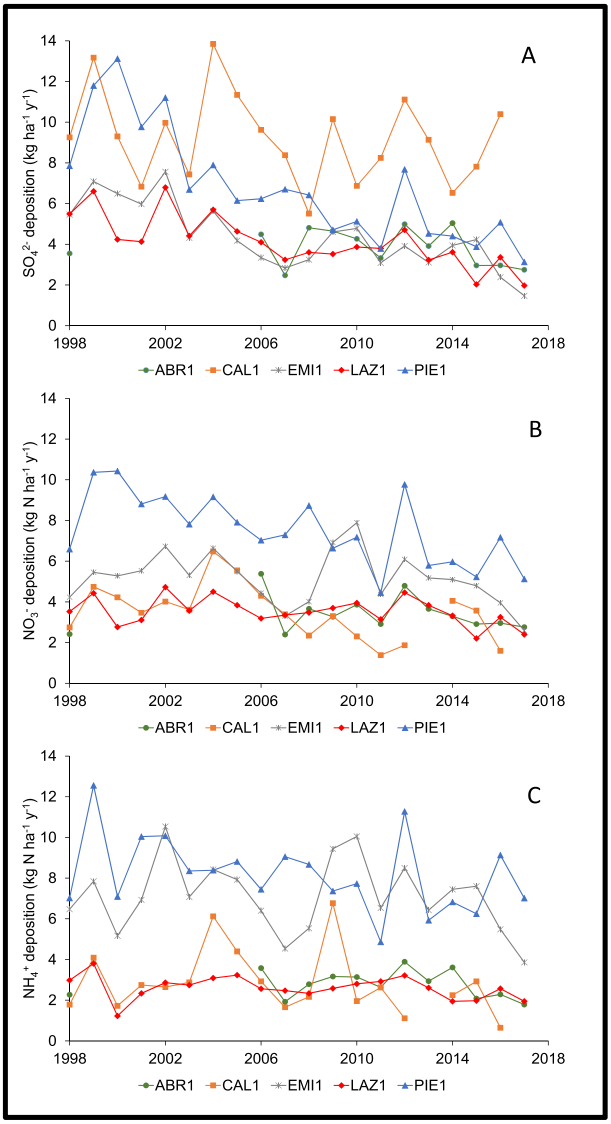


Fig. 3. Variation of sulphate (SO_4^{2-}) (A), nitrate (NO_3^-) (B) and ammonium (NH_4^+) (C) deposition (bulk open field) at 5 ICP Forest sites over the period 1998–2017.

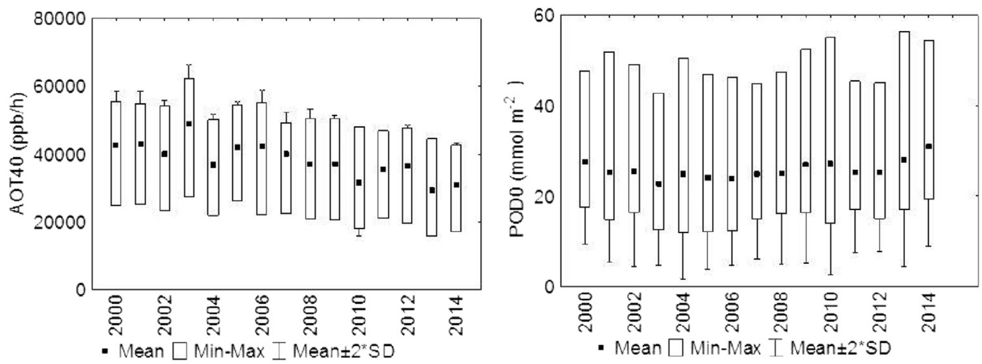
reduction of the net primary production (NPP) at ecosystem level (Fares et al., 2013), ii) reduce biomass and leaf area, and iii) cause premature leaf senescence and visible foliar injury (Ainsworth et al., 2012). While the effects of O_3 have been investigated using controlled experiments such as Open-Top Chambers or ozone-FACE devices (Watanabe et al., 2012; Feng et al., 2015; Paoletti et al., 2017), large-scale epidemiological analysis of O_3 effects on forest ecosystems and the resulting feedbacks with the atmospheric chemistry and physics (Anav et al., 2011) represent a challenge for the scientific community. As an example of epidemiological analysis into the field, Sicard et al. (2016) found positive correlations between annual mean of hourly concentrations of O_3 and crown defoliation in Northern Italy.

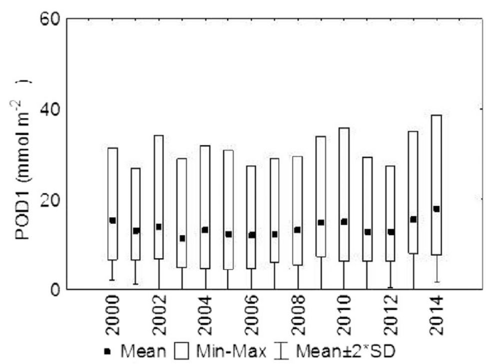
Another important indicator of forest condition is tree growth that is defined as the growth of trees and stands within five years expressed as increment of diameter, basal area, height and/or volume (D'Elia et al., 2017). A case study performed on the stand volume growth of *Fagus sylvatica* in 728 plots distributed across Italy over the period 2001–2005 to analyze O_3 effect (as annual average of hourly concentrations, accumulated exposure over a threshold of 40 ppb "AOT40" and total stomatal flux) showed that beech growth increased with increasing solar radiation and air temperature while effects of soil water content (SWC) and O_3 were not statistically significant (Paoletti et al., 2018). Because *Fagus sylvatica* is sensitive to drought and O_3 (Lebourgeois et al., 2005; De Marco et al., 2014), a possible explanation is that the stand volume growth averaged over 5 years does not capture the high frequency signals of SWC and O_3 (Paoletti et al., 2018). For this reason, dendrometers, a method for measuring tree radial growth at high temporal resolution, may be more suitable to link pollutant exposure to growth dynamics (Paoletti et al., submitted).

In the frame of CLRTAP, different impact metrics and critical loads/levels (CL) have been established, in order to protect forests from adverse effects of N deposition and O_3. Critical levels are defined as the "*concentration, cumulative exposure or cumulative stomatal flux of atmospheric pollutants above which direct adverse effects on sensitive vegetation may occur according to present knowledge*" (CLRTAP, 2015). The O_3 exposure index AOT40 is the current European standard to assess whether vegetation is at risk for O_3 pollution and it is widely applied in Europe (CLRTAP, 2015). However, after decades of studies based on the accumulated exposure to phytotoxic O_3 concentrations, now it is widely recognized that the best metrics for O_3-risk assessment should be based not only on atmospheric concentrations, but also on O_3 uptake through the stomata (Matyssek et al., 2007; Fares et al., 2018). Therefore, CLRTAP has introduced stomatal flux-based metrics and critical levels (CLef) for protecting vegetation against O_3 effects (e.g., Mills et al., 2011b; CLRTAP, 2015; Anav et al., 2016; Sicard et al., 2016). This metric, namely Phytotoxic Ozone Dose above a detoxification threshold Y (PODy, where POD0 and POD1 have Y equal to hourly values of 0 and 1 mmol $m^{-2} s^{-1}$, respectively), considers the influences of air temperature, air-to-leaf water vapor pressure deficit (VPD), light (irradiance), soil water content (SWC), O_3 concentrations and plant phenology to calculate the stomatal O_3 flux, based on the DO_3SE model (Mills et al., 2011b). In combination with dose-response functions, PODy can be used to assess and quantify O_3 impacts to forest trees (Sicard et al., 2016). To calculate PODy, hourly O_3 and meteorological data are needed. Fig. 4 shows average values of AOT40, POD0 and POD1 calculated by a multi-model system composed by WRF (Weather Research and Forecast Model), a regional climate model, and CHIMERE, a chemistry transport model, extrapolated at 17 selected level II plots of Italy (Anav et al., 2016). AOT40 presents average values well above the limits for forest protection (i.e. 5000 ppb h; UNECE, 2010) in all considered years; minimum AOT40 value was found in 2013 while the maximum value was in 2003, mainly due to the heat wave in Europe. AOT40 showed a decreasing trend (Fig. 4) with an annual change of 966 ppb per year. On the contrary, POD_0 showed an increasing trend (0.23 mmol m^{-2} $year^{-1}$, $p < 0.05$), while we did not find a significant trend for POD_1. POD_0 and POD_1 ranged from 22.72 and 11.50 mmol m^{-2} in 2003 to 31.08 and 18.06 mmol m^{-2} in 2014, respectively (Fig. 4).

3.2. Climate change and forest ecosystems

The Mediterranean area can be considered a test area for studying climate change (Sicard and Dalstein-Richier, 2015). Italy is located within a region characterized by a medium-latitude at the boundary between temperate and sub-tropical circulation regimes. Observed values of climate change in the Mediterranean basin outweigh global trends for most variables (Giorgi and Lionello, 2008). The annual average of temperatures is already today of 1.4 °C above the levels of the late 19th century, and this gap is particularly evident during summer (Salvati et al., 2008). More specifically, a temperature increase by 0.8 °C was observed in Italy between the late decades of the 19th century and the early 2000s. This increase was even more evident (1.0 ± 0.1 °C per century) when considering time series data between 1865 and 2003 (Brunetti et al., 2006). Minimum temperatures have increased more than maximum temperatures, particularly in northern Italy (Brunetti et al., 2006). Heat waves occur more frequently, and the frequency and intensity of droughts have increased significantly since





Variable	Period	Annual avarages change
AOT40	2000-2014	-966.02 ***
POD0	2000-2014	0.23*
POD1	2000-2014	0.16

Fig. 4. Mean AOT40, POD_0 and POD_1 from 2000 to 2014 at 17 level II plots along Italy and estimated annual change (*$p < 0.05$; ***$p < 0.001$).

the 1950s (Salvati et al., 2011). The most recent event occurred in summer 2017 when Italy, and especially Central Italy, was affected by a severe drought episode with persistent heat waves, which caused diffuse damages to forest ecosystems, including early foliar shedding, desiccation of leaves and branches, tree mortality (Pollastrini et al., 2018). Future warming in the Mediterranean region is expected to exceed 25% overall increase rates, particularly with a summer heating at a rate of growth by 40% above the global average (Ferrara et al., 2017). Even assuming to be able to contain global warming by 1.5 °C, in compliance with the Paris agreements, it has been predicted that the Mediterranean area will experience a larger increase, likely higher than 2.0 °C. It is plausible to assume that this increase will be increasingly associated with extreme heat wave events, with precipitation decreasing up to 30% (Gualdi and Navarra, 2005). The intermediate position between African tropical and sub-tropical climate and European mid-latitude climate results in fluctuation among different climate regimes for Italy (Salvati et al., 2016), and creates a complex patchwork of climate and vegetation (Blasi et al., 2001).

The high variability of weather-climatic characteristics in the Mediterranean region outlines the importance of accurate site-specific information for an extended period. Italian forests undergo global trends in temperature and precipitation although they can affect local climate by gas exchanges and impacting various components of the energy balance including evapotranspiration. Data measured since 1998 at 13 sites of the ICP-Forest network (level I) show an important spatio-temporal heterogeneity in both rainfall and temperature trends (Marchi et al., 2017). The most evident trend is a slight increase in air temperature over time, in line with the most recent patterns observed on a continental scale (Ferrara et al., 2017). As far as precipitation is concerned, the main trends identify particularly variable trends over space, with a decrease in total rainfalls for specific forest ecosystems and a generalized, slight increase in extreme events, consistent with long-term trends more evident at national and European level. In about 20 years of measurements, a temperature increase was detected, around 0.4 ± 0.2 °C and a moderate decrease in total precipitation, generally < 5% of the annual rainfall regime. In addition to soil drought and summer heat, there are other climatic events with significant impact on forests. Recently, late frosts have been registered in the Apennine region of Italy for two consecutive years (2016 and 2017), which severely affected e.g. beech forests. Finally, windstorms have caused extensive damages in the deciduous forests of the Apennines and coniferous forests of Alps, the last episode being recorded in October 2018 (Motta et al., 2018).

3.3. Freshwater ecosystems

The effects of air pollution on freshwater ecosystems (rivers and lakes) have been monitored since the 1980s through the ICP Waters (International Cooperative Programme for assessment and monitoring of the effects of air pollution on rivers and lakes). Within ICP Waters, > 170 sites in Europe and North America have been monitored with the specific objective of assessing the degree to which atmospheric pollution has affected surface waters, particularly about issues such as acidification, heavy metals and persistent organic pollutants (Skjelkvåle et al., 2005; Garmo et al., 2014).

In Italy, a network of sites exists, consisting of subalpine rivers and lakes and high-altitude lakes in the Central Alps (Mosello et al., 2002). Monitoring at these sites aims at evaluating the long-term response of sensitive ecosystems to changing atmospheric deposition.

The main pressures related to air pollution identified for the Italian sites are acidification and nitrogen enrichment due to high N deposition. The most relevant air pollutants are S and N oxides (SO_2, NO_x) and reduced N (NH_3), resulting in SO_4^{2-}, NO_3^- and NH_4^+ in precipitation affected by the emission sources. It should be highlighted that sensitive sites in Italy are in a limited geographic area, namely the alpine and subalpine areas in North-western Italy. This is because sensitivity to acid deposition strongly depends on the geological background (dominance of acid, low-weatherable rocks; Marchetto et al., 1994); therefore, a long-term monitoring program on acidification was established on selected key sites in this area (Mosello et al., 2002).

Most of the acid-sensitive sites which underwent acidification in the 1980s partially recovered as response to the decreasing of acidifying compounds' deposition, mainly as sulphate (SO_4^{2-}). The majority of sites showed an increase of pH and Acid Neutralizing Capacity (ANC)

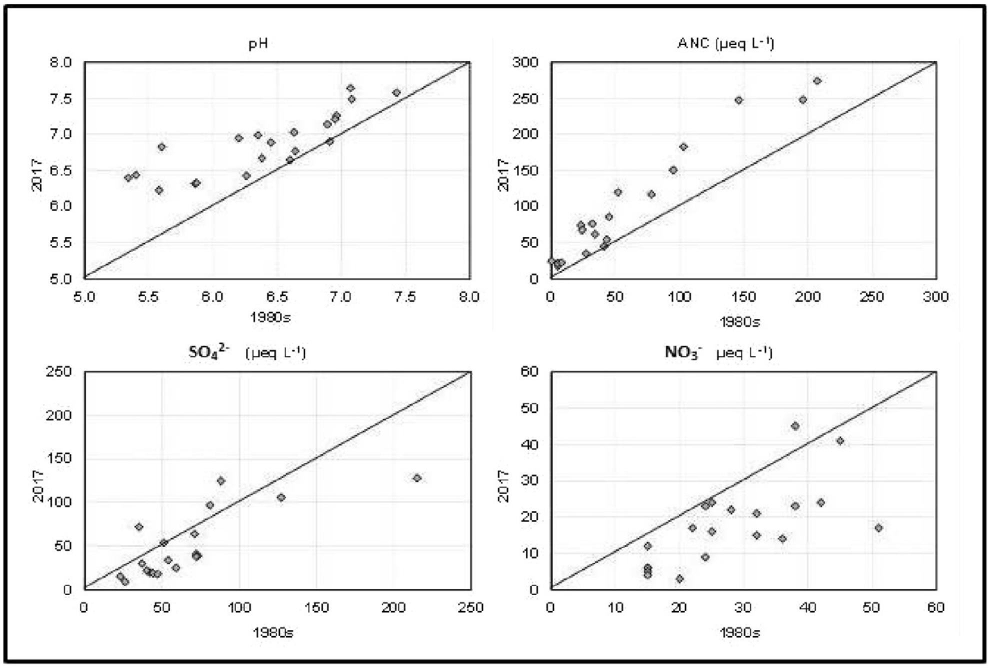


Fig. 5. Comparison of pH, Acid Neutralizing Capacity (ANC), SO_4^{2-} and NO_3^- concentrations measured in the 1980s and in 2017 at sensitive freshwater sites in Italy, including ICP Waters sites (n = 21).

and a decrease in SO_4^{2-} and NO_3^- concentrations (Fig. 5). The sites considered are mainly high altitude lakes, for which deposition is the main vehicle of N: the slight and recent decrease of NO_3^- deposition occurred in Northern Italy has been enough to cause a response in freshwater concentrations (Rogora et al., 2016). For SO_4^{2-}, beside deposition also weathering of sulphur bearing minerals may contribute to surface water concentrations. A climate-related increase of weathering rates (and of SO_4^{2-} export from the catchment) has been detected in some alpine catchments and this may partly counteract the effect of decreasing atmospheric input of SO_4^{2-} (Rogora et al., 2013).

First signs of biological recovery were also identified, such as change in diatom flora and appearance of sensitive species among benthic insects (Marchetto et al., 2004). However, a few sites among high altitude lakes continued to be acidic or still showed a high sensitivity to acidification. This is particularly evident at the snowmelt, when ANC may be fully depleted by the incoming waters rich in acidifying compounds (Rogora et al., 2013).

Beside acid deposition, a further threat to freshwater ecosystems relies in the excessive atmospheric input of N with respect to the retention capacity of catchment soils, leading to NO_3 leaching to surface waters (Rogora, 2007). The area of Lake Maggiore watershed, Northwestern Italy, is strongly affected by N deposition, due to its location north of the Po Plain, one of the most industrialized and urbanised areas of Europe. Therefore, very high N depositions being caused by a combined effect between high air pollutants concentration and high rainfall due to orographic effects (Rogora et al., 2016).

This huge flux of N from the atmosphere caused N saturation of terrestrial catchments and increasing levels of NO_3 in some rivers and lakes in the 1980s and 1990s (Rogora and Mosello, 2007; Rogora, 2007). According to pan-European studies, the Italian ICP Waters sites were among the few sites in Europe showing a significant increase of NO_3 concentrations in that period (Skjelkvåle et al., 2005). Detailed N budgets performed for river catchments stated that atmospheric input of N could not be fully retained by soil and vegetation, with N retention varying from 60 to 70% to 20–30% or even lower in the southern, where catchments were much more affected (Rogora and Mosello, 2007).

Nitrogen deposition in the study area showed a slight tendency to decrease since 2006. The decrease was more evident for oxidized N than for reduced N (Rogora et al., 2016). As an effect, monitoring data for both rivers and lakes showed a decrease in NO_3 concentrations in recent years (Fig. 5). However, N deposition remains a critical issue: an empirical critical load of nitrogen (CLN) of 3–10 kg N ha^{-1} year^{-1} has been identified for permanent oligotrophic lake, ponds and pools (Bobbink and Hettelingh, 2011), and this limit is still exceeded for some sensitive sites like high altitude lakes (Rogora et al., 2016). About ANC, a critical limit of 20 µeq L^{-1} has been identified as the minimum level required for ecosystem protection under UNECE protocols. Raddum and Skjelkvåle (2001) proposed a critical level of 30 µeq L^{-1} in the high Alps due to lithological characteristics of their catchments. However, critical levels should be set differently, depending on the biological target group used (e.g. macroinvertebrates), the sensitivity of the typical fauna and their adaptive capacity to native water chemistry. Furthermore, the proposed critical limits can be hardly reached in high altitude sensitive lakes in the Alps, even under the most optimistic deposition scenarios (Rogora et al., 2003). To conclude, in Italy, the atmospheric deposition of nitrogen is still too high with respect to critical levels and further reductions are needed to sustain acidification recovery and prevent N saturation of aquatic ecosystems.

Climate change also affects the response of freshwater ecosystems to changing deposition of N and acidifying compounds (e.g. Mast et al., 2011; Houle et al., 2010). As an example, long-term studies performed on river catchments in Northern Italy revealed that both increasing temperature and change in precipitation regime were important factors in the long- and short-term NO_3 dynamics in river water (Rogora, 2007). The role of meteorological factors was particularly important in mountain areas, where acid-sensitive freshwater sites are located: temperature and snow cover, as both amount and duration, were important drivers in the recovery trajectory of alpine lakes in the Central Alps (Rogora et al., 2013). Furthermore, recent studies highlighted the prominent role of the cryosphere (retreating glaciers, permafrost thawing) in shaping the chemical and biological features of high altitude ecosystems (Rotta et al., 2018), prompting the need for further studies on the interaction between climate–related changes and air pollution.

4. Measurements and impacts of air pollution on human health in Italy

Air pollution is the largest environmental risk for human health, with > 450.000 premature deaths and 1.6 billion US$ of economic cost due to mortality and disease in Europe in 2010 (WHO, 2015). Cardiovascular and cerebrovascular diseases are the most common reasons for premature death attributable to air pollution and account for 80% of the total premature deaths attributable to air pollution; lung diseases and lung cancer are also common (WHO, 2015).

For Italy, the most recent assessment by the European Environmental Agency calculated 60,600 premature deaths caused by $PM_{2.5}$, 20,500 by NO_2, and 3200 by O_3 in 2015 (EEA, 2018).

Few papers report local effects of air pollution on exposed population in Italian cities or Regions and possible improvements associated to a reduction of pollution. Lagravinese et al. (2014) analyzed the impact of air pollution, considering data from local monitoring stations, on hospital admissions for chronic obstructive pulmonary diseases (COPD) in 103 Italian provinces, and found a clear association between high levels of PM and hospitalization for children, while O_3 explained the hospital admissions for elderly people. Carugno et al. (2017) focused on selected areas of the Lombardy Region and evaluated the PM_{10}-attributable deaths over a 13-year period (2003–2014). The authors reported the estimated PM_{10} effect as about 1% of all natural deaths (min-max range: 0.86%–1.42%) with clear heterogeneity across areas. Noteworthy, the population-weighted exposure levels, calculated using the validated data from the monitoring stations of the Regional agency for environment protection (ARPA Lombardia), decreased during the analyzed period, except for a peak in 2011, and consequently decreased the number of deaths associated to PM exposure, which declined from 343 annual deaths in the period 2003–2006, to 253 in 2007–2010, and to 208 in 2011–2014.

Giannini et al. (2017) reported the estimated attributable deaths in Emilia Romagna region (north-central Italy) over the period 2006–2010 for PM_{10} and $PM_{2.5}$ by using both the data from the regional air quality monitoring network data and the simulation from a modeling suite. The results showed that for the entire region the number of deaths was 4.4 and 2.8 per 100,000 inhabitants for PM_{10} and $PM_{2.5}$ in 2010. In Rome in 2016, 0.6% and 1.5% of hospital admissions due to cardiovascular and respiratory diseases, and 18.1% and 9.2% of mortality for ischemic heart disease and COPD were attributed to $PM_{2.5}$ (De Marco et al., 2018).

Significantly, two projects (VIIAS and EU LIFE+ MED HISS) estimated the effects of air pollution on human health at national scale providing estimations of the air pollution impacts. The health gains of Italian people were quantified in VIIAS by carrying out baseline assessment scenarios on years 2005 and 2010, after changes in targets and national policies. The health impact assessment results highlighted the relevant impact of $PM_{2.5}$ and NOx on mortality, ranging from 23,000 to 34,000 attributable deaths. Northern Italy showed the highest number of deaths related to $PM_{2.5}$ (22,485) and NO_x (14,008) with respect to Central and Southern Italy (Ancona et al., 2015). Remarkable differences were also found between urban and non-urban areas, with deaths attributable to NO_2 of 16,736 and 6651, respectively. The possible benefits for exposed population were evaluated considering 2020 as the target year and modeling different future scenarios (Current Legislation Scenario (CLE), compliance with EU and Italian air quality standards – CLE+ Target 1, CLE concentrations reduced by 20% - CLE+ Target 2). In terms of expected mean exposure concentration, the project estimated a mean population weighted exposure of 15.8 μg/m3 and 18.1 μg/m3 in 2010 and 2020, respectively. This difference in the expected exposure to fine PM determined a 32.8% increase of PM attributable deaths in 2020, compared to $PM_{2.5}$-associated deaths in 2010. Significant reductions of pollutant concentrations and related effects were observed for the other modeled scenarios (CLE+ Target 1 and CLE+ Target 2) (Ancona et al., 2015).

MED HISS project aimed to set up a surveillance system of long-term effects of air pollution in four Countries (Italy, France, Slovenia and Spain) based on air quality and health data provided by: the National Health Interview Surveys (NHIS), mortality and hospital admissions registries and air pollution models (Gandini et al., 2018). MED HISS results for Italy were consistent with VIIAS project, and the highest mortality related to pollution was found in Northern Italy and urban areas. It was important to note that 33,353 (20,429–41,368) deaths were related to $PM_{2.5}$ for 2010, 14 years of life were lost on average for each death and 9.2 months of reduction of life expectancy for everyone are found (Ancona et al., 2015). These results are in line with data reported by Kiesewetter et al. (2015) and highlight relevant benefits for human health because of policy application to reduce air pollution.

Though there is limited quantitative evidence of the health benefits due to emission reduction strategies in urban areas of Italy (Schiavon et al., 2015; De Marco et al., 2018), it is well-known that the main sources of primary air pollution are road traffic, residential heating and industry. Nonetheless it is worthy to underline that NH_3 and NMVOC emissions, from agriculture and industrial productions respectively, play an important role in determining secondary pollution in several areas of Italy. Therefore, a comprehensive approach for reducing PM concentrations for health protection should consider all the cited sectors, aiming at activating integrated reduction measures, such as low traffic emission zones in city centers, ban of older vehicles and heating devices, active mobility, and smart use of fertilizers.

5. Future developments of the monitoring network

In Europe, the Mediterranean basin is the region with the highest variability of biogeographic, geomorphological and climatic features (Wigley, 1992). In particular, the Italian territory has inherently high level of naturalness (sensu Blasi et al., 2007), with an extraordinary concentration of species and habitat diversity. At world level, Italy is recognized as part of Priority eco-regions (Capotorti et al., 2012). Italy hosts important hot spots of biodiversity with about 6711 vascular plant species (MATTM, 2005) that represent about half of the plant species of Europe. In this context, to identify monitoring sites which are representative of all Italian ecosystems is a challenge.

Specific guidelines for the selection of the sites to be included into the network were indicated by the EC, according to climate condition and ecosystem type (MAES, 2016). The location, number and density of sites depend on the sensitivity of ecosystems, the number of different habitats, the intensity of air pollution pressure and the ecosystem area affected by air pollution (NECD Guidance, 2017). The chosen monitoring sites should fulfill the following principles:

- ✓ the site characteristics should be such that impacts of aerial deposition can be distinguished from other pressures;
- ✓ the site should be sensitive to a given pressure such that if there are any impacts they would be readily identifiable;
- ✓ the site should be typical for the ecosystem and habitat to be monitored.

The European long-term monitoring programs on natural ecosystems already existing over a Member State, such as ICP Forests, ICP Waters, Long-Term Ecological Research (LTER), and national network, are an ideal framework for transposing the NECD requirements into National legislation, as Italy did by the legislative decree n. 81 on 30 May 2018. Also, sites where the NECD parameters are monitored in the long-term may be considered for inclusion in the national monitoring network, even though they are not (yet) included into the European long-term monitoring programs cited above. Usually, these sites are established by other European-funded programs, such as the LIFE (https://ec.europa.eu/easme/en/life) and INTERREG (https://www.interregeurope.eu/) programs. An implementing decree will establish the structure of the monitoring network, defining the number and

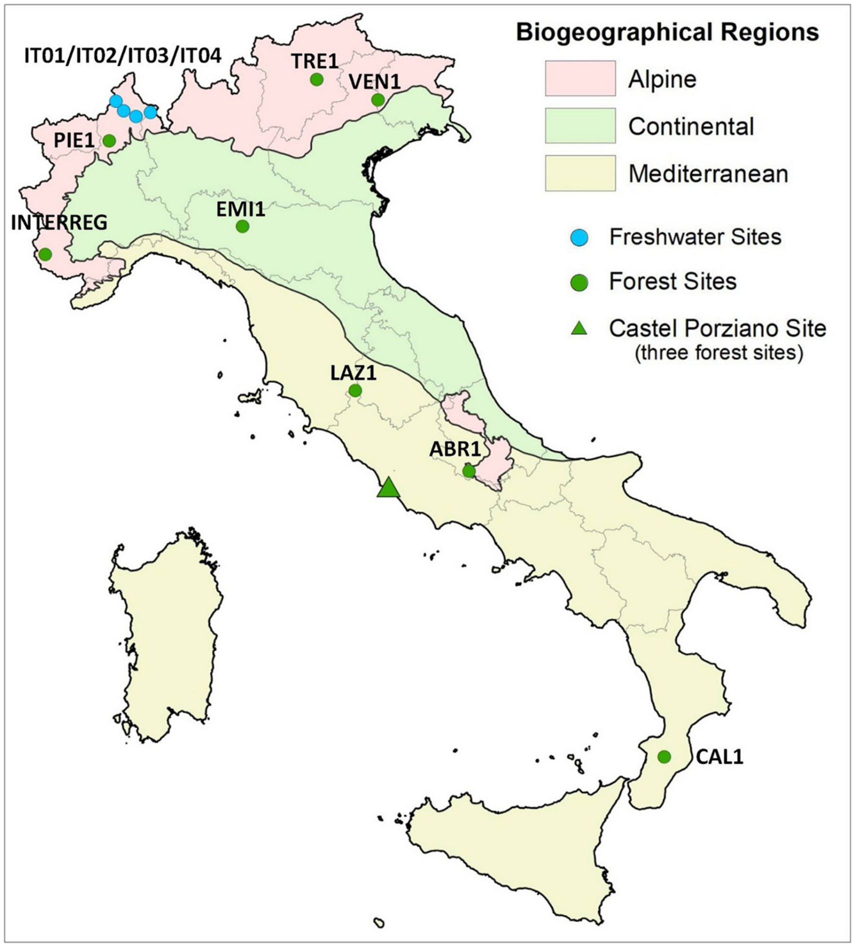


Fig. 6. Location of the sites identified for the NEC Directive implementation in Italy.

location of the selected sites, the parameters to be assessed and the related methodologies, reporting obligations.

Following these guidelines, a selection of the most interesting ecosystems and species for Italy was done based on representativeness in terms of vegetation type (Paoletti, 2006) and climate (Brunetti et al., 1999), in order to cover the whole territory and the different biogeographic regions (https://www.eea.europa.eu/data-and-maps/data/biogeographical-regions-europe-3) (Fig. 6). The strength of this selection was based on four main elements: 1) the long experience of monitoring in the framework of CLRTAP; 2) the solid scientific basis and well-known institutions doing the job; 3) the possibility of coordination among public Institutions; 4) the exchange of experience at European/Mediterranean level.

The monitoring network identified for Italy contains sites distributed over the territory and will produce a high number of monitored parameters. The numbers of sites selected is not high, but the parameters monitored are in strict agreement with the list provided by the NECD. In detail, four sites sensitive to both acidification and nitrogen deposition where long-term data are collected (ICP Waters/LTER) were identified for water bodies monitoring, all of them located in the north alpine region, because this is considered a pristine area in Italy not affected by other anthropogenic sources of air pollution, where the contribution of transboundary air pollution can be distinguished from other pressures; 6 sites for terrestrial ecosystem monitoring (ICP Forests), for both liquid and solid phases monitoring, distributed on a north to south gradient (4 sites for a latitudinal transect of *Fagus sylvatica* and the other two sites in the Mediterranean area characterized by *Quercus petrea* and *Quercus cerris*); 11 sites for ozone and meteorology (LIFE/INTERREG/ICP Forests), distributed in consideration of the different biogeographic areas and habitat distribution (in addition of the species listed before for terrestrial ecosystem, three typical Mediterranean species: *Phyllirea latifolia*, *Pinus pinea* and *Quercus ilex*).

There are of course some critical aspects: mainly in terms of representativeness of the complex and high diverse territory at National scale (i.e. only one point in the South of Italy) and in terms of species. Therefore, an update of the network will be necessary in the future to improve the information we will get from monitoring and obtain a more complete dataset on the impacts of air pollution on different natural ecosystems. In the future, to improve the representativeness of the monitoring network, the collaboration between researchers working on nature protection and biodiversity conservation should be enhanced, to realize more complete and detailed mapping of natural habitats and ecosystems that could be relevant to fulfill the aims of the NECD. Indeed, it would be noteworthy to evaluate a possible integration of other existing measurement activities and networks (in particular ICOS) towards the creation of integrated key-sites at national level.

An increase of representativeness could be provided by a higher density of monitoring sites, thus ensuring an adequate consistency of data. The main constraint to create a larger monitoring network is the economic constraint, as the philosophy was to maximize the results while minimizing the expenses. The main concerns were the low available resources at the moment and the low possibility to assure long term monitoring activities into the future. However, different EU funded projects such as the LIFE project SMART4Action (http://ec.europa.eu/environment/life/project/Projects/index.cfm?fuseaction=search.dspPage&n_proj_id=5035&docType=pdf) showed that saving maintenance costs of the stations and keeping a high level of accuracy in the measured values is possible (Ferrara et al., 2017). The merging between long-term monitoring programs and high-resolution air-quality model could be a possible and desirable solution, especially in a context of changing climate (De Marco, 2009).

Regarding O_3 and carbon fluxes, for the moment, due to lower costs, the choice was to include into the monitoring just O_3 sites. The monitoring of carbon flux is carried out in Italy at 9 active sites candidate to be included into the ICOS network.

For water bodies, to extend the spatial representativeness of the network, a higher number of sites would be included, covering a wider range of deposition inputs and sensitivities. Furthermore, a proper sampling frequency should be identified, to accomplish for the seasonal variability of water chemistry in relation to meteorological factors. As an example, a monitoring network of about 30 high altitude lakes and streams, which have proved to be most sensitive sites to air pollution, should be established, covering a range of ANC from highly sensitive (0–20 $\mu eq\,L^{-1}$) to moderately sensitive sites (20–200 $\mu eq\,L^{-1}$). These sites should be monitored in different periods of the year, including the snowmelt period, to catch the most critical condition for the biota (Stoddard, 1995; Lepori et al., 2003).

Indeed, historically, when looking at the impacts of air pollution on ecosystems, climate is generally not very well considered for the estimation of effects of air pollution on ecosystems (Paoletti et al., 2010). For example, local temperature, soil moisture, solar radiation and relative humidity can change the stomatal conductance of the leaves, making plants more or less sensitive to gaseous pollutants uptake (Manes et al., 2007; Fares et al., 2013; Anav et al., 2018). For this reason, air pollution impacts on ecosystems cannot be estimated neglecting the contribution of climate and, in some cases, the beneficial effects of policies aimed at reducing air pollution could be counteracted by climate change (Anav et al., in preparation). For this reason, we propose to involve climate stakeholders and experts into the discussion of the effects of air pollution on ecosystems. The policies to reduce air pollution impacts should consider the effects of climate to be more effective.

6. Conclusions

Air pollution issue is a well-established environmental policy area at national and European level. The establishment of legal limits for ambient concentration of air pollutants and the implementation of mitigation strategies on air-free emissions, in terms of both national totalities and/or specific sources or sectors, resulted in decreased emissions of air pollutants and noticeable improvements in air quality in Europe (EEA, 2018). This picture is in line with observations reported in this study for Italy. However, in Italy as well as in Europe, air pollution is still matter of concern, as clearly depicted, for human and ecosystem health leading to market and non-market costs (OECD, 2016).

The implementation of the NECD (2016/2284/EU) and of Article 9, ensures that the European countries will carry out monitoring activities able to provide risk assessment and quantification for the pollutant effects covered by the Directive itself (van Grinsven et al., 2016). In Italy, the monitoring network identified for NECD implementation, will fulfill the requests by Article 9 of the Directive, giving long-term information on the impacts of air pollution on ecosystems, and consequently on human health.

Moreover, as the European Commission stated (COM, 2018), the effect of the NECD in reducing the air pollutant impact in 2030 will depend on the full implementation by the Member States of all the measures and on the definition of robust National Air Pollution Control Programmes. Even with these measures, the current analysis shows that the EU will be far from its long-term objective of no exceedance of eutrophication critical loads, in spite of improvements. D'Elia et al. (2018) showed that the attainment of the NEC ceilings in Italy could not guarantee the compliance with EU air quality limits (especially of daily PM10, annual PM2.5 and daily maxima of 8-h running mean of O3 concentrations), suggesting that integrated and effective strategies should be designed to allow the compliance of both emission ceilings and health and environment protection.

The experimental data collected in the framework of European Programs (CLRTAP, LIFE, INTERREG, H2020) allowed to find dose-response relationship evidences, a key step for defining the critical loads/levels for each compound and plant species (Braun et al., 2017; Manes et al., 2003; Salvatori et al., 2013; Sicard et al., 2016). However, much work is required, for instance, to estimate the interaction with climate change, or to include other environmental impacts on ecosystems not covered by the NECD (e.g. materials, cultural heritage) and for the integration of experimental and modeled data.

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