# Air quality in Europe — 2013 report







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# **Executive summary**

'There are still major challenges to human health from poor air quality. We are still far from our objective to achieve levels of air quality that do not give rise to significant negative impacts on human health and the environment.' Janez Potočnik, European Commissioner for the Environment (EU, 2013).

Air quality continues to be a very important issue for public health, the economy and the environment. Europe has significantly cut emissions of several air pollutants in recent decades, greatly reducing emissions and exposure to substances such as sulphur dioxide  $(SO_2)$ , carbon monoxide (CO), benzene  $(C_6H_6)$  and lead (Pb). Despite improvements over several decades, air pollution continues to damage human health and the environment. Particulate matter (PM), ozone (O<sub>2</sub>), reactive nitrogen substances and some organic compounds still pose a significant threat. This leads to ill health, premature deaths, and damage to ecosystems, crops, and buildings. These constitute real losses for the European economy, the productivity of its workforce, and the health of its natural systems (EC, 2011a). 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 adverse effects on public health;
- 2) in ecosystems, where the pressures of air pollution impairs vegetation growth and harms biodiversity.

Emissions of air pollutants derive from almost all economic and societal activities. Policies implemented at the European, national and sectoral level have over time resulted in decreased emissions of many air pollutants and have led to acceptable air quality levels across Europe for some pollutants, e.g. CO and Pb. Nevertheless, road transport, industry, power plants, households and agricultural activities continue to emit significant amounts of air pollution. Combustion of biomass by households — burning fuels such as wood and coal — is an important source of directly emitted PM and polycyclic aromatic hydrocarbons (PAHs: a type of carcinogenic substances). Agriculture is mainly responsible for the ammonia (NH<sub>3</sub>) emissions which exert pressure on both human health and the ecosystems. Such emissions, as well as PM from the combustion of fuels to produce energy for domestic needs, have 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. This is because wood burning is often relatively cheap, and is considered as an environmentally friendly source of energy since it is renewable and carbon-neutral.

Cross border, or trans-boundary pollution is also a challenge in Europe. For many European countries, less than 50 % of the observed fine particulate matter ( $PM_{2.5}$ ) concentrations derive from their own emissions (EU, 2013). Many air pollutants are transported over long distances. Countries and continents are both emitters and receivers of trans-boundary pollution. For example, contributions from intercontinental transport influence the O<sub>3</sub> and PM concentrations in Europe.

Air pollution in Europe, as well as being a cross border issue, can also be viewed as a local and regional problem caused by the emission of specific pollutants, which either directly or through chemical reactions lead to negative impacts. Each pollutant produces a range of effects from mild to severe as concentration or exposure increases. The main effects of air pollution are:

- damage to human health caused by exposure to air pollutants, or by intake of pollutants transported through the air, deposited and then accumulated in the food chain;
- acidification of ecosystems (both terrestrial and aquatic), which leads to loss of flora and fauna;
- eutrophication in ecosystems on land and in water, which can lead to changes in species diversity;

- damage and yield losses affecting agricultural crops, forests and other plants due to exposure to ground-level O<sub>3</sub>;
- impacts of heavy metals or toxic metalloids and persistent organic pollutants on ecosystems, due to their environmental toxicity and due to bioaccumulation;
- contribution to climate forcing and indirect effects on climate;
- reduction of atmospheric visibility;
- damage to materials and buildings due to soiling and exposure to acidifying pollutants and O<sub>3</sub>.

## Purpose and scope of this report

This report presents an overview and analysis of air quality in Europe from 2002 (or later, pending data availability) to 2011. It reviews progress towards meeting the requirements of the air quality directives (EU, 2004b; EU, 2008c) and gives an overview of policies and measures introduced at European level to improve air quality and minimise air pollution impacts on public health and ecosystems. An overview of the latest findings and estimates of the effects of air pollution on health and its impacts on ecosystems 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 (1), including EU Member States and other EEA member countries as of 2011, i.e. EU-27 and EEA-32 respectively.

The report analyses each regulated pollutant at a time, following the single-pollutant approach currently adopted by EU air quality legislation and the World Health Organization (WHO) in its air quality guidelines. In reality, air pollution constitutes a complex mixture of pollutants, which may interact in terms of their impacts on human health and vegetation. Exposure to air pollution is largely a multi-pollutant process.

# Existing air quality legislation made simple

The Air Quality Directives 2008/50/EC and 2004/107/ EC set legally binding limits for ground-level concentrations of outdoor air pollutants.

Key elements of EU air quality legislation are:

- EU limit values are legally binding concentration thresholds that must not be exceeded. Limit values are set for individual pollutants and are made up of a concentration limit, an averaging time over which a pollutant is to be measured or estimated, the number of exceedances allowed per year (if any), and a date by which the limit value must be achieved. Some pollutants have more than one limit value covering different endpoints or averaging times. Limit values are legally binding on EU Member States.
- **Target values** are to be attained where possible by taking all necessary measures not entailing disproportionate costs. Target values are not legally binding.
- Exposure reduction obligation concentrations are to be reduced by a given per cent depending on the mean triennial PM<sub>2.5</sub> urban background concentrations from 2008–2010 to 2018–2020.

This report also refers to WHO Air Quality Guidelines (AQG), which are often more stringent than EU limit and target values.

## The most problematic pollutants

At present, PM and  $O_3$  are Europe's most problematic pollutants in terms of harm to human health. European anthropogenic emissions are the most important contributors to  $O_3$  and PM concentrations levels over Europe, but intercontinental transport of pollution also contributes to increased impacts on health, ecosystems and our economy (particularly crop productivity).

<sup>(1)</sup> The 38 countries are the EU-27 Member States: Austria, Belgium, Bulgaria, Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, Sweden and the United Kingdom, the new (since 2013) Member State Croatia and the remaining five EEA member countries, Iceland, Liechtenstein, Norway, Switzerland and Turkey), as well as five cooperating countries (Albania, Bosnia and Herzegovina, the former Yugoslav Republic of Macedonia, Montenegro, and Serbia). The EU-27 Member States and the five additional EEA member countries make up the EEA-32.

## **Impacts on population**

European citizens often breathe air that does not meet the European standards. The current pollution levels clearly impact on large parts of the urban population. Table ES.1 gives an overview (<sup>2</sup>) of the proportion of the EU urban population exposed to pollutant concentration levels above the limit and target values set in the EU legislation and the air quality guidelines WHO AQG in recent years (2009–2011).

Current pollution levels, especially of PM,  $O_y$ , and an important PAH — benzo(a)pyrene (BaP) clearly impact on large numbers of the urban population. This is particularly evident in the population exposure estimates based on the WHO AQG.

### Impacts on European ecosystems

Air pollution's most important effects on European ecosystems are damage to vegetation resulting from exposure to  $O_{3}$  eutrophication and acidification. As SO<sub>2</sub> emissions have fallen, ammonia (NH<sub>3</sub>) emitted from agricultural activities, and nitrogen oxides (NO<sub>x</sub> – a family of gases that includes nitrogen dioxide – NO<sub>2</sub> and nitrogen oxide – NO) emitted 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.

# Table ES.1Percentage of the urban population in the EU exposed to air pollutant<br/>concentrations above the EU and WHO reference levels (2009–2011)

Pollutant	EU reference value	Exposure estimate (%)	WHO AQG	Exposure estimate (%)
PM <sub>2.5</sub>	Year (20)	20-31	Year (10)	91-96
PM <sub>10</sub>	Day (50)	22-33	Year (20)	85-88
0 <sub>3</sub>	8-hour (120)	14-18	8-hour (100)	97-98
NO <sub>2</sub>	Year (40)	5-13	Year (40)	5-13
BaP	Year (1)	22-31	Year (0.12)	76-94
SO <sub>2</sub>	Day (125)	< 1	Day (20)	46-54
СО	8-hour (10)	< 2	8-hour (10)	< 2
Pb	Year (0.5)	< 1	Year (0.5)	< 1
Benzene	Year (5)	< 1	Year (1.7)	12-13

 Colour coding:
 < 5 %</th>
 5 - 50 %
 50 - 75 %
 > 75 %

Note: The pollutants are ordered in terms of their relative risk for health damage — highest on top.

This estimate refers to a recent three year period (2009–2011) and includes variations due to meteorology, as dispersion and atmospheric conditions differ from year to year.

The reference levels included EU limit or target levels and WHO air quality guidelines (AQG).

The reference levels in brackets are in  $\mu g/m^3$  except for CO which is in mg/m<sup>3</sup> and BaP in ng/m<sup>3</sup>.

For some pollutants EU legislation allows a limited number of exceedances. This aspect is considered in the compilation of exposure in relation to EU air quality limit and target values.

The comparison is made for the most stringent EU limit or target values set for the protection of human health. For  $PM_{10}$  the most stringent standard is for 24-hour mean concentration.

For  $PM_{2.5}$  the most stringent EU standard is the 2020 indicative annual limit value (20  $\mu$ g/m<sup>3</sup>).

As the WHO has not set AQG for BaP and  $C_6H_6$ , the WHO reference level in the table was estimated assuming an additional lifetime risk of 1 x 10<sup>-5</sup>.

The Pb concentrations reported by France for 2009 have been corrected in the exposure estimates given above.

**Source:** EEA, 2013e (CSI 004); AirBase v. 7; ETC/ACM.

<sup>(&</sup>lt;sup>2</sup>) This estimate refers to a recent three-year period (2009–2011) and includes variations due to meteorology, as dispersion and atmospheric conditions differ from year to year.

## **Impacts on climate**

Several air pollutants are also 'climate forcers', a term used to describe pollutants that also have an impact on the planet's climate and global warming in the short-term (decades). Ground-level  $O_{3'}$  particles, and black carbon (a constituent of PM) are examples of air pollutants that are climate forcers and contribute directly to positive or negative changes in global radiative forcing.

In addition, some air pollutants have indirect effects on climate. For example, particles can also cause climate-forcing indirectly, through the changes they are causing on cloud properties, including cloud reflectivity and precipitation, and cloud formation and dynamics. Vegetation is an important terrestrial carbon sink and ozone impairs vegetation growth. It is estimated that such indirect impact of ozone on global warming via its negative impact on vegetation is of similar magnitude to its direct impact as a greenhouse gas (Sitch et al., 2007).

Measures to cut black carbon and other pollutants leading to  $O_3$  formation (among them methane –  $CH_4$  which is itself a greenhouse gas), will have twin benefits, reducing both global warming and pollution effects to human health and ecosystems. Air quality and climate change can be cost-effectively tackled together by using an integrated approach when defining policies and measures.

## Main findings

Emissions of the main air pollutants in Europe declined in the period 2002–2011. This resulted in improved air quality across the region — at least with respect to certain pollutants. Certain individual sectors have seen emissions of some pollutants increase during this period. For example, PM emissions from fuel combustion in the commercial, institutional and household sector, has increased by around 7 % since 2002. This sector is now the most important contributor to total European Union PM emissions. In addition, in 2011 eight Member States exceed (based on provisional reporting of emissions) one or more ceilings (limits) set under EU legislation, when these ceilings should have been reached in all countries by 2010 (EEA, 2013b).

The 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 (which include emission heights, chemical transformations, reactions to

sunlight, additional natural and hemispheric contributions and the impact of weather and topography), emission reductions do not always produce a corresponding drop in atmospheric concentrations, especially for PM and  $O_3$ . For example, while reductions of  $O_3$  forming substances ( $O_3$  precursor gases) have been substantial in Europe, ozone concentrations (in relation to the target value for the protection of health) have generally decreased slowly but have increased in places between 2002 and 2011.

A significant proportion of Europe's population live in cities, where exceedances of air quality standards regularly occur. Particulate matter (PM) and ozone ( $O_3$ ) pollution are particularly associated with serious health risks, and exposure to high levels of organic pollutants, in particular PAHs is a growing health concern in Europe.

## Main findings by air pollutant

### 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. 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;
- 'secondary' particles produced as a result of chemical reactions involving PM forming (precursor) gases after their emission: SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and non-methane volatile organic compounds (NMVOC). When these gases react, they produce PM.

The size of PM is expressed in micrometers. The largest particles of concern are 10 microns in diameter or smaller ( $PM_{10}$ ). But the group of particles of most concern is 2.5 microns in diameter or smaller ( $PM_{2.5}$ ). Some of these are small enough to pass from the lung into the bloodstream just like oxygen molecules. By comparison, the diameter of a human hair is 50–70 microns (Figure 2.1). The health effects of PM are caused after their inhalation and penetration into the lungs and blood stream, leading to adverse effects in the respiratory, cardiovascular,

immune, and neural systems. A fraction of ultrafine particles (with a diameter less than 0.1 microns) may even enter the brain directly through the nose.

## Emissions of primary PM

Emissions of primary  $PM_{10}$  and  $PM_{2.5}$  decreased by 14 % and 16 % respectively in the EU between 2002 and 2011. The reductions in the same period for the EEA-32 member countries were 9 % for  $PM_{10}$  and also 16 % for  $PM_{2.5}$ .

## Emissions of PM precursor gases

PM precursor emissions decreased between 2002 and 2011.

In the EU:

- sulphur oxides (SO<sub>x</sub>) emissions fell by 50 %;
- $NO_x$  emissions fell by 27 %;
- NH<sub>3</sub> emissions fell by 7 %;
- NMVOCs emissions fell by 28 %.

In the EEA-32 countries:

- SO<sub>x</sub> emissions fell by 34 %;
- NO<sub>x</sub> emissions fell by 23 %;
- NH<sub>3</sub> emissions fell by 5 %;
- NMVOCs emissions fell by 27 %.

Despite these emission reductions, 22–44 % of the EU urban population (<sup>3</sup>) was exposed to concentrations of  $PM_{10}$  in excess of the EU air quality daily limit value in the period 2002–2011. Although ambient concentrations of  $PM_{10}$  have slightly decreased during the past decade, there was no discernible downward trend in this particular indicator (Figure ES.1). Between 41 % and 49 % of the urban population in EEA-32 countries was exposed to  $PM_{10}$  concentrations in excess of the daily limit value in the same period.

The EU limit and target values for PM were exceeded widely in Europe in 2011, with the  $PM_{10}$  24-hour limit value being exceeded in 22 European countries. The non-legally binding WHO guidelines for  $PM_{10}$  and  $PM_{2.5}$  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.

## Key observations relating to PM

- Slight reductions were generally observed in ambient PM concentrations over the period 2002–2011. These are the result of the generally declining emissions of primary PM and various precursors to PM. Increasing ambient PM concentrations were also observed at some locations.
- 33 % of the EU urban population lives in areas where the EU air quality 24-hour limit value for particulate matter (PM<sub>w</sub>) was exceeded in 2011 (Figure ES.1). For EEA-32 countries the estimate is 49 %.
- EU urban population exposure to PM<sub>10</sub> levels exceeding the WHO AQG is significantly higher, comprising 88 % of the total urban population in 2011 (Figure ES.1).

## Ground-level O<sub>3</sub>

Ozone is a secondary pollutant (meaning it is not emitted directly by any emission source) formed in the troposphere, the lower part of the atmosphere, from complex chemical reactions following emissions of precursor gases such as  $NO_x$  and NMVOCs. At the continental scale, methane (CH<sub>4</sub>) and carbon monoxide (CO) also play a role in O<sub>3</sub> formation. Ozone is a powerful and aggressive oxidising agent, elevated levels of which cause respiratory health problems and lead to premature mortality. High levels of O<sub>3</sub> can also damage plants, leading to reduced agricultural crop yields and decreased forest growth. Ground-level O<sub>3</sub> contribute also directly and indirectly to global warming.

## Emissions of O<sub>3</sub> precursors

Ozone precursor gas emissions decreased considerably between 2002 and 2011.

In the EU:

- NO<sub>x</sub> emissions decreased by 27 %;
- NMVOC emissions decreased by 28 %;
- CO emissions decreased by 32 %;
- CH<sub>4</sub> emissions decreased by 15 %.

<sup>(&</sup>lt;sup>3</sup>) The range partly reflects variations caused by meteorology, as dispersion and atmospheric conditions differ from year to year.









% of urban population exposed to air pollution exceeding WHO air quality guidelines

Source: EEA, 2013e (CSI 004).

In the EEA-32:

- NO<sub>x</sub> emissions decreased by 23 %;
- NMVOC emissions decreased by 27 %;
- CO emissions decreased by 27 %.

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  $O_3$ .

There is a discrepancy between the past reductions in emissions of  $O_3$  precursor gases in Europe and the change in observed average  $O_3$  concentrations in Europe. One of the reasons for this is increasing inter-continental 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. Moreover, the relationship of  $O_3$  concentrations in Europe to the emitted precursors in Europe is not linear (1 to 1).

Population and crop exposure to  $O_3$  has had a very small decrease since 2002. This excludes the estimated exposures in 2003 and 2006. Variations between years are influenced by meteorological factors. Summers in 2003 and 2006 had favourable meteorological conditions for  $O_3$  formation, resulting in exceptionally high concentrations. In other words, while emissions of gases that contribute to the formation of  $O_3$  dropped significantly in Europe,  $O_3$  concentrations decreased only slightly. Larger reductions in emissions of  $O_3$  precursor gases are necessary to achieve reductions in  $O_3$  concentrations.

Between 14 % and 65 % (<sup>4</sup>) of the EU urban population was exposed to  $O_3$  concentrations above the EU target value for protecting human health in the period 2002–2011 (Figure ES.1). Furthermore, 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 from 2002 to 2010 (Figure 3.7). High  $O_3$  concentrations are most pronounced in southern Europe.

## Key observations relating to $O_3$

• There is no clear trend for O<sub>3</sub> concentrations (when considering the target value for the

protection of health) between 2002 and 2011 in 80 % of the monitoring stations. 18 % of the stations registered a statistically significant decreasing trend, while 2 % registered an increasing trend, most of them in the Iberian Peninsula. It can therefore be concluded that concentrations in the period 2002–2011 do not reflect the general European reductions in emissions of  $O_3$  precursors in the same period.

- Fourteen per cent of the EU urban population live in areas where the EU O<sub>3</sub> target value for protecting human health was exceeded in 2011 (Figure ES.1).
- The percentage of the EU urban population exposed to O<sub>3</sub> levels exceeding the WHO AQG (which is stricter than the EU target value) is significantly higher, comprising 98 % of the total urban population (Table ES.1).
- Europe's sustained ambient O<sub>3</sub> concentrations continue to cause considerable damage to vegetation growth and crop yields, resulting in serious costs to the Europe's economy and reducing plant uptake of carbon dioxide.

## Nitrogen oxides

Nitrogen oxides are 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.  $NO_x$  also contributes to the formation of secondary inorganic PM and  $O_3$  with associated effects on health, ecosystems and climate.

Nitrogen (N) compounds, emitted as  $NO_x$  but also as  $NH_y$  are now the principal acidifying components in our air and cause eutrophication of ecosystems. The sensitive ecosystem area in Europe affected by eutrophication due to excessive atmospheric N was reduced by 23 % from 1990 to 2010 (EEA, 2012a). The area of sensitive ecosystems affected by excessive acidification from air pollution has shrunk even faster, down by 92 % (EEA 2012a) in the same two decades, mainly due to the strong reduction in  $SO_2$  emissions.

<sup>(&</sup>lt;sup>4</sup>) The range partly reflects variations caused by meteorology, as dispersion and atmospheric conditions differ from year to year.

## Key observations relating to NO<sub>2</sub>

- Some cities in Europe show an increase in concentrations of NO<sub>2</sub> measured close to traffic. This reflects the increasing numbers of newer diesel vehicles. Such vehicles emit less CO and NMVOCs than petrol vehicles, but may emit more PM and NO<sub>2</sub>.
- The decrease in NO<sub>x</sub> emissions (34 % from road traffic sources between 2002 and 2011) is considerably greater than the fall in NO<sub>2</sub> annual mean concentrations (ca. 8 % measured at stations close to traffic, between 2002 and 2011). This is attributed primarily to the increase in NO<sub>2</sub> emitted directly into the air from diesel vehicles (the proportion of NO<sub>2</sub> in the NO<sub>x</sub> emissions of a diesel vehicle is far higher than the proportion of NO<sub>2</sub> in the NO<sub>x</sub> emissions of a conventional-petrol vehicle).
- 5 % of the EU urban population lives in areas where the annual EU limit value and the WHO AQG for NO<sub>2</sub> were exceeded in 2011 (Figure ES.1).

## Sulphur dioxide (SO<sub>2</sub>)

Sulphur dioxide is emitted when fuels containing sulphur are burned, or originates 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. Sulphur dioxide can affect the respiratory system and reduce lung function. It is also a major precursor to PM, which is associated with significant health effects.

### Key observations relating to SO<sub>2</sub>

• In the period 2002–2011, EU Member States cut their SO<sub>x</sub> emissions by 50 %, leading to a fall in SO<sub>2</sub> concentrations of about one third. The corresponding emission reduction in the EEA-32 countries in the same period was 34 %.

- Large sensitive ecosystem areas of Europe are no longer exposed to acidification, due mainly to reductions in SO<sub>x</sub> emissions.
- Based on the available SO<sub>2</sub> measurements at urban background stations, the urban population in the EU was not exposed to SO<sub>2</sub> concentrations above the EU 24-hour limit value (Figures ES.1 and 5.3). In the EEA-32 countries, 2.7 % of the urban population were exposed to SO<sub>2</sub> concentrations above the EU 24-hour limit value in 2011.
- The EU urban population exposed to SO<sub>2</sub> levels exceeding the WHO AQG is significantly higher, amounting to 46–54 % of the total urban population in recent years, between 2009 and 2011 (Table ES.1).

## Carbon monoxide (CO)

Carbon monoxide is emitted due to incomplete combustion of fossil fuels and biofuels. Exposure to CO can reduce the oxygen-carrying capacity of blood, thereby reducing oxygen delivery to the body's organs and tissues.

The atmospheric lifetime of CO is about three months. This relatively long lifetime allows CO to slowly oxidise into carbon dioxide, also contributing to the formation of  $O_3$  during this process, the latter having associated effects on the health of humans and ecosystems.

### Key observations relating to CO

- The observed EU average reduction in CO daily 8-hour maxima concentrations in the period 2002–2011 was 35 %. This reflects declining CO emissions of 32 % in the EU over the last decade.
- Exposure of the European population to CO concentrations above the EU limit value and WHO AQG is very localised and sporadic.

## **Heavy metals**

The heavy metals are emitted mainly as a result of various combustion processes and industrial activities. Heavy metals can reside in or be attached to PM. As well as polluting the air, heavy metals are deposited on terrestrial or water surfaces and subsequently buildup in soils or sediments. They are persistent in the environment (meaning they cannot be broken down over time) and may bio-accumulate in food-chains. Bio-accumulation occurs where a substance is not excreted by an animal causing it to accumulate over time in the animal's body. This means the substance can eventually poison the animal, and any other animals further up the food chain that consume it. This overview covers arsenic (As), cadmium (Cd), mercury (Hg), nickel (Ni) and lead (Pb). Arsenic is not a heavy metal per se but it is regularly added to the list of heavy metals, based on its toxicity.

### Emissions of heavy metals

In the period 2002–2011 emissions of heavy metals in the EU were reduced as follows:

- As by 3 %;
- Cd by 27 %;
- Hg by 26 %;
- Ni by 43 %;
- Pb by 20 %.

## Key observations relating to heavy metals

- 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 heavy metal levels in soils, sediments and organisms.
- Despite cuts in estimated emissions of heavy metals since 2002, in the EU a significant share of the EU ecosystem area was still at risk of heavy metal contamination. Exceedances of Hg critical loads (<sup>5</sup>) were estimated to occur at 54 % of the sensitive ecosystem area in the EU in 2010, while for Pb the estimated area in exceedance is 12 %. (Slootweg et al., 2010).

• 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 (countries do not have to monitor certain pollutants as rigorously if these pollutants have already been found to be less widespread). An even smaller number of these stations have been operating for five or more years. 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.

## Benzene (C<sub>6</sub>H<sub>6</sub>) and benzo(a)pyrene (BaP), a polycyclic aromatic hydrocarbon (PAH)

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  $C_6H_6$  exposure in humans. Benzene is a carcinogenic pollutant. The most significant adverse effects from prolonged exposure are damage to the genetic material of cells, a phenomenon that can cause cancer.

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, and for this reason it is used as an indicator of exposure to other harmful PAHs.

## Emissions of $C_6H_6$ and BaP

- Benzene is not included as an individual pollutant in European emissions inventories that cover VOCs. Concentrations of benzene measured at traffic and urban background stations decreased steadily until 2007, after which time they stabilised.
- Emissions of BaP in the EU 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

<sup>(&</sup>lt;sup>5</sup>) The general definition of a critical load is a quantitative estimate of an exposure to pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge.

problem, especially in areas where domestic coal and wood burning is, or becomes more common.

### Key observations relating to C<sub>6</sub>H<sub>6</sub> and BaP

- Exceedances of the limit value for C<sub>6</sub>H<sub>6</sub> were limited to very few locations in Europe in 2011.
- 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 was exposed to BaP concentrations above the target value (1 ng/m<sup>3</sup>) 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 urban population was exposed to BaP concentrations above the WHO reference level in 2011. In large parts of Europe the concentrations are declared to be below the lower assessment threshold and there is no requirement for monitoring BaP. Notably, the WHO reference level (0.12 ng/m<sup>3</sup>) is below the lower assessment threshold (0.4 ng/m<sup>3</sup>).

• The increase in BaP emissions from domestic combustion in Europe over the last years is a matter of concern, as it is aggravating the exposure of the European population to BaP concentrations, especially in urban areas.

# **1** Introduction

## 1.1 Background

Air quality is an important issue for public health, the economy and the environment. Poor air quality as a result of air pollution is a major environmental health risk, contributing to respiratory disease, cardiovascular disease, and lung cancer. In addition to the health effects, air pollution has considerable economic impacts, cutting short lives, increasing medical costs, and reducing productivity through lost working days across the economy.

Air pollution also has several environmental impacts, affecting the quality of fresh water, soil, and the ecosystem services they host. For example, ground-level ozone damages agricultural crops, forests, and plants, by reducing their growth rates. Other pollutants, such as nitrogen oxides (a family of gases referred to collectively as NO<sub>x</sub>), sulphur dioxide  $(SO_2)$  and ammonia  $(NH_3)$  contribute to the acidification of soil, lakes and rivers, causing the loss of animal and plant life. In addition to their acidification effects, NH<sub>2</sub> and NO<sub>y</sub> emissions also disrupt land and water ecosystems by introducing excessive amounts of nutrient nitrogen leading to eutrophication, an oversupply of nutrients that can lead to changes in species diversity and invasions of new species. Air pollution can also damage materials and buildings, including Europe's most culturally significant buildings. Finally, air pollution has a clear impact on the climate, as some air pollutants behave like greenhouse gases. Table 1.1 summarises the key effects of the major air pollutants on health, the environment and the climate.

European air quality policy has achieved considerable successes in the past in reducing air pollution. The most recent package of European air quality policy was launched in 2005 with the Thematic Strategy on Air Pollution designed to make substantial progress towards the long-term EU objective: 'to achieve levels of air quality that do not result in unacceptable impacts on, and risks to, human health and the environment.' This objective is re-confirmed in European Commission's new 'general Union Environment Action Programme' for the period 2014–2020. Unfortunately, Europe is still far from achieving this objective. Despite improvements over several decades, air pollution continues to damage human health and the environment in Europe, leading to premature deaths, ill health, and substantial damage to ecosystems, crops and buildings. These constitute real losses for the European economy, the productivity of its workforce, the health of its natural systems (EC, 2011a). The effects of poor air quality have been felt the most strongly in two main areas. Firstly, these effects have been strongly felt in urban areas, where they cause health problems. Secondly, these effects have been felt in ecosystems, where air pollution impairs vegetation growth and where eutrophication due to air pollution has led to biodiversity loss.

Against the backdrop of these impacts of air pollution, the *Air quality in Europe* — 2013 report is produced by the EEA to assess the status of air quality and recent air quality trends. This annual report provides a more regularly-updated account of air quality than the EEA's less frequent five-yearly 'State of the environment' reports (SOER). The report aims to support policy development and implementation in the field of air quality at both European and national levels.

## 1.2 Objectives and coverage

This report presents an overview and analysis of air quality in Europe from 2002 (or later, pending data availability) to 2011. The evaluation of the status and trends of air quality is based on ambient air measurements, in conjunction with anthropogenic emissions and their trends. It also includes an overview of the latest findings and estimates of the effects of air pollution on health and its impacts on ecosystems.

The report reviews progress towards meeting the requirements of the two air quality directives presently in force (EU, 2004b; EU, 2008c). It also gives an overview of the policies and measures introduced at European level to improve air quality

Pollutant	Health effects	Environmental effects	Climate effects	
Particulate matter (PM)	Cancause or aggravate cardiovascular and lung diseases, heart attacks and arrhythmias, affect the central nervous system, the reproductive system and cause cancer. The outcome can be premature death.	Can affect animals in the same way as humans. Affects plant growth and ecosystem processes. Can cause damage and soiling of buildings. Reduced visibility.	Climate effect varies depending on particle size and composition: some lead to net cooling, while others lead to warming. Can lead to changed rainfall patterns. Deposition can lead to changes in surface albedo (the ability of the earth to reflect radiation from sunlight).	
Ozone (O <sub>3</sub> )	Can decrease lung function; aggravate asthma and other lung diseases. Can lead to premature mortality.	Damages vegetation, impairing plant reproduction and growth, and decreasing crop yields. Can alter ecosystem structure, reduce biodiversity and decrease plant uptake of CO <sub>2</sub> .	Ozone is a greenhouse gas contributing to warming of the atmosphere.	
spleen and blood. Can aggravate and eutrophication of so water, leading to change in species diversity. Acts increased susceptibility to a precursor of ozone and respiratory infection. associated environmenta		Contributes to the acidification and eutrophication of soil and water, leading to changes in species diversity. Acts as a precursor of ozone and particulate matter, with associated environmental effects. Can lead to damage to buildings.	Contributes to the formation of ozone andparticulate matter, with associated climate effects.	
Sulphur oxides (SO <sub>x</sub> )	Aggravates asthma and can reduce lung function and inflame the respiratory tract. Can cause headache, general discomfort and anxiety.	Contributes to the acidification of soil and surface water. Causes injury to vegetation and local species losses in aquatic and terrestrial systems. Contributes to the formation of particulate matter with associated environmental effects. Damages buildings.	Contributes to the formation of sulphate particles, cooling the atmosphere.	
Carbon monoxide (CO)	Can lead to heart disease and damage to the nervous system;can also cause headache, dizziness and fatigue.	May affect animals in the same way as humans. Acts as a precursor of ozone.	Contributes to the formation of greenhouse gases such as $\rm CO_2$ and ozone.	
carcinogen. It can lead to a damage in the blood, heart, liver a and kidney. May also damage the peripheral nervous system. a ir		Highly toxic to aquatic life, birds and land animals. Soil with high arsenic content, reduces plant growth and crop yields. Organic arsenic compounds are persistent in the environment and subject to bioaccumulation.	No specific effects.	
Cadmium (Cd)	Cadmium, especially cadmium oxide, is likely to be a carcinogen. It may cause damage to the reproductive and respiratory systems.	Toxic to aquatic life. Cadmium is highly persistent in the environment and bioaccumulates.	No specific effects.	
Lead (Pb)	Can affect almost every organ and system, especially the nervous system. Can cause premature birth, impaired mental development and reduced growth.	Bioaccumulates and adversely impacts both terrestrial and aquatic systems. Effects on animal life include reproductive problems and changes in appearance or behaviour.	No specific effects.	
Mercury (Hg)	Can damage the liver, the kidneys and the digestive and respiratory systems. It can also cause brain and neurological damage and impair growth.	Bioaccumulates and adversely impacts both terrestrial and aquatic systems. Can affect animals in the same way as humans. Very toxic to aquatic life.	No specific effects.	
Nickel (Ni)	Several nickel compounds are classified as human carcinogens. It may cause allergic skin reactions, as well as affecting the respiratory, immune and defence systems.	Nickel and its compounds can have highly acute and chronic toxic effects on aquatic life. Can affect animals in the same way as humans.	No specific effects.	
Benzene (C <sub>6</sub> H <sub>6</sub> )	A human carcinogen, which can cause leukaemia and birth defects. Can affect the central nervous system and normal blood production, and can harm the immune system.	Has an acute toxic effect on aquatic life. It bioaccumulates, especially in invertebrates. Leads to reproductive problems and changes in appearance or behaviour. It can damage leaves of agricultural crops and cause death in plants.	Benzene is a greenhouse gas contributing to the warming of the atmosphere as it contributes to the formation of ozone and secondary organic aerosols, which can act as climate forcers.	
PAHs, in particular Benzo-a-pyrene (BaP)	Carcinogenic. Other effects may be irritation of the eyes, nose, throat and bronchial tubes.	Is toxic to aquatic life and birds. Bioaccumulates, especially in invertebrates.	No specific effects.	

## Table 1.1 Effects of air pollutants on human health, the environment and the climate

and minimise air pollution impacts on public health and ecosystems.

The report analyses each regulated pollutant at a time, following the single-pollutant approach currently adopted by both EU air quality legislation and the World Health Organization (WHO) in its air quality guidelines. In reality, air pollution constitutes a complex mixture of pollutants, which interact with each other and the atmosphere to produce a variety of effects on human health and vegetation. Therefore, exposure to air pollution is largely a multi-pollutant process.

## 1.3 Effects of air pollution on health

There is a large body of evidence on the health impacts of air pollution, as knowledge in this area has increased considerably in recent decades. The latest WHO review on the health effects of air pollution (WHO, 2013) concludes that a considerable amount of new scientific information on the health effects of particulate matter (PM), ozone  $(O_3)$  and nitrogen dioxide  $(NO_2)$ , observed at levels commonly present in Europe, has been published in the recent years. This new evidence supports the scientific conclusions of the WHO air quality guidelines, last updated in 2005, and indicates that health effects can occur at air pollution concentrations lower than those used to establish the 2005 guidelines. It also provides scientific arguments for decisive action to improve air quality and reduce the burden of disease associated with air pollution in Europe.

Most of the health impact studies reviewed by the WHO are focused on respiratory and cardiovascular effects attributed to exposure to air pollution (WHO, 2005, 2006a, 2006b, 2007, 2008). But evidence is also growing for a range of other effects, caused by exposure to air pollutants at different times of life, ranging from prenatal exposure all the way through childhood and adult life. For example, exposure to air pollutants during pregnancy has been associated with reduced foetal growth, pre-term birth and spontaneous abortions (WHO, 2005). Maternal exposure to air pollution during pregnancy increases the risk of the child developing allergies and asthma later in life (Jedrychowski et al., 2010; Baïz et al., 2011). Furthermore, the mechanisms by which air pollution may act on the nervous system have recently been documented (Genc et al., 2012), and a few epidemiological studies report positive associations between exposure to air pollution and impaired cognitive function (van Kempen

et al., 2012), but more research is needed to better understand these effects.

Health effects are related both to short-term and long-term exposure to air pollution. Short-term (exposure over a few hours or days) is linked with acute health effects, while long-term exposure (over months or years) is linked with chronic health effects. Health impacts of air pollution can be quantified and expressed as mortality and morbidity. Mortality reflects reduction in life expectancy due to air pollution exposure, while morbidity relates to illness occurrence, ranging from minor effects such as coughing to serious conditions that may require hospitalization.

Epidemiological studies attribute the most severe health effects of air pollution to PM. The evidence base for an association between PM and short-term (as well as long-term) health effects has become much stronger. Recent long-term studies show associations between PM and mortality at levels well below the current annual WHO air quality guideline level for  $PM_{25}(10 \ \mu g/m^3)$ . This corroborates earlier scientific evidence, and the WHO has therefore suggested that exposure to PM – even in very small amounts - causes adverse health effects (WHO, 2006a, 2006b, 2013). The latest study from the World Health Organization (WHO, 2013) links long-term exposure to fine particles (PM<sub>2.5</sub>) with cardiovascular and respiratory deaths, as well as increased sickness, such as childhood respiratory diseases.

Ozone  $(O_3)$  also has a marked effect on human health, with recent epidemiological studies indicating considerably larger mortality effects than previously thought (WHO, 2013). High concentration levels of ozone cause breathing problems, reduce lung function, and lead to lung diseases like asthma (WHO, 2008). Short-term exposure to current summer ozone concentrations in Europe has adverse health effects on pulmonary function, leading to lung inflammation and respiratory symptoms. These symptoms in turn result in increased medication usage, morbidity and mortality. New evidence has also emerged detailing the negative effects of long-term exposure to ozone on mortality and reproductive health.

Many studies, reviewed by the WHO (2013) and not previously considered, or published since 2004, have documented associations between short-term and long-term exposure to  $NO_2$  with mortality and morbidity. Both short- and long-term studies have found these associations with adverse effects at concentrations that were at or below the current EU limit values (WHO, 2013). Some polycyclic aromatic hydrocarbons (PAHs) are potent carcinogens, and they are often attached to airborne particles. WHO (2013) found new evidence linking PAH exposure to cardiovascular morbidity and mortality, but at present these effects of PAH exposure cannot be separated from the effects of particles. The WHO (2013) still recommends BaP as an indicator for carcinogenic PAHs, even if it may only explain about half of the PAH overall carcinogenic potency.

Figure 1.1 summarises the key health effects for major air pollutants. Of particular concern in Europe are particulate matter (PM), ground-level ozone  $(O_3)$ , benzo(a)pyrene (BaP) and nitrogen dioxide (NO<sub>3</sub>).

It is important to note that the proportion of the population exposed to lower levels of air pollution

and affected by less severe health impacts is much larger than the proportion of the population affected by the more severe events leading to more serious health impacts (see Figure 1.2). Nevertheless, even these less severe health effects may have strong public health implications. This is because air pollution affects whole populations, especially in major cities, where large populations are continuously exposed. The overall costs of the less severe health impacts may therefore be higher than the sum of the most severe effects. In spite of this, it is the severe outcomes (such as increased risk of mortality and reduced life expectancy) that are most often considered in epidemiological studies and risk analysis. This is usually because of the better availability of data on these severe effects (EEA, 2013a).

#### Figure 1.1 Health impacts of air pollution



Source: EEA, 2013f.





Number of people affected

Source: Based on US EPA.

# 1.4 Effects of air pollution on ecosystems

Air pollution also damages the environment and it is estimated that two-thirds of the protected sites in the EU Natura 2000 network are currently under severe threat from air pollution (EC, 2013). For example, ozone can damage crops and other vegetation, impairing its growth. The atmospheric deposition of sulphur and nitrogen compounds has acidifying effects on soils and freshwaters. The deposition of nitrogen compounds can also lead to eutrophication, an oversupply of nutrients that may lead to changes in species diversity and invasions of new species. In addition, heavy metals and persistent organic pollutants may lead to severe impacts on ecosystems. This is mainly due to their environmental toxicity, but in some cases it is also due to their tendency to 'bioaccumulate', a process whereby the toxin cannot be digested and excreted by animals, and therefore slowly accumulates in the animal's system, causing chronic health problems (<sup>6</sup>).

The impacts of air pollution on the environment depend not only on the air pollutant emission rates but also on the location and conditions of the emissions. Factors such as meteorology and topography are also important, as these determine the transport, chemical transformation and deposition of air pollutants. Furthermore, the environmental impacts of air pollution also depend on the sensitivity of ecosystems to acidification, eutrophication, and heavy metals.

<sup>(6)</sup> Bioaccumulation refers to the accumulation of substances in organisms. While some toxins can be excreted by plants or animals, a bioaccumulating toxin will remain in a plant or animal and can gradually build up over time if the plant or animal continues to be exposed to the bioaccumulating toxin. In the case of toxic substances, bioaccumulation increases the risk of poisoning, even if environmental levels of the toxin are not very high.

### Acidification

Acidification damages plant and animal life both on land and in water, and it damages materials and buildings by corrosion.

The process of soil acidification works as follows: nitrogen and sulphur emissions into the atmosphere create nitric acid and sulphuric acid. This nitric acid and sulphuric acid falls to the earth as acid rain, and in so doing leads to a build-up of hydrogen ions in the soil. This build-up of hydrogen ions leads to a reduction in the soil's pH level, a sign that the soil is becoming more acidic. Acidification also occurs when positively charged ions (also known as 'cations') of calcium, magnesium, potassium and sodium are leached and lost from the soil through the action of acid rain. Nitrogen compounds, often added as fertiliser, contribute further to the acidification of the soil through the production of ammonium. Soils and waters with poor buffering (neutralising) capacity are the most sensitive to acid rain.

It is estimated that 9 % of Europe's forest area and 25 % of the European lakes were exposed to air pollution levels that exceeded EU limits for acidification in 2010 (EC, 2013).

### Eutrophication

Eutrophication refers to an excess of nutrients in water or soil. It threatens biodiversity through the excessive growth of a few species, which thrive in the presence of the added nutrients, to the detriment of a larger number of species, which have long been part of the ecosystems but are accustomed to a lower-nutrient environment. The two major causes of eutrophication are excess nutrient nitrogen (mainly nitrates and ammonium) and excess phosphates in ecosystems. Air pollution contributes to this excess of nutrient nitrogen, as the nitrogen emitted to the air, mostly from  $NO_x$  emissions (mainly from combustion of fuels) and  $NH_3$  emissions (mostly from agricultural fertiliser), deposits on soils and waters.

In water, eutrophication often leads to algae 'blooms', the rapid growth of algae, forming dense patches near the surface of the water and preventing light from penetrating into deeper layers of the water. The fact that light is unable to penetrate into the water may lead to the reduction and sometimes extinction of aquatic plants, as they are unable to survive without this light. Another serious problem arises when the algae begin to die and deposit on the floor of lakes and rivers. Bacteria then take over the ecosystem, decomposing the organic material of the dead algae and using up large amounts of dissolved oxygen in the process. This high biological oxygen demand due to the increased bacterial activity may lead to a severe reduction in oxygen available to other forms of life, and in severe cases causes many fish to suffocate. As more fish die, the number of these bacteria increases even more, intensifying the problem in a vicious cycle.

Eutrophication is still a widespread problem that affects most European ecosystems. In the last decade, the improvement in the acidification problem has not been matched with a parallel improvement in the problem of eutrophication. It is estimated that 62 % of European ecosystem areas and 71 % of the area covered by Natura 2000 protected sites were exposed to air pollution levels that exceeded eutrophication limits in 2010 (EC, 2013).

#### Vegetation damage by ground-level ozone

Ground-level ozone is an air pollutant that is harmful to vegetation, but it is also harmful to the respiratory systems of humans and animals, and causes corrosion of materials and buildings. Ozone is formed near the ground due to the emissions of precursor gases which can come from both human activity and natural processes. Downward transport of ozone that exists in the stratosphere or intercontinental transport of ozone may also contribute to higher background ozone concentrations at ground level. The most important mechanism for removing ozone from the atmosphere is deposition on the earth's surface, in particular through absorption by plants. This absorption damages plant cells impairing their ability to grow, a phenomenon known as 'necrosis'. In some sensitive plants, ozone can cause the leaves to exhibit what appear to be burn-marks.

By impairing plants' reproduction and growth, high levels of ozone can thus lead to reduced agricultural crop yields, decreased forest growth, and reduced biodiversity.

## 1.5 Effects of air pollution on climate

Atmospheric pollution and climate change are distinct problems, but they are linked in several important ways. Greenhouse gases (GHGs), which cause climate change, generally have long lifetimes in the atmosphere, with methane (CH<sub>4</sub>) lasting about 12 years, and carbon dioxide (CO<sub>2</sub>) lasting about 100 years. Air pollutants, like SO<sub>2</sub>, PM, O<sub>3</sub> and NO, have lifetimes of a few days to weeks. Some of these air pollutants (certain constituents of PM, and  $O_3$ ) also have an effect on climate and are therefore called Short-Lived Climate Pollutants. Ground-level ozone is an example of such an air pollutant. It has severe impacts on public health and ecosystems, but it also contributes to global warming as it absorbs some of the infrared energy emitted by the earth and creates warming effects in its immediate surroundings. Ozone is a Short-Lived Climate Pollutant with very strong radiative forcing (7) effects on regional scales. In addition, ozone's effects on vegetation decrease photosynthesis, thereby also reducing plant uptake of carbon dioxide. Fine PM, the other major air pollutant, also has important climate impacts. Black carbon is one of the constituents of fine PM and has a warming effect, while other PM constituents (for instance sulphates and nitrates) may cool the climate.

Particles can also cause climate forcing indirectly, by changing the properties of clouds, such as cloud reflectivity, cloud distribution, cloud formation, and precipitation.

Air pollutants and GHGs (including  $CO_2$ ) are often emitted by the same sources, so certain GHG reduction measures (e.g. on power generation and transport) can also deliver reductions in air pollutants such as  $NO_2$ ,  $SO_2$ , and  $O_3$ . However, decarbonisation does not always reduce emissions of PM, one of the air pollutants of highest concern. For example, substituting biomass burning for fossil fuel combustion is often considered a switch to a climate neutral fuel by the climate convention. However, increased biomass burning leads to increased emissions of PM and other carcinogenic substances such as PAHs (poly-aromatic hydrocarbons), including BaP (benzo(a)pyrene).

## 1.6 Policy instruments and legislation

### Thematic strategy on air pollution

Within the European Union, the Sixth Environment Action Programme (EU, 2002) called for the development of a thematic strategy on air pollution with the objective of achieving levels of air quality that do not result in unacceptable impacts on, and risks to, human health and the environment. Formulated in 2005, the thematic strategy (EC, 2005b) defined the pathway towards achieving this objective through integrated actions across relevant policy areas. It set specific long-term environmental objectives for improvements in 2020 relative to the situation in 2000, specifically (EC, 2005c):

- a 47 % reduction in loss of life expectancy as a result of exposure to PM;
- a 10 % reduction in acute mortalities from exposure to O<sub>3</sub>;
- a 74 % reduction in excess acid deposition in forest areas and a 39 % reduction in surface freshwater areas;
- a 43 % reduction in areas or ecosystems exposed to eutrophication.

To achieve these objectives, it was estimated that key emissions would have to fall significantly in the period 2000–2020, specifically:

- SO<sub>2</sub> emissions to decrease by 82 %;
- NO<sub>x</sub> emissions to decrease by 60 %;
- VOC (volatile organic compounds) emissions to decrease by 51 %;
- NH<sub>3</sub> emissions to decrease by 27 %;
- Primary PM<sub>2.5</sub> (fine particles emitted directly into the air) to decrease by 59 %.

In the 'Roadmap to a Resource Efficient Europe' the European Commission has proposed the following

<sup>(&</sup>lt;sup>7</sup>) Radiative forcing is defined as the difference between radiant energy received by the earth and energy radiated back to space. In the case of ground-level ozone it is a positive forcing, contributing to the increase in temperature.

milestone for the policy: 'By 2020, the EU's interim air quality standards will have been met, including in urban hot spots, and those standards will have been updated and additional measures defined to further close the gap to the ultimate goal of achieving levels of air quality that do not cause significant impacts on health and the environment' (EC, 2011a).

## Legal instruments

In recent decades, the EU has introduced and implemented various legal instruments to improve air quality. There are three different legal mechanisms for air quality management. The first mechanism is the creation of limits or targets for ambient concentrations of air pollutants. The second mechanism is the placing of limits on total pollutant emissions (e.g. national totals). The third mechanism is the regulation of emissions from specific sources or sectors either by setting emission standards (e.g. for vehicle emissions) or by setting requirements on product quality (e.g. sulphur and benzene in fuel). In addition to these three mechanisms, several legal instruments in the EU, intended to minimise environmental impacts from different activities or promote environmental friendly behaviour, also contribute indirectly to minimise air pollution. In the paragraphs that follow below, we deal with each of these mechanisms in turn and give some examples of legal instruments with an indirect positive effect on air quality.

With regard to the first mechanism, the European directives currently regulating ambient air concentrations of the main pollutants are designed to avoid, prevent or reduce the harmful effects of air pollutants on human health and the environment. They comprise:

- Directive 2008/50/EC on ambient air quality and cleaner air for Europe, which regulates ambient air concentrations of SO<sub>2</sub>, NO<sub>2</sub> and other nitrogen oxides, PM<sub>10</sub> and PM<sub>2.5</sub>, Pb, C<sub>6</sub>H<sub>6</sub>, CO, and O<sub>3</sub> (EU, 2008c);
- Directive 2004/107/EC relating to arsenic (As), cadmium (Cd), mercury (Hg), nickel (Ni) and polycyclic aromatic hydrocarbons PAHs, (including BaP) in ambient air (EU, 2004b).

In the case of non-compliance with the air quality limit and target values stipulated in European legislation, air quality management plans must be developed and implemented in the areas where exceedances occur. The plans aim to bring concentrations of air pollutants to levels below the limit and target values.

With regard to the second mechanism, several EU directives regulate anthropogenic emissions of pollutants to air, including precursors of key air pollutants such as  $O_3$  and PM. The National Emission Ceilings Directive (EU, 2001b) and the Gothenburg Protocol (UNECE, 1999) to the UN Convention on Long-range Transboundary Air Pollution (LRTAP, which was revised in 2012), set national emission limits for SO<sub>2'</sub> NO<sub>x'</sub> NMVOCs and NH<sub>3</sub> in order to abate acidification, eutrophication and ground-level ozone. The revised Gothenburg Protocol also includes ceilings for PM<sub>2.5</sub> emissions.

With regard to the third mechanism, several directives and international conventions regulate emissions of the main air pollutants from specific sources and sectors, either by setting emission standards, by requiring the use of the best available technology, or by setting requirements on fuel composition. These directives and international conventions include:

- Directive 2010/75/EU on industrial emissions (integrated pollution prevention and control) (EU, 2010), targets certain industrial, agricultural, and waste treatment installations.
- The Euro Directives set standards for road vehicle emissions. The Euro 5 and 6 standards are set in Regulations (EC) No 692/2008 (EU, 2008a) and No 595/2009 (EU, 2009b).
- Directive 94/63/EC on the control of VOC emissions resulting from the storage of petrol and its distribution from terminals to service stations (EU, 1994) and Directive 2009/126/EC on Stage II petrol vapour recovery during refuelling of motor vehicles at service stations (EU, 2009a).
- Directive 1999/13/EC on the limitation of emissions of VOCs due to the use of organic solvents in certain activities and installations (EU, 1999a).
- Directive 2012/33/EU (EU, 2012) amending Directive 1999/32/EC as regards the sulphur content of marine fuels, Directive 1999/32/EC on reduction of sulphur content of certain liquid fuels (EU, 1999b), and Directive 2003/17/EC

(amending Directive 98/70/EC) relating to the quality of petrol and diesel fuels (EU, 2003a).

- The Marine Pollution Convention, MARPOL73/78 (IMO, 1973), which is the main international convention on preventing pollution by ships from operational or accidental causes. Annex VI sets limits on air pollution from ships for SO<sub>x</sub>, NO<sub>x</sub>, VOC and PM from ship exhausts and prohibits deliberate emissions of ozone-depleting substances.
- The 2004 and 2008 air quality directives do not specify an air quality objective for NH<sub>3</sub>. The Gothenburg Protocol (UNECE, 1999) under the LRTAP convention and the National Emission Ceilings Directive (EU, 2001b) set emission reduction targets for NH<sub>3</sub> with the aim of reducing acidification and eutrophication. Reporting of NH<sub>3</sub> emissions is also required under the Integrated Pollution Prevention and

Control (IPPC) Directive (EU, 2008b), now replaced by Directive 2010/75/EU on industrial emissions (EU, 2010).

Table 1.2 summarises the coverage of the European directives and international conventions regulating air pollutant emissions (either directly or indirectly by regulating emissions of precursor gases). The list is not exhaustive. Annex 2 provides a more detailed description of the directives regulating fuel quality and emissions to air.

In addition to these three mechanisms described above, there are several EU directives intended to minimise environmental impacts, including on climate change, and/or promote environmental friendly behaviour, which also contribute indirectly to minimise air pollution. Examples are:

• the implementation of the Nitrates Directive (91/676/EEC) concerning the protection of

	Pollutants Policies	РМ	0,3	NO <sub>2</sub> NO <sub>x</sub> NH <sub>3</sub>	SO <sub>2</sub> SO <sub>x</sub> S	CO	Heavy metals	BaP PAH	VOCs
Directives	2008/50/EC	PM	03	NO <sub>2</sub>	SO <sub>2</sub>	CO	Pb		C <sub>6</sub> H <sub>6</sub>
regulating ambient air quality	2004/107/EC						As, Cd, Hg, Ni	BaP	
Directives regulating	2001/81/EC	(ª)	(b)	NO <sub>x</sub> , NH <sub>3</sub>	SO <sub>2</sub>				NMVOC
emissions of air pollutants	2010/75/EU	PM	( <sup>b</sup> )	NO <sub>x</sub> , NH <sub>3</sub>	SO <sub>2</sub>	CO	Cd, Tl, Hg, Sb, As, Pb, Cr, Co, Cu, Mn, Ni, V		VOC
	Euro standards on road vehicle emissions	РМ	(b)	NO <sub>x</sub>		CO			HC (hydrocarbons), NMHC (non-methane hydrocarbons)
	94/63/EC	(a)	( <sup>b</sup> )						VOC
	2009/126/EC	(a)	(b)						VOC
	1999/13/EC	(a)	(b)						VOC
	91/676/EEC			NH3					
Directives	1999/32/EC	(a)			S				
regulating fuel quality	2003/17/EC	(a)	( <sup>b</sup> )		S		Pb	PAH	C <sub>6</sub> H <sub>6</sub> , HC (hydrocarbons), VOCs
International	MARPOL 73/78	PM	(b)	NO <sub>x</sub>	SO <sub>x</sub>				VOC
conventions	LRTAP	PM (ª)	( <sup>b</sup> )	NO <sub>2</sub> , NH <sub>3</sub>	SO <sub>2</sub>	CO	Cd, Hg, Pb	BaP	NMVOC

# Table 1.2Legislation in Europe regulating emissions and ambient concentrations of air<br/>pollutants

**Note:** (<sup>a</sup>) Directives and conventions limiting emissions of particulate matter precursors, such as SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and VOCs, indirectly aim to reduce particulate matter ambient air concentrations.

(b) Directives and conventions limiting emissions of ozone precursors, such as  $NO_x$ , VOC and CO, indirectly aim to reduce troposphere ozone concentrations.

waters against pollution caused by nitrates from agricultural sources (EU, 1991), in particular the implementation of agricultural practices that limit fertiliser application and prevent nitrate losses, leads to a decrease of agricultural emissions of nitrogen compounds to air.

 the Energy Taxation Directive (2003/96/EC; EU, 2003b) establishing minimum taxes on motor fuels, heating fuels and electricity, depending on the energy content of the product and the amount of CO<sub>2</sub> it emits. It aims at promoting energy efficiency and less polluting energy products.

### Policy developments

The EU's air policy is currently under a comprehensive review process, and already two near-term measures have been adopted:

- for international shipping, tighter shipping fuel standards and emission standards at the IMO/ MARPOL level resulted in the recent revision of the Sulphur Content of Fuel Directive (adopted as 2012/33/EU); and
- international initiatives within the UNECE CLRTAP on hemispheric transport of air

pollution culminated in the revision of the Gothenburg Protocol in May 2012.

The revised UNECE Gothenburg Protocol establishes new international air pollution emission ceilings for those pollutants already listed in the NEC Directive (NO<sub>x</sub>, VOC, SO<sub>2</sub> and NH<sub>2</sub>), and primary fine particulate matter  $(PM_{25})$  to be met by 2020. However, the revised Gothenburg Protocol on its own does not ensure the achievement of the specific long-term environmental objectives set by the thematic strategy for 2020 (EC, 2005b), except for the objective on reduction in acute mortalities from exposure to  $O_3$  (EC, 2013). Thus, the objectives of the thematic strategy can be met by combining the EU's obligations to reduce air pollution as agreed under the 2012 amendment of the Gothenburg Protocol with additional reductions agreed within the EU. These two sets of emissions reductions will form the basis of a revised National Emission Ceilings Directive and various source control measures at EU and national level.

Other on-going policy measures include the recent Cars 2020 Communication (EC, 2012) setting out a timetable for the successful implementation of the Euro 6 vehicle standards in real-world driving conditions, and the revision of the Non-road Mobile Machinery legislation.

# 2 Particulate matter (PM)

## 2.1 Sources and effects of PM

### 2.1.1 Origins of PM in air

Particulate matter is the general term used for a mixture of particles (solid and liquid) suspended in the air, with a wide range of sizes and chemical compositions.  $PM_{2.5}$  refers to 'fine particles' that have a diameter of 2.5 micrometres or less.  $PM_{10}$  refers to particles with a diameter of 10 micrometres or less (see Figure 2.1).  $PM_{10}$  includes the 'coarse particles' fraction in addition to the  $PM_{2.5}$  fraction.

Particulate matter — also collectively known as aerosols — can be further categorised as either primary particulate matter or secondary particulate matter. Primary particulate matter enters the atmosphere directly (e.g. from chimneys). Secondary particulate matter is formed in the atmosphere from the oxidation and transformation of primary gaseous emissions. These gaseous emissions that contribute to particle formation are also known as precursor gases. The most important precursor gases for secondary particles are SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and VOCs (volatile organic compounds, a class of chemical compounds whose molecules contain carbon). The main precursor gases  $SO_{\gamma}$ ,  $NO_{\gamma}$  and NH<sub>2</sub> react in the atmosphere to form ammonium, sulphate compounds, and nitrate compounds. These compounds then condense into liquid form and form new particles in the air, called secondary inorganic aerosols (SIAs). Certain VOCs are oxidised to form less volatile compounds, which form secondary organic aerosols (SOAs). The formation of SIA and SOA in the atmosphere depends on a variety of chemical and physical factors. The first of these factors is the concentrations of the main precursors. The second of these factors is the reactivity of the atmosphere, which depends on the concentrations of highly reactive substances such as O<sub>3</sub> and the hydroxyl radical (called a radical because it contains an unpaired electron). The third of these factors is meteorological conditions, like solar radiation, relative humidity and cloud cover.



## Figure 2.1 Illustration of PM<sub>2.5</sub> and PM<sub>10</sub> particle size

Source: EPA, 2010.

When all of the previously mentioned main chemical components of an aerosol (including crustal material, sea salt, black carbon, dust, SIAs and SOAs) are measured, they account for about 70 % or more of the mass of  $PM_{10}$  and  $PM_{2.5}$ . The remaining 30 % is thought to be made up of water (Putaud et al., 2004).

Particulate matter can come from natural sources or anthropogenic sources. Natural sources include sea salt, naturally suspended dust, pollen, and volcanic ash (see EEA, 2012c). Anthropogenic sources include fuel combustion in thermal power generation, incineration, domestic heating for households, and fuel combustion for vehicles. In cities, important local sources include vehicle exhausts, road dust re-suspension, and the burning of wood, fuel or coal for domestic heating. These are all low emitters, below 20 meters, leading to significant impacts on the concentration levels close to ground.

## 2.1.2 Effects of PM

Epidemiological studies attribute the most severe health effects from air pollution to PM and, to a lesser extent, ozone. Even at concentrations below current air quality guidelines PM poses a health risk. The evidence for an association between PM and short-term health effects (as well as long-term health effects) has become much stronger. Recent long-term studies show associations between PM and mortality at levels well below the current annual WHO air quality guideline level for  $PM_{2.5}$  (10 µg/m<sup>3</sup>). Scientific evidence does not suggest a threshold of exposure to PM below which no adverse health effects would be anticipated (WHO, 2006a, 2006b, 2013).

The health effects of PM are caused after their inhalation and penetration into the lungs and blood stream, leading to adverse effects in the respiratory, cardiovascular, immune, and neural systems. Ultrafine particles (with diameters of 0.1 micrometres or less) can also penetrate into the brain through the nose (Breysse et al., 2013). Both chemical and physical interactions between PM and lung tissues can induce irritation or damage. The smaller the particles, the deeper they penetrate into the lungs. PM's mortality effects are clearly associated with the  $PM_{2.5}$  fraction, which in Europe represents 40-80 % of the PM<sub>10</sub> mass concentration in ambient air. However, the 'coarser' 2.5-10 µm fraction of PM<sub>10</sub> also has health impacts and affects mortality.

The current levels of PM exposure experienced by most urban and rural populations have harmful effects on human health. Chronic exposure to PM contributes to the risk of developing cardiovascular and respiratory diseases, as well as lung cancer. Mortality associated with air pollution is about 15–20 % higher in cities with high levels of pollution compared to relatively cleaner cities. In the European Union, average life expectancy is estimated to be 8.6 months lower due to exposure to  $PM_{25}$  resulting from human activities (WHO, 2006b).

In addition to its effects on the human health, PM can also have adverse effects on climate change and ecosystems, as indicated in Table 1.1. PM also contributes to soiling of buildings, and can even have a corrosive effect on buildings, depending on the PM composition.

# 2.2 European air quality standards for PM

The EU PM<sub>10</sub> and PM<sub>2.5</sub> limit and target values for health protection are shown in Table 2.1. The deadline for Member States to meet the PM<sub>10</sub> limit values was 1 January 2005. The deadline for meeting the target value for PM<sub>2.5</sub> (25  $\mu$ g/m<sup>3</sup>) was 1 January 2010, while the deadlines for meeting the other limit and 'obligation' values for PM<sub>2.5</sub> (20  $\mu$ g/m<sup>3</sup>) are 2015 or 2020.

For  $PM_{10}$  there are limit values for short-term (24-hour) and long-term (annual) exposure, while for  $PM_{25}$  there are only values for long-term (annual) exposure. In Europe, the short-term limit value for  $PM_{10}$  (i.e. not more than 35 days per year with a daily average concentration exceeding 50 µg/m<sup>3</sup>) is the limit value most often exceeded in European cities and urban areas.

The WHO AQG, shown in Table 2.2, are stricter than the EU air quality standards. The WHO (2008) explains the reasoning behind its limit values as follows:

> 'The 2005 AQG set for the first time a guideline value for PM. The aim is to achieve the lowest concentrations possible. As no threshold for PM has been identified below which no damage to health is observed, the recommended value should represent an acceptable and achievable objective to minimise health effects in the context of local constraints, capabilities and public health priorities.'

# Table 2.1Air quality limit and target values for PM10and PM2.5as given in the Air QualityDirective

Size fraction	Averaging period	Value	Comments	
$PM_{10}$ , limit value	One day	50 µg/m³	Not to be exceeded on more than 35 days per year. To be met by 1 January 2005	
PM <sub>10</sub> , limit value	Calendar year	40 µg/m <sup>3</sup>	To be met by 1 January 2005	
PM <sub>2.5</sub> , target value	Calendar year	25 µg/m³	To be met by 1 January 2010	
PM <sub>2.5</sub> , limit value	Calendar year	25 µg/m³	To be met by 1 January 2015	
PM <sub>2.5</sub> , limit value ( <sup>a</sup> )	Calendar year	20 µg/m <sup>3</sup>	To be met by 1 January 2020	
PM <sub>2.5</sub> , exposure concentration obligation ( <sup>b</sup> )		20 µg/m³	2015	
PM <sub>2.5</sub> exposure reduction target ( <sup>b</sup> )	0-20 % reduction in exposure (depending on the average exposure indicator in the reference year) to be met by 2020			

**Note:** (a) Indicative limit value (Stage 2) to be reviewed by the Commission in 2013 in the light of further information on health and environmental effects, technical feasibility and experience of the target value in Member States.

(b) Based on a three-year average.

Source: EU, 2008c.

#### Table 2.2WHO air quality guidelines

	24-hour mean (μg/m³)	Annual mean (µg/m³)			
PM <sub>2.5</sub>	25	10			
PM <sub>10</sub>	50	20			

Source: WHO, 2006a.

## 2.3 Europe-wide survey of PM

#### 2.3.1 Exceedances of limit and target values

The EU limit values (applying from 2005) and target value (applying from 2010) for PM (the former for  $PM_{10}$  and the latter for  $PM_{2.5}$ ) were exceeded widely in Europe in 2011, as the data of the European air quality database, AirBase (Mol and Hooydonk, 2013), and Map 2.1, Map 2.2 and Figure 2.2 show.

The annual limit value for  $PM_{10}$  (applying from 2005) was exceeded most often (red and dark red dots in Map 2.1) in Poland, Italy, Slovakia, the Balkan region, Turkey and also in several urban regions. Concentrations of above 31 µg m<sup>-3</sup> as an annual mean correspond to exceedances of the daily limit value (also applying from 2005) for  $PM_{10}$ .

The daily limit value was exceeded (orange dots in Map 2.1) in other cities in those countries, as well as in many other countries in central, western and southern Europe. Cities in Latvia, Sweden and the United Kingdom also exceeded the daily limit value for  $PM_{10}$ . Notably, when assessing compliance, the Air Quality Directive allows EU Member States to remove quantified contributions from natural sources over which Member States have no control. The data presented here is before such a correction would be taking place.

There are more monitoring stations measuring  $PM_{10}$ than measuring  $PM_{2.5}$ . For  $PM_{2.5}$  in 2011 there were 902 stations fulfilling the criterion of more than 75 % data coverage. (The data coverage gives the fraction of the year for which valid concentration data are available at each location). This marks an increase of 148 stations compared to the 754 stations that in 2010 measured  $PM_{25}$  with 75 % data coverage or more. In 2011, the  $PM_{25}$  concentrations were higher than the annual target value to be met by 2010 (dark red, red and orange dots in Map 2.2) at several stations in Bulgaria, the Czech Republic, France, Italy, Poland and Slovakia. The stricter value of the WHO guidelines for annual mean PM were exceeded (pale green, yellow, orange, red and dark red dots in Maps 2.1 and 2.2) at most of the monitoring stations across continental Europe, but were exceeded less in Nordic countries.



## Map 2.1 Annual mean concentrations of PM<sub>10</sub> in 2011

Note: The red and dark red dots indicate stations reporting exceedances of the 2005 annual limit value (40 μg/m<sup>3</sup>), as set out in the Air Quality Directive (EU, 2008c).

The orange dots indicate stations reporting exceedances of a statistically derived level (31  $\mu g/m^3$ ) corresponding to the 24-hour limit value.

The pale green dots indicate stations reporting exceedances of the WHO air quality guideline for  $PM_{10}$  of less than 20  $\mu$ g/m<sup>3</sup>.

The dark green dots indicate stations reporting concentrations below the WHO air quality guideline of 20  $\mu$ g/m<sup>3</sup> for PM<sub>10</sub>.

Source: AirBase v. 7.

# 2.3.2 Rural PM background level and secondary PM from precursor gases

The rural background concentration of PM represents the concentration of PM in rural areas. Contributions to PM from urban emissions build on the rural 'background' level to produce the concentrations occurring in urban areas (more generally called urban background concentrations). However, while local control efforts can reduce urban contributions to PM, they will have limited effects on the rural background level, a portion of which is also the result of natural factors.

The rural background concentration level of PM constitutes a substantial part of the PM concentrations measured in cities. Rural concentrations vary across Europe. The highest measured  $PM_{10}$  and  $PM_{25}$  annual mean concentrations at rural background sites in 2011



### Map 2.2 Annual mean concentrations of PM<sub>2.5</sub> in 2011

Note: The dark red dots indicate stations reporting exceedances of the 2010 annual target value (25 μg/m<sup>3</sup>) plus at least 5 μg/m<sup>3</sup>. The red dots indicate stations reporting exceedances of the 2010 annual target value (25 μg/m<sup>3</sup>), as set out in the Air Quality Directive (EU, 2008c).

The orange dots indicate stations reporting exceedances of the 2020 indicative annual limit value (20  $\mu$ g/m<sup>3</sup>), as set out in the Air Quality Directive (EU, 2008c).

The pale green dots indicate stations reporting exceedances of the WHO air quality guideline for  $PM_{2.5}$  of less than 10  $\mu$ g/m<sup>3</sup>.

The dark green dots indicate stations reporting concentrations below the WHO air quality guideline for PM2.5.

Source: AirBase v. 7.

were in Italy and the Czech Republic, with annual means above the  $PM_{10}$  limit value of 40 µg/m<sup>3</sup> and the  $PM_{25}$  target value of 25 µg/m<sup>3</sup>. In addition to primary PM emissions (natural and anthropogenic), rural PM concentrations are determined by contributions from secondary particles, both secondary inorganic aerosols (SIAs) and secondary organic aerosols (SOAs). The latter are partly formed from organic gases emitted from anthropogenic sources and natural sources relating primarily to terrestrial vegetation. The SIA and SOA contribution varies

substantially across Europe and from season to season. The SIA contribution is higher in winter, due to increased emissions from combustion in the cold season, and the SOA contribution is generally higher in summer, when emissions from terrestrial vegetation are larger, increasing from the northern parts to the southern parts of the continent.

The chemical composition of PM is quite different across Europe: on average there is more carbonaceous matter (PM made up of carbon in



# Figure 2.2 Concentration status for daily limit value of PM<sub>10</sub> (left) and for annual target value of PM<sub>2.5</sub> (right), 2011

**Note:** The left figure is based on the 90.41 percentile of the  $PM_{10}$  daily concentrations, corresponding to the 36th highest daily  $PM_{10}$  concentration, used to compare with the  $PM_{10}$  daily limit value of 50 µg/m<sup>3</sup>.

**Source:** AirBase v. 7.

#### Box 2.1 The chemistry of PM formation

#### **Inorganic PM**

In Europe, about one third of the  $PM_{10}$  concentration and half of the  $PM_{2,5}$  concentration measured at regional 'background' stations (sampling stations located in rural areas, away from traffic or other direct sources of pollution) consist of inorganic chemical substances, such as ammonium  $(NH_4^+)$ , nitrate  $(NO_3^-)$  and sulphate  $(SO_4^{-2})$ . These substances are the result of chemical reactions in the atmosphere involving the PM precursor gases:  $NH_3$ ,  $NO_x$  and  $SO_x$ .

Sulphate-forming reactions involve the gas-phase conversion of SO<sub>2</sub> to sulphuric acid and aqueous phase chemical reactions, which may occur in cloud and fog droplets or in liquid films on atmospheric particles. The rate of some of these reactions is enhanced by the presence of metals, such as iron and manganese. The NO<sub>2</sub> portion of NO<sub>x</sub> can be converted to nitric acid. Both sulphuric and nitric acids thus formed react with NH<sub>3</sub> and form ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) and ammonium sulphate (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, both of which are components of PM. In addition, nitric acid may react with chemical substances in coarse particles (those particles between 2.5 µm and 10 µm in diameter) and add nitrate to the coarse particle fraction.

#### Organic PM

Organic substances make upon average about 30 % of the PM<sub>2.5</sub> concentrations and 20 % of the PM<sub>10</sub> concentrations measured at European regional background stations. Organic PM is composed of hundreds of individual chemical substances. Some of the organic substances are semi-volatile, such that they can exist both as condensed in PM and as gases. This ability to exist as particles or as a gas complicates the sampling process. Consequently, it is difficult to obtain complete chemical information of the organic substances.

Organic particulate matter is formed by numerous chain reactions involving a number of gaseous VOC compounds (such as alkanes, olefins, aromatics), isoprene and terpene. There is a clear link between  $O_3$  episodes and formation of organic PM since  $O_3$  participates in some of those chemical reactions.

different forms) in PM<sub>10</sub> in central Europe, more nitrate in it in north-western Europe, and more mineral dust in it in southern Europe (EMEP, 2011; Putaud et al., 2010). The contribution of sea salt to aerosol mass is highly dependent on distance to the sea, i.e. it varies from about 0.5 % of aerosol mass at some inland sites to around 15 % at sites close to the coast (Tørseth et al., 2012). Wind-blown desert dust from Africa is the largest PM<sub>10</sub> component in rural background southern sites of the Mediterranean, where it makes up between 35 % and 50 % of  $PM_{10}$  (Pey et al., 2013). Carbonaceous matter is a significant component of the atmospheric aerosol mass, accounting for between 10 % and 40 % of the  $PM_{10}$  at the EMEP sites (Yttri et al., 2007), and between 35 % and 50 % of the  $PM_{10}$  in southern sites of the Mediterranean. Furthermore, PM chemical composition measurements show that there is a clear decrease in the relative contribution of  $SO_4^{2-}$  and  $NO_3^{-1}$  to  $PM_{10}$  when one moves away from rural sites and toward urban and traffic sites. In contrast, the contribution of carbon particles to the total  $PM_{10}$ ratio increases as one moves from rural to traffic sites (Putaud et al., 2010).

## 2.3.3 Distance to target

The difference between current PM levels and the EU's limit and target values for PM is known as the 'distance to target'. To indicate the distance to target for PM, Figure 2.2 shows the extent of the exceedances in 2011 of the 24-hour limit value for  $PM_{10}$  (to be met by 2005) and of the target value for  $PM_{2.5}$  (to be met by 2010) within the EU. The analysis here is based on measurements at fixed sampling points and does not account for the fact that the Air Quality Directive (EU, 2008c) provides the Member States with the possibility of subtracting the contribution of natural sources (<sup>8</sup>) and winter road sanding/salting when limits are exceeded (EEA, 2012c).

Fixed sampling points in Europe are situated at four types of sites:

- traffic-related locations;
- urban and sub-urban background (non-traffic) locations;

- industrial locations (or other less defined locations);
- rural background sites.

In 2011, the  $PM_{10}$  24-hour limit value (that the relevant European directive declared should have been met by 2005) was exceeded at 43 % of traffic sites, 38 % of urban background sites, 26 % of 'other' sites (mostly industrial) and even at 15 % of rural sites within the EU. The percentage of stations in exceedance at rural background sites has more than doubled from 2009 to 2011. Countries like Austria, the Czech Republic, Germany, Italy, the Netherlands and Poland have had a significant average increase in  $PM_{10}$  concentrations from 2009 to 2011 (in terms of the 36th highest daily concentration, corresponding to the indicator used for the daily limit value).

The  $PM_{10}$  24-hour limit value is more stringent than the annual limit value and is more frequently exceeded. Figure 2.3 shows for all Member States the attainment of the  $PM_{10}$  24-hour limit value in 2011. It clearly indicates that exceedance of the daily limit value was observed in 22 Member States at one or more stations in 2011, with only Estonia, Finland, Ireland, Malta and Luxembourg recording no exceedance. The only country with  $PM_{10}$ concentration data for 2001, 2005, 2010 and 2011, which did not register an exceedance of the  $PM_{10}$ 24-hour limit value in any of the years, was Ireland.

Figure 2.4 shows a similar plot, but this time it is for the annual mean  $PM_{2.5}$  values for 2011 in the EU Member States. It shows that exceedance of the target value for  $PM_{2.5}$  threshold (25 µg/m<sup>3</sup>, and that the relevant European directive declared should have been met by 2010) was observed in eleven Member States at one or more stations in 2011, mostly in eastern Europe. The only country with  $PM_{2.5}$  data for 2001, 2005, 2010 and 2011 that did not register an exceedance of this target value for  $PM_{2.5}$ in any of the four years was Finland.

The  $PM_{2.5}$  target value threshold was exceeded in 2011 at 10 % of traffic sites, 18 % of urban background sites, 7 % of 'other' (mostly industrial) sites, and 5 % of rural sites. Particular sites in Bulgaria and Poland registered annual mean concentrations of  $PM_{2.5}$  concentrations close to or above double the target value threshold.

<sup>(8)</sup> Examples of natural sources the directive allow to subtract are: volcanic eruptions, seismic activities, geothermal activities, wild-land fires, high-wind events or the atmospheric resuspension or transport of natural particles from dry regions and sea spray.



### Figure 2.3 Attainment situation for PM<sub>10</sub> in 2011

**Note:** The graph is based on the 90.41 percentile of PM<sub>10</sub> daily mean concentration values corresponding to the 36th highest daily mean for each Member State; the boxes present the range of concentrations at all stations types (in μg/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the limit value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

Source: ETC/ACM.



### Figure 2.4 Attainment situation for PM<sub>2.5</sub> in 2011

**Note:** The graph is based on PM<sub>2.5</sub> annual mean concentration values; they present the range of concentrations at all stations types (in µg/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the target value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

Source: ETC/ACM.

These findings demonstrate that PM concentrations must be reduced substantially in large areas of Europe for the limit and target values to be met.

### 2.3.4 Trends in PM concentrations

The average change in  $PM_{10}$  annual mean concentrations since 2002 is presented in Figure 2.5,

for traffic, urban background, rural background and other (mostly industrial) stations. On average, all station types show decreasing concentrations since 2002, but some stations in all station types have registered an increase. Map 2.3 shows the average changes in  $PM_{10}$  annual mean concentrations between 2002 and 2011 (<sup>9</sup>). Most of the stations registering a trend (<sup>10</sup>) recorded decreasing annual mean concentrations of  $PM_{10}$  by more than 1 µg/m<sup>3</sup>

# Figure 2.5 Trends in PM<sub>10</sub> (top graph, 2002–2011) and PM<sub>2.5</sub> (bottom graph, 2006–2011) annual concentrations (in µg/m<sup>3</sup>) per station type



**Ste:** The graphs are based on annual mean concentration trends for  $PM_{10}$  (top) and  $PM_{2.5}$  (bottom); they present the range of concentration changes per year (in µg/m<sup>3</sup>) per station type (urban, traffic, rural and other — mostly industrial). The trends are calculated based on the officially reported data by the EU Member States with a minimum data coverage of 75 % of valid data per year, for at least 8 out of the 10 years period for  $PM_{10}$  and for at least 5 out of the 6 years period for  $PM_{2.5}$ . In 2006, France introduced a nation-wide system to correct  $PM_{10}$  measurements. French  $PM_{10}$  data prior to 2007 have been corrected here using station-type dependent factors (de Leeuw and Fiala, 2009).

The diagram indicates the lowest and highest trends, the means and the lower and upper quartiles, per station type. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

Source: ETC/ACM.

<sup>(9)</sup> A consistent set of 953 stations with data for 2002 to 2011 was used in the trend analysis in the EEA-32 countries. Of these, 430 stations registered a trend (i.e. a statistically significant trend, using the Mann-Kendall test). The remaining 523 stations had no significant trend.

 $<sup>(^{10})</sup>$  226 stations, which is the equivalent of 53 % of the stations with a trend.


#### Map 2.3 Annual changes in concentrations of PM<sub>10</sub> in the period 2002–2011

**Note:** The data presented were derived from a consistent set of stations in all years. In 2006, France introduced a nation-wide system to correct  $PM_{10}$  measurements. French  $PM_{10}$  data prior to 2007 have been corrected using station-type dependent factors (de Leeuw and Fiala, 2009).

Statistically significant trends (level of significance 0.1) are calculated by applying the Mann-Kendall test. Increasing concentrations are indicated with red dots and decreasing concentrations with green dots, when statistically significant. The applied method is described in de Leeuw, 2012.

Source: ETC/ACM.

per year during this period. Only 2 % of the stations registered a positive trend (meaning increasing concentrations) from 2002 to 2011 (<sup>11</sup>). Table A1.1 and Table A1.2 (Annex 1) show the average trends by countries and by station type for PM<sub>10</sub>/ for the period 2002–2011. The tables show that the 6 stations in Bulgaria with data from 2002 to 2011 have registered an increase in PM<sub>10</sub> concentrations, but no significant trend (<sup>12</sup>). Hungary and Poland have also registered increasing trends (<sup>13</sup>) for some of the station types.

The number of  $PM_{2.5}$  stations operational over the last six years is still limited (<sup>14</sup>). Concentrations, on average, tended to decrease during the period 2006–2011 for traffic and other (mostly industrial) stations, but increase for urban and rural background stations (Figure 2.5). Table A1.3 (Annex 1) shows the trends for mean annual  $PM_{2.5}$  by country and by station type for the period 2006–2011. Several countries have registered increasing trends of  $PM_{2.5}$  annual mean

<sup>(&</sup>lt;sup>11</sup>) 2 % is equivalent to ten stations.

<sup>&</sup>lt;sup>(12)</sup> The statistical significance in trend was calculated using the Mann-Kendall test.

<sup>(&</sup>lt;sup>13</sup>) Both significant and non-significant trends, according to the Mann-Kendall test.

<sup>(&</sup>lt;sup>14</sup>) A consistent set of 163 stations with data for 2006 to 2011 was used in the trend analysis. Of these, only 21 % (34 stations) registered a trend (i.e. a significant trend, using the Mann-Kendall test). The remaining 129 stations had no significant trend.

concentrations for one or more station types in the period 2006–2011. This is the case for France, Germany, Belgium, Austria, Hungary, the Czech Republic, Slovakia, Denmark and Poland. The available data for PM<sub>2.5</sub> are too limited to draw firm conclusions about the observed trends, as in some cases they were based on measurements from only one or two stations, but the development is clearly not satisfactory across Europe.

#### Emissions of primary PM and precursor gases

When explaining trends in PM concentrations in the air, emission trends for both primary PM and precursor gases must be considered. In addition to anthropogenic and natural emissions, meteorology plays an important role. A certain fraction of the emitted precursor gases forms particles in the air, depending on atmospheric conditions (temperature, sunlight, humidity, reaction rate). Dispersion and atmospheric conditions differ also from year to year. These meteorological conditions have not been taken into consideration in the present analysis.

The European anthropogenic emissions inventory (a record of what types of emissions are being released by what sources and in what quantities) of primary PM is almost complete, with the exception of nonexhaust emissions (tyre and road wear), which have not been fully reported by all countries. Natural primary emissions of PM (primarily sea salt and naturally suspended soil dust including desert dust) are not part of this inventory. The EU emissions inventory for the period 1990–2011 was published by the EEA (2013b).

Emissions of primary PM fell in the EU by 14 % for PM<sub>10</sub> and 16 % for PM<sub>2.5</sub> between 2002 and 2011 (Figure 2.6). The reductions in the same period for the 32 EEA member countries were 9 % for PM<sub>10</sub> and also 16 % for PM<sub>2.5</sub>. Emissions of the precursor gases  $SO_x$  and  $NO_x$  declined by 50 % and 27 % respectively in the period 2002 to 2011 in the EU, and by 34 % and 23 % in the EEA-32 countries. Emissions of NH<sub>3</sub>, another precursor gas, have fallen less, declining by only about 7 % in the EU and 5 % in the 32 EEA member countries between 2002 and 2011.

Organic precursor gases of secondary organic aerosols (SOAs) are dominated by natural organic emissions but also include an anthropogenic component. Natural VOC emissions are not included in the present emission inventories.

Depending partly on the atmospheric conditions, SIAs contribute on average about one third (35 % in 2010) of the  $PM_{10}$  mass in rural air in Europe, while central Europe had the highest SIA contribution to rural background concentrations of  $PM_{10}$  in 2010 with around 50 % (EMEP, 2012). SIAs account for a lower percentage of PM in urban air because local emissions of primary particles add to the urban PM mass concentrations.



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Figure 2.7 Contributions to EU emissions from main source sectors of primary PM,  $NO_{x'} SO_{x'}$ NMVOCs and  $NH_{3'} 2002-2011$  (Gg/year = 1 000 tonnes/year)

Source: EEA.

#### Sectoral output of primary PM and precursor gases

Various source sectors contribute to the primary anthropogenic PM and precursor gases (Figure 2.7). Commercial, institutional and household fuel combustion dominates emissions of primary  $PM_{10}$  and  $PM_{2.5}$  and has slightly increased its emissions since 2002. This sector's share of the total primary PM emissions has also increased from 33 % in 2002 to 39 % in 2011 for  $PM_{10}$  and from 42 % to 50 % of the total  $PM_{2.5}$  primary emissions.

The second largest source of emissions of primary PM<sub>10</sub> is industry, followed by transport. For PM<sub>25</sub> both sectors have had roughly similar levels of emissions, with the transport sector having only slightly higher emissions than the industrial sector. Non-exhaust emissions from road traffic, which are not included in Figure 2.7, add to the total road traffic emission contribution. Non-exhaust emissions are estimated to equal about 50 % of the exhaust emissions of primary PM<sub>10</sub> and about 22 % of the exhaust emissions of primary  $PM_{2.5}$ (Hak et al., 2009). It has been shown that even with zero tail-pipe emissions, traffic will continue to contribute to fine and ultrafine particles through non-exhaust emissions (Dahl et al., 2006; Kumar et al., 2013) and it is estimated that nearly 90 % of the total PM emissions from road traffic will come from non-exhaust sources by the end of the decade (Rexeis and Hausberger, 2009). The transport sector is clearly the largest contributor to NO<sub>x</sub> emissions, while the energy production and industrial sectors dominate the SO<sub>v</sub> emissions. The agricultural sector was responsible for 93 % of the total NH<sub>3</sub> emissions in the EU in 2011 and has only decreased its NH<sub>3</sub> emissions by 7 % between 2002 and 2011. European policies have cut PM precursor gas emissions significantly, with the exception of NH<sub>3</sub>. It is estimated that current European policies cut NO<sub>x</sub> emissions from road vehicles by 55 % and from industrial plants by 68 % in the period 1990–2005, compared to a hypothetical situation with no directives in force. The policy-induced reduction in SO<sub>x</sub> emissions from the industrial plant sector is estimated at 70 % in the same period (EEA, 2010b).

# Relationship of emissions to ambient PM concentrations

Emissions of primary PM from commercial, institutional and household fuel combustion have increased slightly since 2002 (Figure 2.7), even if there has been a reduction in officially reported emissions between 2010 and 2011. This means that this source may contribute to keeping PM concentrations elevated in both rural and urban areas, despite emission reductions in other sectors. Contrastingly, diminishing primary PM emissions from transport may compensate for that increase, especially in urban areas.

The reductions in emissions of the PM precursors  $NO_x$  and  $SO_x$  were much larger than the reductions in primary PM from 2002 to 2011. Meanwhile the reduction in  $NH_3$  emissions was small (about 7 %) between 2002 and 2011 in the EU and even smaller (5 %) in the EEA-32. There was a slight upward tendency observed in  $PM_{2.5}$  concentrations at rural sites between 2008 and 2010 (Figure 2.5 in EEA, 2012b). This increase was not reversed in 2011 when average  $PM_{2.5}$  levels across Europe were maintained at 2010 levels.

There is a conundrum in the relationship between PM concentrations on the one hand and emissions of primary PM and PM precursors on the other hand. Sharp falls in emissions have not led to equally sharp falls in concentrations of PM. Part of this conundrum can be explained by uncertainties in the reported emissions of primary PM from the commercial, institutional and household fuel combustion sector. In addition, and as discussed in EEA (2012b), intercontinental transport of PM and its precursor gases from outside Europe may also influence European ambient levels, pushing up PM levels in spite of falling emissions in Europe.

Beauchamp et al. (2013) have modelled the sensitivity of PM concentrations across Europe to reductions in NH<sub>3</sub> emissions from agriculture. The results, from three different chemistry transport models, show that as currently planned, the revised Gothenburg Protocol will only reduce the number of exceedances of PM<sub>10</sub> daily limit values in Europe by 14 to 22 % in 2020 compared to 2009 and by 19 to 28 % for the exceedances of the  $PM_{25}$  annual limit value, pointing at a need for further measures to comply with the EU limit values. The same study shows also that PM concentrations and the number of exceedances can be considerably reduced if NH<sub>3</sub> emissions from agriculture would be reduced beyond the emission targets for 2020 set in the revised Gothenburg Protocol. As an example, a further reduction (above and beyond the reduction planned in the revised Gothenburg Protocol) of 30 % in NH<sub>2</sub> agriculture emissions in the EU would result in a further reduction of between 5 % and 9 % in the number of stations in exceedance of the PM<sub>10</sub> daily limit value. Such a further reduction in NH<sub>3</sub> would also result in a reduction of between 3 % and 10 %

in the number of stations in exceedance of the  $PM_{2.5}$ limit value of 20 µg/m<sup>3</sup> (which the relevant European directive declares should be met by 1 January 2020). Finally, this further reduction would also reduce the annual mean  $PM_{2.5}$  concentrations by up to 11 % in central and western Europe, compared to the Gothenburg Protocol scenario for 2020.

#### 2.4 Exposure to PM pollution in Europe

The  $PM_{10}$  monitoring data in AirBase provide the basis for estimating the exposure of the urban-dwelling European population to exceedances of the  $PM_{10}$  daily limit value (50 µg/m<sup>3</sup> not to be exceeded on more than 35 days a calendar year). This estimate is shown in Figure 2.8 for the period 2002–2011. The exposure is estimated based upon  $PM_{10}$  measured at all urban background (non-traffic) monitoring stations. For each city an average concentration is calculated. It is considered that the entire population in cities is potentially exposed to these concentrations, since people move freely within the city. In 2011 about 33 % of the urban population in the EU was exposed to  $PM_{10}$  above the daily limit value. The extent of exposure above the limit value has varied between 20 % and 44 % since 2002 and there is no apparent trend over this period. For EEA-32 countries the estimate of the urban population exposed to  $PM_{10}$  above the daily limit value is 49 % in 2011 and the variation was between 28 % and 49 % during the period 2002–2011. The range partly reflects variations caused by meteorology and changes in the subset of cities and stations included in the analysis.

For PM<sub>2.5'</sub> the 2008 Air Quality Directive (EU, 2008c) introduced a target value, to be attained by 2010, which will become a limit value starting in 2015 (Table 2.1). In 2011 about 15 % of the urban population in the EU and EEA-32 was exposed to PM<sub>2.5</sub> above the target value threshold. The percentage of the population exposed to annual levels above the target value has varied between 6 % and 15 % in the period 2009–2011. The same directive also established the national exposure reduction target for human exposure to PM<sub>2.5</sub> based on the average exposure indicator (AEI) set at the





Source: EEA, 2013e (CSI 004).





 $PM_{2.5}$  (three-year running mean,  $\mu g/m^3$ ) at (sub)urban stations

**Note:** The three-year running mean of PM<sub>2.5</sub> concentrations (2009–2011) is calculated as the average over all operational (sub) urban background stations within a Member State in the period 2009–2011. The orange dots correspond to AEI-values as provided by the EU Member States in the air quality questionnaire (reporting year 2011; reference period 2009–2011 except Poland: reference period 2010–2011).

national level. The AEI is an averaged level of concentrations (in space — per country and time), measured at urban background monitoring stations (representative of general population exposure) over a three-year period. Figure 2.9 indicates that in at least eight Member States the average urban concentrations in the period 2009–2011 were above  $20 \ \mu g/m^3$ . This is the legally binding level for this exposure concentration obligation to be met in the EU by 2015. The presented levels are not based on a stable set of stations. For a number of countries results are based on data for less than three years.

Table ES.1 shows the percentage of the EU urban population exposed to concentrations above the EU limit values and the WHO AQG levels between 2009 and 2011. Between 22 % and 33 % of the urban population were exposed to PM<sub>10</sub> concentrations exceeding the EU daily limit value in this time, while up to 88 % (in 2011) of the same urban population was exposed to concentrations exceeding the stricter WHO AQG value for PM<sub>10</sub>. Here too the range in the percentage of the population affected partly reflects variations caused by meteorology.

#### 2.5 Impacts on health

The recent Global Burden of Disease study indicates that worldwide, 3.1 million deaths and almost 3 % of all DALYs (Disability Adjusted Life Years) could be attributed to exposure to ambient  $PM_{2.5}$ . In western, central and eastern Europe it was 430 000 premature deaths, and over 7 million years of DALYs (Lim et al., 2012).

Source: ETC/ACM.

### 2.6 Responses

Because PM is made up of both primary and secondary PM, European efforts to reduce rural and urban concentrations must address emissions of both primary PM and also precursor gases. Key anthropogenic sources of these compounds are road vehicles, industrial installations and agriculture. The following section contains a discussion of the European-level policy responses to emissions from each of these sources in turn. Annex 2 contains more information on each of the policy instruments discussed below.

#### 2.6.1 Road transport sector

For the road transport sector, the 'Euro' standards regulate exhaust emissions of CO,  $NO_{x'}$  NMVOCs and primary PM.  $NO_x$  and PM emissions are directly relevant for PM concentrations in the air.

The 'Euro' emission limits for type approval of new cars are as follows:

- for total PM mass exhaust emissions, the Euro 4 emission limits (in force since 2005) are 75 % lower than the 1996 Euro 2 emission limits for light-duty diesel (passenger) vehicles, while for heavy-duty diesel vehicles the Euro IV 2005 PM emission limit is 92 % lower than Euro II (from 1996);
- for NO<sub>x</sub>, the 2005 Euro 4 emission limits are 50 % lower than the 1996 Euro 3 emission limits for diesel passenger cars and 58 % lower for heavy-duty (i.e. non-passenger) diesel vehicles;
- Euro 5 (from 2009) requires a further substantial drop in emission limits, in particular for total PM mass exhaust emissions (80 % reduction compared to Euro 4 for diesel cars).

These reductions in permissible emission limits have resulted in substantial reported declines in  $NO_x$  and PM emissions from vehicles over the last decade despite the large increase in the number of vehicles and total traffic activity over the same period. The decrease in transport emissions in the period 2002–2011 was 31 % for  $NO_x$ , 24 % for  $PM_{10}$  and 27 % for  $PM_{2.5}$  in the EU. Under real-world driving conditions, emissions from vehicles often exceed the test-cycle limits specified in the Euro emission standards (EEA, 2011a). Reported emissions data from countries are based on the best knowledge available concerning real-world emissions. However, the lack of vehicle exhaust measurements, especially for newer vehicle technology types, often means the reported emission estimates are of relatively high uncertainty, and may not always reflect the actual, potentially higher, on-road driving emissions.

Non-exhaust vehicle emissions, such as tyre and brake wear, and road abrasion, are currently not regulated.

New policy measures to deal with transport emissions are still being formulated. These include the recent Cars 2020 Communication setting out a timetable for the successful implementation of the Euro 6 vehicle standards in real-world driving conditions, and the assessment of the Non-road Mobile Machinery legislation, carried out in 2013.

#### 2.6.2 Large combustion plants

European directives related to industrial emissions the Large Combustion Plant (LCP) Directive (EU, 2001a) and the IPPC Directive (EU, 2008b) have resulted in a substantial reduction in emissions from large combustion and industrial plants. Both directives, together with several other sectoral directives, have been replaced by the Industrial Emissions Directive (EU, 2010) but will remain in force for several years. In addition and even prior to these directives, the National Emission Ceilings Directive (EU, 2001b) and the Gothenburg Protocol (UNECE, 1999) lead to the implementation of abatement technologies and a shift towards less polluting fuels in large combustion plants. Although the effects of the directives and international protocols on PM emissions have not been fully assessed, the EEA (2010b) estimated that they delivered reductions in NO<sub>x</sub> and SO<sub>x</sub> emissions (PM precursor gases) of about 50 % and 75 % respectively in the period 1990-2005.

#### 2.6.3 NEC Directive on total emissions

The NEC Directive (EU, 2001b) includes limits on total national emissions of the acidifying or eutrophying gases  $SO_{2'} NO_x$  and  $NH_{3'}$  all of which are also PM precursors. The ceilings were to be met by 2010, and the 2011 emission data provided by the EU Member States estimate that  $NO_x$  emissions in that year exceeded the ceiling by 5 % while  $SO_x$ emissions were 44 % beneath the ceiling (Annex II emission ceilings in the NEC Directive) and  $NH_3$ emissions were 15 % below their ceiling (see Annex I of the NEC Directive) (EEA, 2013b). The NEC Directive is currently under revision. The revised version will establish emission ceilings to meet, as a minimum, the obligations set by the newly revised UNECE Gothenburg Protocol, which established new international air pollution emission ceilings to be met by 2020 for those pollutants already listed in the NEC Directive ( $NO_x$ , VOC, SO<sub>2</sub> and  $NH_3$ ), as well as for primary fine particulate matter ( $PM_{25}$ ).

### 2.6.4 Air quality plans

The air quality directives in force require that air quality plans are developed as an additional policy instrument and implemented in air quality management zones and agglomerations where ambient concentrations of pollutants exceed the relevant air quality limit or target values. To ensure coherence between different policies, the air quality plans should be consistent (where feasible) and integrated with plans and programmes pursuant to the directives regulating air pollutant emissions. The air quality plans may additionally include specific measures aiming to protect sensitive population groups, e.g. children.

A recent pilot project, which aimed at improving the knowledge on implementation of air quality legislation, has carried out a review of the main measures adopted to manage PM concentration levels by 12 participating cities. It found that most of the measures targeted traffic emissions, e.g. the creation of Low Emission Zones; improvement of public transport; promotion of cycling; management of traffic flow; and change in speed limits. The commercial and residential combustion sector was also targeted by measures, as it was identified as the second most important contributor to PM<sub>10</sub> exceedances in almost every city. Some of the measures that were considered successful by the cities include: ensuring compliance with new low-sulphur standards for shipping fuels in the port areas; a ban on the marketing, sale, and distribution of bituminous coal; fuel conversion in domestic heating and the creation of district heating (EEA, 2013g).

# 3 Ozone $(O_3)$

#### 3.1 Sources and effects of O<sub>3</sub>

#### 3.1.1 Origins of $O_3$ in the air

Unlike primary air pollutants, which are emitted directly into the air, ground-level (tropospheric)  $O_3$  is not directly emitted into the atmosphere. Instead, it is formed from complex chemical reactions following emissions of precursor gases such as nitrogen oxides (a family of gases also known as  $NO_x$  that includes NO and  $NO_2$ ) and non-methane VOCs (NMVOCs). At the continental scale, methane (CH<sub>4</sub>) and carbon monoxide (CO) also play a role in  $O_3$  formation.

#### 3.1.2 Effects of $O_3$

Ground-level ozone is a powerful and aggressive oxidising agent, which can have a marked effect on human health. Excessive  $O_3$  in the air can lead to respiratory health problems, such as breathing problems, asthma, reduced lung function, and

other lung diseases. Short-term studies show that current  $O_3$  concentrations in Europe — especially in the summer — have adverse health effects on pulmonary function, causing lung inflammation, respiratory symptoms, increased usage of respiratory medication, morbidity, and mortality. Several European studies have reported that daily mortality rises in line with increases in  $O_3$  exposure (WHO, 2008).

There is also new evidence linking long-term exposure to ozone with deterioration in reproductive health and greater mortality effects than previously thought (WHO, 2013).

Ozone in the ambient air can also react with other components occurring indoors to produce short-lived pollutants that are highly irritating (and even more irritating than their precursors), and may also have long-term health effects (Chen et al., 2012). Known products of indoor ozone reactions include compounds such as formaldehyde, acetaldehyde, and other organic acids.

#### Box 3.1 Ozone: a photochemically formed pollutant

Ozone is not emitted directly into the air. Virtually all of it is formed by chemical reactions involving primarily NO,  $NO_2$  and VOC.

NO and NO<sub>2</sub> are emitted during fuel combustion, for example by industrial facilities and road transport.

Volatile organic compounds are emitted from a large number of sources including paint, road transport, refineries, dry-cleaning of fabrics, and other activities that involve the use of solvents. Volatile organic compounds are also emitted by vegetation, with the amount of VOCs coming from vegetation dependent on temperature. Methane ( $CH_4$ ), also a VOC, is released from coal mining, natural gas extraction and distribution, landfills, wastewater, ruminants (grass-eating animals), rice cultivation and biomass burning.

The chemistry of  $O_3$  formation and its decay are complex and are driven by energy from the sun. Therefore,  $O_3$  is labelled as photochemical pollutant. The main features of this can be summarised as follows:  $NO_2$  can efficiently absorb sunlight and dissociate, producing atomic oxygen (O) and NO. The atomic oxygen in turn reacts rapidly with molecular oxygen ( $O_2$ ) to form  $O_3$  (provided a third molecule such as molecular oxygen or nitrogen absorbs the excess energy released in this reaction). On the other hand, NO, typically emitted by combustion processes, reacts rapidly in the air with  $O_3$  to form  $NO_2$ and  $O_2$  and therefore contributing to the decay of  $O_3$  concentrations. The latter is known as the titration reaction.

The chemical mechanism outlined above describes the equilibrium state in the atmosphere, in the absence of other gaseous substances, a situation in which the amount of  $O_3$  would be controlled by the relative amounts of  $NO_2$  and NO as well as the intensity of sunlight. But this equilibrium is being disturbed by other pollutants, which are increasing the overall level of ozone. This is because polluted air also contains VOCs. Nitrogen oxides and VOCs take part in hundreds of chemical reactions. VOCs are also degraded to produce substances that react with NO to produce  $NO_2$  without consuming  $O_3$ , thus disturbing the equilibrium of the titration reaction. The net result of these reactions is that more than one  $O_3$  molecule is formed for each VOC molecule degraded.

Fire plumes from wild forest and other biomass fires contain CO and can contribute to  $O_3$  formation. There is also a global background concentration of  $O_3$  in air, partly resulting from photochemical  $O_3$  formation ( $O_3$  created by sunlight) globally and partly from the downward transport of stratospheric  $O_3$  ( $O_3$  existing many miles up in the atmosphere) to the troposphere (air that is closer to ground level).

High levels of  $O_3$  can also damage plants, impairing reproduction and growth, leading to reduced agricultural crop yields, decreased forest growth, and reduced biodiversity. Ozone decreases photosynthesis, thereby reducing plant uptake of carbon dioxide (EEA, 2010a). Ozone also increases the rate of degradation of buildings.

Relatively few studies have investigated the impacts of both ozone and nitrogen on vegetation. Evidence suggests that ozone and nitrogen can have both synergistic and antagonistic effects on species health and ecosystem processes, and that they may interact in unpredictable ways to affect plant communities (Harmens and Mills, 2012).

In addition to its effects on human health, plants and crops,  $O_3$  is a greenhouse gas that contributes to the warming of the atmosphere. Ozone is currently considered to be the third-most important greenhouse gas, after  $CO_2$  and  $CH_4$ . In contrast to  $CO_2$ ,  $O_3$  is a short-lived greenhouse gas, meaning that any reductions in ground-level ozone production will reduce atmospheric ozone concentrations within months and hence reduce its contribution to global warming relatively quickly.

# 3.2 European air quality standards for O<sub>3</sub>

European air quality objectives for  $O_3$  are shown in Table 3.1. The 2008 Air Quality Directive (EU, 2008c) sets out values for the protection of human health and for the protection of vegetation.

For health protection, a daily maximum eight-hour average threshold is specified ( $120 \ \mu g/m^3$ ). The target value, to be applied by Member States from 1 January 2010, is that the threshold should not

be exceeded at a monitoring station on more than 25 days per year, determined as a three-year average starting from 2010. The long-term objective (LTO) is that the threshold level should not be exceeded at all. For health protection, there are also two other types of thresholds, i.e. 'public information' threshold and 'alert' threshold. When the public information threshold is breached, the authorities in that country are expected to notify their citizens, using a public information notice on the website of the country's relevant air quality authority. When the alert threshold is exceeded, the country affected is requested to draw up a short-term action plan according to specific provisions defined in the 2008 Air Quality Directive.

The updated WHO air quality guideline for  $O_3$  is an 8-hour mean concentration of 100 µg/m<sup>3</sup> (WHO, 2006a). The WHO (2008) explains the rationale for the guideline as follows:

The previously recommended limit, which was fixed at 120  $\mu$ g/m<sup>3</sup> 8-hour mean, has been reduced to 100  $\mu$ g/m<sup>3</sup> based on recent conclusive associations between daily mortality and O<sub>3</sub> levels occurring at O<sub>3</sub> concentrations below 120  $\mu$ g/m<sup>3</sup>.'

The EU has the objective of protecting vegetation from high  $O_3$  concentrations accumulated over the growing season (defined as the summer months May to July). The vegetation protection value is specified as 'accumulated exposure over threshold', AOT40. This is calculated as the sum of all hourly  $O_3$ values over 40 micrograms per cubic metre ( $\mu$ g/m<sup>3</sup>) during the daylight period of the most intense growing season (May to July). The target value for 2010 is 18 000 ( $\mu$ g/m<sup>3</sup>).hour. The long-term objective is 6 000 ( $\mu$ g/m<sup>3</sup>).hour, as shown in Table 3.1.

#### Table 3.1 Air quality standards for ozone as defined in the Air Quality Directive

Objective	Period	Target or threshold value	Number of allowed exceedances
Human health	Maximum daily 8-hour mean	120 µg/m³ (ʰ)	25 days per year averaged over three years
Vegetation	AOT40 accumulated over May–July	18 000 (µg/m³).h averaged over five years	
LTO health	Maximum daily 8-hour mean	120 µg/m <sup>3</sup>	
LTO vegetation	AOT40 accumulated over May–July	6 000 (µg/m³).h	
Information	One hour	180 µg/m³	
Alert (ª)	One hour	240 µg/m <sup>3</sup>	

**Note:** (<sup>a</sup>) To be measured over three consecutive hours.

(b) Target value to be met by 1 January 2010.

Source: EU, 2008c.

In addition to the EU target value, the UNECE Convention on Long-range Transboundary Air Pollution (UNECE, 1979) defines a 'critical' level for the protection of forest. This critical level is a function of the accumulated exposure over threshold AOT40 during the full summer (April–September) and is set to 10 000 ( $\mu$ g/m<sup>3</sup>).hour.

#### 3.3 Europe-wide survey of O<sub>3</sub>

# 3.3.1 Exceedance of the target values for protection of health

The target value threshold for  $O_3$  of 120 µg/m<sup>3</sup> (daily maximum of running 8-hour mean values) was exceeded on more than 25 days per year at a large number of stations across Europe in 2011 (the red and dark red dots in Map 3.1).

Since the formation of  $O_3$  requires sunlight,  $O_3$  concentrations show a clear increase as one moves from the northern parts to the southern parts of the continent, with the highest concentrations in some Mediterranean countries. The concentration of  $O_3$  typically increases with altitude in the first kilometres of the troposphere. Close to the ground,  $O_3$  is depleted due to surface deposition and the titration reaction (see Box 3.1). Higher concentrations of ozone can therefore be observed at high altitude stations.

#### 3.3.2 Ozone in rural, urban and traffic locations

In contrast to other pollutants,  $O_3$  levels are generally highest at rural locations. This is because at short distances from  $NO_x$  sources, as is the case at urban and traffic stations,  $O_3$  is depleted through

# Map 3.1 26th-highest daily maximum 8-hour average O<sub>3</sub> concentration recorded at each monitoring station in 2011



**Note:** The map shows the proximity of recorded  $O_3$  concentrations to the target value. At sites marked with red and dark red dots, the 26th-highest daily  $O_3$  concentration exceeded the 120  $\mu$ g/m<sup>3</sup> threshold, implying an exceedance of the threshold and the number of allowed exceedances by the target value.

**Source:** AirBase v. 7.

the titration reaction by the freshly emitted NO (see Box 3.1). Figure 3.1 (top) shows this gradient from higher concentrations at rural sites towards lower concentrations at urban sites and even lower concentrations at traffic locations. The high O<sub>2</sub>

concentrations occurring at a few urban stations shown in Map 3.1 are due to the O<sub>3</sub> formation that occurs at times in large urban areas during episodes of high solar radiation and temperatures.

Distribution of AOT40 concentrations for the protection of forests

#### Figure 3.1 Concentration status for the target value threshold for protection of human health (top) and objectives for protection of crops (bottom left) and forests (bottom right), in 2011



Distribution of AOT40 concentrations for the protection of vegetation (acccumulated over May to July)



0 to 80 (µg/m<sup>3</sup>)

corresponding to the 26th highest as defined by the target value, and for the various types of stations (top); AOT40 concentrations (in (µg/m<sup>3</sup>).h) measured in rural stations in the EU for the protection of vegetation (accumulated over May to July) (bottom left); and AOT40 concentrations (in (µg/m<sup>3</sup>).h) measured in rural stations in the EU for the protection of forests (accumulated over April to September) (bottom right). The CLRTAP critical level for the protection of forests (Mills et al., 2011) is set at 10 000 ( $\mu$ g/m<sup>3</sup>).h for AOT40 (the accumulated dose of O<sub>3</sub> over a threshold of 40 ppb, equivalent to 80  $\mu$ g/m<sup>3</sup>, from 1 April to 30 September).

Source: AirBase v. 7.

Differences in the distribution and magnitude of  $O_3$  precursor emission sources, the chemical composition of the air and climatic conditions along the north-south and east-west directions in Europe result in considerable regional differences in summer  $O_3$  concentrations. Year-to-year differences in the  $O_3$  levels are also induced by meteorological variations. Hot, dry summers with long-lasting periods of high air pressure over large parts of Europe lead to elevated  $O_3$  concentrations.

#### 3.3.3 Distance to target

The health-related threshold of the  $O_3$  target value (applicable from 2010) was exceeded more than 25 times in 2011 at 40 % of the rural stations, 24 % of urban background stations, 21 % of industrial sites (labelled as 'other' in Figure 3.1 (top)), and 9 % of traffic sites.

Figure 3.1 (bottom left) shows that the threshold used for the target value (applicable from 2010) set for protection of vegetation was exceeded to a substantial degree (31 % of the rural stations) in 2011. The highest measured values (in Italy) exceeded 55 000  $\mu$ g/m<sup>3</sup>.h, which is more than three times the target threshold.

Figure 3.1 (bottom right) shows that the UNECE – CLRTAP (<sup>15</sup>) critical level of 10 000 ( $\mu$ g/m<sup>3</sup>).h set for protection of forests (UNECE, 2011) was exceeded at 89 % of the rural stations in 2011. For this forest-protection objective, there is a substantially higher number of exceedances, than for the protection of vegetation.

Conformity with the WHO AQG value for  $O_3$  (8-hour mean of 100 µg/m<sup>3</sup>) set for the protection of human health was observed only at three out of 513 rural background stations in 2011. Only 2 % and 3 % of (sub)urban background and traffic stations, respectively, measured concentrations that did not exceed the WHO AQG in 2011.

Although the EU target value ( $120 \ \mu g/m^3$ , 25 exceedances allowed) is less ambitious than the WHO AQG, non-attainment situations (i.e. not having achieved the less ambitious EU air quality standard) are widely found in most of the EU Member States as shown in Map 3.1 and Figure 3.2.

#### 3.3.4 Trends in O<sub>3</sub> concentrations

Changes in O<sub>3</sub> concentrations can be different for the different station types and for different indicators



### Figure 3.2 Attainment situation for O<sub>3</sub> in 2011

Note: The graph is based on the 93.15 percentile of maximum daily 8-hour mean concentration values corresponding to the 26th highest daily maximum of the running 8-h mean for each Member State; the boxes present the range of concentrations at all stations types (in μg/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the target value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

<sup>(15)</sup> UNECE-CLRTAP: United Nations Economic Commission for Europe — Convention on Long-range Transboundary Air Pollution.

(Mol et al., 2011). In the past ten years, there has been a decline in the number of episodic high  $O_3$ concentrations (also called 'peak concentrations') (EEA, 2010a). However, often the data does not paint a uniform and steady trend. For example, lately, in the summer of 2012 the information threshold (a one-hour average  $O_3$  concentration of 180 µg/m<sup>3</sup>) was exceeded at approximately 28 % of all operational stations. This was a much higher percentage than in 2011, when 18 % of operational stations registered these exceedances, but it was still among the lowest percentages since 1997. The long-term objective (LTO) for the protection of human health was exceeded in all EU Member States except Estonia during summer (April–September) 2012. The average number of exceedances in 2012 was comparable with 2009–2011 years (EEA, 2013c).

Map 3.2 shows the average changes in the maximum daily 8-hour  $O_3$  values for stations that registered a trend over the period 2002 to 2011 (<sup>16</sup>). At 49 % of the stations registering a trend, a slight negative trend (of less than 2 µg/m<sup>3</sup> per year) is apparent, while 42 % of the stations had a more pronounced negative trend (equal to or above 2 µg/m<sup>3</sup> per year) (<sup>17</sup>). 9 % of the stations registered a positive trend from 2002 to 2011 (<sup>18</sup>). Most of the stations having a large positive trend (19 in total) are located in the Iberian Peninsula (<sup>19</sup>), where 14 stations registered a large positive trend. Six out of those 19 stations are

## Map 3.2 Annual changes in annual mean of the maximum daily 8-hour O<sub>3</sub> concentrations in the period 2002–2011



Note: The data presented were derived from a consistent set of stations in all years.

Statistically significant trends (level of significance 0.1) are calculated by applying the Mann-Kendall test. Increasing concentrations are indicated with red dots and decreasing concentrations with green dots, when statistically significant. The applied method is described in de Leeuw, 2012.

<sup>(&</sup>lt;sup>16</sup>) A consistent set of 1 325 stations with data for 2002 to 2011 was used in the trend analysis. Of these, only 266 stations registered a trend (a statistically significant trend, using the Mann-Kendall test). The remaining 1 059 stations had no significant trend.

 $<sup>\</sup>binom{17}{42}$  42 % is equivalent to 111 stations.

<sup>(&</sup>lt;sup>18</sup>) 9 % is equivalent to 24 stations.

<sup>(19)</sup> Nineteen stations registered a trend with an increase of 2 µg/m<sup>3</sup> per year or more, 14 of which located in the Iberian Peninsula.

classified as being traffic stations, six are urban, five are industrial and two are rural background stations. Consequently, the Iberian Peninsula registered both increasing and decreasing trends over different types of stations and regions.

Table A1.4 (Annex 1) shows the average trends by country and by station type for the maximum daily 8-hour mean  $O_3$  concentrations over the period 2002–2011. Increasing trends were registered at traffic stations in several countries: Bulgaria, Denmark, Lithuania, Portugal, Spain, and the United Kingdom. The increasing  $O_3$  levels at traffic locations are mainly attributable to a reduced depletion of

 $O_3$  by NO as a result of the decrease in traffic NO<sub>x</sub> emissions (de Leeuw, 2012). Bulgaria and Hungary also registered an increase in the maximum daily 8-hour mean  $O_3$  concentrations in urban background stations. The trends registered at rural stations were on average decreasing in all Member States.

Figure 3.3 shows the trends of the maximum daily 8-hour mean  $O_3$  concentrations at different station types over the period 2002–2011 (<sup>20</sup>). This indicator is directly related to the target value for  $O_3$ , as 25 days per year are allowed to have exceedances of the target value threshold of 120 µg/m<sup>3</sup>. Figure 3.3 does not show a clear trend at the aggregated EU level.

## Figure 3.3 Trends in the maximum daily 8-hour $O_3$ concentrations (in $\mu$ g/m<sup>3</sup>) (2002–2011) per station type



**Note:** The graph is based on the 93.15 percentile of the maximum daily 8-hour  $O_3$  concentration trends; it presents the range of concentration changes per year (in  $\mu g/m^3$ ) per station type. The trends are calculated based on the officially reported data by the EU Member States with a minimum data coverage of 75 % of valid data per year for at least 8 years out of the 10 years period.

The diagram indicates the lowest and highest trends, the means and the lower and upper quartiles, per station type. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

 $<sup>(^{\</sup>rm 20})$  Including both significant and non-significant trends.

### Emissions of O<sub>3</sub> precursors

The EU emissions of the air pollutants primarily responsible for the formation of harmful ground level  $O_3$  fell significantly in the period 2002–2011. Carbon monoxide emissions were cut by 32 % (Figure 3.4), NMVOCs by 28 %, NO<sub>x</sub> by 27 %, and CH<sub>4</sub> by 15 % (<sup>21</sup>). Nevertheless, in 2011 NO<sub>x</sub> emissions remained 5 % above the NEC Directive ceiling (Annex II) to be attained by 2010, mainly due to road transport emissions (EEA, 2013b).

The transport and the energy sectors are the main sectors responsible for emissions of  $NO_{x'}$ , followed by industry (Figure 3.5). The transport sector reduced its  $NO_{x}$  emissions by 31 % between 2002

and 2011, and the energy and industry sectors reduced their NO<sub>x</sub> emissions in the same period by 27 % and 26 %, respectively. The agriculture sector was the only sector to register an increase (of 5 %) in NO<sub>v</sub> emissions between 2002 and 2011. As Figure 3.5 shows, several sectors have cut their NMVOC emissions in the last decade, with the exception of the agriculture sector, which increased its emissions by 91 %. The transport sector, which was the second largest emitter in 2002, secured the largest reduction with a 60 % cut in the period 2002–2011. The 'solvent and product use' sector has been the largest source of NMVOC emissions since 1999; it has reduced its emissions by 16 % from 2002 to 2011, the same reduction registered by the industry sector. Non-methane volatile organic compound emissions



**Note:**  $CH_4$  emissions are total emissions (IPPC sectors 1–7) excluding sector 5, LULUCF (land use, land use change and forestry). **Source:** EFA.

 $NO_x$ , CO and 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-Irtap).

 $CH_4$  emissions were downloaded from EEA greenhouse gas data viewer (http://www.eea.europa.eu/data-and-maps/data/data-viewers/greenhouse-gases-viewer).

<sup>(&</sup>lt;sup>21</sup>) EEA-32 countries registered emission reductions as follows between 2002 and 2011: 27 % for CO, 27 % for NMVOC, 23 % for NO<sub>x</sub>.

energy sectors (Figure 3.5). While the waste and

cut its CH<sub>4</sub> emissions by 6 %.

energy sectors have cut their emissions in 2002-2011

by 22 % and 23 %, respectively, agriculture has only

from the waste sector were reduced by 17 % and emissions from the energy sector fell by 26 % in the period from 2002–2011.

Agriculture is the main sector responsible for  $CH_4$  emissions in Europe, followed by the waste and

Figure 3.5	Contributions to EU emissions of O <sub>3</sub> precursors from main source sectors
	of NO <sub>x</sub> , NMVOC, CO and CH <sub>4</sub> , 2002–2011 (Gg/year = 1 000 tonnes/year)



**Note:** CH<sub>4</sub> emissions excluding sector 5, LULUCF (land use, land use change and forestry).

 $NO_x$ , CO and 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).

 $CH_4$  emissions were downloaded from EEA greenhouse gas data viewer (http://www.eea.europa.eu/data-and-maps/data/data-viewers/greenhouse-gases-viewer).

Source: EEA.

# Relationship of $O_3$ precursors to ambient $O_3$ concentrations

The relationship of O<sub>3</sub> concentration to the emitted precursors is not linear. There is a discrepancy between the cuts in O<sub>3</sub> precursor gas emissions and the change in observed O<sub>3</sub> concentrations in Europe (Figure 3.3). The reasons for this include increasing inter-continental transport of O<sub>3</sub> and its precursors in the northern hemisphere (EEA, 2010a; EEA, 2012b) and other factors that are likely to mask the effects of European measures to reduce O<sub>3</sub> precursor emissions. These other factors include climate change/variability, NMVOC emissions from vegetation the magnitude of which is difficult to quantify, and fire plumes from forest and other biomass fires (EEA, 2010a). Formation of tropospheric O<sub>3</sub> from increased concentrations of CH<sub>4</sub> may also contribute to the sustained O<sub>3</sub> levels in Europe. Methane concentrations increased continuously during the 20th century, before growth slowed after 1990. Then, between 1999 and 2007 CH<sub>4</sub> concentrations levelled off. Since 2007, however, measurements suggest that concentrations of CH have started to rise again (Dlugokencky et al., 2009). Methane is a slowly-reacting pollutant that is well mixed across the world. Isolated local and regional abatement of CH<sub>4</sub> emissions may therefore have limited impact on local O<sub>3</sub> concentrations.

Clearly,  $O_3$  concentrations are not only determined by precursor emissions but also by meteorological conditions. Sunlight and high temperatures favour  $O_3$  formation. Episodes of elevated  $O_3$  levels occur during periods of warm, sunny weather. However, independent of the episodic nature of  $O_3$  pollution that is strongly influenced by meteorological conditions, emissions of  $O_3$  precursor gases are sustaining a baseline of exceedances of legal concentration thresholds. The  $O_3$  pollution problem requires further mitigation efforts.

### 3.4 Exposure to O<sub>3</sub> pollution in Europe

#### 3.4.1 Human exposure

The  $O_3$  monitoring data in AirBase provide the basis for estimating the urban exposure of the European population to exceedances of the EU's  $O_3$  target value (applicable from 2010) for the protection of human health. This estimation is shown in Figure 3.6 for the period 2002–2011. The exposure is estimated based on  $O_3$  measured at all urban background monitoring stations. For each city an average concentration is calculated. It is considered that the entire population of a city is exposed to this average concentration.

# Figure 3.6 Percentage of the EU urban population exposed to ozone concentrations over the target value threshold set for protection of human health, 2002–2011



**Source:** EEA, 2013e (CSI 004).

It is noteworthy that people in rural areas are exposed to higher  $O_3$  levels than people in cities. In urban areas with fresh inputs of NO from traffic emissions, some of the  $O_3$  is depleted while oxidising NO to NO<sub>2</sub>. In 2011 about 14 % of the EU population in urban areas was exposed to  $O_3$  concentrations above the EU target value. The percentage of the urban population exposed to ozone levels above the target value has varied between 14 % and 65 % since 2002. The same exposure levels were estimated for the urban population of the EEA-32.There is no apparent trend over this period. The range partly reflects variations caused by meteorology.

The EU urban population exposed to  $O_3$  levels exceeding the WHO AQG value (which is stricter than the EU's target value) is significantly higher, representing about 98 % of the total urban population in 2011 (Table ES.1).

### 3.4.2 Exposure of ecosystems

The target value for protecting vegetation from high  $O_3$  concentrations is 18 000 µg/m<sup>3</sup>.hour (accumulated exposure above 40 µg/m<sup>3</sup> for the summer months of May to July, averaged over five years. The long-term objective is no more than 6 000 µg/m<sup>3</sup>.hour of accumulated exposure to AOT40.

Since 2002, the target value threshold for protection of vegetation has been exceeded in a substantial part of the agricultural area in the EU countries. For example, in 2010 (the latest year available for this particular assessment) the threshold was exceeded in about 21 % of this area (Figure 3.7 and Map 3.3). Exceedances of the target values have notably been observed in southern and central Europe (Map 3.3). The long-term objective for vegetation protection was met in 15 % of the total agricultural area in 2010, mainly in the United Kingdom, Ireland and the Nordic countries. When Map 3.3 (displaying the ozone exposure of vegetation in 2010) is compared to its counterparts from 2009, 2008 and 2007, an increase in the extent of areas with the highest AOT40 levels (red and dark red) can be seen specifically in the south-western regions of Europe (Horálek et al., 2012).

The variations between years (Figure 3.7) are influenced by meteorological factors. Summers in 2003 and 2006 had favourable meteorological conditions for  $O_3$  formation resulting in

exceptionally high concentrations. In fact, average  $O_3$  concentrations in 2006 were only slightly higher than in 2005 but June and July 2006 were characterised by a large number of  $O_3$  episodes resulting in a much higher AOT40 value compared to 2005 (EEA, 2011b).

Regarding the UNECE–CLRTAP critical level for the protection of forest (10 000 ( $\mu$ g/m<sup>3</sup>).h), Figure 3.7 shows that this critical level was exceeded in 61 % of the total forest area in the EEA-32 member countries in 2010 and in 98 % of the EEA-32's forest area in 2004. Map 3.4 shows clearly that the areas in 2010 that met these protection criteria were in the northern part of Europe, while the highest exceedances occurred in southern France and northern Italy.

### 3.5 Impacts

### 3.5.1 Impacts on health

Once exposed to ozone, our bodies try to prevent it from entering our lungs. This reflex reduces the amount of oxygen we inhale. Inhaling less oxygen makes our hearts work harder. So for people already suffering from cardiovascular diseases or respiratory diseases like asthma, high-ozone episodes can be debilitating and even fatal. Section 1.3 describes the impact on human health from both short-term and long-term exposure (EEA, 2013f).

### 3.5.2 Impacts on ecosystems and climate

Sitch et al. (2007) have shown that the indirect impacts of ozone on global warming potential via ozone's negative impacts on vegetation are of similar magnitude to ozone's direct impacts on global warming as a greenhouse gas.

Trees are an important carbon sink and many studies have shown that ozone reduces tree growth. Harmens and Mills (2012) estimated that between 1990 and 2000 the reduction in carbon stored in vegetation that can be accounted for by ozone concentrations was 6.2 % globally and almost 4 % in Europe.

Harmens and Mills (2012) concluded that today's levels of ozone exposure in northern and central Europe have the potential to reduce the rate of increase in forest living biomass by roughly 10 %, as compared to pre-industrial ozone exposure levels.







**Note:** Top figure: In the Air Quality Directive (2008/50/EC) the target value for protection of vegetation is set to 18 000 ( $\mu$ g/m<sup>3</sup>).h while the long-term objective is set to 6 000 ( $\mu$ g/m<sup>3</sup>).h. Due to lack of detailed land cover data and/or rural O<sub>3</sub> data, Iceland and Norway were not included until 2006. Switzerland has not been included in the analysis for the period 2004–2007 due to the same reasons. Due to lack of data, Turkey is not included during the entire period (2002–2010).

Bottom figure: LRTAP/UNECE has set a critical level for protection of forest to 10 000 ( $\mu$ g/m<sup>3</sup>).h. Since 2004 a growing number of EEA member countries have been included. In 2004 Bulgaria, Greece, Iceland, Norway, Romania, Switzerland and Turkey have not been included. In 2005–2006 Iceland, Norway, Switzerland and Turkey are still excluded in the analyses due to lack of detailed land cover data and/or rural O<sub>3</sub> data. In 2007 data for Switzerland and Turkey are still missing. Since 2008 only Turkey is not included. Calculations of forest exposure are not available for years prior to 2004.

**Source:** EEA, 2013d (CSI 005).

< 6 000

6 000-12 000 12 000-18 000 18 000-27 000 > 27 000 Non-mapped countries

Poor data coverage

Rural background station



#### Exposure of European agricultural areas to ozone (AOT40) Map 3.3

Source: EEA, 2013d (CSI 005)/(Horalek et al., 2012).



Map 3.4 Exposure of European forest areas to ozone (AOT40)

Source: EEA, 2013d (CSI 005)/(Horalek et al., 2012).

Ozone-induced growth reductions also result in an economic loss for the forest owners. For example the annual economic loss for owners of Swedish forests has been estimated to be approximately EUR 40 m (Karlsson et al., 2005).

Mills and Harmens (2011) calculated that assuming soil moisture is not limiting to production, ozone impacts on wheat resulted in losses in production of 27 million tonnes of grain in 2000. The study showed that effects would be greatest in parts of central Europe (e.g. Germany, France and Poland), as well as in some Mediterranean countries (e.g. Italy, Spain).

#### 3.6 Responses

The transboundary nature of ozone pollution requires international as well as national efforts to effectively reduce emissions of nitrogen oxides and volatile organic compounds and hence ozone impacts. Such emission reductions would have co-benefits for climate change, ecosystems and human health. The goal of reducing ozone impacts on vegetation and health has been identified as a priority area in the long-term strategy of the UNECE LRTAP Convention, translated in the reviewed Gothenburg Protocol (UNECE, 2012).

Current policy measures to reduce  $O_3$  concentrations mainly target emissions of the precursors  $NO_x$  and NMVOCs.

The relevant  $NO_x$ -reducing measures are described in Section 2.6 of this report (since  $NO_x$  is also a precursor of PM). As noted in Section 2.3.4, the directives for road vehicle emissions and the LCP and IPPC Directives for industrial sources and power plants are estimated to have reduced  $NO_x$ emissions from road vehicles by 55 % and from power plants and large industrial plants by 68 %, compared to a hypothetical situation with no directives implemented (EEA, 2010b).

The Euro emission standards also limit NMVOCs emissions from road vehicles. Specifically, the introduction of the three-way catalyst (a type of catalytic converter) in road vehicles has led to considerable NMVOCs emission reductions.

Vapour emissions of NMVOCs at motor fuel service stations and petrol storage terminals are regulated and limited by the Vapour Recovery Directives (EU, 1994 and 2009a, discussed in Annex 2).

Directives limiting emissions of NMVOCs from the industrial sector include the Paints Directive (EU, 2004a), the Solvents Directive (EU, 1999a) and the IPPC Directive (EU, 2008b) - the latter two replaced by Directive 2010/75/EU on industrial emissions (EU, 2010). Each is described in Annex 2. The Solvents Directive limited emissions of VOCs from a number of activities and installations, including coating, dry cleaning, varnish and adhesives manufacturing, pharmaceutical manufacturing, printing, surface cleaning, vehicle refinishing and others. The Paints Directive regulated the maximum VOC contents in paints and varnishes and in vehicle-refinishing products. The Directive on Industrial Emissions (EU, 2010), which replaced the previous two directives, also adds new elements that were not present in either the Paints Directive or the Solvents Directive. For example, it regulates emission permits and requires the use of best available techniques (BAT) in production facilities and cleaning equipment.

The UNFCCC Kyoto Protocol addresses emissions of  $CH_{\mu}$  as one of the six main GHGs.

Implementing air quality plans can determine the extent of progress nationally and locally towards the air quality targets and long-term objectives for  $O_3$  (EU, 2008c).

# 4 Nitrogen dioxide (NO<sub>2</sub>)

#### 4.1 Sources and effects of NO<sub>x</sub>

#### 4.1.1 Origins of $NO_x$ in air

Nitrogen dioxide is a reactive gas that is mainly formed by oxidation of nitrogen monoxide (NO). High temperature combustion processes (e.g. those occurring in car engines and power plants) are the major sources of NO and NO<sub>2</sub>. These two gases are collectively known as NO<sub>x</sub>. Nitrogen monoxide accounts for the majority of NO<sub>x</sub> emissions. A small part of  $NO_x$  emissions is directly emitted as  $NO_{2'}$ typically 5–10 % for most combustion sources. Diesel vehicles are an exception, typically emitting a higher proportion of NO<sub> $\gamma$ </sub>, up to as much as 70 % of their  $NO_{y}$  is  $NO_{2}$  (e.g. Grice et al., 2009) because their exhaust aftertreatment systems increase the direct  $NO_2$  emissions (see Section 4.3). There are clear indications that for traffic emissions the direct NO, fraction is increasing significantly due to increased penetration of diesel vehicles, especially newer diesel vehicles (Euro 4 and 5). This may lead to more frequent breaching of the NO<sub>2</sub> limit values in traffic hotspots.

#### 4.1.2 Effects of $NO_x$

Like ozone, NO<sub>2</sub> is an air pollutant which primarily affects the respiratory system. Short-term exposure to NO<sub>2</sub> can result in adverse health effects such as changes in lung function in sensitive population groups, while long-term exposure can lead to more serious effects such as increased susceptibility to respiratory infection. Epidemiological studies have shown that symptoms of bronchitis in asthmatic children increase in association with long-term exposure to NO2. Reduced lung function is also linked to NO<sub>2</sub> at concentrations currently found in cities in Europe and North America (WHO, 2008). It should be noted that as NO<sub>2</sub> is highly correlated with other pollutants (in particular PM) it is difficult to differentiate the effects of NO<sub>2</sub> from those of other pollutants in epidemiological studies. However, some epidemiological studies do suggest an association that is independent of PM mass metrics between long-term NO<sub>2</sub> exposure and respiratory

and cardiovascular mortality. It also found an association between long-term NO<sub>2</sub> exposure and respiratory symptoms and reduced lung function in children (WHO, 2013).

 $NO_2$  is one of the reactive nitrogen compounds that can also have adverse effects on ecosystems, as they have acidifying effects but are also important nutrients. Excess deposition of reactive nitrogen can lead to a surplus of nutrient nitrogen in ecosystems, causing eutrophication (nutrient oversupply) in terrestrial and aquatic ecosystems. Excess nitrogen supply can lead to changes in unique terrestrial, aquatic or marine animal and plant communities, including biodiversity loss (EEA, 2010a).

Nitrogen oxides  $(NO_{\chi})$  play a major role in the formation of  $O_3$ . They also contribute to the formation of secondary inorganic aerosols (SIAs), through nitrate formation, contributing to  $PM_{10}$  and  $PM_{25}$  concentrations.

# 4.2 European air quality standards for NO<sub>2</sub> and NO<sub>x</sub>

European air quality standards for NO<sub>2</sub> and NO<sub>x</sub> as set by the 2008 Air Quality Directive (EU, 2008c) are shown in Table 4.1. For NO<sub>2</sub>, two limit values and an alert threshold are defined for the protection of human health. The limit values are specified using criteria of shortterm (one-hour) and long-term (annual) exposure, and EU Member States were obliged to meet them by 1 January 2010. The one-hour value can be exceeded up to 18 times per year before the limit value is breached. A critical level is set for the annual mean of NO<sub>x</sub> for the protection of vegetation, defined as the sum of NO and NO<sub>2</sub> expressed in units of mass concentration of NO<sub>2</sub>.

The 2008 Air Quality Directive (EU, 2008c) also defines an 'alert' threshold value of 400  $\mu$ g/m<sup>3</sup>. When this threshold is exceeded over three consecutive hours in areas of at least 100 km<sup>2</sup> or an entire air quality management zone, authorities have to implement short-term action plans. These

# Table 4.1Limit and threshold values for NO2 and NOx as set out in the 2008 Air QualityDirective

Objective	Averaging period	Limit or threshold value	Number of allowed exceedances
Human health	One hour	200 µg/m³	18 hours per year
Human health	Calendar year	40 µg/m³	
Alert ( <sup>a</sup> )	One hour	400 µg/m <sup>3</sup>	
Vegetation (b)	Calendar year	30 µg/m³	

**Note:** (<sup>a</sup>) To be measured over three consecutive hours at locations representative of air quality over at least 100 km<sup>2</sup> or an entire zone or agglomeration, whichever is smaller.

(<sup>b</sup>) As oxides of nitrogen (NO<sub>x</sub>), expressed as  $\mu$ g NO<sub>2</sub>/m<sup>3</sup>.

Source: EU, 2008c.

NO <sub>2</sub>	Table 4.2	WHO air quality guideline for NO <sub>2</sub>
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	1-hour mean µg/m³	Annual mean µg/m³
NO <sub>2</sub>	200	40

Source: WHO, 2006.

action plans may include measures in relation to motor-vehicle traffic, construction works, ships at berth, and the use of industrial plants or products and domestic heating. The framework of these plans may also consider specific actions aiming at the protection of sensitive population groups, including children, by reducing their exposure to high NO<sub>2</sub> levels.

The threshold values used in the human health objectives set by the 2008 Air Quality Directive are identical to the WHO air quality guidelines for  $NO_{2'}$  shown in Table 4.2 (WHO, 2006).

#### 4.3 Europe-wide survey of NO<sub>2</sub> and NO<sub>x</sub>

#### 4.3.1 Exceedances of limit values for NO,

The limit value of the annual mean  $NO_2$  concentration is set at 40 µg/m<sup>3</sup> and EU Member States were obliged to meet this by 2010. In 2011,

21 of them recorded exceedances of the limit value at one or more stations (red and dark red spots in Map 4.1; Figure 4.2). Figure 4.1 shows that the lowest concentration levels and fewest exceedances occur at rural stations, and that the highest concentrations and exceedances are at traffic stations. Guerreiro et al. (2010) provide a thorough discussion of NO<sub>2</sub> concentrations at hotspots close to traffic and also in the urban background.

#### 4.3.2 NO, in rural, urban and traffic locations

Nitrogen dioxide concentrations vary between rural, urban and traffic sites in a different manner from PM and O<sub>3</sub>. Nitrogen dioxide concentrations are higher close to the sources and at traffic stations, decreasing in quantity in urban background areas. The lowest concentrations are found in rural areas (Figure 4.1). While secondary PM and O<sub>3</sub> are formed regionally from precursor gases, chemical reactions are less likely to create NO<sub>2</sub> on this geographical scale. For most  $NO_x$  sources, the share of NO in  $NO_x$  emissions is much greater than that of  $NO_{2'}$ typically 10-20 times higher (22). Reactions between NO and O<sub>3</sub> then create more NO<sub>2</sub>, reducing the amount of NO. In traffic and urban areas with fresh inputs of NO, some of the O<sub>3</sub> present is depleted while oxidising NO to NO2. In rural areas, relatively limited fresh NO emissions are available, except near highways and near combustion plumes. The reaction between NO, NO<sub>2</sub> and O<sub>3</sub> leads to chemical equilibrium in the absence of VOCs (Box 3.1 on O<sub>3</sub> in Chapter 3).

<sup>(&</sup>lt;sup>22</sup>) An exception is emissions from motor vehicles produced after 1990 (i.e. complying with Euro standards). Due to the effect of catalytic converters on gasoline-powered vehicles and particle filters on diesel vehicles, the NO<sub>2</sub> fraction in emissions from these vehicles is much higher, making up between 20 % and 70 % of NO<sub>x</sub> depending upon the technology (e.g. Grice et al., 2009).



Map 4.1 Annual mean concentration of NO, in 2011

Note: Red and dark red dots correspond to exceedances of the annual limit value (40 μg/m<sup>3</sup>). Dark red dots correspond to exceedances of the annual limit value + 10 μg/m<sup>3</sup>.
Source: AirBase v. 7.

### 4.3.3 Distance to target

Although the annual limit value was exceeded in 2011 at only one rural background station, it was exceeded at 3 % (38) of all urban background stations. Exceedance of the limit value was reported at 42 % of traffic stations, with a maximum observed concentration of 103  $\mu$ g/m<sup>3</sup> in 2011, i.e. 2.5 times the threshold.

Figure 4.2 shows the attainment of annual mean NO<sub>2</sub> values for 2011 for all Member States. It clearly indicates that exceedance of the annual limit value and the WHO AQG value was observed in most Member States at one or more stations in 2011, with only Estonia, Lithuania, Ireland, Slovenia, Cyprus and Malta not registering any exceedance. The only countries, with complete NO<sub>2</sub> data for the years

2001, 2005, 2010 and 2011, which did not register an exceedance of the  $NO_2$  annual limit value in any of the four years were Estonia and Ireland.

The hourly limit value threshold for  $NO_2$  is less stringent. About 1 % of urban background stations and 5 % of traffic stations reported exceedances.

These findings demonstrate that NO<sub>2</sub> concentrations must be reduced substantially in large areas of Europe (focusing on traffic and urban locations) for the annual limit value to be met. Exceedances of the annual limit value are rather persistent: 8 % of the stations operational in the period 2007–2011 in the EU showed exceedances in each of the five years. These long-lasting exceedances are mostly observed at traffic stations.







Distribution of hourly NO<sub>2</sub> concentrations

**Note:** The graphs are based on the annual mean NO<sub>2</sub> concentrations (in  $\mu$ g/m<sup>3</sup>) (left) and the 99.78 percentile of the NO<sub>2</sub> hourly concentrations, corresponding to the 19th highest as defined by the limit value (right), for the various types of stations.

Source: AirBase v. 7.





# **Note:** The graph is based on the annual mean concentration values for each EU Member State; the boxes present the range of concentrations at all stations types (in µg/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the limit value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

The NO<sub>x</sub> annual limit value for the protection of vegetation ( $30 \ \mu g/m^3$ , expressed as  $\mu g \ NO_2/m^3$ ) has been exceeded at 26 rural background stations, mainly in Italy (16 stations), but also in Austria, Belgium, France, Germany and the Netherlands.

### 4.3.4 Trends in NO, concentrations

The trends in NO<sub>2</sub> concentrations over the period 2002–2011 are summarised in Figure 4.3 and Map 4.2. A consistent set of stations was used to compile these figures (<sup>23</sup>). Figure 4.3 shows very slowly decreasing NO<sub>2</sub> concentrations. Most of the stations (58 %) with data in the period 2002–2011 did register a significant trend. Of those that had a statistically significant trend (42 %), 95 % had a decreasing trend (Map 4.2).

Table A1.5 and Table A1.6 (Annex 1) show the calculated trends by country and by station type for NO<sub>2</sub> annual mean and NO<sub>2</sub> hourly concentrations, respectively, in the period 2002–2011. For each type of station, around 5 or 6 countries have registered increasing NO<sub>2</sub> annual mean concentrations. These trends are mostly statistically non-significant, but some stations registered statistically significant increasing trends in Portugal, Poland and Luxembourg, There are also increasing trends in several countries for NO<sub>2</sub> hourly mean concentrations, mostly at urban background and traffic stations. Only Poland and Estonia registered an increase in this indicator at rural stations (with both statistically significant and non-significant trends respectively). Ten out of 25 countries had increasing hourly concentrations at traffic stations. The most rapidly increasing average trend was registered in Norway, followed by Portugal.

### NO<sub>x</sub> emissions

As shown in Figure 3.4, EU emissions of  $NO_x$  fell by 27 % in the period 2002–2011 and by 3 % from 2010 to 2011. Nevertheless, total  $NO_x$  emissions in 2011 were about 5 % higher than the emissions ceiling for 2010 for the EU as a whole set in the NEC Directive (EU, 2001b).

Transport is the sector that emits the most  $NO_{x'}$  accounting for 47 % of the total in 2011, followed by the energy sector, which contributed 21 % of

total NO<sub>x</sub> emissions (Figure 3.5). These two sectors have substantially reduced their emissions since 2002. Over the period 2002–2011, emissions from transport decreased by 31 %, and emissions from industry fell by 26 %. The energy sector also saw steep declines with emissions of NO<sub>x</sub> falling by 27 %. The agriculture sector is the only sector that has increased its NO<sub>x</sub> emissions in the period, with an increase of 5 % since 2002.

Actual emissions from vehicles (often termed 'real world emissions') may exceed the allowed test cycle emissions specified in the Euro emission standards for each vehicle type. This is particularly the case for  $NO_x$  emissions from light-duty diesel vehicles. EU Member States regularly update the emission 'factors' (values used to estimate how much of a particular pollutant is present in emissions of a particular type) used in their emission inventories and their previously reported emissions. Reported developments in emissions should therefore include 'real world' emission factors.

# Relationship of $NO_x$ emissions and $NO_2$ concentrations

Nitrogen oxides emissions primarily comprise NO but also include some directly emitted NO<sub>2</sub>, which countries are not currently required to report as a separate compound under the relevant EU legislation (EU, 2001b). The concentrations of NO<sub>2</sub> found in air originate both from directly emitted NO<sub>2</sub> and from chemical reactions forming NO<sub>2</sub> in the atmosphere, predominantly between NO and O<sub>3</sub>.

Figure 4.3 shows the development in NO<sub>2</sub> concentrations recorded at rural, urban and traffic stations. At urban background locations the situation is relatively clear: NO<sub>2</sub> levels are decreasing at 96 % of the stations registering a trend (<sup>24</sup>). At traffic and rural locations, NO<sub>2</sub> concentrations are decreasing at fewer stations, but still at 90 % of the stations registering a trend. Despite the fact that the great majority of stations registered a decrease in annual mean concentrations, the average decrease per year for stations with a statistically significant trend (638 stations) is only 0.67  $\mu$ g/m<sup>3</sup>. When considering all stations (in total 1 545), the average decrease per year is even lower, of  $0.38 \,\mu\text{g/m}^3$ . The average decrease in NO<sub>2</sub> concentrations measured over Europe between 2002 and 2011 (ca. 7 % decrease)

<sup>(&</sup>lt;sup>23</sup>) A consistent set of 1 545 stations with data for 2002 to 2011, with a minimum data coverage of 75 % of valid data per year for at least 8 years out of the 10 years period, was used.

<sup>(&</sup>lt;sup>24</sup>) Of the total of 1 545 stations with available data for the trend analysis, 638 stations registered a trend (a significant trend, using the Mann-Kendall test). Of the 638 stations with a trend, 595 had a decreasing trend.





**Note:** The graphs are based on annual mean concentration trends (top) and the trends in percentile 99.78 of  $NO_2$  hourly values (bottom); they present the range of concentration changes per year (in  $\mu g/m^3$ ) per station type (urban, traffic, rural, and other — mostly industrial). The trends are calculated based on the officially reported data by the EU Member States with a minimum data coverage of 75 % of valid data per year for at least 8 years out of the 10-year period.

The diagram indicates the lowest and highest trends, the means and the lower and upper quartiles, per station type. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

Source: ETC/ACM.

is much slower than the decrease in  $NO_x$  emissions (27 % decrease) over the same period. The main reason for this is attributed to the increase in the share of  $NO_2$  in the  $NO_x$  emissions from traffic (Guerreiro et al., 2010).

Map 4.2 shows the spatial distribution of  $NO_2$  trends at station locations between 2002 and 2011, based on the same data and trend analysis as presented in

Figure 4.3. Although some countries have clusters of stations reporting an upward trend, most regions have stations with both upward and downward trends with the latter dominating. It is important to note that the number of stations with data covering the period 2002 to 2011 is very low in some parts of Europe, notably in parts of eastern Europe and Scandinavia.



Map 4.2 Annual changes in concentrations of NO, in the period 2002–2011

Note: The data presented were derived from a consistent set of stations in all years.

Statistically significant trends (level of significance 0.1) are calculated by applying the Mann-Kendall test. Increasing concentrations are indicated with red dots and decreasing concentrations with green dots, when statistically significant. The applied method is described in de Leeuw, 2012.

Source: ETC/ACM.

### 4.4 Exposure to NO<sub>2</sub> pollution in Europe

#### 4.4.1 Human exposure

The NO<sub>2</sub> monitoring data in AirBase provide the basis for estimating the exposure of the European population to exceedances of the NO<sub>2</sub> annual mean limit value of 40 µg/m<sup>3</sup>. Figure 4.4 presents this data for the period 2002–2011, based on NO<sub>2</sub> measured at urban background monitoring stations. For each city, an average concentration is calculated. It is considered that the whole population is potentially exposed to the calculated city average concentration, since people move freely within the city. Close to traffic people are in reality exposed to higher NO, concentrations than in urban background areas. In some European cities many people live in close proximity to traffic. This renders an underestimation of the estimated impact of exposure. According to this method, about 5 % of the EU and EEA-32

urban population was exposed to NO<sub>2</sub> above the EU annual limit value and the WHO AQG for NO<sub>2</sub> in 2011. The fraction of urban population exposed to concentration above the annual limit value varied between 5 % and 23 % between 2002 and 2011, with the same percentages estimated for the EEA-32. There is a trend of decreasing NO<sub>2</sub> exposure over this period with a decrease also observed between 2010 and 2011. The range partly reflects variations caused by meteorology.

#### 4.4.2 Exposure of ecosystems

Nitrogen compounds emitted as  $NO_x$  and  $NH_3$ are now the principal acidifying components in in ecosystems. However, acidification is not only caused by emissions of  $NO_x$  and  $NH_3$  directly. It is also caused by emissions of  $SO_x$ . In addition to the acidification caused by  $NO_x$  and  $NH_y$  these two



Figure 4.4 Percentage of the EU urban population potentially exposed to NO<sub>2</sub> concentration over the limit value set for protection of human health, 2002–2011

Source: EEA, 2013e (CSI 004).

nitrogen compounds are also the main causes of eutrophication in ecosystems.

The acidification and eutrophication effects are estimated using the concept of 'critical load', a term that describes the ecosystem's ability to absorb acidifying and eutrophying pollutants that have been deposited from the atmosphere without negative effects on the natural environment. Exceedance of these spatially determined critical loads present a risk of damage. Such exceedances are estimated based upon measurement data and model calculations.

The area of sensitive ecosystems in the EU where the critical load of acidity was exceeded in 2010 was estimated to have declined by 92 % compared with the 1990 base year (EEA, 2012a). This improvement is primarily attributed to sharp reductions in  $SO_x$ emissions in the past two decades.

Concerning eutrophication, calculated exceedances for 2010 cover most of continental Europe as well as Ireland and southern areas of the United Kingdom and Sweden (EEA, 2010a). The sensitive ecosystem area in Europe affected by eutrophication due to excessive atmospheric nitrogen was estimated to have been reduced by 23 % from 1990 to 2010 (EEA, 2012a). The risk of ecosystem eutrophication and its geographical coverage have diminished only slightly over the last decade and is still widespread over Europe. This conflicts with the EU's long-term objective of not exceeding critical loads of airborne acidifying and eutrophying substances in European ecosystem areas (EU, 2001b; EU, 2002; EC, 2005b).

### 4.5 Responses

The most relevant EU legislative instruments addressing NO<sub>x</sub> emissions and concentrations of NO<sub>x</sub> and NO<sub>2</sub> relate to motor vehicle emissions (the Euro emission standards) and emissions from combustion of fuel in industry and power production (the LCP and IPPC Directives). As described in the preceding sections, the legislation has resulted in an overall reduction of NO<sub>x</sub> emissions. However, 'real-world emissions' of NO<sub>x</sub> from diesel passenger cars have decreased very little over the last decade (both per-vehicle emissions and total emissions) and are considered to be the main driver of the exceedances of the NO<sub>2</sub> concentration limit value found across the EU. The upcoming Euro 6 standard will focus on real-world emissions.

As explained above, concentrations of  $NO_2$  in the atmosphere and at rural, urban and traffic locations originate partly from direct  $NO_2$  emissions, and partly from NO emissions transformed into  $NO_2$ . A negative effect of some technologies used in diesel vehicles to meet the Euro emission standards is that the fraction of direct NO<sub>2</sub> emissions in total NO<sub>x</sub> emissions from these vehicles is increasing.

A pilot project has done a review of the main measures adopted to manage NO<sub>2</sub> concentration levels by 12 participating cities. It found that most of the measures target traffic emissions, e.g. the creation of a Low Emission Zones; improvement of public transport; promotion of cycling; management

of traffic flow; and change in speed limits. The commercial and residential combustion sector, also considered by the cities as a main sector responsible for exceedances of  $NO_2$  limit values, was also targeted by several measures (EEA, 2013g).

The policy responses for  $NO_x$  mitigation are presented in more detail in Section 2.6. The use of air quality plans is described in Section 2.6.4.

# 5 Sulphur dioxide (SO<sub>2</sub>)

#### 5.1 Sources and effects of SO<sub>2</sub>

#### 5.1.1 Origins of SO, in air

Sulphur dioxide is emitted when fuels containing sulphur are burned. The key manmade contributions to ambient SO<sub>2</sub> derive from sulphur containing fossil fuels and biofuels used for domestic heating, stationary power generation, and transport. Volcanoes are the most important natural source.

### 5.1.2 Effects of SO<sub>2</sub>

Epidemiological studies suggest that  $SO_2$  can affect the respiratory system and lung functions, and causes irritation of the eyes. Inflammation of the respiratory tract causes coughing, mucus secretion, aggravation of asthma and chronic bronchitis, and makes people more prone to infections of the respiratory tract. Mortality and hospital admissions for cardiac disease increase on days with higher  $SO_2$ levels (WHO, 2008).

Sulphur dioxide is a major precursor to  $PM_{2.5'}$  which is associated with significant health effects, as described in Section 2.1.

Sulphur dioxide contributes to acidic deposition, causing adverse effects on aquatic ecosystems in

rivers and lakes, damage to forests, and acidification of soils. The major effects of deposited sulphur compounds are the loss of acid neutralisation capacity in soils and waters, loss of nutrients such as potassium or magnesium from soils, and the release of aluminium to the soil and waters. In certain biogeochemical conditions, sulphur can initially be stored in soils with subsequent slow release, a process known as postponed acidification. Thus, SO<sub>2</sub> emission reduction measures can take many decades before they have a positive effect.

# 5.2 European air quality standards for SO<sub>2</sub>

Table 5.1 presents the European air quality limit values for SO<sub>2</sub> defined in the 2008 Air Quality Directive (EU, 2008c). Values are given for health protection and vegetation protection. Health protection limit values are specified for 1-hour averages, and for 24-hour averages. Countries were obliged to meet the vegetation protection limits as well as both health protection limits by 2005. There is also an 'alert' threshold value of 500  $\mu$ g/m<sup>3</sup>. When this alert threshold is exceeded over three consecutive hours, authorities have to implement action plans to remedy the high levels of SO<sub>2</sub>. As shown in Table 5.2 (WHO, 2006), the WHO air quality guidelines for SO<sub>2</sub> are significantly more stringent than the limit values set by the 2008 Air Quality Directive.

#### Table 5.1 Air quality standards for SO, as given in the 2008 Air Quality Directive

Objective	Averaging period	Limit or threshold value	Number of allowed exceedances
Human health	One hour	350 µg/m³	24 hours per year
Human health	One day	125 µg/m³	3 days per year
Alert (ª)	One hour	500 µg/m³	
Vegetation	Calendar year	20 µg/m³	
Vegetation	Winter (1 October-31 March)	20 µg/m³	

Note: (a) To be measured over three consecutive hours at locations representative of air quality over at least 100 km<sup>2</sup> or an entire zone or agglomeration, whichever is the smaller.

Source: EU, 2008c.

Table 5.2

	10-minute mean µg/m³	24-hour mean µg/m³
SO <sub>2</sub>	500	20

WHO air quality guidelines for

**Source:** WHO, 2006.

### 5.3 Europe-wide survey of SO,

#### 5.3.1 Exceedances of limit values

SO,

The hourly limit value for the protection of human health was only exceeded in 2011 at five stations in the EU (two urban) in Italy, Spain, Bulgaria and Romania. The daily limit value was exceeded at seven stations in the same countries (Figure 5.2). None of those exceedances occurred at rural locations.

Map 5.1 shows annual mean  $SO_2$  concentrations in 2011. As in previous years, the highest concentrations and exceedances of the annual limit value for protection of vegetation occurred in the Balkan countries and Turkey, and at some stations in Silesia in southern Poland.

### 5.3.2 Distance to target

Figure 5.1 is the concentration status graph for the daily and hourly limit values of  $SO_2$  for health protection.  $SO_2$  concentrations are generally well below the limit values.





Note: The red and dark red dots correspond to exceedances of the limit value (20 µg/m<sup>3</sup>) for the protection of vegetation. Most of the stations on the map are, nevertheless, not located in rural areas, where vegetation exposure is important.
Source: AirBase v. 7.



## Figure 5.1 Concentration status for the daily (left) and hourly (right) limit values of SO<sub>2</sub> for health protection, EU, 2011

**Note:** The graphs are based on:the 99.18 percentile of the daily mean  $SO_2$  concentrations (in  $\mu g/m^3$ ), corresponding to the 4th highest as defined by the daily limit value (left); and the 99.73 percentile of the  $SO_2$  hourly concentrations, corresponding to the 25th highest as defined by the hourly limit value (right), for the various types of stations.

Source: AirBase v. 7.





Note: The red line corresponds to the EU limit value of 125 µg/m<sup>3</sup>, EU Member States only.

The graph is based on the 99.18 percentiles of the daily mean concentration values for each Member State; the boxes present the range of the 99.18 percentiles at all stations types (in  $\mu$ g/m<sup>3</sup>) with data officially reported by the EU Member States and how they relate to the daily limit value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

The limit value set for the protection of vegetation  $(20 \ \mu g/m^3 annual mean)$  was exceeded at twenty stations in the EU in 2011. None of those exceedances occurred at rural locations where there is more vegetation that needs to be protected than in urban areas. The limit value for the protection of vegetation set for the winter period ( $20 \ \mu g/m^3$ ) was not exceeded at rural stations within the EU during the winter 2010/2011, but it was exceeded at two rural industrial stations: one in Norway, close to the Russian border, and one in Turkey. On the other hand, within the EU, it was exceeded at 27 urban, 8 traffic and 6 industrial stations.

Figure 5.2 shows for all EU Member States the attainment of the daily limit value for SO<sub>2</sub> in 2011.

#### 5.3.3 Trends in SO, concentrations and emissions

Reported SO<sub>2</sub> concentrations decreased steadily in the period 2002 to 2011, falling on average by about one third in the EU. At nearly all urban background and traffic stations a statistically significant trend is observed from 2001 to 2010 (de Leeuw, 2012). In last year's overview, an analysis showed that a decade ago the average concentration at traffic stations was about 1  $\mu$ g/m<sup>3</sup> higher than at urban background stations. This gap between traffic stations and urban background stations narrowed to only 0.3  $\mu$ g/m<sup>3</sup> in 2010, suggesting a decreasing contribution of SO<sub>2</sub> emissions from road traffic.

EU emissions of  $SO_x$  (a family of gases that includes SO and SO<sub>2</sub>) have fallen substantially since 2002 (Figure 2.6). Total EU emissions of  $SO_x$  in 2011 were 50 % less than in 2002. The reduction of EEA-32 emissions of  $SO_x$  in the same period was 34 %. Sulphur dioxide emissions in 2011 in the EU were approximately 42 % lower than the aggregated emissions ceiling for the EU set for 2010 in the NEC Directive. Observed SO<sub>2</sub> concentrations fell by 35 % at traffic stations during the period 2002–2011, and by 27 % and 35 % at urban and rural stations, respectively. The fall observed at industrial stations was even higher, with a 46 % decrease in average concentrations between 2002 and 2011. These data correspond well with the reported emissions reductions.

The energy sector is still the main source of  $SO_x$  emissions, accounting for 59 % of EU emissions in 2011 (Figure 2.7), although its emissions have

fallen by 57 % since 2002. The next largest sector is industry, accounting for 25 % of EU SO<sub>x</sub> emissions in 2011, and a reduction of 30 % of its emissions between 2002 and 2011.

### 5.4 Exposure to SO<sub>2</sub> pollution in Europe

#### 5.4.1 Human exposure

Monitoring data from AirBase provide the basis for estimating the European population's exposure to exceedances of the  $SO_2$  limit value of 125 µg/m<sup>3</sup> as a daily average. According to the relevant directive, this limit value should not to be exceeded on more than three days in a year and was to be met by 2005. An analysis of this data is shown in Figure 5.3 for the EU in the period 2002–2011. The exposure is estimated based on  $SO_2$  measured at urban background locations. It is considered that the entire population is potentially exposed to these concentrations, since people move freely within the city. For each city an average concentration is calculated.

In 2011 none of the EU urban population (and 0.2 % of the EEA-32 urban population) was exposed to  $SO_2$  above the 24-hour average limit value. The amount of 'excess' exposure when this limit value was exceeded has varied in the EU and EEA-32 between 0.4 % and 4.5 % since 2002. The range partly reflects variations caused by meteorology. There is a trend of decreasing exposure to  $SO_2$  over this period. The stations in the EU measuring  $SO_2$  concentrations above the limit values are mostly industrial stations.

The EU urban population exposed to  $SO_2$  levels exceeding the WHO AQG in 2011 was significantly higher than the EU urban population exposed to  $SO_2$  levels above the EU's own 24-hour limit value, amounting to 45 % of the total urban population. However, here too a declining trend can be observed: this percentage has declined from 81 % to 45 % between 2002 and 2011 (Figure ES.1).

### 5.4.2 Exposure of ecosystems

Sulphur dioxide emissions and subsequent deposition of sulphur (via wet or dry deposition) contribute to acidification of the natural environment. The exposure of European ecosystems to acidifying compounds is described in Section 4.4.2


Figure 5.3 Percentage of the EU urban population exposed to SO<sub>2</sub> concentrations over the daily average limit value for protection of human health, 2002–2011

Source: EEA, 2013e (CSI 004).

### 5.5 Responses

The directives most relevant for the reduction of  $SO_2$  in the air are those relating to emissions from combustion of fuels in power plants and industry, i.e. the LCP and IPPC Directives (EU, 2001a and EU, 2008b). These directives led to significantly reduced  $SO_x$  emissions from these sources.

The Sulphur Content of Liquid Fuels Directive (EU, 1999b) has limited the sulphur content of

heavy fuel oil and gas oils since 2003, contributing to  $SO_2$  emission reductions and subsequent reductions of  $SO_2$  concentrations in the air.

The Fuel Quality Directive (EU, 2003a) cut the sulphur contents of fuels from 150 mg/kg for petrol and 350 mg/kg for diesel before 2005 to 50 mg/kg for each by 2005 and to 10 mg/kg by 2009. Air quality plans, as described in Section 2.6.4 are additional policy instruments to reduce exposure to  $SO_2$ .

# 6 Carbon monoxide (CO)

# 6.1 Sources and effects of CO

### 6.1.1 Origins of CO in air

Carbon monoxide is a gas emitted due to incomplete combustion of fossil fuels and biofuels. Road transport was once a significant source of CO emissions, but the introduction of catalytic converters reduced these emissions significantly. CO concentrations tend to vary with traffic patterns during the day. The highest CO levels are found in urban areas, typically during rush hours at traffic locations.

### 6.1.2 Health effects of CO

Carbon monoxide enters the body through the lungs. In the blood it is strongly bound to haemoglobin. Exposure to CO can reduce blood's oxygen-carrying capacity, thereby reducing oxygen delivery to the body's organs and tissues. Those suffering from cardiovascular disease are the most sensitive towards CO exposure. Such people already have a reduced capacity for pumping oxygenated blood to the heart, which can cause them to experience myocardial ischemia (reduced oxygen to the heart), often accompanied by angina (chest pain), when exercising or under increased stress. Short-term CO exposure further affects the already-compromised ability of their bodies to respond to the increased oxygen demands of exercise or exertion. At extremely high levels, CO can cause death.

The atmospheric lifetime of CO is about three months. It slowly oxidises into carbon dioxide, also forming  $O_3$  during this oxidation process. CO therefore contributes to the atmospheric background concentration of  $O_{3'}$  with associated effects on health and ecosystems.

# 6.2 European air quality standards for CO

Table 6.1 sets out the European air quality limit value and the WHO air quality guideline for CO. The European limit value for health protection is the maximum allowable daily eight-hour mean, to be met by 2005.

# 6.3 Europe-wide survey of CO

#### 6.3.1 Exceedances of limit values

Four out of 1 003 operational stations with more than 75 % data coverage (i.e. each station produced valid data at least 75 % of the time) in the EEA-32 countries reported exceedances of the EU's CO limit value: two traffic stations (located in Austria and Bulgaria), one urban background station (located in Italy) and one industrial station (located in Sweden).

The annual averages of the daily 8-hour maxima (Map 6.1) show elevated levels in some of those countries.

# Table 6.1Air quality limit values set by<br/>the Air Quality Directive and the<br/>WHO air quality guideline for CO

	Hourly CO mg/m <sup>3</sup>	8-hour average CO mg/m <sup>3</sup>
EU	-	10
WHO	30	10

**Source:** EU, 2008c; WHO, 2000.

≤ 0.5

No data

process

0.5-1.0 1.0-2.0 2.0-2.5 > 2.5

Countries/regions not included in

the data exchange

.



#### Annual mean of maximum daily 8-hour mean CO concentrations in 2011 Map 6.1

Source: AirBase v. 7.

# Figure 6.1 Concentration status for the CO limit value, 2011

Distribution of the max 8-hour mean CO concentrations Percentage of stations (%)



Source: AirBase v. 7.

### 6.3.2 Distance to target

Figure 6.1 shows that, except at very few stations, measured CO concentrations in Europe are well below the EU limit value.

Figure 6.2 shows for all EU Member States the status of the maximum daily 8-hour mean value of CO for 2011. It shows that exceedance of the EU limit value and the WHO AQG value occurred in four EU Member States in 2011.

In contrast to the situation for the NO<sub>2</sub> annual limit value, CO exceedance situations are not widespread.

### 6.3.3 Trends in CO concentrations

More than 80 % of urban background and traffic stations show a downward trend in CO concentrations between 2001 and 2010 (de Leeuw, 2012).

The concentration of CO at rural stations is very low - close to the detection limit. At these stations



Figure 6.2 Attainment situation for the maximum daily 8-hour mean value of CO in 2011

**Note:** The graph is based on the maximum daily 8-hour mean value of CO concentrations (in mg/m<sup>3</sup>) for each Member State; the boxes present the range of concentrations at all stations types officially reported by the EU Member States and how the concentrations relate to the limit value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

there is a large contribution from the hemispheric background of CO and no apparent trend can be seen. The CO emission reduction in the period 2002–2011 was 32 % in the EU and 27 % in the EEA-32 (Figure 3.4). Commercial, institutional and household fuel combustion was Europe's largest CO source in 2011, following the very significant reduction in transport sector emissions that have resulted from the application of the Euro standards.

Carbon monoxide concentrations are on average higher at traffic stations, compared to urban background stations.

Average CO concentrations have decreased at all station types between 2002 and 2011. The observed average reduction in CO daily 8-hour maxima concentrations in this period was 35 % for all stations in the EU. The reduction registered at traffic stations in the period was 37 % and while for urban background stations it was 18 % (<sup>25</sup>). These reductions in concentrations are 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. It is worth noting that instrument measurement uncertainties at these low concentration levels affect the accuracy of the measured concentrations, and therefore also affect the accuracy of trend estimates.

# 6.4 Exposure to CO pollution in Europe

Based on the available measurements, it can be concluded that the European population's exposure to CO ambient concentrations above the limit value (8-hour maximum) is very localised and infrequent, and is limited to very few areas near traffic and industry.

# 6.5 Responses

Carbon monoxide emissions are mainly regulated by the IPPC Directive (EU, 2008b) - now replaced by the Industrial Emissions Directive (EU, 2010) and the 'Euro' standards for motor vehicles, which set CO emission limits for gasoline and diesel vehicles (see Annex 2). The emission limits set out in both of these legal instruments have been more than halved since the early 1990s. Over the same period, the CO emissions from transport have been reduced by more than 75 %, falling even faster than the reduction in emission limits. The largest CO emission sector is now 'commercial, institutional and household fuel combustion', which is currently unregulated with respect to CO emissions. CO emissions in the EU from this sector have increased by about 7 % between 2007 and 2011, with some reduction in 2011 compared to 2010 (Figure 3.5).

<sup>(25)</sup> The estimate is based on a consistent set of stations (305 stations in total) with at least 75 % data coverage in both years of 2002 and 2011. The number of stations data for the analysis was 165 traffic stations and 95 urban background stations.

# 7 Heavy metals

Arsenic (As), cadmium (Cd), lead (Pb), mercury (Hg) and nickel (Ni) are mainly emitted as a result of various industrial activities and the combustion of coal. Although the atmospheric concentrations of these metals are low, they still contribute to the deposition and build-up of heavy metal contents in soils, sediments and organisms. Heavy metals do not break down in the environment, and some bioaccumulate, i.e. they gradually accumulate in plants and animals and cannot be excreted by them. This means that plants and animals can be poisoned by heavy metals over a long period of time through long-term exposure to even small amounts of heavy metals. If a heavy metal has bioaccumulated in a particular place in the food chain — for example in a fish — then human consumption of that fish presents a serious health risk.

Arsenic exposure is associated with increased risk of skin and lung cancer. Arsenic is not a heavy metal per se but is regularly added to the list of heavy metals, based on its toxicity. Arsenic is a metalloid a chemical element that has properties that are in between or a mixture of those of metals and non-metals. Cadmium is associated with kidney and bone damage and has also been identified as a potential human carcinogen, causing lung cancer. Lead exposure has developmental and neuro-behavioural effects on foetuses, infants and children, and can also elevate blood pressure in adults. Mercury is toxic in the elemental and inorganic forms, but the main concern is associated with mercury's organic compounds, especially methyl mercury. Methyl mercury accumulates in the food chain, for example in predatory fish in lakes and seas and passes through ingestion to humans. Nickel is a known carcinogen and also has other non-cancerous effects, for example on the endocrine system.

Air pollution is only one source of exposure to these metals, but their persistence and potential for long-range atmospheric transport means that atmospheric emissions of heavy metals affect even the most remote regions (WHO, 2007).

# 7.1 Sources and effects of heavy metals

# 7.1.1 As

# Origins of As in air

Arsenic is released into the atmosphere from both natural and anthropogenic sources. Most man-made emissions are released from metal smelters and the combustion of fuels. Pesticides used to be an important source of As, but restrictions in various countries have reduced its role. Tobacco smoke may contain As, making it a source of As exposure in ambient air.

Arsenic in the air is usually a mixture of atomic As and arsenate ( $AsO_4^{3-}$  a compound that combines arsenic and oxygen), with organic As compounds. These organic varieties are usually of negligible importance except in areas where there is substantial application of methylated As pesticides.

# Effects of As

The non-cancerous effects of inhaling air with high As levels include increased mortality from cardiovascular diseases, neuropathy, and gangrene of the extremities. There is evidence that inorganic As compounds cause cancer of the skin and lungs in humans. Lung cancer is the critical effect following exposure by inhalation.

Arsenic is highly toxic to aquatic life and also very toxic to animals in general. Plant growth and crop yields may be reduced where soil As content is high. Organic As compounds are very persistent in the environment (they are not broken down over time by environmental processes) and bioaccumulate in the food chain.

# 7.1.2 Cd

# Origins of Cd in air

Cadmium is released into the atmosphere from natural and anthropogenic sources. Soil dust and fires are considered the main natural sources of Cd in the atmosphere, while minor amounts are also emitted with sea salt spray or from volcanic eruptions. The anthropogenic sources of Cd include non-ferrous metal production, stationary fossil fuel combustion, waste incineration, iron and steel production, and cement production.

# Effects of Cd

Cadmium is highly 'persistent' in the environment and bioaccumulates. In heavily contaminated areas, resuspended dust (caused by vehicles or wind blowing cadmium particles off the ground) can constitute a substantial part of the exposure for the local population.

In Europe, air pollution and fertilisers (both mineral and organic) contribute almost equally to annual exposure. Both of these increase the relatively large accumulations of Cd in topsoil, thereby increasing the risk of future exposure through food. The levels of Cd in non-smokers have not decreased over the last decade.

The kidneys and bones are the critical organs affected by chronic environmental exposure to Cd. The main effects of this exposure include impaired kidney function and increased risk of osteoporosis. An increased risk of lung cancer has also been reported following inhalation exposure to Cd.

Cadmium is toxic to aquatic life as it is directly absorbed by organisms in water. It interacts with cellular components, causing toxic effects in the cells of all organisms.

# 7.1.3 Pb

# Origins of Pb in air

Lead is released into the atmosphere from natural and anthropogenic sources. Natural emissions typically include soil dust and sea spray containing Pb, as well as particles found in ashes from volcanoes and forest fires. These emissions are not entirely natural but contain some deposits of anthropogenic Pb. Major anthropogenic emission sources of Pb on a global scale include traffic-related fossil fuel combustion, waste incineration and production of non-ferrous metals, iron, steel and cement. The contribution to emissions from Pb in petrol fuel has now been eliminated in Europe. This followed a complete phase-out through legislation and a complete switch-over to un-leaded petrol.

# Effects of Pb

Lead is a neurotoxic metal that also accumulates in the body and damages organs, such as the kidneys, liver, brain, and nerves. Exposure to high levels of Pb causes serious brain damage, including mental retardation, behavioural disorders, memory problems and mood changes. Impairment of neurodevelopment in children is the most critical effect. Exposure in utero, during breastfeeding, or in early childhood may lead to such health problems. Lead accumulates in the skeleton which is potentially dangerous during pregnancy. Hence, previous exposure to a woman before she becomes pregnant is important in determining the health of her child.

Inhalation exposure may be significant when Pb levels in the air are high. Elevated exposures are generally due to local sources rather than being the result of long-range transport from remote sources. However, air pollution may contribute significantly to the Pb content of crops, through direct deposition. Although uptake via plant roots is relatively limited, rising Pb levels in soils over the long term are a matter for concern and should be addressed because of the possible health risks of low-level exposure to Pb.

Lead bioaccumulates and adversely impacts both terrestrial and aquatic systems. As with humans, the effects on animal life include reproductive problems and changes in appearance or behaviour.

# 7.1.4 Hg

# Origins of Hg in air

The largest anthropogenic source of Hg emissions to air on a global scale is the combustion of coal and other fossil fuels. Others sources include metal production, cement production, waste disposal and cremation. In addition, gold production makes a significant contribution to global air emissions of Hg. The main natural sources of Hg emissions are diffusion from the Earth's mantle though the lithosphere, evaporation from the sea surface and geothermal activity. Mercury emitted in inorganic forms is converted biologically to methyl mercury in soil and water.

# Effects of Hg

Mercury can damage the liver, the kidneys and the digestive and respiratory systems. It can also cause brain and neurological damage and impair growth. Methyl mercury is a potent neurotoxin. Unborn children are the most vulnerable population group in terms of exposure to Hg.

Mercury bioaccumulates and adversely impacts both terrestrial and aquatic systems. It can affect animals in the same way as humans and is very toxic to aquatic life.

# 7.1.5 Ni

### Origins of Ni in air

Nickel occurs in soil, water, air and in the biosphere. Nickel emissions to the atmosphere may come from natural sources such as windblown dust, volcanoes and vegetation. The main anthropogenic sources of Ni emissions into the air are combustion of oil for the purposes of heating, shipping or power generation; Ni mining and primary production; incineration of waste and sewage sludge; steel manufacture; electroplating; and coal combustion.

### Effects of Ni

Nickel exposure can result from breathing ambient air. In very small quantities Ni is essential to humans. However, a large uptake can be a danger for human health as several Ni compounds are carcinogenic, increasing the risk of developing, for example, cancers of the lung, nose, larynx or prostate. Non-cancerous effects on health include allergic skin reactions (generally not caused by inhalation), disruption of endocrine regulation, and damage to the respiratory tract, and the immune systems. The most common harmful health effect of Ni in humans is an allergic reaction. Approximately 10–20 % of the population is sensitive to Ni. As is the case for humans, Ni is an essential element for animals in small amounts. But in high concentrations, Ni and its compounds can be acutely and chronically toxic to aquatic life and may affect animals in the same way as humans. It is known that high Ni concentrations in sandy soils can damage plants, and that high concentrations in surface waters can diminish the growth rates of algae. Microorganisms can also suffer from growth decline. Nickel is not known to accumulate in plants or animals.

## 7.2 European air quality standards for heavy metals

Table 7.1 shows the European air quality target values for As, Cd, and Ni, and the limit value for Pb. The values specified are maximum annual averages, which countries are obliged to meet by 2013, except for the limit value for Pb which was to be met by 2005. Table 7.1 also shows the WHO air quality guidelines as annual mean concentrations.

No EU target or limit value has been set for Hg concentrations in air. A protocol on heavy metals including Hg was adopted in 2003 within the framework of the UNECE LRTAP. It aimed at limiting emissions of Hg.

### 7.3 Europe-wide survey of heavy metals

Barrett et al. (2008) reviewed the concentrations of Pb and the pollutants covered by Directive 2004/107/EC (EU, 2004b), i.e. As, Cd, Ni and BaP, and pointed to the lack of concentration measurements. Compared to 2006 (the year reviewed by Barrett et al.), the number of monitoring stations has increased in 2011, but monitoring data for parts of Europe is still missing. This might be the reason why values for these pollutants are reported for a relatively small number of stations: As concentrations of these pollutants are frequently below the lower assessment threshold (LAT) in the EU, techniques other than monitoring such as modelling — can be used for assessing air quality. Following the data quality objectives set in EU legislation, for indicative measurements a criterion on data coverage of 14 % (i.e. at least 14 % of the data produced by each monitoring stations in the course of a year has to be valid before it can be used in the study) is applied here on the heavy metal data. A problem in analysing the data of these pollutants is that it is not always certain (from the data made

# Table 7.1Air quality limit and target values for As, Cd, Ni and Pb regulated by EU, and WHO<br/>air quality guidelines

Pollutant	EU target or limit value (°) ng/m <sup>3</sup>	WHO AQG ng/m <sup>3</sup>
Arsenic	6 ( <sup>b</sup> )	-
Cadmium	5 ( <sup>b</sup> )	5 (ª)
Nickel	20 ( <sup>b</sup> )	-
Lead	500 (°)	500

**Note:** (a) Annual mean, measured as contents in PM<sub>10</sub>.

(<sup>b</sup>) Target value, entering into force on 31 December 2012.

(<sup>c</sup>) Limit value to be met by 1 January 2005. The limit value to be met only by 1 January 2010 in the immediate vicinity of specific industrial sources situated on sites contaminated by decades of industrial activities. In such cases, the limit value until 1 January 2010 is 1 000 ng/m<sup>3</sup>.

(d) WHO AQG set to prevent any further increase of cadmium in agricultural soil, likely to increase the dietary intake of future generations.

Source: EU, 2004b; and WHO, 2000.

available by the countries) whether the heavy metals have been measured on the  $PM_{10}$ -particle size fraction (as required by the directive) or on another (undefined) size fraction, e.g. particles of all sizes.

Map 7.1 presents annual mean ambient concentrations of As, Cd, Pb and Ni reported across Europe. The maps show that the air pollution problem of these heavy metals is highly localised: problems are related to specific industrial plants or areas covered by monitoring stations.

The results from the reported 2011 data can be summarised as follows:

- Arsenic concentrations below the lower assessment threshold (2.4 ng/m<sup>3</sup>) were reported at nearly 90 % of the stations in 2011. At fifteen stations (out of 631 operational stations) the reported concentrations exceeded the target value set for 2013 (6 ng/m<sup>3</sup>). Exceedances of the target value were observed in Belgium, the Czech Republic, Germany, Poland, Finland and Bulgaria. In five exceedance situations, As has been reported as belonging to the 'aerosol' category, i.e. an undefined size fraction.
- **Cadmium** concentrations in the air exceeded the target value at 1 % of the stations in Europe in 2011, i.e. at 8 stations in total. Exceedances beyond the 5 ng/m<sup>3</sup> target value were observed in two countries (Belgium and Bulgaria). At the majority of the other stations (94 %), Cd

concentrations were below the lower assessment threshold  $(2 \text{ ng/m}^3)$ .

- Lead concentrations exceeded the 500 ng/m<sup>3</sup> limit value at two stations in 2011. 99 % of the stations reported Pb concentrations below the lower assessment threshold of 0.25 µg/m<sup>3</sup>.
- Nickel concentrations exceeded the target value of 20 ng/m<sup>3</sup> at eight out of the 632 operational stations (1 % of the total operational stations). These stations are located in Belgium, the Czech Republic, Germany, Italy, and the United Kingdom. Most of the exceedances are related to industry.
- Mercury concentrations recorded in AirBase are very few, despite the fact that Directive 2004/107/ EC (EU, 2004b) requests EU Member States to perform (indicative) measurements of Hg at least at one background station. Background concentrations of Hg in air in 2011 ranged from 0.4 to 6.9 ng/m<sup>3</sup> (stations in Germany, Finland, Latvia, Malta, Poland, Sweden, Slovenia and the United Kingdom). One industrial station in the United Kingdom registered a concentration of 21.1 ng/m<sup>3</sup> Hg in air. Measurements of deposited mercury (mercury that is no longer airborne but is now deposited on the soil or ground) are reported at 25 stations (of which 18 are located in Germany and the United Kingdom). Averaged over all stations, the annual deposition flux is 0.11 g/ha/year.

# Map 7.1 Annual mean concentrations of heavy metals (arsenic, cadmium, lead and nickel), 2011



Source: AirBase v. 7.

# 7.4 Trends in concentrations and emissions of heavy metals

There is no requirement in EU legislation for EU Member States to report emissions for different heavy metals. Nevertheless, a number of EU Member States have signed and ratified the 'Heavy Metals Protocol' under the UNECE LRTAP Convention, under which the reporting of certain information is required. Meanwhile, other countries voluntarily report emission data for these metals to the Convention. Nevertheless, the reported estimates of heavy metal emissions are of relatively high uncertainty compared to the main air pollutants, as the data is not always complete, and because emissions estimates are sometimes based on relatively few measurements.

# 7.4.1 As

The number of As monitoring stations has increased rapidly in recent years, to more than 600 stations in 2010 and 2011, including background, traffic and industrial stations.

The majority of stations recorded a slight reduction in As concentrations in the period 2006–2011, although it is worth stressing that the sampling and measurement method was not reported for some stations and the methods might have changed during the period. The short period of the assessment makes it impossible to undertake a statistical analysis of the concentration trend and its statistical significance.

Most countries decreased their average As concentrations between 2006 and 2011. Poland was in 2011 responsible for 21 % of the EU's total As emissions, but has registered a decrease in emissions of 6 % from 2006 to 2011. Poland has also observed a decrease in As concentrations in the same time period in all but two of its monitoring stations. Bulgaria, which was responsible for 7 % of the EU's As emissions in 2011, decreased its total As emissions between 2006 and 2011 by 11 %. However, this overall decline in emissions did not translate into a uniform decline in concentrations: As concentrations at one station in Bulgaria increased significantly between 2006 and 2011, while at another station a decrease could be observed over the same period. Slovakia, responsible for 11 % of EU As emissions in 2011, reduced its As emissions by 14 % between 2006 and 2011, but registered an increase of 5 % between 2010 and 2011. The changes in Slovak emissions up to 2010 are reflected in generally decreasing As concentrations between

2006 and 2010. The impact of the increase in emissions between 2010 and 2011 is unknown, as Slovakia did not report data on As concentrations for 2011.

Figure 7.1 shows the development in As (and other heavy metals) emissions reported by the EU Member States between 2002 and 2011 as a percentage of 2002 emissions. Arsenic emissions were reduced on average by about 3 % from 2002 to 2011 in the EU and in the EEA-32 countries.

The time series of changes in As concentrations are at present too short and the geographical coverage too limited to support a Europe-wide comparison with emission trends.

# 7.4.2 Cd

About half of the stations with measurements since 2006 recorded a reduction in concentrations of Cd during the last six years, while 13 % of stations registered some increase. The number of stations measuring Cd concentrations in the air has increased significantly over recent years, i.e. from 37 in 2002 and 239 in 2006, reaching 665 in 2011 and covering 23 EU Member States, as well as Norway and Switzerland.

Concentrations have decreased in 13 out of 14 stations in Switzerland over the last ten years (2002–2011), although two stations registered an increase from 2010 to 2011. The United Kingdom (with 19 stations recording data from 2006 to 2011) registered a decrease in concentrations between 2006 and 2011 in 14 stations. Spain registered a clear general decrease in concentrations between 2006 and 2011. In Austria, Belgium, Bulgaria, the Czech Republic and Germany the stations recorded both increasing and decreasing concentrations since 2006.

Cadmium emissions in both the EU and the EEA-32 decreased by 27 % between 2002 and 2011 (Figure 7.1). The Cd concentration time series are limited to several countries. The limited geographical coverage in Europe does not support a Europe-wide comparison with emission changes.

# 7.4.3 Pb

For the period 2006–2011, data on Pb concentrations in 10 countries were available in AirBase. While concentrations have remained almost constant at traffic and rural stations since 2006, there was a clear reduction in measured concentrations at industrial





Source: EEA.

stations between 2002 and 2009. There was some increase in Pb concentrations between 2009 and 2011 in 50 % of the stations reporting data in 2009 and 2011.

Lead emissions decreased in the EU and EEA-32 by 20 % between 2002 and 2011 (Figure 7.1). The Pb concentration time series from 2002 to 2011 are limited to four countries (Austria, Bulgaria, Portugal and Switzerland). The low geographical coverage does not support a Europe-wide comparison with changes in Pb emissions.

### 7.4.4 Hg

Various compounds of Hg are measured at a number of stations in the EMEP network, using a variety of methods. In view of the limited data available in AirBase, trends in Hg concentrations in air in Europe are not evaluated here.

Mercury emissions in the EU and in the EEA-32 decreased by 26 % between 2002 and 2011 (Figure 7.1).

### 7.4.5 Ni

Nickel concentrations were reported for the period 2006–2011 by nine countries: Austria, Belgium, Bulgaria, the Czech Republic, Germany, Italy, Spain, Switzerland, and the United Kingdom, while there is limited data for earlier years. The average concentrations measured between 2006 and 2011 have continuously decreased in all these countries, with the exception of Germany. Nevertheless, reported total Ni emissions in Germany have decreased by 29 % from 2006 to 2011.

Nickel emissions decreased in the EU and EEA-32 countries by 43 % between 2002 and 2011 (Figure 7.1). As outlined above, Ni concentration time series for the period 2002–2011 are very limited and the geographical coverage is too low to support a Europe-wide comparison with emission changes.

## 7.5 Exposure to heavy metal pollution in Europe

Human exposure to Pb, As, Cd, and Ni ambient air concentrations above the limit or target values is a local problem, restricted to a few areas in Europe, and is typically caused by specific industrial plants.

On the other hand, atmospheric deposition of heavy metals into the environment contributes to the exposure of ecosystems and organisms to heavy metals and bioaccumulation in the food chain, thus affecting human health. Some ecosystem areas are at risk due to atmospheric deposition of Cd, Pb or Hg.

The share of national ecosystem area in Europe, exceeding critical loads for Cd is below 1 % in most countries, except countries which have set lower critical loads than other countries (e.g. Bulgaria) (Slootweg et al., 2010).

For Pb the area and extent of the exceedances of critical loads are much higher. Atmospheric deposition of Pb exceeds the critical loads in over 12 % of the EU ecosystem area (Slootweg et al., 2010).

The largest exceedances of heavy metal critical loads involve Hg. More than half of all EEA-32 countries (<sup>26</sup>) have exceedances of critical loads for Hg across nearly 90 % or more of their ecosystem area. In total, atmospheric deposition of Hg exceeds the critical loads across 54 % of the EU ecosystem area (Slootweg et al., 2010).

# 7.6 Responses

Emissions of heavy metals are regulated by the Fuels Quality Directive (EU, 2003a) and the Industrial Emissions Directive (EU, 2010) (which replaced both the former IPPC Directive (EU, 2008b) and the Waste Incineration Directive (EU, 2000)). The Industrial Emissions Directive includes metals and their compounds in its list of polluting substances to be regulated. It obliges industries to use best available techniques to limit the emissions of heavy metals as much as possible.

The Fuels Quality Directive (2003/17/EC) requires all motor fuel sold in the EU after 1 January 2002 to be lead-free. This has eliminated the contribution from road traffic to Pb concentrations in the air.

The European Commission's Strategy on Hg (EC, 2005a), launched in 2005 and reviewed in 2010, addresses most aspects of the Hg life cycle. Its key aim is to reduce Hg levels both in relation to human exposure and in relation to exposure to the environment. The strategy identifies 20 priority actions undertaken, both within the EU and internationally, to reduce Hg emissions, cut supply of and demand of Hg, and protect against exposure to Hg, with special attention paid to reducing the exposure of fish to methyl mercury. As a result, restrictions were set on the sale of measuring devices containing Hg, a ban on exports of Hg from the EU came into effect in March 2011, and new rules on safe storage of Hg were adopted.

Furthermore, international conventions control emissions and transport of Hg. One of these conventions is the UNECE Convention on Long-range Transboundary Air Pollution, which aims to cut emissions of Hg and other heavy metals from industry, combustion, and waste. Another agreement that seeks to control Hg is the Basel Convention (<sup>27</sup>) on the Control of Transboundary Movements of Hazardous Wastes and their Disposal, which aims to protect human health and the environment from the movement of waste to eastern Europe and developing countries. The Basel Convention prohibits waste contaminated by Hg from being exported from the EU for disposal, recovery, or recycling in other countries.

<sup>(&</sup>lt;sup>26</sup>) Albania, Bosnia and Herzegovina, Bulgaria, Croatia, Denmark, Greece, Hungary, Italy, Latvia, Lithuania, Luxembourg, the former Yugoslav Republic of Macedonia, the Netherlands, Poland, Romania, Slovenia and Spain.

<sup>(27)</sup> http://www.basel.int.

# 8 Benzene (C<sub>6</sub>H<sub>6</sub>) and benzo(a)pyrene (BaP)

# 8.1 Sources and effects

8.1.1  $C_6 H_6$ 

# Origins of $C_6H_6$ in air

Incomplete combustion of fuels is the largest source of  $C_6H_6$ . Benzene is an additive to petrol, and 80–85 % of  $C_6H_6$  emissions are due to vehicle traffic in Europe. Other sources of  $C_6H_6$  include domestic heating and oil refining, as well as the handling, distribution and storage of petrol. In general the contributions to  $C_6H_6$  emissions made by domestic heating are small (about 5 % of total emissions) but with sharp differences depending on the region. In areas where wood burning accounts for more than half of domestic energy needs, wood combustion can be an important local source of  $C_6H_6$  (Hellén et al., 2008).

Removal of  $C_6H_6$  from the atmosphere mainly occurs through the photochemical degradation of  $C_6H_6$ . This degradation also contributes to  $O_3$  formation, although the chemical reactivity of  $C_6H_6$  is relatively low. Benzene can often survive un-degraded in the atmosphere for several days, a period of time that is sufficient for it to be transported over long distances.

# Health effects of $C_6H_6$

Inhalation is the dominant pathway for  $C_6H_6$ exposure in humans. Benzene is a carcinogenic pollutant. The most significant adverse effect from prolonged exposure is damage to the genetic material of cells. This damage can cause cancer. Chronic exposure to  $C_6H_6$  can damage bone marrow and cause haematological effects such as decreased red and white blood cell counts.

# 8.1.2 Benzo(a)pyrene (BaP)

### Origins of BaP in air

Benzo(a)pyrene is a five-ring polycyclic aromatic hydrocarbon (PAH) and is found in fine particulate

matter that originates in incomplete combustion of various fuels. The main sources of BaP in Europe are waste burning, coke and steel production, vehicles, and domestic home heating, in particular wood burning. Other sources include outdoor fires and rubber tyre wear.

# Health effects of BaP

The International Agency for Research on Cancer (IARC) considers BaP a carcinogen. While laboratory studies show that BaP is a known carcinogen in animals, epidemiological studies have only been able to assess the effect of a mixture of PAHs (including BaP) found in soot, tars and oils.

Prenatal exposure to PAHs has been linked to significantly reduced birth weight in an international study from Poland (Krakow) and USA (New York) (Choi et al., 2006), and it is suggested that pre-natal exposure also adversely affects cognitive development in young children (Edwards et al., 2010).

# 8.2 European air quality standards for $C_6H_6$ and BaP

The limit value for  $C_6H_6$  and the target value for BaP for the protection of human health (both of which are set by EU legislation) are shown in Table 8.1.

# Table 8.1Air quality limit and target values<br/>for annual mean concentration<br/>as set out in EU legislation

	EU
Benzene µg/m³	5 (ª)
Benzo(a)pyrene ng/m <sup>3</sup>	1 ( <sup>b</sup> )

Note: (a) Limit value to be met by 2010.

(<sup>b</sup>) Target value to be met by 2013.

Source: EU, 2004b.

# 8.3 Europe-wide survey of C<sub>6</sub>H<sub>6</sub> and BaP

8.3.1  $C_6 H_6$ 

### Exceedances of limit value

Benzene is measured at a relatively small number of stations. At many locations, annual mean concentrations of  $C_6H_6$  are below the lower assessment threshold of 2 µg/m<sup>3</sup> (Barrett et al., 2008). When concentrations are below the lower assessment threshold, European legislation allows for air quality to be assessed by means of indicative or discontinuous measurements. The 2008 Air Quality Directive (EU, 2008c) sets an annual average concentration limit value of 5  $\mu$ g/m<sup>3</sup> for C<sub>6</sub>H<sub>6</sub> in ambient air, to be met by 2010.

Map 8.1 presents the annual average  $C_6H_6$  concentrations at stations with at least 50 % data coverage. The limit value was exceeded at four stations within the EU (in the Czech Republic, Italy and Poland), as well as in Serbia. The exceedances were observed in urban (2), traffic (2) and industrial (1) stations, with no exceedances of the limit value observed at rural background stations.

#### Map 8.1 Annual mean benzene concentrations, 2011





Note:Dark green dots correspond to concentrations under the WHO reference level (1.7 μg/m³).Pale green dots correspond to concentrations under the lower assessment threshold (2 μg/m³).Orange dots correspond to concentrations between the lower and the upper assessment threshold (3.5 μg/m³).Red dots correspond to concentrations under the limit value of 5 μg/m³ but higher than the upper assessment threshold.Dark red dots correspond to concentrations exceeding the limit value of 5 μg/m³.The data coverage criterion has been set to 50 % by an European Commission working group on benzene (Mol et al., 2011).

Source: AirBase v. 7.



# Figure 8.1 Concentration status for the benzene limit value, 2011

Source: AirBase v. 7.

### Distance to target

Figure 8.1 shows that, except at a few stations, measured  $C_6H_6$  concentrations in the EU are well below the limit value.

Figure 8.2 shows for all EU Member States the status of the annual mean  $C_6H_6$  values for 2011. It shows that in 2011  $C_6H_6$  annual concentrations were, on average, well below the limit value.

### Trends in $C_6H_6$ concentrations

Annual mean concentrations of  $C_6H_6$  were highest in 2011 at traffic stations, as petrol is still one of the most important sources of  $C_6H_6$ . Concentrations measured at traffic and urban background stations decreased steadily from 2002 until 2007, after which point they stabilised. Benzene concentrations at urban and rural stations show a less sharp decrease during the same period than traffic stations.

Whether  $C_6H_6$  emissions recorded a similar stabilisation after 2007 is unclear because  $C_6H_6$  is not included as an individual pollutant in European emissions inventories covering VOC. This means that  $C_6H_6$  emissions are not recorded.



### Figure 8.2 Attainment situation for annual limit value of benzene in 2011

Note: The graph is based on the annual mean concentration values for each Member State; the boxes present the range of concentrations at all stations types (in μg/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the limit value set by EU legislation (marked by the red line).

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

### 8.3.2 BaP

### Exceedances of target value

Benzo(a)pyrene measurements in 2011 were above the target value threshold (1 ng/m<sup>3</sup> annual average to be met by 2013) at 35 % of monitoring stations in the EU (<sup>28</sup>) (Map 8.2). This was the case mainly at urban and suburban background stations (50 % of stations in urban and suburban locations exceeded the target value) and, to a lesser extent, at rural, traffic and industrial stations. Exceedances are most predominant in central and eastern Europe (Austria, the Czech Republic, Hungary, Italy (the Po Valley), Latvia, Poland and Slovakia) although there are also exceedances in Bulgaria, Germany, Finland, France, and the United Kingdom (the Midlands and Northern Ireland). In addition, France and Greece also report exceedances of the target value in one or more air quality management zones.



Map 8.2 Annual mean concentrations of benzo(a)pyrene (ng/m<sup>3</sup>), 2011

Note: Dark green dots correspond to concentrations under the WHO reference level (0.12 ng/m<sup>3</sup>).
Pale green dots correspond to concentrations under the lower assessment threshold (0.4 ng/m<sup>3</sup>).
Orange dots correspond to concentrations between the lower and the upper assessment threshold (0.6 ng/m<sup>3</sup>).
Red dots correspond to concentrations above the upper assessment threshold but under the target value of 1 ng/m<sup>3</sup>.
Dark red dots correspond to concentrations exceeding the target value of 1 ng/m<sup>3</sup>.

Source: AirBase v. 7.

<sup>(28)</sup> With 14 % or more of valid data.

# Figure 8.3 Concentration status for the benzo(a)pyrene target value in 2011



#### **Source:** AirBase v. 7.

### Distance to target

Figure 8.3 shows that many stations are approaching and exceeding the target value for BaP for rural, urban, traffic and other (including industrial) station types. As the figure shows, exceedances occurred at all station types, and the highest concentrations are predominantly measured at urban stations. Figure 8.4 shows for all EU Member States a box plot of the annual mean BaP values for 2011. It shows that average annual concentrations of BaP exceeded the target value in twelve countries (Austria, Bulgaria, the Czech Republic, Finland, France, Germany, Hungary, Italy, Latvia, Poland, Slovakia and the United Kingdom). The average concentration measured at Polish stations is five times higher than the target value.

#### Trends in BaP concentrations and emissions

Trends in BaP concentrations over the last five years (2007–2011) can be sourced from a limited number of stations. In total, there is data on these concentrations from 117 stations in the following 13 countries: Austria, Belgium, Bulgaria, the Czech Republic, Denmark, Estonia, Germany,

# ng/m<sup>3</sup> 20.00 15.00 10.00 5.00 0.00 Netherlands Clech Republic Belgium Estonia Bulgaria Jovenia United Kingdo' HUNDBAN Poland Luxemb ithu

Figure 8.4 Attainment situation for annual mean concentration of BaP in 2011

**Note:** The graph is based on the annual mean concentration values for each Member State; the boxes present the range of concentrations at all stations types (in ng/m<sup>3</sup>) officially reported by the EU Member States and how the concentrations relate to the limit value set by EU legislation (marked by the red line). The countries not included in the graph have not reported measured BaP concentrations.

The diagram indicates the lowest and highest observations, the means and the lower and upper quartiles. The lower quartile splits the lowest 25 % of the data and the upper quartile splits the highest 25 % of the data.

Italy, Lithuania, Poland, Spain, Switzerland, and the United Kingdom (<sup>29</sup>). The annual mean BaP concentration averaged over these 117 stations increased by 36 % from 2007 to 2011, registering a considerable increase (25 %) from 2007 to 2008, and a small decrease (4 %) between 2010 and 2011.

For the shorter three-year period leading up to 2011, 22 countries (with 320 stations in all) reported BaP measurements, allowing an analysis of persistent exceedances over this period. About one third of these stations exceeded the EU target value in 2011, and of these stations, 80 % were persistent exceeders (that is they exceeded the target value during all three years from 2009 to 2011).

Emissions of BaP in the EU and the EEA-32 countries have increased by 11 % between 2002 and 2011. The main emission sector is the 'commercial, institutional and household fuel combustion' sector, responsible for 84 % of the total emissions of BaP in 2011 in the EU. This sector increased its emissions of BaP by 24 % between 2002 and 2011. In spite of this increase over the ten-year period, this sector appears to have made improvements more recently. After increasing by 16 % between 2006 and 2010, emissions in the sector fell by 7 % between 2010 and 2011. The emissions data for 2011 are still preliminary.

# 8.4 Exposure to C<sub>6</sub>H<sub>6</sub> and BaP pollution in Europe

While exposure to  $C_6H_6$  in Europe is limited to a few local areas — often close to traffic or industrial sources — exposure to BaP pollution is quite

significant and widespread. As Map 8.2 shows, people across Europe, and especially in central and eastern Europe, are exposed to ambient BaP concentrations above the target value of 1 ng/m<sup>3</sup> (to be met by 2013). Between 22 % and 31 % of the urban population in the EU was exposed to BaP concentrations above the target value in the period 2009 to 2011. As much as 94 % of the EU urban population was exposed to BaP concentrations above the WHO reference level over the same period. The increase in BaP emissions and concentrations in Europe over the last five years is therefore a matter of concern, as it is aggravating the exposure of the European population to BaP concentrations, especially in urban areas.

# 8.5 Responses

The Fuels Quality Directive (EU, 2003a) limits the  $C_6H_6$  content in petrol to below 1 %.

Regarding BaP, the Industrial Emissions Directive (EU, 2010) regulates emissions from a large range of industrial sources. The list of regulated compounds includes 'substances and preparations which have been proved to possess carcinogenic or mutagenic properties'. As a proven carcinogen, BaP is thus included in the list of compounds regulated by this directive.

The UNECE Protocol on Persistent Organic Pollutants (POPs) obliges parties to reduce their emissions of PAHs to below their levels in 1990 (or an alternative year between 1985 and 1995). For the incineration of municipal, hazardous and medical waste, it lays down specific limit values.

<sup>(29)</sup> With 14 % or more of valid data.

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# Annex 1 Trends in PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub> and NO<sub>2</sub> by country and station type

# Table A1.1Average trends of $PM_{10}$ annual mean concentrations and 95 % confidence limits<br/>(2 sigma) (in $\mu$ g.m<sup>-3</sup>/year) by country and by station type, period 2002–2011

		(Su	b)urban	backgro	ound		(Sub)urt	oan traffi	c		Rural ba	ckgroun	d
Country code	Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma
AT (ª)	Austria	25	11	- 0.67	0.18	17	9	- 0.58	0.24	10	6	- 0.58	0.26
BE	Belgium	11	4	- 1.04	0.47	6	4	- 0.96	0.39	4	0	- 0.73	1.02
BG	Bulgaria	4	0	0.57	1.44	2	0	0.78	4.01				
СН	Switzerland	11	5	- 0.50	0.17	7	5	- 0.55	0.21	7	5	- 0.58	0.18
CZ	Czech Republic	31	11	- 1.05	0.31	13	3	- 1.13	0.48	7	2	- 0.95	0.60
DE	Germany	112	55	- 0.56	0.08	65	42	- 0.92	0.13	46	20	- 0.44	0.11
ES	Spain	26	18	- 1.25	0.30	25	12	- 1.11	0.38	12	7	- 0.32	0.13
FI	Finland	5	2	- 0.21	0.11	12	7	- 0.33	0.10	1	0	- 0.05	0.32
FR	France	141	52	- 0.49	0.07	28	16	- 0.75	0.17	7	4	- 0.78	0.26
GR	Greece	2	2	- 2.33	1.17	1	1	- 1.61	2.14				
HU	Hungary	5	1	0.36	1.11	4	1	- 0.46	1.05				
IE	Ireland	3	3	- 0.48	0.27	2	1	- 0.90	0.83				
IS	Iceland					1	0	0.29	0.72				
IT	Italy	30	10	- 0.98	0.34	31	11	- 0.77	0.31	2	1	- 0.44	0.86
LT	Lithuania	2	0	- 0.34	0.87	4	1	- 1.07	0.90				
NL	Netherlands	4	3	- 0.69	0.41	8	7	- 0.81	0.30	14	6	- 0.54	0.21
NO	Norway					2	2	- 0.97	0.46				
PL	Poland	16	3	- 0.19	0.53	2	0	0.20	1.67	2	1	1.36	1.16
PT	Portugal	12	5	- 1.09	0.44	12	10	- 2.04	0.44	3	0	- 1.06	1.45
RS	Serbia	1	0	- 1.73	3.86								
SE	Sweden	3	2	- 0.50	0.32	4	3	- 1.34	0.58				
SI	Slovenia	3	2	- 1.17	0.70	1	0	- 1.25	1.78	1	0	- 0.27	0.78
SK	Slovakia	12	2	- 0.31	0.60	3	0	- 0.49	1.24				
UK	United Kingdom	4	1	- 0.68	0.64	2	1	- 0.82	0.34				

**Note:** (a) At three stations, parallel measurements have been made using gravimetry and beta attenuation; the time series using the non-reference method have been excluded.

Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 8 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

		(Su	b)urbaı	n backgr	ound		(Sub)urł	oan traffi	с	Rural background				
Country code	Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	
AT (ª)	Austria	25	6	- 1.26	0.39	17	4	- 0.99	0.54	10	4	- 1.06	0.52	
BE	Belgium	11	3	- 1.27	0.92	6	3	- 1.23	0.78	4	0	- 1.13	2.01	
BG	Bulgaria	4	0	2.29	3.76	2	0	4.32	8.41					
СН	Switzerland	11	4	- 0.81	0.41	7	3	- 0.87	0.47	7	3	- 0.94	0.39	
CZ	Czech Republic	31	6	- 1.44	0.71	13	2	- 1.78	1.04	7	1	- 1.23	1.31	
DE	Germany	112	45	- 0.87	0.18	65	31	- 1.42	0.26	46	16	- 0.75	0.25	
EE	Estonia													
ES	Spain	26	22	- 2.29	0.47	25	13	- 1.99	0.65	12	9	- 0.83	0.27	
FI	Finland	5	2	- 0.36	0.31	12	6	- 0.71	0.25	1	0	0.05	0.66	
FR	France	141	42	- 0.56	0.13	28	11	- 0.74	0.31	7	4	- 1.01	0.50	
GR	Greece	2	2	- 4.11	2.14	1	1	- 2.15	3.23					
HU	Hungary	5	0	0.62	2.27	4	1	- 1.14	1.65					
IE	Ireland	3	0	- 0.75	0.89	2	1	- 1.54	1.45					
IS	Iceland					1	0	- 0.15	1.79					
IT	Italy	30	9	- 1.49	0.69	31	11	- 0.99	0.69	2	0	- 0.71	2.02	
LT	Lithuania	2	0	- 0.41	1.90	4	1	- 1.75	1.50					
NL	Netherlands	4	1	- 1.13	0.86	8	4	- 1.09	0.57	14	5	- 0.78	0.45	
NO	Norway					2	2	- 2.43	0.85					
PL	Poland	16	5	0.11	1.10	2	0	1.58	3.57	2	0	3.42	3.14	
PT	Portugal	12	7	- 2.37	0.88	12	11	- 3.83	0.84	3	0	- 1.95	2.60	
RS	Serbia	1	0	- 1.41	8.44									
SE	Sweden	3	2	- 0.77	0.56	4	3	- 3.20	1.35					
SI	Slovenia	3	0	- 1.27	1.40	1	0	- 0.53	3.22	1	0	0.62	2.06	
SK	Slovakia	12	2	- 0.15	1.04	3	0	- 0.53	2.40					
UK	United Kingdom	4	2	- 0.76	0.86	2	2	- 1.50	0.84					

# Table A1.2Average trends of PM10 90.41 percentile concentrations and 95 % confidencelimits (2 sigma) (in μg.m<sup>-3</sup>/year) by country and by station type,period 2002–2011

**Note:** (a) At three stations parallel measurements have been made using gravimetry and beta attenuation; the time series using the non-reference method have been excluded.

Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 8 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

# Table A1.3Average trends of PM

		(Su	ıb)urban	backgro	und		(Sub)url	oan traffi	с	Rural background				
Country code	Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	
AT	Austria	1	0	0.10	1.60	3	1	- 0.55	1.14	1	0	0.51	1.63	
BE	Belgium	2	0	0.60	1.06	3	0	0.39	1.00	1	1	1.32	1.20	
BG	Bulgaria	1	0	- 1.91	6.13					1	0	- 0.07	1.16	
CZ	Czech Republic	16	1	0.18	0.86	6	2	- 0.18	1.28	4	0	0.93	2.22	
DE	Germany	10	0	0.05	0.71	7	0	0.36	0.80	4	0	0.30	0.82	
DK	Denmark	1	0	- 0.12	2.10	1	0	0.61	3.77					
EE	Estonia	1	0	- 0.34	3.82									
ES	Spain	2	2	- 1.57	0.87	2	1	- 0.28	1.77	9	3	- 0.44	0.60	
FI	Finland	2	1	- 0.20	0.51	2	0	- 0.19	0.53	2	0	0.22	0.84	
FR	France	15	5	1.19	0.64	2	0	0.98	1.84					
HU	Hungary	1	1	1.90	2.69	2	0	- 0.30	2.39					
IT	Italy	9	3	- 1.08	1.09	9	3	- 0.33	0.89	1	0	0.22	2.68	
LT	Lithuania					2	2	1.09	1.36					
MT	Malta					1	0	- 0.63	1.81					
NO	Norway	1	0	- 0.33	1.17	7	1	- 0.53	0.67					
PL	Poland					1	0	0.38	1.34					
РТ	Portugal	1	0	- 0.26	2.64	2	1	- 2.19	2.01	4	0	- 0.49	1.14	
SE	Sweden	2	1	- 0.29	0.92	3	1	- 1.11	1.15	1	0	- 0.45	0.88	
SK	Slovakia	2	0	0.65	2.54					1	1	2.30	2.00	
UK	United Kingdom					1	1	- 1.13	1.38	1	0	- 0.62	0.83	

**Note:** Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 5 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

# Table A1.4Average trends of $O_3$ 93.25 percentile concentrations and 95 % confidence limits<br/>(2 sigma) (in µg.m<sup>-3</sup>/year) by country and by station type, period 2002–2011

		(Su	b)urba	n backgı	round	(	(Sub)ur	ban traf	fic	Rural background					
Countr code	y Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma		
AT	Austria	44	9	- 0.85	0.35	6	2	- 1.15	0.86	44	18	- 1.05	0.31		
BE	Belgium	10	0	- 1.04	1.04	4	0	- 0.74	1.85	15	0	- 1.22	0.84		
BG	Bulgaria	4	1	0.29	2.21	2	1	2.30	6.17	1	0	- 1.57	3.73		
СН	Zwitzerland	13	2	- 0.81	0.80	6	0	- 0.36	0.97	10	2	- 0.96	0.75		
CY	Cyprus									1	0	- 1.01	3.14		
CZ	Czech Republic	22	8	- 1.29	0.50	4	1	- 1.00	1.37	17	8	- 1.87	0.67		
DE	Germany	139	26	- 0.99	0.23	24	4	- 0.81	0.52	73	19	- 1.21	0.30		
DK	Denmark	3	1	- 0.17	1.17	2	0	0.50	1.25	2	0	- 0.71	1.60		
EE	Estonia	1	0	0.14	3.14					3	2	- 1.39	0.89		
ES	Spain	50	14	- 0.64	0.38	70	17	0.27	0.40	34	7	- 0.86	0.39		
FI	Finland	4	0	- 0.37	0.83					10	3	- 1.31	0.66		
FR	France	236	29	- 0.95	0.19					51	12	- 1.40	0.42		
GR	Greece	6	3	- 2.45	1.39	4	0	- 0.59	2.03						
HU	Hungary	6	0	0.78	2.16					1	0	- 1.80	3.77		
IE	Ireland	1	0	- 0.06	1.91	1	1	- 1.82	2.03	6	0	- 0.42	1.18		
IT	Italy	55	21	- 1.33	0.57	16	1	- 0.89	1.27	17	7	- 1.27	1.18		
LT	Lithuania	1	0	- 1.58	4.88	2	0	0.16	4.12	3	0	- 0.54	0.82		
LU	Luxembourg	1	0	- 1.17	11.68					1	0	- 0.34	3.29		
LV	Latvia									1	0	1.86	2.38		
МК	former Yugoslav Republic of Macedonia					1	0	- 1.27	3.53						
NL	Netherlands	5	0	- 0.82	1.68	3	0	- 0.66	1.84	20	0	- 0.79	0.67		
NO	Norway									6	3	- 1.11	0.55		
PL	Poland	7	0	- 0.95	1.00					10	2	- 0.95	0.81		
PT	Portugal	18	2	- 0.52	0.83	5	1	0.44	1.11	4	0	- 1.70	3.17		
RO	Romania	1	1	- 7.15	8.68										
SE	Sweden	3	1	- 1.61	1.48					6	2	- 1.27	0.83		
SI	Slovenia	4	0	- 0.69	1.33	2	0	- 0.10	1.65	3	0	- 0.81	1.46		
SK	Slovakia	5	1	- 1.64	1.29					6	0	- 0.99	0.86		
UK	United Kingdom	34	4	- 0.76	0.53	2	0	0.60	1.63	16	2	- 0.78	0.68		

Note: Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 8 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

		(Sı	ıb)urba	n backgr	ound		(Sub)ur	ban traf	iic	Rural background				
Country code	Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	
AT	Austria	47	11	- 0.23	0.10	33	6	- 0.19	0.16	30	4	- 0.01	0.06	
BA	Bosnia and Herzegovina	1	1	- 1.94	1.14									
BE	Belgium	13	9	- 0.54	0.16	10	7	- 0.69	0.18	9	7	- 0.52	0.13	
BG	Bulgaria	4	0	0.26	1.21	1	0	0.36	2.95					
СН	Switzerland	13	6	- 0.36	0.15	7	4	- 0.34	0.23	10	5	- 0.24	0.11	
CZ	Czech Republic	32	15	- 0.28	0.10	14	7	- 0.70	0.30	15	3	- 0.11	0.11	
DE	Germany	150	50	- 0.29	0.05	91	39	- 0.53	0.11	66	34	- 0.22	0.05	
DK	Denmark	3	3	- 0.60	0.27	5	3	- 0.56	0.39	1	0	- 0.31	0.38	
EE	Estonia	1	1	- 0.29	0.32					3	0	- 0.02	0.05	
ES	Spain	42	20	- 0.47	0.17	48	17	- 0.50	0.24	18	3	- 0.01	0.10	
FI	Finland	4	3	- 0.47	0.22	8	3	- 0.65	0.37	2	0	- 0.08	0.16	
FR	France	229	130	- 0.50	0.04	51	24	- 0.47	0.15	21	8	- 0.21	0.09	
GR	Greece	5	2	- 1.08	0.47	5	2	- 1.87	1.16					
HU	Hungary	5	0	- 0.04	0.54	6	3	- 0.40	0.72	1	0	0.31	0.41	
IE	Ireland	1	0	0.06	0.42	3	0	0.33	1.33	2	0	0.03	0.16	
IS	Iceland	1	1	- 1.05	0.89	1	1	- 1.04	0.92					
IT	Italy	68	26	- 0.66	0.22	62	14	- 0.50	0.32	15	6	- 0.56	0.35	
LT	Lithuania					1	1	- 1.68	1.23	1	1	- 0.13	0.10	
LU	Luxembourg					1	1	0.72	0.75	1	1	0.21	0.52	
LV	Latvia	2	0	- 0.22	1.12					2	1	- 0.13	0.19	
NL	Netherlands	7	4	- 0.70	0.27	6	4	- 0.72	0.27	20	13	- 0.36	0.09	
NO	Norway	1	0	- 0.40	1.02	2	0	0.16	0.99	4	1	- 0.03	0.03	
PL	Poland	14	1	0.03	0.24	3	1	- 0.07	1.01	8	3	0.11	0.13	
PT	Portugal	18	4	0.06	0.29	12	3	0.01	0.27	3	1	0.00	0.44	
RS	Serbia	3	1	- 0.93	1.51									
SE	Sweden	3	1	- 0.32	0.27	3	1	- 0.49	0.45	3	2	- 0.07	0.05	
SI	Slovenia	3	0	0.13	0.47	1	0	- 0.16	0.80	1	0	0.22	0.46	
SK	Slovakia	4	2	- 0.11	1.30	2	0	0.32	2.06					
UK	United Kingdom	34	11	- 0.16	0.16	14	2	- 0.39	0.45	5	4	- 0.36	0.18	

# Table A1.5Average trends of NO2 annual mean concentrations and 95 % confidence limits<br/>(2 sigma) (in µg.m-3/year) by country and by station type, period 2002–2011

**Note:** Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 8 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

		(Su	b)urbaı	n backgr	ound		(Sub)ur	ban traff	ïc	Rural background				
Country code	Country	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	Ntot	Nsig	Trend	2 sigma	
All MS	All member countries	708	92	- 0.50	0.19	394	74	- 0.15	0.31	227	48	- 0.48	0.19	
AT	Austria	47	3	- 0.96	0.55	33	5	0.61	0.75	30	4	- 0.14	0.46	
BE	Belgium	13	0	0.51	1.04	10	0	0.17	1.36	8	2	- 0.69	0.62	
BG	Bulgaria	4	0	0.06	12.92	2	0	- 0.53	10.41					
СН	Switzerland	13	1	- 0.28	0.77	7	0	0.21	1.06	10	1	- 0.41	0.69	
CZ	Czech Republic	32	2	- 1.19	0.80	14	3	- 1.54	1.33	15	2	- 0.41	0.85	
DE	Germany	150	11	- 0.51	0.26	91	15	- 0.43	0.42	66	17	- 0.56	0.31	
DK	Denmark	3	0	- 1.03	1.44	5	1	1.66	1.44	1	0	- 0.17	2.67	
EE	Estonia	1	0	- 1.00	3.86					3	0	0.32	0.70	
ES	Spain	43	6	- 0.70	0.76	50	15	0.23	1.00	18	7	- 0.28	0.57	
FI	Finland	4	4	- 2.31	1.03	8	1	- 0.84	1.87	2	0	- 1.25	2.17	
FR	France	230	36	- 0.36	0.20	51	13	0.39	0.75	21	8	- 0.73	0.56	
GR	Greece	6	5	- 5.32	1.77	5	4	- 7.39	3.06					
HU	Hungary	6	0	- 3.29	4.56	6	2	- 2.19	3.92					
IE	Ireland	1	0	0.05	1.39	3	0	- 0.10	4.90	2	0	- 0.31	2.74	
IS	Iceland	1	0	- 0.25	8.62	1	0	- 5.16	7.88					
IT	Italy	68	13	- 1.49	0.95	62	9	- 0.78	1.15	15	3	- 1.27	1.34	
LT	Lithuania					1	1	- 9.10	6.88					
LU	Luxembourg					1	0	1.67	7.01	1	0	- 0.24	3.34	
LV	Latvia	3	0	1.96	3.23									
NL	Netherlands	7	0	- 0.01	1.06	7	0	- 0.96	1.65	20	1	- 0.18	0.48	
NO	Norway	1	0	1.15	9.97	2	1	7.49	10.02					
PL	Poland	14	1	0.51	1.66	3	0	- 0.01	3.71	6	2	0.32	1.43	
PT	Portugal	18	2	1.73	1.34	12	2	2.79	1.83	3	1	- 0.36	2.18	
RO	Romania													
SE	Sweden	3	0	- 0.55	1.63	3	1	1.43	2.36					
SI	Slovenia	3	0	- 1.06	3.10	1	0	- 0.97	4.16	1	0	- 0.33	1.45	
SK	Slovakia	4	2	0.74	3.49	2	0	- 1.01	5.05					
UK	United Kingdom	33	6	1.11	1.27	14	1	0.98	1.53	5	0	- 1.47	1.59	

# Table A1.6 Average trends of NO<sub>2</sub> 99.78 percentile concentrations and 95 % confidence limits (2 sigma) (in µg.m<sup>-3</sup>/year) by country and by station type, period 2002–2011

**Note:** Ntot is the number of monitoring stations used to calculate the average trend;

Nsig is the number of monitoring stations where a significant trend (p < 0.1) has been observed.

A consistent set used in the calculations: that is, station operation during at least 8 years and having for each year a data coverage of 75 % or more.

National trends are calculated by averaging the trends estimated at individual stations.

Stations located outside continental Europe (French overseas department, Canary Island, Azores, Madeira are excluded). Only countries included in the consistent set have been listed in the table.

# Annex 2 European policies and measures on air pollutant emissions

# Background

During the period addressed in this report (2002–2011), environmental policies and measures at the European level have affected the development of air pollutant emissions and the occurrence of air pollution.

The EU has developed a series of six Environment Action Programmes (EAPs) (<sup>30</sup>), starting in 1973. The 5th EAP (1993–2000) and 6th EAP (2002–2012) are most relevant for the period addressed here. The 5th EAP, under the theme of 'air pollution' concentrated on acidification and air quality, with particular attention given to:

- a strategy to ensure that critical loads of acidifying, eutrophying and photochemical air pollutants are not exceeded;
- establishing or amending air quality objectives for specific pollutants;
- developing common procedures for assessing and monitoring air quality.

The 6th EAP had as its theme 'environment and health and quality of life'. The 6th EAP's Article 7 (f) on air quality, states that: 'development and implementation of the measures in Article 5 in the transport, industry and energy sectors should be compatible with and contribute to improvement of quality of air'. Further measures envisaged by the 6th EAP include:

- improving monitoring and assessment of air quality, including the deposition of pollutants, and the providing of information to the public, including the development and use of indicators;
- a thematic strategy to strengthen a coherent and integrated policy on air pollution to cover priorities for further actions, the review and

updating where appropriate of air quality standards and national emission ceilings with a view to reaching the long-term objective of no-exceedence of critical loads and levels, and the development of better systems for gathering information, modelling and forecasting;

- adopting appropriate measures concerning ground-level O<sub>3</sub> and particulates;
- considering indoor air quality and the impacts on health, with recommendations for future measures where appropriate.

Thus, the 5th EAP and 6th EAP set the scene for developing specific policies and directives to control air pollution and improve air quality in the last two decades.

During the 1990s, the EU developed and adopted a series of directives on air quality management and assessment (<sup>31</sup>), setting, for example, limit values and target values for air quality, and methods to monitor and assess air quality. These directives have paved the way for the effective exchange of data on air quality from a network of air quality monitoring stations. This has enabled the overview of European air quality as presented in this report.

The setting of health-related air quality limit and target values specified in the air quality directives benefited from the work and studies carried out under the EU's Clean Air for Europe (CAFE) Programme (<sup>32</sup>), in cooperation with the World Health Organization (WHO), on the health effects of air pollutants.

The 6th EAP specified that the Commission should develop thematic strategies on a series of environmental problems, including air pollution. The Thematic Strategy on Air Pollution (<sup>33</sup>) was formulated as the final result of the CAFE

<sup>(&</sup>lt;sup>30</sup>) http://ec.europa.eu/environment/archives/env-act5/envirpr.htm.

<sup>(&</sup>lt;sup>31</sup>) http://ec.europa.eu/environment/air/quality/legislation/existing\_leg.htm.

 $<sup>({}^{\</sup>rm 32}) \ http://ec.europa.eu/environment/archives/cafe/general/keydocs.htm.$ 

 $<sup>({}^{33}) \</sup> http://ec.europa.eu/environment/archives/cafe/pdf/strat\_com\_en.pdf.$ 

Programme. It considers the complex interaction between pollutants, impacts, and pollutant 'receptors' (i.e. the humans and nature affected by pollution). It deals with PM in air, acidification, eutrophication and ground-level  $O_3$ . It also covers impacts on human health, nature and biodiversity, materials, and crops. The Strategy sets goals for reduced impacts on human health and the natural environment in 2020.

# **Current European policies and measures**

This report focuses primarily on the policies and measures that have affected the changes (i.e. improvements) in the air quality during the period of this overview (2002–2011).

# **Overview of directives**

# Regulations of emissions from the road traffic sector

Standards set in Europe have been used to limit emissions from motor vehicles since about 1970, through the socalled ECE R15/01-15/04 regulations for petrol-powered passenger cars. Since 1992, regulation of emissions continued under the so-called Euro 1–6 regulations for light-duty vehicles (conventional petrol-powered and diesel-powered) and the Euro I–VI regulations for heavy-duty diesel engines in trucks and buses.

# Petrol vapour recovery directives (34)

### The Stage I Petrol Vapour Recovery Directive

(1994/63/EC) aims to prevent emissions to the atmosphere of VOCs during the storage of petrol at terminals and subsequent distribution to service stations. It entered into force on 20 December 1994. The directive contains measures that terminals should employ, such as floating roofs and reflective coatings to reduce evaporative losses from storage tanks. In addition, the directive ensures that when petrol is loaded onto tankers and transported to service stations any vapours are recovered and returned to the tanker or terminal. Implementation was obligatory from 31 December 1995 for new service stations, and with a delay of three, six, or nine years depending upon the size of existing service stations, with shorter times for larger stations. This means that the directive has influenced VOC emissions since 2000.

# The Stage II Petrol Vapour Recovery Directive

(2009/126/EC) aims to ensure the recovery of petrol vapour that would otherwise be emitted to the air during the refuelling of vehicles at service stations. Member States had until 31 December 2011 to transpose the directive into national law.

The minimum level of vapour recovery of the systems employed under this directive should be 85 %. New service stations should comply with the directive from 2012, while existing stations have a longer timeline. All stations with a throughput greater than 3 000 m<sup>3</sup>/year must comply by end-2018 at the latest. The directive has not yet had an effect on VOC emissions in Europe.

# Directives on fuel quality

The **Sulphur Contents of Liquid Fuels Directive**, 1999/32/EC (<sup>35</sup>) regulates the sulphur (S) in fuel oils, establishing the following limits for sulphur:

- In heavy fuel oil, the maximum S content is 1 % by weight, to be implemented by 1 January 2003. Derogation is provided for installations that come under the IPPC Directive requiring emission reduction technology.
- In petrol the maximum S content is:
  - 0.20 % by weight, to be implemented by July 2000. Derogation is provided for certain external waterways;
  - 0.10 % by weight, to be implemented by 1 January 2008.

**Directive 2012/33/EU** (EU, 2012) amends Directive 1999/32/EC as regards the sulphur content of marine fuels, and Directive 1999/32/EC on reduction of sulphur content of certain liquid fuels (EU, 1999b) and brings EU regulations into line with current International Maritime Organization (IMO) regulations for sulphur emissions from ships under MARPOL Annex VI. Currently, the maximum sulphur content of marine fuels is limited to 3.5 %, but this latest directive will limit this figure progressively further for member states to 0.1 % in the North Sea and English Channel by 2015 and to 0.5 % in other European sea areas by 2020.

The **Fuels Quality Directive** (2003/17/EC) (<sup>36</sup>), amends the previous Fuels Directive (98/70/EC). It

<sup>(34)</sup> http://ec.europa.eu/environment/air/transport/petrol.htm.

<sup>(35)</sup> http://ec.europa.eu/environment/air/transport/sulphur.htm.

<sup>(&</sup>lt;sup>36</sup>) http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2003:076:0010:0019:EN:PDF.

regulates the contents of S, Pb and  $C_6H_6$  in motor fuels, as well as other fuel quality parameters. The limits set in this directive are:

- for S, 10 mg/kg for petrol and diesel from 1 January 2009;
- for Pb in petrol, 0.005 g/l (in practice lead-free gasoline);
- for  $C_6H_6$  in petrol, 1 % v/v;
- for PAH in diesel fuel, 11 % m/m.

Since 1 January 2002, all petrol sold in the EU is unleaded. Between 1 January 2005 and 1 January 2009 the limit on the S content of petrol and diesel was 50 mg/kg.

The **Fuel Quality Directive** (2009/30/EC) (<sup>37</sup>), amends Directive 98/70/EC as regards the specification of petrol, diesel and gas-oil and introducing a mechanism to monitor and reduce greenhouse gas emissions; it amends Directive 1999/32/EC as regards the specification of fuel used by inland waterway vessels; and repeals Directive 93/12/EEC.

It regulates the reduction of sulphur content of inland waterway fuel in one step to 10 ppm by 1 January 2011. Further, it amends a number of elements relating to the petrol and diesel specifications as well as introducing a requirement on fuel suppliers to reduce the greenhouse gas intensity of energy supplied for road transport (Low Carbon Fuel Standard). In addition the directive establishes sustainability criteria that must be met by biofuels if they are to count towards the greenhouse gas intensity reduction obligation.

The directive regulates vapour pressure waiver for petrol containing bioethanol (up to 10 % v/v). It regulates the contents of S, Pb, hydrocarbons including  $C_6H_6$ , Oxygenates (VOCs), PAHs, in motor fuels, as well as other fuel quality parameters. The limits set in this directive are:

- for S, 10 mg/kg for petrol and diesel from 1 January 2009;
- for Pb in petrol, 0.005 g/l (in practice lead-free gasoline);

- for  $C_6H_6$  in petrol, 1 % v/v;
- for PAH in diesel fuel, 8 % m/m.

The **Energy Taxation Directive** (2003/96/EC; EU, 2003b) is not a directive on fuel quality, but establishes a financial mechanism to promote the use of less polluting fuels and increases incentives to use energy more efficiently. It establishes minimum taxes on motor fuels, heating fuels and electricity, depending on the energy content of the product and the amount of  $CO_2$  it emits.

# **Regulation of industrial emissions**

**Directive 2010/75/EU** aims to recast the seven pre-existing directives related to industrial emissions into a single clear and coherent legislative instrument. These seven pre-existing are: the IPPC Directive, the LCP Directive, the Waste Incineration Directive, the Solvents Emissions Directive and three directives on titanium dioxide. The Commission proposed that minimum emission limit values in certain industrial sectors should be tightened — particularly for large combustion plants where progress to reduce pollution is considered insufficient.

The aforementioned legal instruments are briefly described below:

The Solvents Directive (38) (1999/13/EC) regulates the use of solvents and sets limits on emissions of VOCs due to the use of organic solvents in certain activities and installations. The expressed objective of the directive is to limit the formation of  $O_3$  in air. The list of activities and uses of solvents regulated by the directive includes adhesive coating and other coating activities; dry cleaning; manufacturing of varnishes, adhesives, inks, and pharmaceuticals; printing, surface cleaning; vehicle refinishing; and wood impregnation amongst others. The directive sets emission limit values for waste gases or per volume of product, and requires a solvent management scheme for each activity. The general compliance date for existing installations is 31 October 2007, while new installations shall comply when commencing operations.

The **Waste Incineration Directive** (2000/76/EC) (<sup>39</sup>) repealed former directives on the incineration

<sup>(&</sup>lt;sup>37</sup>) http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2009:140:0088:0113:EN:PDF.

<sup>(&</sup>lt;sup>38</sup>) http://ec.europa.eu/environment/air/pollutants/stationary/solvents.htm.

<sup>(&</sup>lt;sup>39</sup>) http://ec.europa.eu/environment/air/pollutants/stationary/wid.htm.

of hazardous waste (Directive 94/67/EC) and household waste (Directives 89/369/EEC and 89/429/ EEC) and replaced them with a single text. The aim of the Waste Incineration Directive is to prevent or reduce as far as possible negative effects on the environment caused by the incineration and coincineration of waste. In particular, it should reduce pollution caused by emissions into the air, soil, surface water and groundwater, and thus lessen the risks that these pose to human health. This is to be achieved through the application of operational conditions, technical requirements, and emission limit values for incineration and co-incineration plants within the EU.

The Waste Incineration Directive sets emission limit values and monitoring requirements for air pollutants such as dust,  $NO_{x'}$  SO2, hydrogen chloride (HCl), hydrogen fluoride (HF), heavy metals, dioxins and furans.

Most types of waste incineration plants fall within the scope of the Waste Incineration Directive, with some exceptions, such as those treating only biomass (e.g. vegetable waste from agriculture and forestry).

The Waste Incineration Directive makes a distinction between:

- incineration plants, which are dedicated to the thermal treatment of waste and may or may not recover heat generated by combustion;
- co-incineration plants, such as cement or lime kilns, steel plants or power plants whose main purpose is energy generation or the production of material products and in which waste is used as a fuel or is thermally treated for the purpose of disposal.

The deadline to bring existing plants into compliance was 28 December 2005.

The Large Combustion Plants (LCP) Directive (<sup>40</sup>) (2001/80/EC) regulates emissions of acidifying pollutants, PM and  $O_3$  precursors (e.g.  $SO_2$ ,  $NO_x$  and PM ('dust')) from large combustion plants for heat end energy production. The directive sets emission limit values (ELVs) (<sup>41</sup>) for  $SO_2$ ,  $NO_x$  and dust (Total Suspended Particles, TSP), which vary according to the age of the plant, the fuel used, and the plant capacity (see EEA Technical report No 8/2010).

Regarding deadlines for implementation, the LCP Directive contains the following provisions:

- plants licensed after 27 November 2002 have to comply with the (stricter) emission limit values for SO<sub>2</sub>, NO<sub>x</sub> and dust fixed in part B of Annexes III to VII to the directive;
- plants licensed after 1 July 1987 and before 27 November 2002, have to comply with the (less strict) emission limit values fixed in part A of Annexes III to VII to the LCP Directive;
- significant emission reductions are required from 'existing plants' (licensed before 1 July 1987) to be achieved by 1 January 2008, either:
  - by individual compliance with the emission limit values established for new plants referred to in bullet 2 above (1987–2002); or
  - through a national emission reduction plan (NERP) that achieves overall reductions calculated using the emission limit values.

The **Paints Directive** (<sup>42</sup>) (2004/42/CE) establishes limit values for the maximum VOC contents of decorative paints and vehicle-refinishing products in order to limit the emissions of VOC. The Paints Directive also amends the Solvents Directive concerning vehicle-refinishing products. The Paints Directive has two phases for the implementation of stricter limits on VOC contents in products: Phase I to be implemented by 1 January 2007, and Phase II by 1 January 2010.

The original Integrated Pollution Prevention and Control (IPPC) Directive was adopted on 24 September 1996, and has since been amended four times. It regulates almost all industrial plants, including those engaged in energy production, metals production, mineral industries, chemical industries, waste management and other sectors. The air pollutants addressed by this directive are SO<sub>2</sub>, NO<sub>x</sub>, CO, VOC, metals, dust, asbestos, Cl, F, As, cyanides and other carcinogenic and mutagenic compounds, as well as some specific dioxins. New installations and existing installations that are subject to 'substantial changes' have been required to meet the requirements of the IPPC Directive since 30 October 1999. Other existing installations had to be brought into compliance by 30 October 2007. This

<sup>(&</sup>lt;sup>40</sup>) http://ec.europa.eu/environment/air/pollutants/stationary/lcp.htm.

<sup>(&</sup>lt;sup>41</sup>) Under the LCP Directive, Member States have certain opt-out provisions (Article 4(4)) and may define and implement national emission reduction plans (NERPs) (Article 4(6)).

<sup>(42)</sup> http://ec.europa.eu/environment/air/pollutants/paints\_legis.htm.

was the key deadline for full implementation of the directive.

In the directive, the concept of 'best available techniques' (BAT) plays a central role. In this context:

- 'techniques' include both the technology used and the way in which the installation is designed, built, maintained, operated and decommissioned;
- 'available 'techniques are those developed on a scale that allows application in the relevant industrial sector, under economically and technically viable conditions, taking into consideration the costs and advantages, whether or not the techniques are used or produced inside the Member State in question, and as long as they are reasonably accessible to the operator;
- 'best' means most effective in achieving a high general level of protection of the environment as a whole.

Operators of relevant industrial installations must apply BAT to prevent and control pollution. Authorities are also obliged to set up a system of issuing integrated permits that will lead to the implementation of BAT in new and existing plants.

Conclusions as to what are considered to be BAT at the EU level for the activities covered by the directive are given in BAT reference documents (BREFs), which are developed under the coordination of the Commission (EU IPPC Bureau, Joint Research Centre), through an exchange of information by expert groups comprised of representatives of the EU Member States, industry, NGOs and other stakeholders.

The definition of industrial combustion facilities used in this study goes beyond that in the LCP BREF. The LCP BREF covers, in general, combustion installations with a rated thermal input exceeding 50 MW. This includes the power generation industry and those industries where 'conventional' (commercially available and specified) fuels are used and where the combustion units are not covered within another sector BREF. In this context, industrial combustion facilities comprise power plants, refineries and those in the manufacturing sector, regardless of capacity.

# Directive on national total emissions

# The National Emissions Ceiling Directive

(NEC Directive) (<sup>43</sup>), adopted on 23 October 2001, sets upper limits for each Member State for the total emissions in 2010 of the four main pollutants responsible for acidification, eutrophication and ground-level  $O_3$  pollution (SO<sub>2</sub>, NO<sub>X</sub>, VOC and NH<sub>3</sub>). The directive leaves it largely to the Member States to decide which measures — in addition to Community legislation for specific source categories — to take in order to comply.

### Directive concerning the protection of waters against pollution caused by nitrates from agricultural sources

Council Directive 91/676/EEC concerning the protection of waters against pollution caused by nitrates from agricultural sources has the aim of providing for all waters a general level of protection against pollution. It's implementation, and in particular the implementation of agricultural practices that limit fertiliser application and prevent nitrate losses, leads to a decrease of agricultural emissions of nitrogen compounds not only to water, but also to air. Member States shall:

- establish a code or codes of good agricultural practice, to be implemented by farmers on a voluntary basis;
- set up where necessary a programme, including the provision of training and information for farmers, promoting the application of the code(s) of good agricultural practice.

Moreover there is a requirement for the establishment of fertiliser plans on a farm-by-farm basis and the keeping of records on fertiliser use.

# LRTAP Convention

The long-range transport of air pollution is an important factor affecting ecosystems and the human population. The United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (LRTAP) aims at reducing and preventing air pollution. In addition to the EU legislation listed above, the LRTAP Convention has a number of legally

<sup>(43)</sup> http://ec.europa.eu/environment/air/pollutants/ceilings.htm.

binding protocols, covering specific categories of air pollutants. The Convention was ratified by the European Community in 1982. Article 2 of the Convention states that 'the Contracting Parties, taking due account of the facts and problems involved, are determined to protect man and his environment against air pollution and shall endeavour to limit and, as far as possible, gradually reduce and prevent air pollution, including long-range transboundary air pollution'.

The Convention has set up a process for negotiating concrete measures to control specific pollutants

through legally binding protocols. Since 1984, eight protocols have come into force. The most recent, the 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, came into force on 17 May 2005.

In 2013, the parties to the Convention were requested to report 2011 emissions data for  $NO_{x'}$ NMVOC,  $SO_{x'}$  NH<sub>3</sub>, CO, heavy metals, persistent organic pollutants and PM, and also associated activity data. The information was copied by Member States to the EEA Eionet Reportnet Central Data Repository.

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