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Air quality status and trends in Europe

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HIGHLIGHTS

- We analyse status and trends of air quality in Europe from 2002 to 2011.
- We estimate the exposure of urban EU population and ecosystems to air pollution.
- Current air pollution levels still have adverse effects on health and ecosystems.
- Particulate matter and ozone are Europe's most problematic pollutants for health.

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ABSTRACT

This paper presents an overview and analysis of air quality in Europe from 2002 to 2011. It reviews progress towards meeting the requirements of the EU air quality directives, as well as the development of European air pollutant emissions over the last decade. An overview of the latest findings and estimates of the exposure of urban population and ecosystems to air pollution in Europe is also given. The evaluation of the status and trends of air quality is based on ambient air measurements, in conjunction with data on anthropogenic emissions and their trends. The analysis covers up to 38 European countries, including EU Member States and member countries of the European Environment Agency (EEA) as of 2011.

1. Introduction

Humans and the environment in Europe are exposed to a complex mixture of many air pollutants emitted from various sources and subject to atmospheric processes that can create new pollutants. Many of these pollutants can cause severe health problems and impact on ecosystems. Understanding the status and developments of air quality levels in Europe is crucial to support European, national and regional policy development and implementation, as well as to inform the research and innovation communities of the most important challenges in air quality assessment and management that needs to be met in the near and medium-term future.

Despite successful legislation seen over several decades and success in dealing with certain emissions to air, a large part of the European population is still breathing air with air pollution levels exceeding the EU standards and the World Health Organisation air quality guidelines (WHO AQG) for the protection of health. The current air pollution levels continue to cause important impacts on human health, as well as on ecosystems in Europe.

Emissions of air pollutants derive from almost all economic and societal activities. In Europe, emissions of many air pollutants have decreased. Much progress has been made in tackling air pollutants such as sulphur dioxide (SO_2), carbon monoxide (CO), lead (Pb) and benzene (C_6H_6). On the other hand, road transport, industry, power plants, households, and agricultural activities continue to emit significant amounts of air pollutants. Combustion of biomass and solid fuels by households is an important source of directly emitted particulate matter (PM) and polycyclic aromatic hydrocarbons (PAHs).

The air quality status and trends presented in this paper were prepared for the European Environment Agency. Additional details on the work presented in this paper can be found in the EEA (2013a) report.

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2. Methodology

2.1. Air quality measurements

The evaluation of the status and trends of air quality is based on ambient air measurements reported by the European countries to the European Environment Agency (EEA) as of 2011, for the period from 2002 to 2011, and available in Airbase v. 7 (EEA, 2013g). The analysis covers all the regulated pollutants in the Air Quality Directives (EU, 2004 and EU, 2008) in the EU-27 and the EEA-32 member countries, i.e. the EU-27 Member States and the remaining five EEA member countries (Iceland, Liechtenstein, Norway, Switzerland and Turkey). In some cases, the analysis cover, in addition to the EEA-32 member countries, the new EU Member State Croatia and five EEA cooperating countries (Albania, Bosnia-Herzegovina, the former Yugoslav Republic of Macedonia, Montenegro, Serbia) where data is available.

Only monitoring stations with at least 75% of data coverage (fraction of the year for which valid concentration data is available) were considered in the analysis of PM with diameters of 10 μm or less (PM10), fine particles with diameters of 2.5 μm or less (PM2.5), ozone (O_3), nitrogen dioxide (NO_2), SO_2, and CO. For the analysis of benzene concentrations, the data coverage criterion was set to 50%, in accordance with the requirements of the European Commission working group on benzene (EU, 1998). For toxic metals (arsenic, cadmium, lead, mercury, and nickel) and benzo(a)pyrene (BaP), a data coverage requirement of at least 14% was applied, corresponding to the data quality objective set in the EU Directives (EU, 2004 and EU, 2008) for indicative measurements.

2.2. Trend analysis

For detecting and estimating the trends of measured ambient concentrations the nonparametric Mann–Kendall's test (Gilbert, 1987) for testing the presence of the monotonic increasing or decreasing trend was used. In addition, the nonparametric Sen's method for estimating the slope of a linear trend (as change per year) was applied. The calculated trends were considered as statistically significant for levels of significance 0.1 (corresponding to 10% chance there is no trend) or lower.

The trends of PM10, O_3, and NO_2 were calculated based on the officially reported data in Airbase v. 7 from a consistent set of stations with a minimum data coverage of 75% of valid data per year for at least 8 years out of the 10 years period. PM concentrations not measured with the reference method (gravimetric) must be corrected with a correction factor determined in a demonstration of equivalence of the non-reference methods (e.g. BAM and TEOM) used by the countries (EU, 2008). de Leeuw and Fiala (2009) discusses the uncertainties caused by using non-reference methods for PM10 and explains the corrections done to the French PM10 data prior to 2007. Due to the fact that the number of stations with available PM2.5 long time series is limited, the calculation of PM2.5 trends was done for the period 2006–2011 and for stations with a minimum data coverage of 75% of valid data per year for at least 5 out of the 6 years period.

National trends are calculated by averaging the trends estimated at individual stations. Stations located outside continental Europe (French overseas departments, Canary Island, Azores, Madeira) are excluded.

For some of the EU air quality standards it is allowed to exceed the standards' threshold concentration during a number hours or days. This is the case for e.g. the daily PM10 limit value (LV) (35 days with concentrations above 50 $\mu g/m^3$ are allowed per year), the O_3 target value (TV) for the protection of human health (25 days per calendar year may exceed 120 $\mu g/m^3$ of maximum daily 8-h mean concentrations), and the hourly NO_2 LV (allowing 18 h in the year with concentrations above 200 $\mu g/m^3$). In order to have a more statistically robust calculation of the trends for these standards, taking into account that up to 25% of data coverage may be missing per year, the trends of the corresponding percentiles were calculated, instead of the Kth highest value (de Leeuw, 2012). For the PM10 daily LV, the 90.4 percentile of daily concentrations was used; for the O_3 TV for the protection of human health, the 93.15 percentile of maximum daily 8-h mean concentrations was used; and for the NO_2 hourly LV, the 99.79 percentile of hourly concentrations was used to calculate the trends.

2.3. Estimation of urban population and ecosystem exposure

The monitoring data in AirBase v. 7 provide the basis for estimating the exposure of the urban European population to concentrations above the EU limit values, target values and long-term objective for the protection of human health, as well as to concentrations above the WHO AQG. For each city included in the Urban Audit (Eurostat, 2013) an average concentration is calculated based upon the measurements carried out at all urban or sub-urban background monitoring stations with a minimum data coverage of 75%. The exposure of the population in that urban area is estimated considering that the entire population in that city is potentially exposed to the calculated average concentrations, since people move freely within the city (EEA, 2013b).

The estimation of exposure of European ecosystems to acidification, eutrophication and ozone is based on the EEA's indicator CSI 005, latest updated for 2010 (EEA, 2013c).

2.4. Emission data

Nitrogen oxides (NO_x), CO and non-methane volatile organic compounds (NMVOC) emissions were downloaded from EEA Air pollutant emissions data viewer (LRTAP Convention) (http://www.eea.europa.eu/data-and-maps/data/data-viewers/air-emissions-viewer-lrtap) (EEA, 2013e) and methane (CH_4) emissions from the EEA greenhouse gas data viewer (http://www.eea.europa.eu/data-and-maps/data/data-viewers/greenhouse-gases-viewer) (EEA, 2013f) for the period 2002 to 2011.

3. Main findings by air pollutant

3.1. Particulate matter (PM)

In terms of potential to harm human health, PM poses the greatest risk, as it penetrates into sensitive regions of the respiratory system and can lead to health problems and premature mortality (WHO, 2013). PM in the air has many sources and is a complex heterogeneous mixture. The sizes and chemical composition of this mixture can change in time and space, depending on emission sources and atmospheric and weather conditions.

PM in the atmosphere originates from primary particles emitted directly and secondary particles produced as a result of chemical reactions involving PM forming (precursor) gases: SO_2, NO_x, NH_3 and non-methane volatile organic compounds (NMVOC). Emissions of primary PM10 and PM2.5 decreased by 14% and 16% respectively in the EU-27 between 2002 and 2011. The reductions in the same period for the EEA-32 member countries were 9% for PM10 and 16% for PM2.5.

PM precursor emissions continued to decrease between 2002 and 2011. In the EU-27 Sulphur oxides (SO_x) emissions fell by 50%; NO_x emissions fell by 27%; NH_3 emissions fell by 7%; NMVOCs emissions fell by 28%. In the EEA-32 countries: SO_x emissions fell by

34%; NO_x emissions fell by 23%; NH_3 emissions decreased by 5%; NMVOCs emissions fell by 27%.

These results notwithstanding, not all economic sectors have reduced emissions during this period. For example, the "commercial, institutional and household fuel combustion" sector, which dominates emissions of primary PM10 (39% of EU-27 in 2011) and PM2.5 (59% of EU-27 in 2011), has slightly increased its PM emissions since 2002. The agricultural sector has also had a slight increase in its primary PM10 emissions since 2002, and only a slight decrease in its PM2.5 and NH_3 emissions. The agricultural sector was responsible for 11%, 3% and 93% of respectively PM10, PM2.5 and NH_3 emissions in EU-27 in 2011. In addition, in 2011 eight EU Member States exceed one or more emission ceilings set under EU legislation. For example, reported emission data for 2011 for NO_x, NMVOC and NH_3 shows emissions higher than the respective ceilings in several Member States, which should have been reached in 2010 (EEA, 2013d). Table S5 in supplementary material shows a comparison between the emissions of NO*x*, NMVOC, SO_2 and NH_3 reported by the Member States and the emission ceilings set by the NEC directive (EU, 2001) for 2010.

Despite the emission reductions, 20%–44% of the EU-27 urban population was exposed to concentrations of PM10 in excess of the EU air quality daily limit value (50 μg/m^3, Table 1) in the period 2002–2011. For the EEA-32 countries exposure is even higher with between 25% and 49% of the urban population exposed to PM10 concentrations in excess of the daily limit value in the same period; the high PM10 levels observed in Turkey are mainly responsible for this increase. In 2011, 33% of the urban population in EU-27 lived in areas where the EU air quality daily limit value for PM10 was exceeded (Fig. 1), while for the EEA-32 countries the estimate is 49%. Although ambient concentrations of PM10 have slightly decreased during the past decade, there was no discernible downward trend in this particular indicator (Fig. 1). Considering the WHO AQG (20 μg/m^3 for PM10 annual mean), 88% of urban dwellers were exposed to PM10 concentrations that exceeded the WHO AQG for the protection of human health in 2011 (Fig. 1).

In terms of exposure to PM2.5, 31% of the EU-27 urban population was exposed to PM2.5 concentration levels above the exposure concentration obligation, to be attained by 2015 (20 μg/m^3 for PM2.5 annual mean, Table 1) and 96% was exposed to levels above the WHO AQG (10 μg/m^3 for PM2.5 annual mean) in 2011.

The EU limit and target values for PM (Table 1) were exceeded widely in Europe in 2011, with the PM10 daily limit value being exceeded in 22 European countries (Fig. 2). Exceedance of the target value for PM2.5 (annual average of 25 μg/m^3 to be met in 2015) was observed in 11 EU Member States at one or more stations in 2011, mostly in Eastern Europe.

Exceedances of the PM10 daily limit value were measured at 43% of traffic sites, 38% of urban and sub-urban background sites, 26% of stations classified as 'other' (mostly industrial), and even at 15% of rural background sites within the EU-27. The percentage of stations in exceedance at rural background sites has more than doubled from 2009 to 2011. The PM2.5 target value was exceeded at 10% of traffic sites, 18% of urban and suburban background sites, 7% of 'other' (mostly industrial) sites and at 5% of rural sites.

The WHO AQGs for PM10 and PM2.5 annual mean concentrations, which are stricter than the limit and target values set by EU legislation, were exceeded at the majority of monitoring stations across continental Europe.

Trends of measured PM10 concentrations were computed for both the annual means and for the daily 90.4 percentile concentrations, for the period 2002–2011. It is important to note that most of the stations with available data for this trend analysis are situated in France and Germany. Most of the stations (62%) did not register significant trends for both PM10 indicators. On the other hand, most of the registered significant trends are decreasing. In general, the largest decrease is observed at traffic stations indicating the efforts taken to reduce road transport emissions. Table S1 in supplementary material shows the average trends and their 95% confidence limits by country and by station type of PM10 daily mean 90.4 percentile, corresponding to the most stringent and frequently exceeded PM10 limit value.

While the trends of PM10 are in average decreasing across Europe, the same is not registered for PM2.5 concentrations. Due to lower availability of data to compute PM2.5 trends, this analysis is more limited both in spatial and temporal coverage, than for PM10 trends, and should therefore be considered with caution and taking into account the considerable uncertainties. Table S2 in supplementary material shows the average trends and their 95% confidence limits by country and by station type of PM2.5 annual mean. 79% of the stations did not register significant trends of PM2.5 in the period 2006–2011. The average trends at background stations are slightly increasing, especially in urban-suburban areas, which is a development of considerable concern. These national increasing trends (e.g. in France) would need to be reversed in order to meet the targets set by the Directive (EU, 2008), particularly the National Exposure Reduction Targets for PM2.5 for the period 2010–2020, based on urban background measurements. On the other hand, the average trend measured at traffic stations was slightly decreasing in the period 2006–2011, but increasing in a few countries.

Table 1
Air quality limit and target values and long-term objectives defined in the directives (EU, 2004; EU, 2008), and WHO air quality guidelines (WHO, 2006, 2000).

Pollutant	Averaging period	Limit or target value		LTO	WHO AQG
		Value[e]	Max. nr. allowed occurrences	Value[e]	Value[e]
PM10	24 h	50	35		50[a]
	Annual	40			20
PM2.5	24 h				25[a]
	Annual	25		8.5 –18	
	Annual	20[c]			10
O_3	8 h daily max	120	25	120	100
	AOT40[d]	18 000		6000	
NO_2	1 h	200	18		200
	Annual	40			40
NO_x	Annual	30			
BaP	Annual	1 ng/m^3			0.12 ng/m^3[b]
SO_2	10 min				500
	1 h	350	24		
	24 h	125	3		20
	Annual & winter	20			
CO	1 h				30 mg/m^3
	8 h daily max	10 mg/m^3			10 mg/m^3
Arsenic	Annual	6 ng/m^3			
Cadmium	Annual	5 ng/m^3			5 ng/m^3
Nickel	Annual	20 ng/m^3			
Lead	Annual	0.5			0.5
Benzene	Annual	5			1.7[b]

[a] 99th percentile (3 days/year).
[b] As the WHO has not set AQG for BaP and benzene the WHO reference level in the table was estimated assuming an additional lifetime risk of 1×10^{-5}.
[c] The exposure concentration obligation for PM2.5, to be attained by 2015, is based on a 3-years average measured at urban background stations.
[d] AOT40 is the accumulated dose of O_3 over a threshold of 40 ppb, equivalent to 80 μg/m^3, May to July. The unit is (μg/m^3) h.
[e] Units in μg/m^3, if not specified.

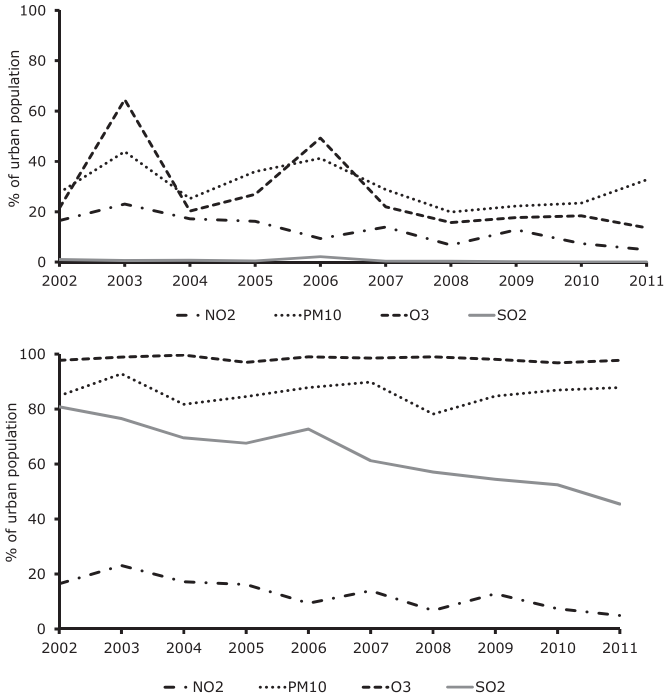

Fig. 1. Percentage of the EU-27 urban population potentially exposed to air pollution exceeding allowable EU air quality standards (top) and WHO air quality guidelines (bottom).

3.2. Ground-level O_3

Ozone is a secondary pollutant formed in the troposphere by complex chemical reactions following emissions of precursor gases such as NO_x and NMVOCs. At the continental scale, CH_4 and CO also play a role in O_3 formation. Ozone is a powerful and aggressive oxidising agent, elevated levels of which cause respiratory health problems and lead to premature mortality (WHO, 2013). High levels of O_3 can also damage plants, leading to reduced agricultural crop yields and decreased forest growth. Tropospheric O_3 contributes also directly and indirectly to global warming.

Ozone precursor gas emissions decreased considerably between 2002 and 2011. In the EU-27: NO_x emissions decreased by 27%; NMVOC emissions decreased by 28%; CO emissions decreased by 32%; CH_4 emissions decreased by 15%. While in the EEA-32 Member Countries: NO_x emissions decreased by 23%; NMVOC emissions decreased by 27%; CO emissions decreased by 27%. CH_4 emission data for all EEA-32 Member Countries was not available. Ozone in Europe results also from precursor gases emitted elsewhere. For example, increased global emissions of CH_4 lead to higher concentrations of CH_4 in Europe, which in turn contribute to the formation of O_3.

Between 14% and 65% of the EU-27 and EEA-32 urban population was exposed to O_3 concentrations above the EU target value for protecting human health (the maximum daily running 8-h mean value may not exceeding 120 μg/m^3 more than 25 times per year) in the period 2002–2011 (Fig. 1). The range partly reflects variations caused by meteorology, as dispersion and atmospheric conditions differ from year to year. The percentage of the EU-27 urban population exposed to O_3 levels exceeding the WHO AQG (8-h mean of 100 μg/m^3, which is stricter than the EU target value) is significantly higher, comprising 98% of the total urban population in 2011 (Fig. 1). High O_3 concentrations are most pronounced in southern

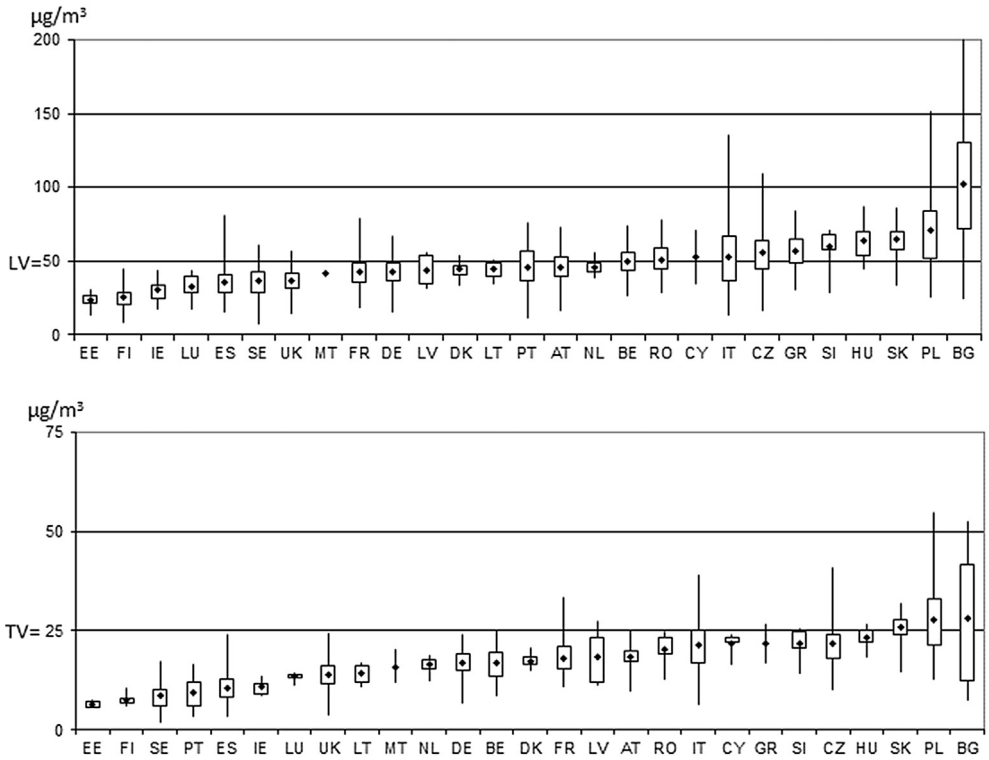

Fig. 2. Attainment situation for PM10 daily limit value (percentile 90.4) (top) and PM2.5 annual target value (bottom) in 2011 for each EU Member State, measured at all stations types. The diagrams indicate the lowest and highest observations, the means and the lower and upper quartiles.

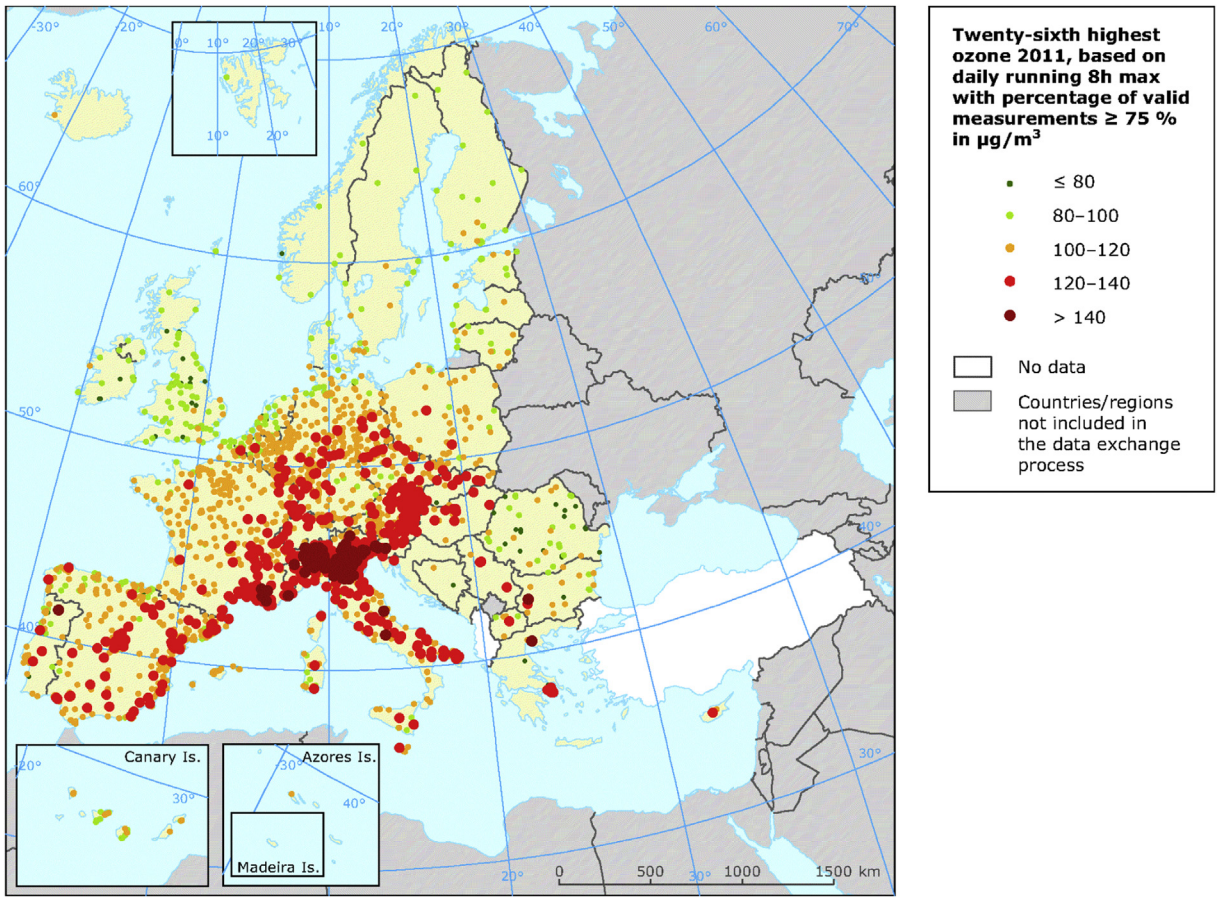

Fig. 3. 26th-highest daily maximum 8-h average O_3 concentration recorded at each monitoring station in 2011 (EEA, 2013a).

Europe, due to the more favourable climatological conditions for its formation (Fig. 3).

The health-related O_3 target value was exceeded more than the allowable 25 times in 2011 at 40% of the rural stations, 24% of urban/suburban background stations, 21% of industrial and 9% of traffic stations, over seventeen EU-27 countries. Conformity with the WHO AQG value for O_3 was observed only at three out of 513 rural background stations in 2011 (stations: Topolniky – Aszod, Slovakia; Glashaboy, Ireland; Great Dun Fell, UK). Only 2% and 3% of (sub)urban background and traffic stations, respectively, measured concentrations which did not exceed the WHO AQG in 2010.

O_3 also damages vegetation, including crops and forests, and O_3 concentrations outside urban areas are higher than in urban areas, due to the titration process of O_3 removal by urban NO emissions. Between 21% and 69% of agricultural crops in the EEA-32 were exposed to O_3 levels above the EU target value for protecting vegetation (18 000 (µg/m^3) h for AOT40, see Table 1) from 2002 to 2010, mostly in southern and central Europe (EEA, 2013c). The exposure to O_3 levels above the long-term objective for protecting vegetation (6000 (µg/m^3) h for AOT40) varied between 76% and 98% of agricultural crops in the same period. This long-term objective was only met in 15% of the total agricultural area in 2010, mainly in the United Kingdom, Ireland and the Nordic countries. In addition, 61% of the EEA-32 forest area was exposed to O_3 levels above the critical level for protection of forest (AOT40 accumulated over April to September) set by the LRTAP/UNECE to 10 000 (µg/m^3) h.

Trends of measured O_3 concentrations were estimated for two health-related statistics: the 93.15 percentile of maximum daily 8-h mean concentrations (as indictor for the EU target value for the protection of health) and for SOMO35,[1] for the period 2002–2011. There is no clear trend for O_3 concentrations (when considering the target value for the protection of health) between 2002 and 2011 in 80% of the monitoring stations (see Table S3 in supplementary material). 18% of the stations registered a statistically significant decreasing trend, while 2% registered a significant increasing trend, most of them in the Iberian Peninsula. In terms of SOMO35 trends, the situation is quite similar, with only 24% of the monitoring stations registering significant trends, mostly decreasing.

This analysis shows that there is a discrepancy between the cuts in emissions of O_3 precursor gases and the change in observed average O_3 concentrations in Europe. This apparent mismatch between the trends in the anthropogenic emission of precursors and the observed trends (or lack of trends) in ozone is also discussed in several publications (e.g. Colette et al., 2011; Wilson et al., 2012). Tropospheric ozone (O_3) interactions with vegetation, atmospheric chemistry and aerosols are complex, and only partially understood. Vegetation is both a source of ozone precursors (VOC, NO*x*) and a sink through deposition processes. One of the reasons for this mismatch in concentration trends may be increasing intercontinental transport of O_3 and its precursors in the northern hemisphere, which is likely to mask the effects of European measures to reduce O_3 precursor emissions. It is also unclear whether the lack of trends can be explained in some cases by the large interannual variability in O_3 concentrations due to meteorology, or

[1] SOMO35 is the accumulated O_3 concentration (daily maximum 8-h) in excess of 35 ppb (70 µg/m^3). It is used as an indicator of health hazards for exposure to ozone.

biomass burning events, or by uncertainties in the measurement data. The length of the period and the choice of O_3 parameter (percentile, mean values, etc) for trend analysis also influence the outcome of the trend estimate (Colette et al., 2011).

3.3. Nitrogen dioxide (NO_2) and nitrogen oxides (NO_x)

NO_x is emitted during fuel combustion, such as by vehicle engines, industrial facilities and domestic heating. Among the chemical species that comprise NO_x, NO_2 is associated with adverse effects on health, as high concentrations cause inflammation of the airways and reduced lung function (WHO, 2013). NO_x may also cause adverse effects on vegetation and contributes to the formation of secondary inorganic PM and O_3 with associated effects on health, ecosystems and climate.

EU emissions of NO_x fell by 27% in the period 2002–2011. Nevertheless, total NO_x emissions in 2011 were about 5% higher than the emissions ceiling for the EU as a whole set in the NEC Directive (EU, 2001) for 2010. Transport is the dominant sector for NO_x emissions, accounting for 47% of the total in 2011, followed by the energy sector, which contributed 21% of the total. These two sectors have substantially reduced emissions between 2002 and 2011; the transport sector by 31% and the energy sector by 27%.

In 2011, 21 of the 27 EU countries recorded exceedances of the NO_2 annual mean limit value at one or more stations. Exceedance of this limit value was reported for 42% of traffic stations and only 3% of all urban background stations in 2011. The estimate of the urban population in the EU-27 and EEA-32 exposed to NO_2 concentrations above the annual limit value and the WHO AQG for NO_2 in 2011 is of 5%. This estimate has varied between 5% and 23% between 2002 and 2011, with a decreasing tendency over this period. Since only concentrations measured at urban background stations are considered and 86% of the exceedances are measured at traffic stations, the percentage of urban population exposure to NO_2 concentrations above the limit value may be underestimated, as more than 20% of the European urban population lives less than 400 m from busy roads (EC, 2006).

Reactive nitrogen (N) compounds, emitted as both NO_x and NH_3, cause eutrophication of ecosystems and are now, after the very strong reduction in SO_2 emissions, the principal acidifying components in the air. The sensitive ecosystem area in Europe affected by eutrophication due to excessive atmospheric N deposition was reduced by 23% from 1990 to 2010. Despite this reduction, in 2010 there was still 44% of the EU-27 total area of sensitive ecosystems at risk of eutrophication and 2% is at risk of acidification (EEA, 2012).

Trends of measured annual mean NO_2 concentrations were computed for the period 2002–2011, by country and station type. For each type of station, around 5–6 countries have registered increasing NO_2 annual mean concentrations (see Table S4 in supplementary material). The trends are mostly non-significant, but some stations register significant increasing trends in Portugal, Poland and Luxembourg. Both Poland and Luxembourg have reported increased total and road traffic NO*x* emissions in the period, which explains the increase in NO_2 concentrations. While Portugal has reduced its NO*x* emissions in the period, its ratio diesel/gasoline consumption for road transport has almost doubled from 2002 to 2011, leading to a considerably higher NO_2 fraction of NO*x* emissions, which may explain the increase in NO_2 concentrations measured at Portuguese stations.

The decrease in NO_x emissions (27%) is considerably greater than the fall in NO_2 annual mean concentrations (ca. 8% measured at traffic stations) between 2002 and 2011. As for Portugal, this is attributed primarily to the increase in the share of NO_2 in the NO_x emissions from traffic, primarily due to the fitting of exhaust aftertreatment systems in diesel vehicles, and the shift in the photostationary state in favour of NO_2 that results from a decrease in NO_x, without an equivalent decrease in O_3 concentrations (Guerreiro et al., 2010).

3.4. Benzo(a)pyrene (BaP), a polycyclic aromatic hydrocarbon (PAH)

Benzo(a)pyrene (BaP) is a polycyclic aromatic hydrocarbon (PAH), formed mainly from the burning of organic material such as wood, and from car exhaust fumes, especially from diesel vehicles. It is a known cancer-causing agent in humans (WHO, 2013), and for this reason it is used as an indicator of exposure to harmful PAHs.

Benzo(a)pyrene measurements in 2011 were above the target value threshold (1 ng/m^3 annual average to be met by 2013) at 35% of the monitoring stations (Fig. 4). This was the case mainly at urban and suburban background stations (50% exceeded the target value) and, to a lesser extent, at rural, traffic and industrial stations. Exceedances are most predominant in central and eastern Europe.

Exposure of the European population to BaP concentrations above the target value is significant and widespread, especially in central and eastern Europe. Between 22% and 31% of the urban population in the EU-27 was exposed to BaP concentrations above the target value (1 ng/m^3) in the period 2009 to 2011, and in this time a tendency of increasing exposure can be observed. As much as 94% of the EU-27 urban population was exposed to BaP concentrations above the WHO reference level (0.12 ng/m^3) over the same period. In large parts of Europe the concentrations are expected to be below the lower assessment threshold (0.4 ng/m^3), and consequently the air quality directive (EU, 2004) does not require monitoring of BaP. As the WHO reference level is below the lower assessment threshold the estimate of the population exposure is highly uncertain.

Emissions of BaP in the EU-27 have increased by 11% between 2002 and 2011, due to the increase in emissions from the 'commercial, institutional and household fuel combustion' sector of 24%. In Europe, BaP pollution is an increasing problem, especially in areas where domestic coal and wood burning is common.

3.5. Sulphur dioxide (SO_2)

Sulphur dioxide is emitted when fuels containing sulphur are burned or from high temperature industrial processes involving raw materials high in sulphur content (such as smelters). It contributes to acidification, the impacts of which can be significant, including adverse effects on aquatic ecosystems in rivers and lakes; damage to forests and terrestrial ecosystems; as well as reduced biodiversity. SO_2 can affect the respiratory system and reduce lung function (WHO, 2013). It is also a major precursor to PM, which is associated with significant health effects.

In the period 2002–2011, EU-27 Member States cut their SO_x emissions by 50%, leading to a fall in SO_2 concentrations of about one third. The corresponding emission reduction in the EEA-32 countries in the same period was 34% and is probably the main reason why concentrations within EU-27 fell in average by one third and not more. Moreover, SO_2 concentrations are very low in large parts of Europe, i.e. around or below instruments detection limit, decreasing the accuracy of the trend calculation.

The area of sensitive ecosystems affected by excessive acidification from air pollution has shrunk by 92% from 1990 to 2010 mainly due to the strong reduction in SO_2 emissions (EEA, 2012).

The EU-27 urban population was not exposed to SO_2 concentrations above the EU daily limit value in 2011, but 46% was exposed to SO_2 levels exceeding the WHO AQG (Fig. 1). Exceedances of the EU daily and hourly limit values for the protection of human health were only registered at very few traffic or industrial stations.

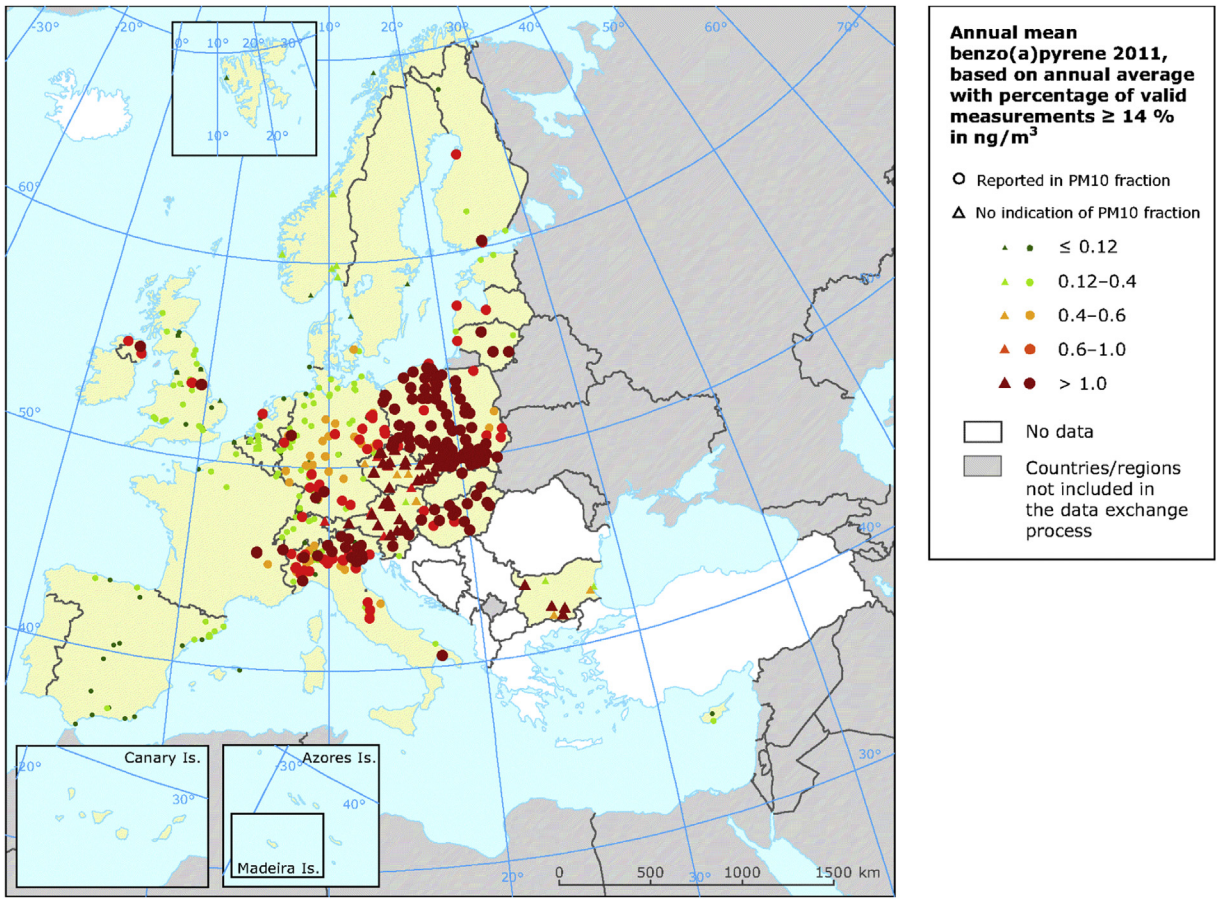

Fig. 4. Annual mean concentrations of benzo(a)pyrene (ng/m^3), 2011 (EEA, 2013a).

3.6. Carbon monoxide (CO)

Carbon monoxide is emitted due to incomplete combustion of fossil fuels and biofuels, and enters the body through the lungs. Exposure to CO can reduce the oxygen-carrying capacity of blood, thereby reducing oxygen delivery to the body's organs and tissues (WHO, 2000). The atmospheric lifetime of CO is about three months. This relatively long lifetime allows CO to slowly oxidise into carbon dioxide (CO_2), also forming O_3 during this process. CO therefore contributes to the atmospheric background concentration of O_3, with associated effects on the health of humans and ecosystems.

The observed EU-27 average reduction in CO daily 8-h maxima concentrations in the period 2002–2011 was 35%. The reduction registered at traffic stations in the period was of 37% and of 18% for urban background stations. These reductions in concentrations are is in line with the reported reduction in total emissions of about 32% over the same period. CO concentrations are now very low most of the time and instrument measurement uncertainties at these levels affect the accuracy of the measured concentrations and therefore also the accuracy of trend estimates.

Exposure of the European population to CO concentrations above the EU limit value and WHO AQG is very localised and sporadic. Only four out of 1003 stations in the EEA-32 registered exceedance of the EU limit value and WHO AQG in 2011.

3.7. Toxic metals

The toxic metals - arsenic (As), cadmium (Cd), lead (Pb), mercury (Hg) and nickel (Ni) – are emitted mainly as a result of various combustion processes and industrial activities. Toxic metals can reside in or be attached to PM and impact on human health when inhaled (WHO, 2013). As well as polluting the air, toxic metals can be deposited on terrestrial or water surfaces and subsequently build-up in soils or sediments. Toxic metals are persistent in the environment and may bio-accumulate in food-chains.

A relatively small number of stations measure concentrations in air of As, Cd, Pb and Ni in Europe, since levels are often below the lower assessment threshold set by EU legislation. An even smaller number of these stations have been operating for five or more years, rendering trend analysis difficult. In the case of Hg, only a few stations report concentrations in the air of different forms of Hg, making an analysis at the European level very difficult. The concentrations of As, Cd, Pb and Ni in air are generally low in Europe with few exceedances of limit values or target values. However, these pollutants contribute to the deposition and build-up of toxic metal levels in soils, sediments and organisms.

In the period 2002–2011 emissions of toxic metals in the EU-27 were reduced as follows: As was reduced by 3%; Cd by 27%; Hg by 26%; Ni by 43%; and Pb by 20%. Despite these cuts in emissions, a significant share of the EU-27 ecosystem area is still at risk of toxic metal contamination. Exceedances of Hg critical loads were estimated to occur at 54% of the sensitive ecosystem area in the EU-27 in 2010, while for Pb the estimated area in exceedance is 12% (Slootweg et al., 2010).

3.8. Benzene (C_6H_6)

Benzene is released during incomplete combustion of fuels used by vehicles. Other sources are domestic heating, oil refining, and

the handling, distribution and storage of petrol. Inhalation is the dominant pathway for benzene exposure in humans. Benzene is a carcinogenic pollutant and the most significant adverse effects from prolonged exposure are damage to the genetic material of cells, a phenomenon that can cause cancer (WHO, 2000).

Exceedances of the limit value for benzene were limited to very few locations (0.9% of stations) in Europe in 2011. Annual mean concentrations of benzene were highest in 2011 at traffic stations, as gasoline is still one of the most important sources of benzene. Concentrations measured at traffic and urban background stations decreased steadily until 2007, after which they stabilised. Benzene concentrations at urban and rural stations show a slower decrease during the same period than traffic stations. Whether benzene emissions recorded a similar stabilisation after 2007 is unclear because benzene is not included as an individual pollutant in European emissions inventories covering VOCs.

4. Discussion and concluding remarks

Emissions of the main air pollutants in Europe declined in the period 2002–2011, resulting, for some of the pollutants, in improved air quality across the region. These results notwithstanding, not all countries and economic sectors have had a satisfactory development in terms of emissions decrease (see Table S5 in supplementary material). While continuous progress has been observed in other sectors, air pollutant emissions from agriculture and the combustion of fuels to produce energy for domestic needs has either decreased very little (the case of agriculture), or not decreased (in the case of domestic fuel combustion) in the last decade. In fact, biomass combustion has become a more important source of air pollution (Saffari et al., 2013; OECD/IEA, 2013). This is because wood burning is often relatively cheap, and is considered as a climate friendly source of energy since it is renewable and carbon-neutral. Agriculture is responsible for 93% of ammonia emissions, which exert pressure on both human health and the ecosystems.

Emission reductions resulted in a notable reduction of ambient concentrations of SO_2, CO, and Pb. However, due to the complex links between emissions and air quality, emission reductions do not always produce a corresponding drop in atmospheric concentrations, especially for secondary pollutants like PM and O_3. For example, there is an apparent mismatch between the trends in the anthropogenic emission of precursors and the observed trends (or lack of trends) in ozone, also reported in several publications (e.g. Colette et al., 2011; Wilson et al., 2012) (see Discussion in chapter 3.2). European emissions are the most important contributors to O_3 and PM concentrations levels over Europe, but intercontinental transport of pollution also plays in and contributes to increased impacts on health, ecosystems and the economy (particularly crop productivity). The Relative Annual Intercontinental Response (RAIR) to emission reductions outside Europe is about 5% for PM and 43% for O_3 (UN, 2010). Hence, a substantial contribution from intercontinental transport affects the O_3 concentrations in Europe and to a lesser extent the PM. Further studies are needed to attribute European air quality exceedance to non-European emissions of air pollutants.

Air pollutant concentrations are still too high and harm human health and ecosystems. The effects of poor air quality have been felt the most strongly in two areas: in urban areas, where the majority of the European population lives, leading to premature mortality and increased morbidity; and in ecosystems, where the pressures of air pollution impairs vegetation growth and harms biodiversity.

Air pollution's most important effects on European ecosystems are damage to vegetation resulting from exposure to O_3, eutrophication and acidification. As SO_2 emissions have fallen, ammonia (NH_3) emitted from agricultural activities, and NO_x emissions from combustion processes have become the predominant acidifying and eutrophying air pollutants.

Concerning eutrophication, calculated exceedances of critical loads in 2010 cover most of continental Europe as well as Ireland and the United Kingdom. 44% of the EU-27 total area of sensitive ecosystems is at risk of eutrophication (EEA, 2012). On the other hand, the total area of sensitive ecosystems in the EU-27 that was in exceedance of critical loads of acidity in 2010 has been reduced to 2% (EEA, 2012).

A significant proportion of Europe's population lives in urban areas where exceedances of air quality standards occur. Table 2 gives an overview of the proportion of the EU-27 urban population exposed to pollutant concentration levels above the most stringent limit and target values for the protection of human health set in the EU legislation and the WHO AQGs in recent years (2009–2011). The current pollution levels, especially of PM, O_3 and BaP clearly impact on large parts of the urban population. This is particularly evident in the population exposure estimates based on the WHO AQG.

At present, PM and O_3 are Europe's most problematic pollutants. PM poses the most serious air pollution health risk in the EU, leading to premature mortality and increased morbidity, followed by O_3. In addition, Europe's sustained ambient O_3 concentrations continue to cause considerable damage to vegetation growth and crop yields, and reducing plant uptake of CO_2. Both effects constitute real losses for the European economy, the productivity of its workforce, as well as the health of its natural systems (EC, 2011).

Benzo(a)pyrene (BaP) is a carcinogen and an indicator for carcinogenic PAHs. A considerable proportion of the urban population in the EU-27 (22–31% between 2009 and 2011) were exposed to concentrations exceeding the EU target value to be met by 2013, and as much as 94% was exposed to BaP concentrations above the WHO reference level (estimated assuming an additional lifetime risk of 1×10^{-5}) over the same period (Table 2). Exposure to high levels of PAHs, including BaPs, which are carcinogenic substances, is a growing health concern in Europe. The increase in BaP emissions from domestic combustion in Europe over the last years is therefore a matter of concern, as it is aggravating the exposure of the European population to BaP concentrations, especially in urban areas.

The annual limit value for NO_2 is widely exceeded across Europe, but 86% of the exceedances in 2011 were measured at traffic

Table 2
Percentage of the urban population in the EU-27 Member States exposed to air pollutant concentrations above the EU and WHO reference levels (2009–2011).

Pollutant	EU reference value	Exposure estimate (%)	WHO reference level	Exposure estimate (%)
PM2.5	Year (20)	20–31	Year (10)	91–96
PM10	Day (50)	22–33	Year (20)	85–88
O_3	8-h (120)	14–18	8-h (100)	97–98
NO_2	Year (40)	5–13	Year (40)	5–13
BaP	Year (1 ng/m^3)	22–31	Year (0.12 ng/m^3)	76–94
SO_2	Day (125)	<1	Day (20)	46–54
CO	8-h (10 mg/m^3)	<2	8-h (10 mg/m^3)	<2
Pb	Year (0.5)	<1[a]	Year (0.5)	<1[a]
C_6H_6	Year (5)	<1	Year (1.7)	12–13

Notes: The reference levels in brackets are in μg/m^3 except for CO which is in mg/m^3 and BaP in ng/m^3. The comparison is made for the most stringent EU limit or target values set for the protection of human health. As the WHO has not set AQG for BaP and benzene, the WHO reference level in the table was estimated assuming an additional lifetime risk of 1×10^{-5}.

[a] The Pb concentrations reported by France for 2009 have been submitted in the wrong units and have therefore been corrected in the exposure estimates given above.

stations. In 2011, more than 5% of Europeans living in cities were exposed to NO_2 urban background levels above the EU limit value for annual average NO_2. In addition to its health effects, NO_2 is a major cause of eutrophication and acidification, and is also contributing to the formation of PM and O_3.

SO_2 concentrations have continued to decrease over the last decade, especially at urban and traffic stations. The most stringent EU limit value has not been exceeded in the urban background in the EU-27 since 2010. On the other hand, the urban population in the EU-27 exposed to SO_2 levels exceeding the WHO AQG is still significant, amounting to 46 %–54% of the total urban population between 2009 and 2011 (Table 2).

CO, benzene and toxic metals (As, Cd, Ni, Pb) concentrations in outdoor air are generally low, localised and sporadic in the EU, with few exceedances of the limit and target values set by EU legislation.

During the period addressed in this paper, 2002–2011, environmental policies and measures at the European level have affected the development of air pollutants emissions and the occurrence of air pollution. In terms of EU-27 total emissions, the NEC directive (EU, 2001) ceilings set for 2010 were met by 2011 for all pollutants except for NO*x* (emission ceiling set in annex II of the directive), which was exceeded by 5% (Table S5 in supplementary material). In contrast, a few countries exceeded their national emission ceilings for NMVOC and NH_3. Only the SO_2 emission ceilings were met by all countries in 2010 and 2011 (EEA, 2013d).

Recent policy measures have been and are under way, recognising the pressing need for action across Europe to improve air quality. EU's air policy has recently been under a comprehensive review process and in December 2013 a new clean air policy package was proposed, including:

- A new Clean Air Programme for Europe with measures to ensure that existing targets are met in the short term, and new air quality objectives for the period up to 2030,
- A revised National Emission Ceilings Directive with stricter national emission ceilings for the six main pollutants, and
- A new Directive to reduce pollution from medium-sized combustion installations, such as energy plants for street blocks or large buildings, and small industry installations.

Improving understanding of air pollution and developing effective policies for its reduction remains therefore a challenge and a priority in Europe.

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Appendix A. Supplementary data

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References

Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B., D'Angiola, A., D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouïl, L., Russo, F., Solberg, S., Stordal, F., Tampieri, F., 2011. Air quality trends in Europe over the past decade: a first multi-model assessment. Atmos. Chem. Phys. 11, 11657–11678. http://dx.doi.org/10.5194/acp-11-11657-2011.

EEA, 2012. Evaluation of Progress under the EU National Emission Ceilings Directive. Progress Towards EU Air Quality Objectives. European Environment Agency, Copenhagen. EEA Technical Report No 14/2012.

EEA, 2013a. Air Quality in Europe – 2013 Report. European Environment Agency, Copenhagen. EEA Report No 9/2013.

EEA, 2013b. Exceedance of Air Quality Limit Values in Urban Areas (Indicator CSI 004). http://www.eea.europa.eu/data-and-maps/indicators/exceedance-of-air-quality-limit-1/exceedance-of-air-quality-limit-5 (last accessed 06.03.14.).

EEA, 2013c. Exposure of Ecosystems to Acidification, Eutrophication and Ozone (Indicator CSI 005). http://www.eea.europa.eu/data-and-maps/indicators/exposure-of-ecosystems-to-acidification-2/exposure-of-ecosystems-to-acidification-5 (last accessed 06.03.14.).

EEA, 2013d. NEC Directive Status Report 2012. European Environment Agency, Copenhagen. EEA Technical Report No 6/2013.

EEA, 2013e. European Union Emission Inventory Report 1990–2011 Under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP). European Environment Agency, Copenhagen. EEA Technical Report No 10/2013.

EEA, 2013f. Annual European Union Greenhouse Gas Inventory 1990–2011 and Inventory Report 2013. European Environment Agency, Copenhagen, EEA. Technical Report No 8/2013.

EEA, 2013g. Airbase. http://www.eea.europa.eu/themes/air/air-quality/map/airbase (last accessed 15.07.14.).

EC, 2006. Development of a Methodology to Assess Population Exposed to High Levels of Noise and Air Pollution Close to Major Transport Infrastructure. European Commission. http://ec.europa.eu/environment/air/transport/pdf/final_report_main.pdf (last accessed 14.03.14.).

EC, 2011. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions 'Roadmap to a Resource Efficient Europe' (COM(2011) 571 final20.9.2011). European Commission, Brussels.

EU, 1998. Council Directive on Ambient Air Quality Assessment and Management Working Group on Benzene. Position Paper. http://ec.europa.eu/environment/air/pdf/ppbenzene.pdf (last accessed 20.03.14.).

EU, 2001. Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on National Emission Ceilings for Certain Atmospheric Pollutants, OJ L 309, 27.11.2001, pp. 22–30.

EU, 2004. Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 Relating to Arsenic, Cadmium, Mercury, Nickel and Polycyclic Aromatic Hydrocarbons in Ambient Air, OJ L 23, 26.1.2005, pp. 3–16.

EU, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe, OJ L 152, 11.6.2008, pp. 1–44.

Eurostat, 2013. Eurostat Regional Yearbook 2013. Eurostat, European Commission, Luxembourg.

Gilbert, R.O., 1987. Statistical Methods for Environmental Pollution Monitoring. Van Nostrand Reinhold, New York.

Guerreiro, C., Horálek, J., de Leeuw, F., Hak, C., Nagl, C., Kurfürst, P., Ostatnicka, J., 2010. Status and Trends of NO_2 Ambient Concentrations in Europe. European Topic Centre on Air and Climate Change, Bilthoven. ETC/ACC Technical Paper 2010/19.

de Leeuw, F., Fiala, J., 2009. Indicators on Urban Air Quality – a Review of Current Methodologies. European Topic Centre on Air and Climate Change, Bilthoven. ETC/ACC Technical Paper 2009/8.

de Leeuw, F., 2012. AirBase: a Valuable Tool in Air Quality Assessments at a European and Local Level. Bilthoven, ETC/ACM Technical Paper 2012/4.

OECD/IEA, 2013. Nordic Energy Technology Perspectives: Pathways to a Carbon Neutral Energy Future. International Energy Agency, Paris.

Saffari, A., Daher, N., Samara, C., Voutsa, D., Kouras, A., Manoli, E., Karagkiozidou, O., Vlachokostas, C., Moussiopoulos, N., Shafer, M.M., Schauer, James J., Sioutas, C., 2013. Increased biomass burning due to the economic Crisis in Greece and its adverse impact on Wintertime air quality in Thessaloniki. Environ. Sci. Technol. 47, 13313–13320. http://dx.doi.org/10.1021/es403847h.

Slootweg, J., Posch, M., Hettelingh, J.-P., 2010. Progress in the Modelling of Critical Thresholds and Dynamic Modelling, Including Impacts on Vegetation in Europe. CCE Status Report 2010. Coordination Centre for Effects, National Institute for Public Health and the Environment, Bilthoven.

UN, 2010. Hemispheric Transport of Air Pollution 2010-Part D: Answers to Policy-relevant Science Questions. United Nations Economic Commission for Europe, Geneva. Air Pollution Studies No. 20.

WHO, 2000. Air Quality Guidelines for Europe. World Health Organization, Regional Office for Europe, Copenhagen.

WHO, 2006. Air Quality Guidelines. Global Update 2005. Particulate Matter, Ozone, Nitrogen dioxide and Sulfur dioxide. World Health Organization, Regional Office for Europe, Copenhagen.

WHO, 2013. Review of Evidence on Health Aspects of Air Pollution – REVIHAAP Project. Technical Report. World Health Organization, Regional Office for Europe, Copenhagen, Denmark.

Wilson, R.C., Fleming, Z.L., Monks, P.S., Clain, G., Henne, S., Konovalov, I.B., Szopa, S., Menut, L., 2012. Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996–2005. Atmos. Chem. Phys. 12, 437–454. http://dx.doi.org/10.5194/acp-12-437-2012.