Evaluation of current limit and target values as set in the EU Air Quality Directive

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Front page picture: *Some examples of daily life's limit values*

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1. Introduction

According to the directive 2008/50/EC on ambient air quality and cleaner air for Europe (AQ Directive, (EC 2008)) the Commission shall review in 2013 the provisions related to $PM_{2.5}$ and, as appropriate, other pollutants. This report aims to inform this review by assessing current information on limit values, target values, information and alert thresholds. After a short discussion of the application area of the AQ Directive (Chapter 2) for each of the components listed in the directive an assessment of the air quality in the European Union is presented. In Chapter 3 for each pollutant the following issues will be discussed:

- the current situation of air quality in Europe; and if more than one standard is defined, which is the more stringent one;
- short comparison of EU values with other international air quality standards.

The results are discussed in Chapter 4 and a number of points which could be considered in the review of the Air Quality Directive are presented.
Information on the current (2009) ambient air quality situation is mainly obtained from AirBase (see Mol et al, 2011) and the air quality questionnaire (Jimmink et al, 2011). In general no reference will be given to these reports; from the context it will be clear which of the two sources (or another source) is used.

2. Application areas of the Directive

The directive on ambient air quality and cleaner air for Europe (Air Quality Directive, (EC 2008) defines and establishes *"… objectives for ambient air quality designed to avoid, prevent or reduce harmful effects on human health and the environment as a whole".* Whereas the first objective (the protection of human health) seems clear, the second objective (protection of the environment as a whole) is more difficult to interpret. Reading the directive it becomes clear that "*the environment as a whole*" must be interpreted as *vegetation and natural ecosystems*, see the points (10) and (12) in the preamble. The definition of *critical levels* and the various annexes where limit or target values are set and where the location of monitoring stations is further specified in the Annexes. Animal life seems not to be included in the protection target.

The spatial extent where the limit or target values have to be attained is also not necessarily clear. In the Directive "ambient air" is defined as *"..outdoor air in the troposphere*"; in particular from the sections where the location of sampling points is described it is clear that only the lowest part, at ground-level or in the boundary layer is meant here. Limiting the harmful ozone levels in the middle and upper troposphere (ozone acts here as a greenhouse gas and it contributes to the concentrations at ground-level) is certainly not one of the objectives.

With respect to the protection of human health, Annex III, A2 of the Directive lists restrictions as to where the limit values apply:

> *"Compliance with the limit values directed at the protection of human health shall not be assessed at the following locations:*
> *(a) any locations situated within areas where members of the public do not have access and there is no fixed habitation;*
> *(b) in accordance with Article 2(1), on factory premises or at industrial installations to which all relevant provisions concerning health and safety at work apply;*
> *(c) on the carriageway of roads; and on the central reservations of roads except where there is normally pedestrian access to the central reservation."*

As Brunekreef and Maynard (2008) indicate, the first two restrictions are reasonable but the exclusion of on-road exposure (with some exceptions for pedestrians but not for cyclists) will make it more difficult to address car and bus commuting exposures in the future.

In the third daughter directive on ozone (EC 2002) the year 2020 has been set as benchmark to review the long-term objectives for protection of human health and vegetation. In the Air Quality Directive similar objectives have been set but without mentioning a date by which the objectives should be met.

With respect to the protection of vegetation and ecosystem, the preamble (point 10) indicates that such assessment *"should focus on places away from built-up areas".* In Annex III, B2, on the macroscale siting of sampling points, a further description in more quantitative terms is given. The restrictions are such that in more densely populated regions no such areas exist. For example, whereas there are more then 150 Natura-2000 areas in the Netherlands widely spread over the country, according to the annual reporting questionnaire on air quality assessments (EC 2004) the critical levels for protection of vegetation and natural ecosystems is only applicable in a small area in the northern part of the Netherlands. Most likely the situation in the Netherlands is not unique. Large parts of Europe's ecosystems might therefore not be covered by the directive.

The term *critical level* has been newly introduced in the Air Quality Directive. In the first Daughter Directive the term limit value has been used both for the protection of human health as for ecosystems/vegetation. Whereas in the definition of *limit value* the requirement ´*to be attained within a given period and not to be exceeded once attained'* this requirement is not included in the definition of *critical level*.

3. Reviewing the current levels

In this section the current air quality (data from 2009) is compared with the values or thresholds as set in the Air Quality Directive. This analysis is based on information extracted from AirBase (Mol et al. 2011) and on the information reported under the air quality questionnaire (decision 2004/461/EC, (EC 2004; Jimmink et al (2011)).

The air quality in relation to the limit or target values is presented in so-called distance-to-target graphs. In these graphs the (relative) frequency distribution of concentrations measured at each station type is shown. In AirBase each monitoring station has been classified according to the type of area surrounding the station and on pollutant source and the distance to these sources. The stations are grouped into four types:

- **rural background** stations: stations located in rural areas (non-urbanised areas with a low building density) so that its pollution level is not influenced significantly by any single source but rather by the integrated contribution from all sources upwind of the station;
- **urban background** stations: station located in urban (continuously built-up urban area) or suburban (largely built-up urban area); its pollution level is not influenced significantly by any single source but rather by the integrated contribution from all urban sources upwind of the station. The levels at urban background stations are assumed to be representative of the exposure of the general urban population;
- **traffic** stations: stations located such that its pollution level is determined predominantly by the emissions from nearby traffic (roads, motorways, highways). Traffic stations may be located in urban, suburban or rural areas;
- **other stations:** this group contains mainly **industrial** stations and a small number of stations with an unknown classification. An industrial station is a station located such that its pollution level is influenced predominantly by emissions from nearby single industry or industrial complexes. Industry source is here defined in its broader meaning including sources like power generation, incinerators and waste treatment plants. Industrial stations may be located in urban, suburban and rural areas.

Information on station type forms is part of the data flow under the Exchange of Information Decision. Station classification is generally provided by the local network managers. Analyses made by the Topic Centre (see e.g. Horalek et al 2009) suggest that not in all cases similar classification criteria have been used. This may hamper the comparability between countries. Mistakes in staion classification will introduce uncertainties in particular when estimating the exposure of humans or ecosystems.

In the distance-to-target graphs the air quality is compared to the EU standards. It should be noted that a number of Member States have set more stringent standards at the national or regional level either by setting lower threshold values or by having a lower number of allowable exceedances. In Annex A a summary of these national standards is presented.

In case the limit (LV) or target value (TV) is expressed as a maximum allowable number of exceedances (N_{exc}) of a specified threshold value, the $(N_{exc}+1)^{th}$ highest value has been evaluated: there is compliance with the limit value if this concentration is below the threshold level.

Alternatively, the distance-to-target could be expressed in term of the number of hours/days above the limit or target value. The number of exceedances and the $(N_{exc}+1)^{th}$ highest value are of course related. However, we prefer the $(N_{exc}+1)^{th}$ highest value as this presentation provides more information for the lower end of the distribution.

In the graphs shown below data from all operational stations has been included. In this report an *operational station* is defined as a station having a data coverage of at least 75%, that is, the number of valid measurements is at least 75% of the annual maximum (8760 hours, 365 days per year). For benzene, the heavy metals and benzo(a)pyrene a lower criterion (50, 14 and 14%, respectively) has been applied, see Mol et al (2011) for further discussion. For one pollutant the number may slightly differ for the different indicators as for a few stations only daily values have been reported.

The spatial distribution of air quality over Europe will not be discussed here. For concentration maps showing the most relevant air quality indicators, the reader is referred to de Smet et al (2009) and references cited therein.

Unless otherwise stated the information on air quality given in the next chapters refer to the EU-27 Member States only.

3.1 Sulphur dioxide, SO_2

For sulphur dioxide (SO_2) two limit values for protection of human health and two critical levels for the protection of vegetation are defined. The levels are in force since January 2005. In addition an alert threshold has been defined (Table 3.1).

Table 3.1 Reference values for sulphur dioxide as given in the Air Quality Directive.

objective	period	Limit or threshold value	Number of allowed exceedances
human health	one hour	350 µg/m^3	24 hours per year
human health	one day	125 µg/m^3	3 days per year
alert[(a)]	one hour	500 µg/m^3	
vegetation	calendar year	20 µg/m^3	-
vegetation	Winter (1 Oct - 31 March)	20 µg/m^3	-

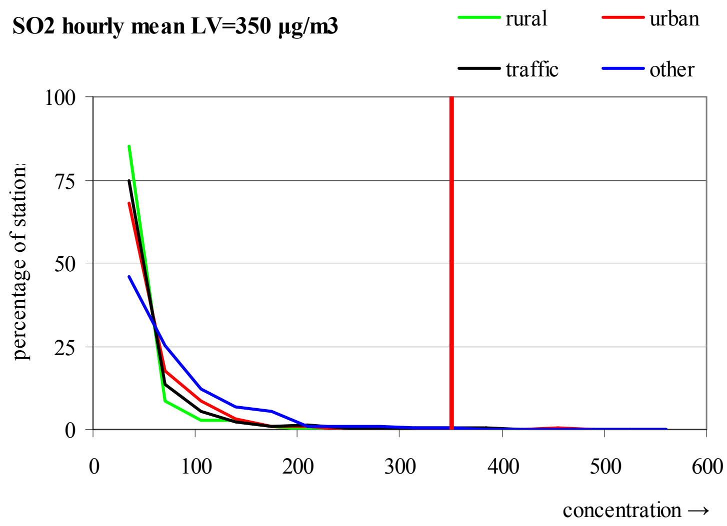
(a) to be measured over three consecutive hours at locations representative of air quality over at least 100 km^2 or an entire zone or agglomeration, whichever is the smaller.

The distance-to-target graphs[1] for the hourly and daily SO_2 limit values using 2009 data are given in Figure 3.1. Exceedances of the limit value are observed at 6 out of 1710 stations (0.4%) for the hourly limit value) and at 5 out of 1759 stations (0.3%) for the daily limit value. The year 2009 is an exception, in general the daily limit value is more frequently exceeded than the hourly limit value. No exceedance of daily and hourly limit value is observed at rural stations. The reporting under the AQ Directive also indicates a slightly more frequent violation of the daily limit value: 4 respectively 3 out of 805 zones are not in compliance. In the air quality questionnaire local industry and power generation have been listed as major reasons for exceedance; 25-30% of the hourly exceedances have been attributed to accidental industrial emissions. The SO_2 concentrations show a steady

[1] More detailed information on the averaged concentrations and number of monitoring stations for each of the station types (*rural, urban traffic* and *other*) is for each of the distance-to-target graphs given in Annex B.

decrease over the period 1999-2009. At all station types (rural, urban, traffic) the concentrations are more than halved in the last 11 years

During the last 11 years (1999-2009) the alert value of 500 µg/m^3 has been exceeded on the average less than 44 times per million monitoring hours[2]; 2003 was the year with the highest frequency: 77 exceedances per million observations. During the years 2007-2009 (4.5 million hourly observations) 701 episodes of one or more hours with in total 1365 exceedances of the alert value have been counted. In 81 cases the episode lasted for 3 or more consecutive hours and an alert situation occurred. Regular and accidental emissions have been quoted as main reason for the exceedances. Most frequently alert situation were reported in Bulgaria (31 times) , France (18 times) and Spain (16 times).



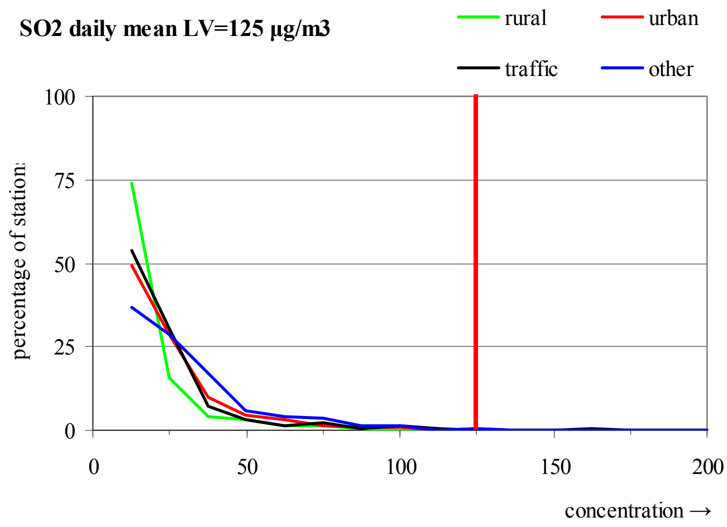


Figure 3.1 Distance to target graphs for hourly (expressed as the 25th highest hourly mean) and daily (expressed as 4th highest daily mean) limit value of SO_2, EU27 Member States, period 2009.

The limit values for the protection of vegetation are only analysed for rural background stations. Both during the calendar year 2009 as well as during the winter 2008/2009 no exceedances were observed. Less than 0.5% of the zones report non-attainment with the critical levels for vegetation. It should be stressed, however, that, according to the information given by the Member States, the SO_2

[2] If we assume that a monitoring station has a data coverage of 90%, one million monitoring hours is realised when 125 stations are operational during a full calendar year.

critical level for vegetation is only applicable in about 40% of the zones, covering about 60% of the EU-27 area.

Based on theoretical considerations (increased emissions and worse dispersion conditions during the winter compared to the full year) the critical level for the winter period will be the most stringent one. The very small number of exceedances precludes such a conclusion based on observations.

A comparison of the EU air quality standards set for the protection of human health with other international standards is given in Table 3.2. The most stringent EU daily limit value is weaker than the corresponding standards in the other countries included in the comparison with exception of the standards at federal level in the USA. Five Member States have set national limit values. It is difficult to evaluate which of the Member States has the most strict limit because limit value, number of allowed exceedance and averaging period differ. It is estimated that Hungary has introduced the most strict values although those set by Austria and Sweden might not be much lower.

Table 3.2 International Air Quality standards set for sulphur dioxide for the protection of human health.

SO2 µg/m^3	hourly	daily	annual
EU	350	125	-
WHO (1)	-	20	-
Switzerland (2)		100	30
USA (3)	-	365	80
California (3)	655	105	-
Japan (4)	266	105	
China, residential (5)	150	50	20
China, commercial (5)	500	150	60
India (6)		80 (80)	50 (20)
EU-MS (7)	250	50	

(1) Air Quality Guidelines as recommended by WHO, source: (WHO 1987, 2000, 2006,)
(2) source: Federal Office of the Environment, http://www.bafu.admin.ch/luft/00632/00634/index.html?lang=en
(3) source: US-EPA, http://www.epa.gov/air/criteria.html
California Air Resource Board, http://www.arb.ca.gov/research/aaqs/caaqs/caaqs.htm
(4) Japan: Ministry of the Environment, http://www.env.go.jp/en/air/aq/aq.html
(5) China has different standards for residential, commercial and industrial areas. The air quality standards for residential and commercial areas are listed here. Source: Clean Air Initiative for Asian Cities (2010) Air Quality in Asia: status and trends, 2010 edition. www.cleanairinitiative.org
(6) India: limit values set for industrial, residential, and rural area; for ecologically sensitive areas more stringent values have been set for SO_2 and NO_2 (given in parentheses).The daily, 8-hourly and hourly values shall be complied with 98% of the time; 2% of the time exceedance is allowed but not on two consecutive days. Source: Gazette of India, November 16, 2009
(7) A number of Member States have set national limit values (see Annex A); for comparison the (estimated) most strict value(s) set by one of the MS are given. For SO_2 the given limit values have been set by Hungary and may be exceeded 24 (hourly LV) and 3 (daily LV) timer each year.

Whereas the World Health Organisation (WHO) recommends only a guideline for 24-hour averages, most countries have set standards for shorter (15 minutes, half-hourly or hourly) and longer (annual) periods. The WHO Air Quality Guideline (AQG) level is substantially lower than the EU limit value. If a similar methodology as in the EEA Core Set Indicator on urban air quality (CSI004, see de Leeuw and Fiala, 2009) is applied, it is estimated that 70-85 % of the urban population within the EU-27 is exposed to levels above the WHO AQG. Based on geographical information (coordinates of the monitoring stations and city boundaries) the (sub)urban background stations located in one of the cities included in the Urban Audit dataset (Eurostat, 2011) are identified. Using the concentrations measured at these (sub)urban background stations the averaged concentration is calculated for each city. Together with the information on population numbers extracted from the Urban Audit database, the Core Set Indicator CSI004 estimates the frequency distribution of the urban population exposure.

3.2 Nitrogen dioxide, NO_2, and oxides of nitrogen, NO_x

For nitrogen dioxide (NO_2) two limit values for protection of human health are defined. In addition an alert threshold has been set. For the protection of vegetation a critical level is set for the annual mean of nitrogen oxides (NO_x), defined as the sum of nitrogen monoxide and nitrogen dioxide expressed in units of mass concentration of NO_2.

Table 3.3 Reference values for nitrogen dioxide and oxides of nitrogen as given in the Air Quality Directive.

objective	period	Limit or threshold value	Number of allowed exceedances
human health	one hour	200 µg/m^3	18 hours per year
human health	calendar year	40 µg/m^3	
alert[(a)]	one hour	400 µg/m^3	
vegetation[(b)]	calendar year	30 µg/m^3	

(a) to be measured over three consecutive hours at locations representative of air quality over at least 100 km^2 or an entire zone or agglomeration, whichever is the smaller.
(b) as oxides of nitrogen, expressed as µg NO_2/m^3

The distance-to-target graphs for the hourly and annual NO_2 limit values are given in Figure 3.2 (2009 data). It becomes clear from this graph that (i) the annual limit value is more stringent than the hourly limit value[3] and (ii) the exceedance of the annual limit value is most frequently observed at traffic stations.
Averaged over all available stations the NO_2 concentrations show a downward tendency. However, looking at the individual stations, the decrease is at a number of stations statistically not significant. Slightly increasing concentrations (mostly statistically not significant) can be observed at a number of stations.

The AQ questionnaire reports show similar results: the annual limit value is the most stringent one, with an exceedance of 40 µg/m^3 level in 29% of the 819 zones; in 24% of the zones the

[3] From a statistical comparison between the short-term (19th highest hourly value) and long-term limit value (annual mean value) it is estimated that an hourly limit value of about 140 µg/m^3 would result in a more equal probability of exceedance for both averaging periods (see also Chapter 4, Figure 4.1). On the other hand, it is estimated that an hourly limit value of 200 µg/m^3 and an annual level of 55-58 µg/m^3 are more or less equivalent.

concentrations exceed the limit value plus margin of tolerance (42 µg/m^3). The hourly limit value is exceeded in 4% of the zones. Exceedance of the annual limit value is a known and persistent problem. In the air quality questionnaire *"local traffic"* has been listed as main reason for exceedance. The increasing number of diesel cars resulting in increasing direct NO_2 emissions is seen as one of the main causes. In Guerreiro et al (2011) a more in-depth study will be presented.

The alert threshold is only occasionally exceeded. Since 1999 on average 5.1 exceedances have been counted per million monitoring hours (with a minimum of 1.1 counts per million in 2002 and a maximum of 18.8 counts per million in 2000). In the period 2007-2009, 175 episodes were observed; most of them (160) lasted for 1 or 2 hours. An alert situation (exceedance during 3 or more consecutive hours) occurred in 15 cases; the maximum length of an episode was 8 hours. Alert situations were most frequently observed in Bulgaria and Spain (both 5 times) and Italy (two times).

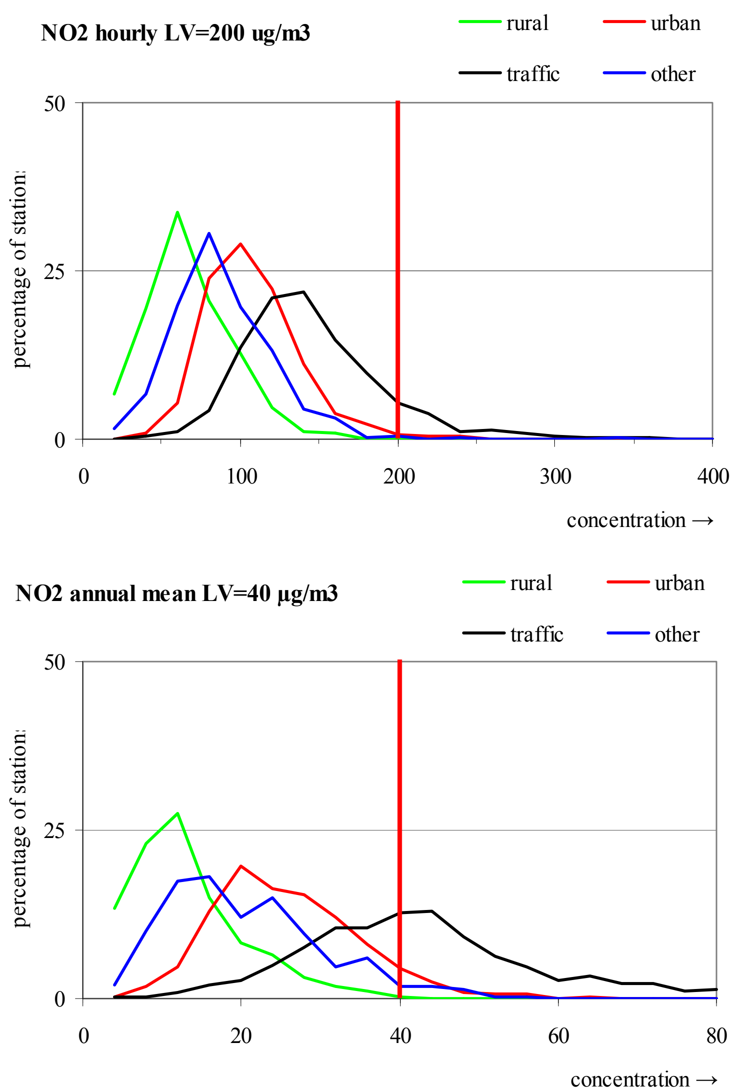


Figure 3.2 Distance to target graphs for hourly (expressed as the 19th highest hourly mean) and annual limit value of NO_2, EU-27 Member States, period 2009.

The NO_x critical level for the protection of vegetation is not widely exceeded at rural background stations. As Figure 3.2 shows there is just a limited number of rural stations where the NO_2 concentrations exceeds 30 µg/m^3; in rural areas the NO_2/NO_x ratio approaches one and no frequent exceedance of the NO_x critical level is observed. According to the information submitted by the

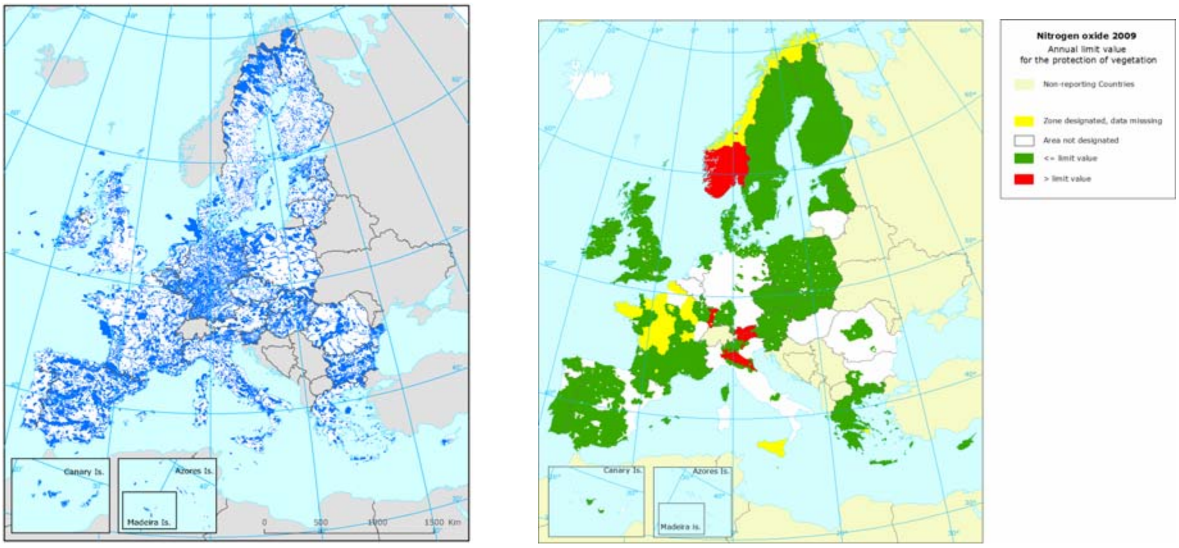


Figure 3.2a. Distribution of Natura 2000 sites across the 27 EU Member States (left, source: EEA, 2011b) and the assessment of the NO_x limit value set for the protection of ecosystems in air quality management zones (right, source: Jimmink et al, 2011).

Member States, the NO_x critical level is applicable in 364 air quality management zones covering about 70% of the area in the EU-27. 28 zones are reported to be above the critical level; information is missing for 41 zones. A visual inspection of the maps showing the zones designated for this limit value and ecosystem maps like Natura-2000 shows that this limit value has possibly not been considered for all relevant ecosystem types (Figure 3.2a).

Table 3.4 shows a comparison of the EU air quality standards set for the protection of human health with other international standards. The (most stringent) EU annual limit value equals the standards set by China and India but is stronger than the standard in the USA and the one set by the Californian Air Resources Board. In 2010 the US-EPA revised the NO_2 air quality standards. A short term (hourly) standard has been added; it will protect against adverse health effects associated with short-term exposure to NO_2, including respiratory effects which may result in increased hospital admissions. The US-EPA concluded that evidence suggesting an association between long-term exposure to NO_2 and adverse health effects is too limited to suggest any change in the annual standard; the existing annual standard of 53 ppb (100 $\mu g/m^3$) is retained. The Swiss NO_2 standard is more stringent than the EU standard. Within the EU the most strict limit values have been set by Sweden (hourly, daily mean) and Austria (annual mean). The EU limit values agree with the AQGs recommended by the WHO.

In population studies and some indoor studies NO_2 has been associated with adverse health effects even when concentrations are below 40 $\mu g/m^3$ (WHO 2006). This might support a lowering of the annual NO_2 guideline value; however, as NO_2 is an important constituent of combustion generated air pollution and is highly correlated with other primary and secondary combustion products, it is unclear whether the health effects found in epidemiological studies are attributable to NO_2 alone or to the mixture of combustion products. The WHO (WHO 2006) judges that current scientific literature has not accumulated sufficient evidence to change the 2000 guideline (WHO 2000)[4].

[4] Note that uncertainties concerning the NO_2 guideline were also expressed in the 2000 report. Here the WHO states: *"…. it is proposed that a long-term guideline for nitrogen dioxide be established. Selecting a well supported value based on the studies reviewed has not been possible, but it has been noted that a prior review conducted for the Environmental Health Criteria document on nitrogen oxides recommended an annual value of 40 $\mu g/m^3$. In the absence of support for an alternative value, this figure is recognized as an air quality guideline."*

Table 3.4 International Air Quality standards set for nitrogen dioxide for the protection of human health (see notes in Table 3.2 for further explanation)

NO_2 µg/m^3	hourly	daily	annual
EU	200		40
WHO	200		40
Switzerland		80 (a)	30
USA	190 (b)		100
California	339		57
Japan		76-115	
China, residential	120	80	40
China, commercial	120	80	40
India		80 (80)	40 (30)
EU-MS (c)	90	60	30

(a) may be exceeded only once per year
(b) the 3-year average of the 98th percentile of the daily maximum 1-hour average may not exceed 100 ppb (190 µg/m3)
(c) The most strict limit value for hourly and daily mean has been set by Sweden; the most strict annual limit value is set by Austria and has to be met by 1-1-2015.

3.3 Particulate matter, PM_{10}, $PM_{2.5}$

For particulate matter (PM_{10}, $PM_{2.5}$) various limit and target values for protection of human health are defined (Table 3.5). For PM_{10} a daily average concentration may exceed 50 µg/m^3 on not more than 35 days per year; the yearly averaged concentration should be below 40 µg/m^3. The distance to target graphs (Figure 3.3) show that violation of the daily limit value is much more frequent than of the annual limit value. A statistical analysis of monitoring data indicates that the daily limit value corresponds to an annual mean concentration of about 28-33 µg/m^3 depending on location (see e.g. Buijsman et al 2005; Stedman et al 2007).

In the annual reporting questionnaire (2009) the daily and annual limit value has been exceeded in 34% and 10% of the 803 zones, respectively. Of the EU-27 population 43% (16%) lives in zones where the daily (annual) limit value is exceeded in 2009. Note that these population numbers refer to the total population in a zone. The fraction actually exposed will be lower as exceedances may occur only in certain areas within a zone. Applying a similar methodology as used for the EEA CSI004 Urban Air Quality Indicator, 9-14% of the urban population is exposed to an annual mean of 40 µg/m^3 or more; 18-40% is exposed to concentrations above the daily limit value.

Table 3.5 Reference values for PM_{10} and $PM_{2.5}$ as given in the AQ Directive.

Size fraction	period	value	comments
PM_{10}, limit value	one day	50 µg/m^3	Not to be exceeded on more than35 days per year
PM_{10}, limit value	calendar year	40 µg/m^3	
$PM_{2.5}$, target value	calendar year	25 µg/m^3	To be met by 1-1-2010
$PM_{2.5}$, limit value	calendar year	25 µg/m^3	To be met by 1-1-2015
$PM_{2.5}$, limit value (a)	calendar year	20 µg/m^3	To be met by 1-1-2020
$PM_{2.5}$, exposure concentration obligation		20 µg/m^3	2015
$PM_{2.5}$ exposure reduction target	0-20% reduction in exposure (depending on the average exposure indicator in the reference year) to be realised in 2020		

(a) indicative limit value (Stage 2) to be reviewed by the Commission in 2013.

In the AQ Directive a $PM_{2.5}$ reference level of 25 µg/m^3 is set, initially as target value to be met by 2010 and as limit value to be met by 2015. Information from $PM_{2.5}$ monitoring stationsis still limited when compared to the available PM_{10} information (in the EU27 the number of operational stations with a data coverage of 75% or more was 2400 (PM_{10}) and 570 ($PM_{2.5}$) in 2009. The distance to target graph in Figure 3.4 shows that at 3%, 9% and 7% of the rural, urban background and traffic stations the target value is exceeded. Exceedance is also observed at 6% of the industrial sites. The limit value plus Margin of Tolerance (for 2009, 29 µg/m^3) is exceeded on 3% of the stations.

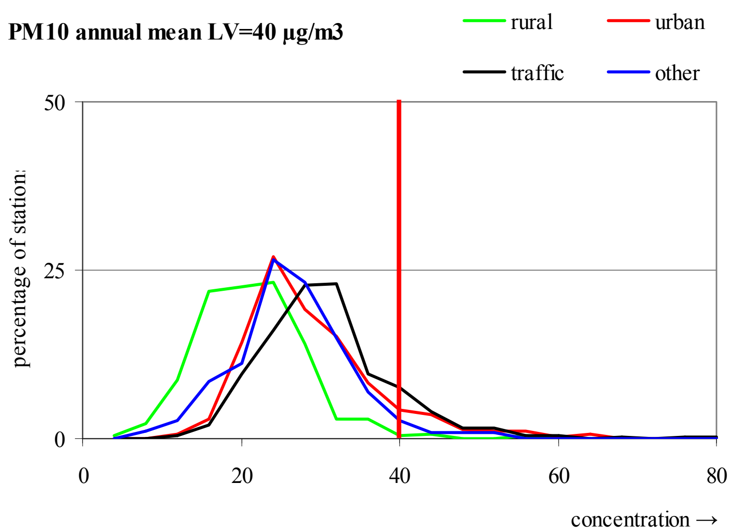


Figure 3.3a Distance to target graphs for annual limit value of PM_{10}, EU27 Member States, period 2009.

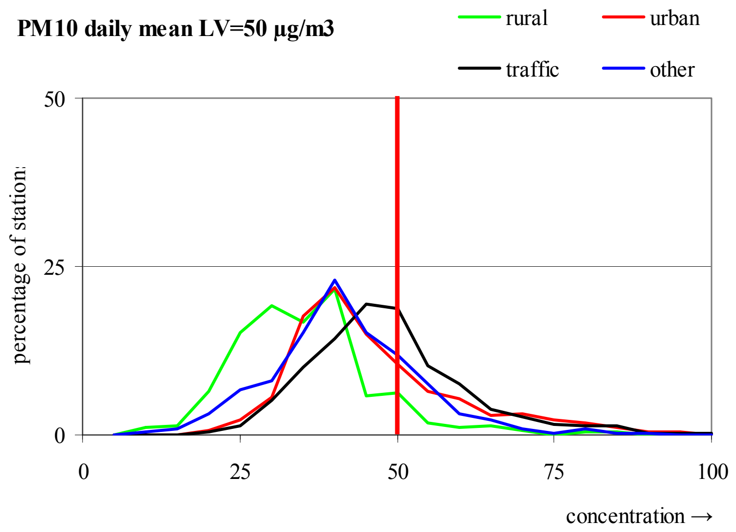


Figure 3.3b Distance to target graphs for daily (expressed as 36th highest daily mean) limit value of PM_{10}, EU27 Member States, period 2009.

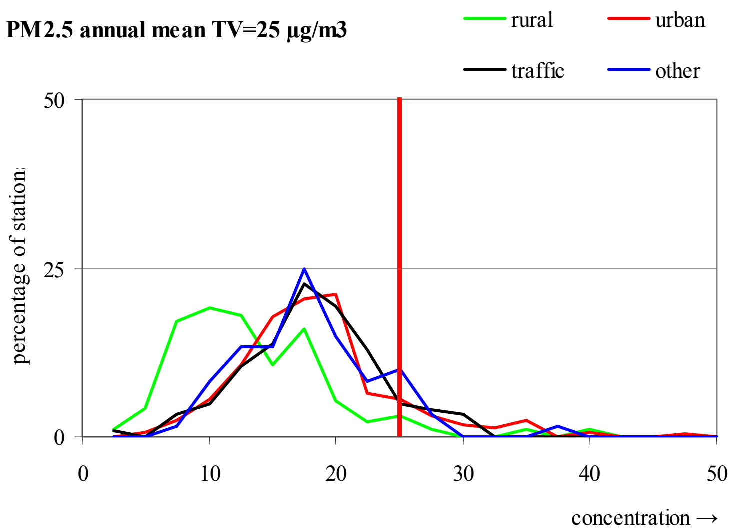


Figure 3.4 Distance to target graphs for annual limit value of $PM_{2.5}$, EU27 Member States, period 2009.

The Air Quality Directive introduced an additional $PM_{2.5}$ objective targeting the exposure of the population to fine particles. This *exposure concentration obligation* is set at the national level and is based on the average exposure indicator (AEI). The AEI is determined as a three-year running annual mean concentration measured at a selected set of stations in urban background locations throughout the territory of a Member State. The AEI reflects the $PM_{2.5}$-exposure of the general (urban) population. Member States provide information on stations and measurement configurations selected for determination of the AEI in the annual air quality reporting questionnaire (EU, 2004). However, in the questionnaires reporting for 2009, only 12 Member States provided information on the selected stations. As a first estimate of the *exposure concentration obligation* we have calculated here the three-year running mean (2007-2009) as the mean of the annual averaged concentration over all operational (sub)urban background stations in each individual year. Please note that the approximated levels (*Figure 3.5*) are not based on a stable set of stations. For a number of countries results are based on data for two or one year only. Figure 3.5 indicates that in 7 Member States current urban concentrations are above 20 µg/m^3, the level legally binding in 2015.

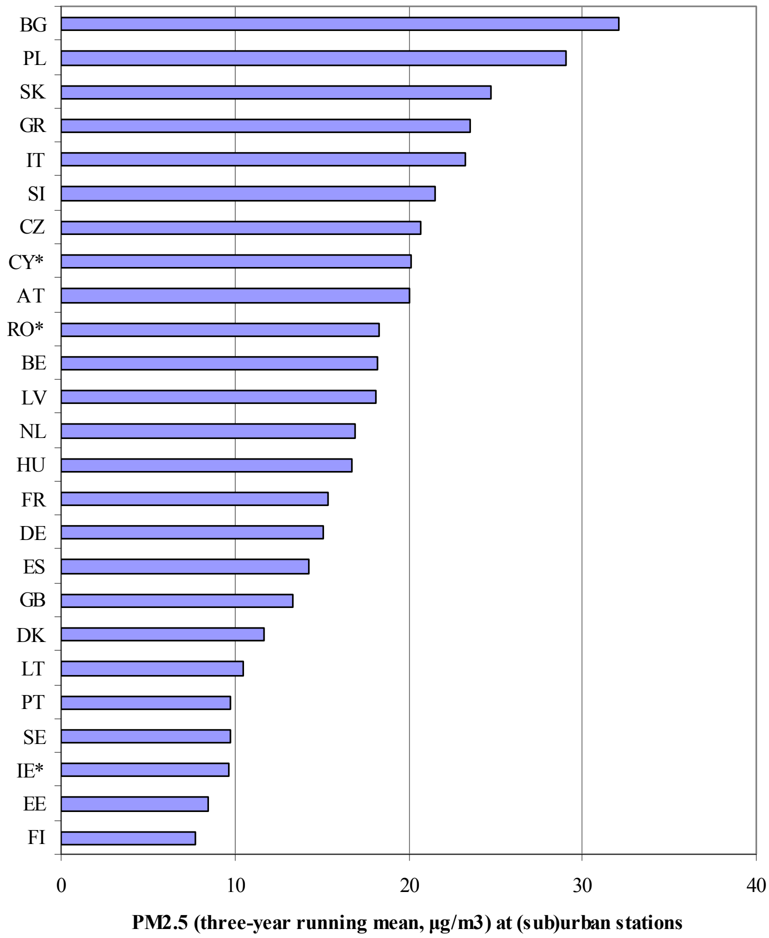


Figure 3.5 Average exposure indicator. Three-year running mean (2007-2009) over all operational (sub)urban background stations. Results for countries marked with an asterisk are based on 2009 data only. .

The second objective is an exposure reduction target; the reduction percentage depends on the AEI at the reference date (a three-year-period ending in 2010 or 2011).

A comparison of the European air quality standards set for the protection of human health with other international standards is given in Table 3.6. Except China and Switzerland, the other countries have defined standards both for PM_{10} as well as for $PM_{2.5}$. Several countries, including the EU, set a daily limit value of 50 µg/m^3 for PM_{10}; the EU limit value is, however, weaker as 35 exceedances per year are allowed. The USA put emphasis on regulation of $PM_{2.5}$ having, together with California, the lowest limit value for the annual mean. At the national level in the EU Austria and Estonia and at the regional level Scotland have set stricter limit values for PM_{10} by allowing less exceedances of a daily mean of 50 µg/m^3. Estonia and Scotland have set the annual limit value at 20 and 18 µg/m^3, respectively. In Finland the $PM_{2.5}$ limit value of 25 µg/m^3 has to be met by 1-1-2010.

The WHO has recommended guidelines for annual mean levels; the given daily limit values are based on the relation between 24-hour and annual PM levels. The EU limit value for PM_{10} falls between the WHO interim target 2 and 3[5], the $PM_{2.5}$ target vales equals the WHO interim target 2. During the period 2006-2009, 80-90% of the urban population in EU-27 was exposed to annual mean PM_{10} levels above the WHO AQG.

Table 3.6 International Air Quality standards for particulate matter set for the protection of human health (see notes in Table 3.2 for further explanation)

PM_{10}/$PM_{2.5}$ (a)	hourly	daily	annual
EU	- / -	50 / -	40 / 25
WHO	- / -	50 / 25	20 / 10
Switzerland	- / -	50 (b) / -	20 / -
USA	- / -	150 / 35	- / 15
California	- / -	50 / -	20 / 12
Japan	200 / -	100 / -	- / -
China residential	- / -	50 / -	40 / -
China commercial	- / -	150 / -	100 / -
India	- / -	100/60	60/40
EU-MS (c)	- / -	50 / -	18 / 12

(a) PM_{10} and $PM_{2.5}$ standards have been separated by a slash.
(b) May be exceeded only once per year.
(c) The most strict limit value for PM_{10} daily mean has been set by Estonia and Scotland: exceedance of the level of 50 µg/m^3 is allowed for 7 days; the most strict PM_{10} and $PM_{2.5}$ annual limit values are set by Scotland.

[5] In addition to a guideline the WHO (2006) has set one or more interim targets indicating different levels of health impact. Air quality guideline and interim targets for annual mean PM_{10} and $PM_{2.5}$ concentrations are given below.

Annual mean level	PM_{10} (µg/m^3)	$PM_{2.5}$ (µg/m^3)	Basis for the selected level
WHO interim target 1 (IT-1)	70	35	These levels are estimated to be associated with about 15% higher long-term mortality than at AQG levels.
WHO interim target 2 (IT-2)	50	25	In addition to other health benefits, these levels lower risk of premature mortality by approximately 6% (2–11%) compared to IT-1.
WHO interim target 3 (IT-3)	30	15	In addition to other health benefits, these levels reduce mortality risk by approximately another 6% (2–11%) compared to IT-2 levels.
WHO air quality guidelines (AQG)	20	10	These are the lowest levels at which total, cardiopulmonary and lung cancer mortality have been shown to increase with more than 95% confidence in response to $PM_{2.5}$ in the ACS study *(323)*. The use of the $PM_{2.5}$ guideline is preferred.

3.4 Lead, Pb

For lead (Pb) in PM_{10} an annual limit value of 0.5 µg/m^3 is set in the AQ Directive for the protection of human health.

In 2009 exceedances of the limit value were observed at two stations (in Bulgaria and Romania; in the EU-27, 573 monitoring stations were operational in 2009). The violation of the limit value seems to be only a local issue. No monitoring data has been received from Greece, Hungary, and Portugal. According to the reporting questionnaire for the Air Quality Directive (EU, 2004b) the concentrations in Greece and Hungary are below the lower assessment threshold (LAT) and other methods than monitoring could be used for assessment. However, Portugal did not provide information on the assessment regime but declared that concentrations are below the limit value. According to the questionnaire there is one station in Portugal measuring lead; data from this station has not been delivered to AirBase.
In the questionnaire (2009 data) lead concentrations have been assessed in 667 zones. In agreement with the monitoring data in two zones (located in Bulgaria and Romania) levels above the limit value have been reported.

A comparison of the European air quality standards set for the protection of human health with other international standards is given in Table 3.7. The EU limit value of 0.5 µg/m^3 corresponds with the WHO guideline. Similar or weaker standards are set in China and India; Japan and Canada have no national lead standard. For the USA the national lead standard of 0.15 µg/m^3 as rolling 3-months average is more strict than the EU standard. Within the EU, Hungary is the only Member State having set a national limit value for lead, see Table 3.7.

Table 3.7 International Air Quality standards for lead set for the protection of human health (see notes in Table 3.2 for further explanation).

Lead µg/m^3	3-monthly	daily	annual
EU			0.5
WHO			0.5
Switzerland			0.5 (b)
USA	0.15 / 1.5 (a)		
Japan			-
China			1 / 1
India		1.0	0.50
EU-MS (c)			0.3

(a) USA: averaged for rolling 3-months, calendar quarter, respectively. California has a 30 day average standard of 1.5 µg/m^3;
(b) in addition a standard of 100 µg/(m^2.day) has been set as deposition flux;
(c) Hungary is the only Member State having a national limit value for lead.

3.5 Benzene, C_6H_6

For benzene an annual limit value of 5 µg/m^3 is set in the AQ Directive for the protection of human health. By 1 January 2010 this limit value had to be met.

At nine stations (out of 561 stations with valid data) an exceedance was observed in 2009 in the EU Member States. None of the exceedances was observed at a rural station (see Figure 3.6); seven exceedances were recorded at traffic stations. For six of the stations reporting an exceedance in 2009, time series are available since 2005. At five stations (located in Czech Republic, Greece, Italy, and Poland) the limit values were exceeded each year since 2005.The Member States reported exceedances in 10 zones on a total of 727 zones in 2009. Clearly, benzene is a more local traffic-related problem.

A comparison of the European air quality standards set for the protection of human health with other international standards is given in Table 3.8. The USA, California, and China have not set a national standard for benzene; the Japanese standard is 40% lower than the EU limit value. Within the EU Hungary has set an additional national limit value for benzene for hourly mean concentrations (10 µg/m^3). In Scotland and Northern Ireland the annual limit has been reduced to 3.25 µg/m^3.

The WHO has not recommended a guideline value. Benzene is carcinogenic to humans and therefore no safe level of exposure can be recommended. The WHO (2000) estimated the excess lifetime risk of leukemia at an air concentration of 1 µg/m^3 to be 6×10^{-6}. The acceptance that the upper limit of the additional lifetime risk should be less than 1×10^{-5} (~1×10^{-6} is generally accepted as an acceptable risk level, (RIVM 2003)), would suggest an ambient air quality standard for benzene of 1.7 µg/m^3. The EU limit value of airborne benzene associates with an excess lifetime risk of 3×10^{-5}.

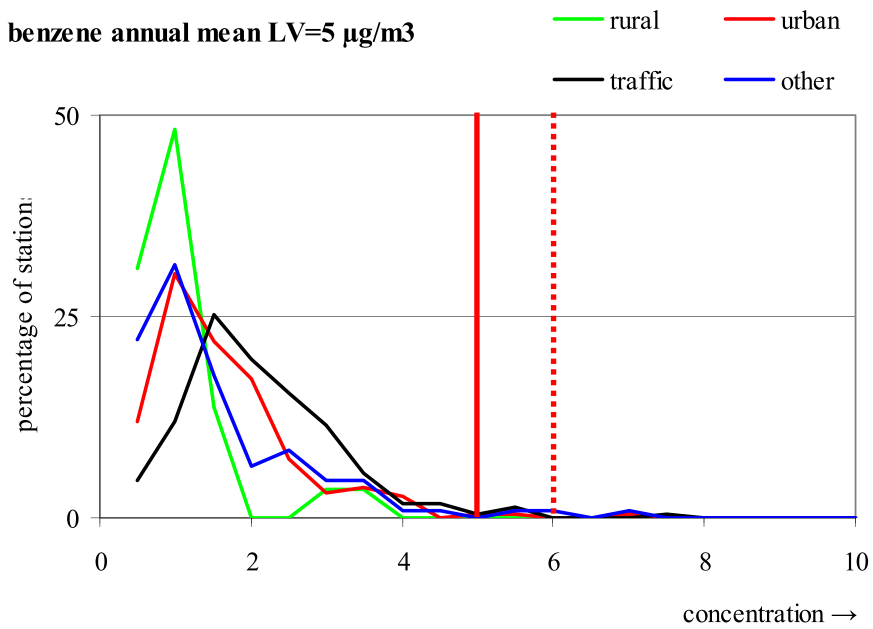


Figure 3.6 Distance to target graph annual limit value of benzene, EU27 Member States, period 2009.

Table 3.8 International Air Quality standards set for the protection of human health (see notes in Table 3.2 for further explanation).

Benzene µg/m^3	hourly	annual
EU	-	5
WHO	-	(1.7) (a)
USA / California	-	-
Japan	-	3
China	-	-
India	-	5
EU-MS (b)	10	3.25

(a) value corresponding to an additional lifetime risk of 10^{-5}, see text;
(b) most strict limit values set in individual Member States.

3.6 Carbon monoxide, CO

For carbon monoxide (CO) a limit value for the maximum daily 8-hour mean of 10 mg/m^3 is set in the AQ Directive for the protection of human health. This limit value is in force since 1 January 2005.
In 009 at five stations (from a total of 1138) the LV has been exceeded; most of the exceedances are observed at traffic stations. Exceedances are observed in Italy (4 stations) and Bulgaria. The AQ questionnaire lists one zone (from a total of 755) which is in exceedance. It can be concluded that non-attainment of CO limit values is mainly a local problem. During the last eleven years (1999-2009) concentrations at traffic and urban background stations showed a decreasing trend.

A comparison of the European air quality standard set for the protection of human health with other international standards is given in Table 3.9. The EU has set a limit value for the maximum daily 8-hour mean only; several other countries, and also the WHO, have set both hourly as well as 8-hourly standards. The EU limit value is in agreement or stricter than the standards in other OECD regions; China and India have a more stringent standard. In the EU both Hungary and Poland have reduced the 8-hourly limit value to 5 mg/m^3. Additionally Hungary introduced an hourly and annual limit value in the national legislation. The WHO hourly guideline is equivalent to the 8-hour guideline. A survey of the data in AirBase showed 10 (in 2007; all located in Italy) and four (in 2008; all located in Italy) stations where the maximum hourly concentration exceeds the WHO-AQG of 30 mg/m^3. No exceedance of the WHO-AQG was reported in 2009. Affected are mainly urban traffic and – to a lesser extent – urban industrial stations.

Table 3.9 International Air Quality standards set for carbon monoxide for the protection of human health (see notes in Table 3.2 for further explanation).

CO mg/m3	hourly	8 hourly	daily
EU	-	10	-
WHO	30	10	-
Switzerland	-	-	8 (a)
USA	40	10	-
California	23	10	-
Japan	-	23	11
China residential	10	-	4
China commercial	10	-	4
India	4	2	-
EU-MS (b)	10	5	3 (annual limit value)

(a) not to be exceeded more than once per year;
(b) national limit values set by Hungary.

3.7 Ozone, O_3

For ozone (O_3) target values as well as information and alert thresholds for the protection of human health have been defined. Further, target values for the protection of vegetation have been set. The target values should have been met by 1 January 2010. Additionally, the AQ Directive defines long-term objectives (LTO) both for the protection of human health and for protection of vegetation. While in the old ozone directive (EC 2002) the LTO should be met by 2020, in the new AQ Directive no date is specified.

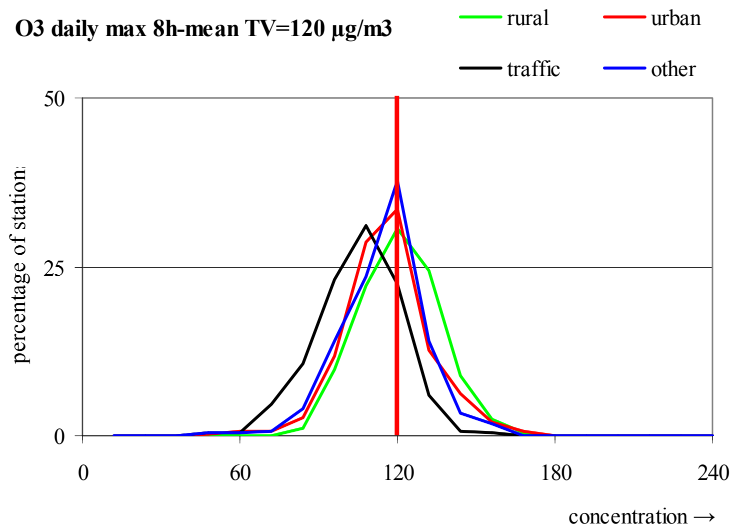
In 2009 the health related target was exceeded at 36% of the 482 operational rural background stations. In urban areas were the observed concentrations at about 22% of the in total 1001 stations above the target value.
Although the AOT40 value[6] averaged over all operational rural background stations in EU-27 is below the target value, the target value was exceeded at about a quarter of the rural stations (see Figure 3.7).
In contrast to the other air pollutants the ozone levels are generally the highest at rural locations. This is due to the depletion of ozone through the reaction with nitrogen monoxide, a pollutant especially emitted by traffic.

Table 3.10 Air quality standards for ozone as given in the AQ Directive.

objective	period	Target or threshold value	Number of allowed exceedances
human health	Maximum daily 8-hour mean	120 µg/m^3	25 days per year averaged over three years
vegetation	AOT40 accumulated over May-July	18 000 (µg/m^3).h averaged over five years	
LTO health	Maximum daily 8-hour mean	120 µg/m^3	
LTO vegetation	AOT40 accumulated over May-July	6 000 (µg/m^3).h averaged over five years	
information	one hour	180 µg/m^3	
alert[(a)]	one hour	240 µg/m^3	

(a) to be measured over three consecutive hours.

[6] The vegetation-related target value has been defined in terms of a total ozone dose during the growing season. The AOT40 (**A**ccumulated **O**zone over a **T**hreshold of **40** ppb) is the sum of the difference between hourly concentrations greater than 80 µg/m^3 (40 ppb) and 80 µg/m^3 over a certain period using only hourly data measured between 8:00 and 20:00 Central European Time. For crops generally the period 1 May to 31 July is selected; for forest protection the period is 1 April to 30 September.



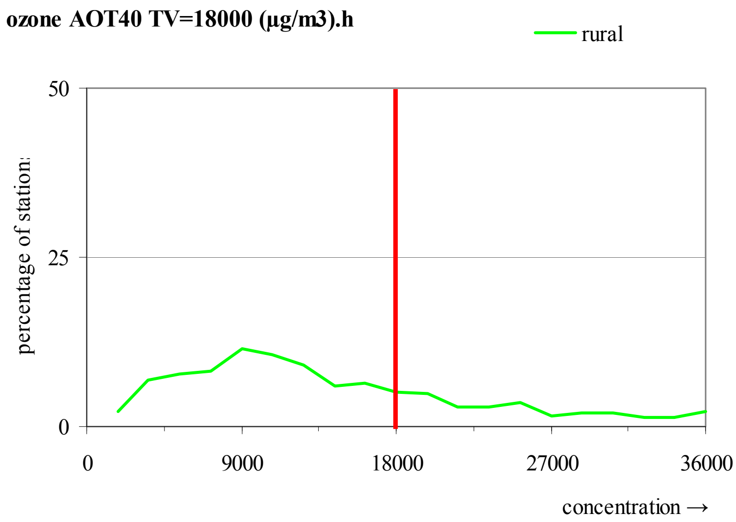


Figure 3.7 Distance to target graphs for target value set for the protection of human health (top, expressed as 26th highest daily mean) and the target value set for the protection of vegetation (bottom, expressed as AOT40; only rural background stations are shown) of ozone, EU-27 Member States, period 2009.

A comparison of the European air quality target values set for the protection of human health with other international standards is given in Table 3.11. Standards for hourly as well as for 8-hourly concentrations have been set. The AirBase data shows a high correlation between the maximum hourly and maximum 8-hourly averaged concentrations with an averaged ratio of the two concentrations of 0.85 – 0.90. The WHO AQG of 100 µg/m^3 (8 hourly mean) corresponds to the Japanese and Chinese hourly standards of 120 µg/m^3. Given the ratio between hourly and 8-hourly concentration the Californian 8 hourly standard will be slightly more strict than the hourly standard. Thestandard set by India and the WHO AQG are the most strict.

In addition to an AQG, the WHO recommends an interim target (IT-1) of 160 µg/m^3 (8-hour). For all countries the national standards are weaker than the WHO AQG but more strict than this interim target. The EU target value of 120 µg/m^3 which might be exceeded during 25 days per year, corresponds to a maximum 8-hour concentration of about 160 and corresponds to the WHO interim target value. According to the analysis presented here, none of the Member States have set limit or target values other than those set by the EU.

Table 3.11. International Air Quality standards set for the protection of human health (see notes in Table 3.2 for further explanation).

Ozone µg/m^3	hourly	8 hour
EU	-	120
WHO	-	100
Switzerland	120 (a)	-
USA	-	150
California	180	140
Japan	120	-
China residential	120	-
China commercial	160	-
India	180	100
EU-MS (b)		100

(a) may be exceeded only once per year
(b) national air quality objective set by the United Kingdom, not to be exceeded more than 10 times a year.

3.8 4th Daughter Directive pollutants: arsenic, cadmium, nickel and benzo(a)pyrene

The target values for the protection of human health set for these pollutants are given in Table 3.12 together with a comparison with international air quality standards.
Concentrations of the pollutants covered by the 4th Daughter Directive (DD), arsenic, cadmium, nickel and benzo(a)pyrene, have been reviewed by Barrett et al. (2008). The submitted monitoring data over the past years are in line with the findings of this study. Compared to 2008, the number of reporting stations increased from 120 to 170 stations depending on the pollutant. However, monitoring data of these pollutants is reported for a relatively small number of stations compared to the other regulated air pollutants. Reason for this is that the concentrations of these pollutants are frequently below the lower assessment threshold and other techniques than monitoring (e.g. modelling) can be used for assessing the air quality.

Table 3.12 Target values for the protection of human health (4th Daughter Directive) as set by the European Union compared with international air quality standards (see notes in Table 3.2 for further explanation).

pollutant	EU Target value (a)	WHO AQG	Other countries (b)
arsenic	6 ng/m^3	6.6 ng/m^3 (c)	India: 6 ng/m^3 Hungary: 10 ng/m^3 as limit value
cadmium	5 ng/m^3	5 ng/m^3 (d)	Switzerland: 1.5 ng/m^3 Flanders: 30 ng/m^3 as limit value Hungary: 5 ng/m^3 as limit value
nickel	20 ng/m^3	25 ng/m^3 (c)	India: 20 ng/m^3 Hungary: 25 ng/m^3 as limit value
benzo(a)pyrene	1 ng/m^3	0.12 ng/m^3 (c)	United Kingdom: 0.25 ng/m^3 Hungary: 0.12 ng/m^3 India: 1 ng/m^3 China: 10 ng/m^3

(a) annual mean; target value enters into force 1-1-2012;
(b) in addition to standards for concentrations in ambient air, several countries have set standards for deposition, see Annex A;
(c) arsenic, nickel compounds and B(a)P are human carcinogens and no safe level for inhalation exposure can be recommended. The given values correspond to an excess lifetime risk level of 10^{-5};
(d) to prevent any further increase of cadmium in agricultural soils likely to increase the dietary intake of future generations, a guideline of 5 ng/m^3 has been established.

Results for the reporting year 2009 can be summarized as follows:
Arsenic: At about 90% of the stations a concentration below the lower assessment threshold has been reported. However, at 11 (from the 534 operational stations) the observed concentration is above the target value set for 2012. A relatively large number of exceedance is observed in Belgium (6 stations of which 4 are located close to one industrial plant in Hoboken, near Anvers (VMM, 2009)). The remaining five exceedances are seen in Czech Republic (3 stations), Germany, and Bulgaria, both at industrial (3 stations) and urban sites (2 stations). According to the AQ questionnaire in 8 zones (from a total of 623) the target value was exceeded in 2009.

Cadmium: Air concentrations are in excess of the target value at 4% of the 581 operational stations. Exceedances are observed in two countries (Belgium, 21 stations; Bulgaria, 3 stations) mainly at industrial and (sub)urban stations but also at two rural background station in Belgium suggesting a more widespread dispersion of high Cd levels. At the majority of the other stations concentrations are below the lower assessment threshold; the AQ-questionnaire indicates concentrations below the LAT in more than two-third of the zones. According to the AQ questionnaire in 4 zones (from a total of 632) the target value was exceeded in 2009.

Nickel: Exceedances of the target value are seen at 8 of the 561 operational stations; these stations are located in the eastern part of Belgium, the German Ruhr area, in France and south Norway. Most of the exceedances are related to industry. According to the AQ questionnaire in 8 zones (from a total of 632) the target value was exceeded in 2009.

In the reports under decision 2004/461/EC (the "questionnaire") "*local industry including power production*" had been listed as mean reason of exceedance for the three pollutants listed above.

Benzo(a)pyrene: the target value was exceeded at 37% of the430 operational monitoring points in 2009. Affected are mainly at (sub)urban background stations and, to a lesser extent, at the other stations types (rural, traffic and industrial stations). There is a concentration of impact in central and eastern Europe (NE-SW corridor from the Baltic States, over Poland, Czech Republic, Slovakia, Hungary and Austria, the Po Valley) although exceedances are also observed in the UK (Midlands, Northern Ireland), the German Ruhr area and Bulgaria.
The wide-spread observed exceedances are in agreement with the reports under the Air Quality Directive, where, in addition to the MS mentioned above, Finland and Greece report exceedance of the target value in one or more zones. The assessment for Greece is based on modelling. From the information provided by the AQ questionnaire it is not clear which method has been used to asses the situation in Finland. According to the AQ questionnaire in 134 zones (from a total of 611) the target value was exceeded in 2009. The total population living in zones reporting an exceedance and potentially exposed to B(a)P concentrations above the target value is estimated as 94 million persons.
Long time series for B(a)P are available for a limited number of stations; 45 stations have reported data for at least four consecutive years since 2005. The time series averaged per country show that the exceedances of the target value are persistent.

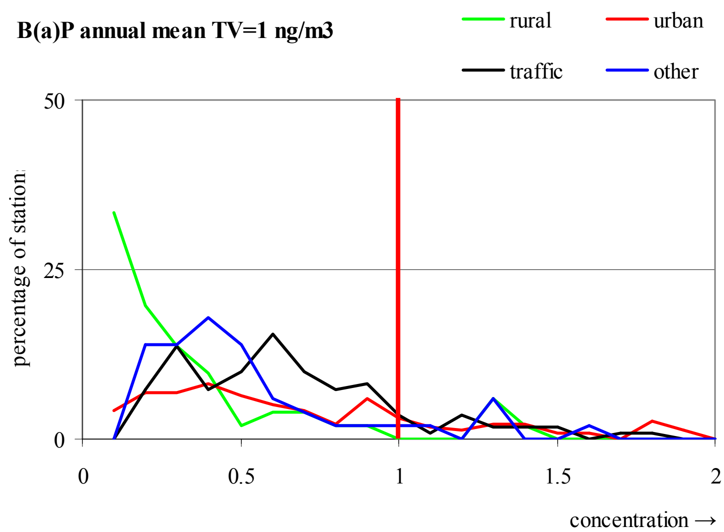


Figure 3.8 Distance to target graphs for target value of B(a)P, EU-27 Member States, period 2009

4. Discussion

Comparability of different threshold values set for one pollutant

For PM_{10}, NO_2 and SO_2 more than one limit value has been set for the protection of human health. In Figure 4.1 data in relation to these limit values is pair-wise compared using AirBase data for the period 2006-2008[7]. With 3 years of data and, depending on the pollutant 1800-2800 operational monitoring stations (including stations outside the EU), 5000 to 8000 data points are shown in each graph.

As discussed above, the number of exceedances of both SO_2 limit values set for the protection of human health is small. Over the period 2006-2008 in 1.6% of the cases an exceedance of the daily limit value is observed; the hourly limit value is slightly less exceeded: in 1.1% of the cases. In general it is the daily limit value that is the most limiting; only in 13 occasions (0.2%) the hourly limit value is exceeded while no exceedance of the daily limit value is observed. Thus, one may conclude that assessment of exceedances of the SO_2 daily limit value will be sufficient in the future.

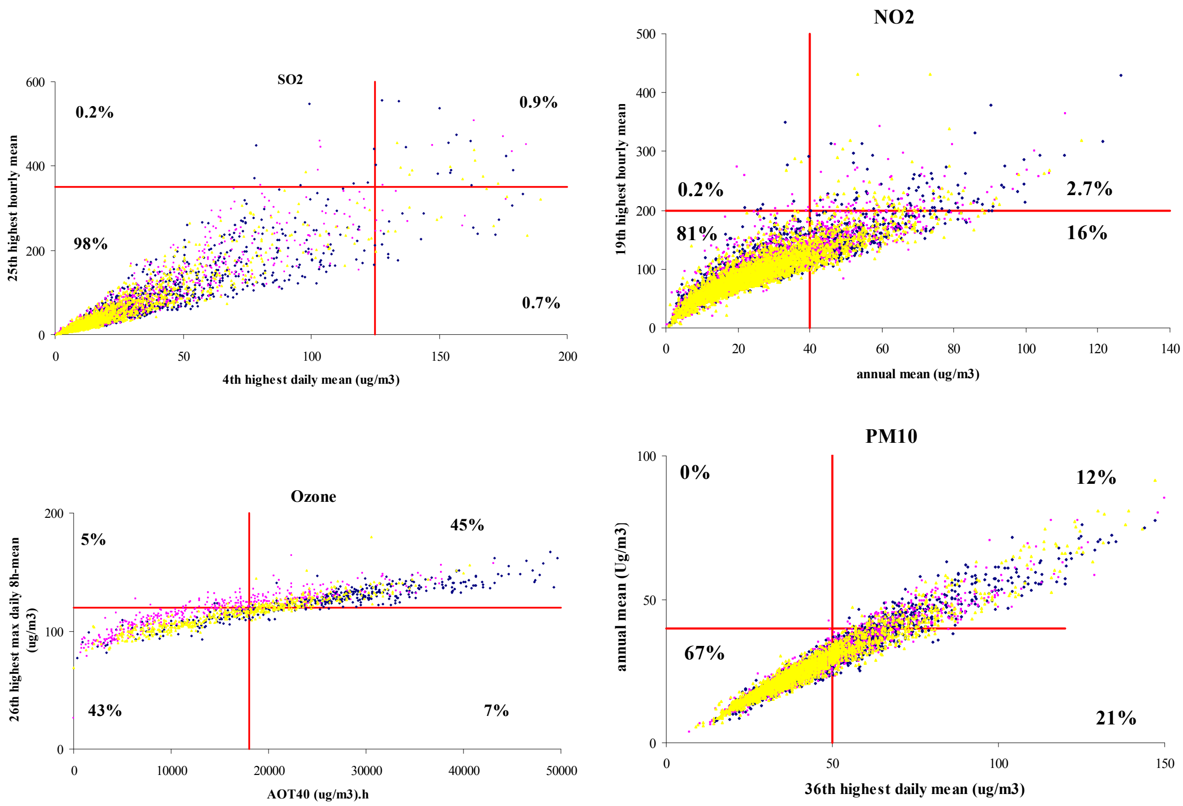


Figure 4.1. Relationship between air quality indicators related to limit or target values. Top left: the 4^{th} highest daily mean and 25^{th} highest hourly mean concentration of SO_2. Top right: annual mean and the 19'th highest hourly mean concentration of NO_2. Bottom left: AOT40 and 26^{th} highest maximum daily 8h- mean concentration of ozone. Bottom right: 36^{th} highest daily mean and annual mean concentration of PM_{10}. Data taken from AirBase (2006-2008). Red lines indicate the respective limit or target values. The fraction of observations in each sector is indicated.

[7] This analysis has been based on a three-year period in order to include a range of meteorological conditions. The period 2006-2008 was selected to include the year 2006 which had rather bad dispersion conditions (relatively high concentrations). The air quality situations in 2008 and 2009 are similar; including 2009 data will not lead to different conclusions.

For NO_2 and PM_{10} the situation is even clearer. For NO_2 in 19% of the cases the annual limit value is exceeded; the hourly limit value is exceeded in less than 3% of the cases. There are rarely exceedances of the hourly mean limit value if there are no exceedances of the annual mean limit value: during 2006-2008 this is observed for 0.2% of the cases.

The scatter plot of PM_{10} shows one single situation where the daily limit value is and the annual limit value is not exceeded (Figure 3.9). If the daily limit value is exceeded, there is a probability of 35% to have a concurrent exceedance of the annual limit value.

In summary, it seems adequate to report in the future only on the most stringent limit value for SO_2, NO_2 and PM_{10}, respectively. This might lower the administrative burden of the Member States. For example, in the air quality questionnaire 12 (sub)forms could be simplified or deleted.

When comparing for ozone the AOT40 (vegetation related) with the 26th highest maximum daily 8h-mean concentration (health related) no clear conclusion can be drawn. In total in 12% of the occasions (5% + 7%, see Figure 3.9) only one of the target values was exceeded (period 2006-2008). As the vegetation related target value (AOT40) is only evaluated at rural stations it can not be concluded that one of the target values is the most limiting. Thus, one may conclude that the evaluation of both of them is necessary.

Table 4.1 gives an overview of the most stringent EU limit or target value set for the protection of human health. The EU value is compared with the AQG recommended by the WHO. A rough estimate of the urban population exposed to concentrations above the EU reference value and the AQG is given. This estimate refers to the situation for the period 2006-2008 and it is based on the methodology for the Urban Air Quality indicator CSI004 (EEA, 2011).

Table 4.1. The most stringent EU limit or target levels compared to the WHO Air Quality Guidelines (average period and concentration, in µg/m^3 except for CO where the LV is given in mg/m^3). Estimate of the fraction of the urban population exposed to level above the reference level.

component	EU reference value	Exposure estimate (%)	WHO AQG	Exposure estimate (%)
SO_2	day (125)	0.3 – 2.3	day (20)	68 - 85
NO_2	year (40)	7 - 19	year (40)	7 – 19
PM_{10}	day (50)	18 – 40	year (20)	80 – 90
lead	year (0.5)	< 1	year (0.5)	< 1
CO	8-hour (10)	0 – 2	8-hour(10)	0 - 2
benzene (a)	year (5)	< 1	year (1.7)	15 - 21
ozone	8-hour (120)	16 - 50	8-hour (100)	> 95

Colour coding of exposure estimates, fraction of urban population exposed to concentrations above the reference level:

< 10%	10-50%	50-90%	>90%	

(a) no AQG recommended by WHO; value estimated assuming an additional lifetime risk of 1 x 10^{-5}, see text in paragraph 3.5.

Protection of human health

For pollutants with a low or zero threshold for adverse effects, it will generally lead to larger health benefits and it will in general also be more cost-effective to have a (small) improvement in air quality for the whole population than having a (larger) improvement for the population in hot-spot areas. In the AQ Directive the concept of an exposure reduction target has been introduced for $PM_{2.5}$. In addition a limit value has been introduced to avoid exposure to excessive concentrations.
Therefore the review of the AQ Directive could consider: The concept of exposure reduction targets might as well be applicable to other pollutants than $PM_{2.5}$.

In its 13th meeting (April 2010) the Task Force on Health (UN/ECE, 2010) noted that so far only a few studies on effects of long term exposure to ozone have been published. However, these papers suggest that chronic exposure increases the risk of adverse health effects, including experiencing asthma symptoms and asthma related hospitalizations. In an analysis of the American Cancer Society cohort, Jerret et al (2009) showed that ozone was significantly associated with death from respiratory causes. A concentration increase of 10 µg/m^3 in averaged 1-hour daily maximum ozone concentration was estimated to increase respiratory mortality by 2%.
The annual mean ozone concentration originates to a some extent from sources outside the European Union. In order to reduce long-term ozone exposure, co-operation at northern-hemispheric scale will be needed.
Therefore the review of the AQ Directive could consider: A long-term (annual) target for ozone could be discussed.

There is an on-going discussion which role chemical and physical properties of airborne particulate matter play in the generation of health effects. A review made by US-EPA (2009) concluded that there is a causal relation between cardiovascular and respiratory effects and short- and long-term exposure to $PM_{2.5}$. The collected evidence from the reviewed studies is suggestive of a causal relationship between exposure to the coarse fraction of PM ($PM_{10-2.5}$) as well as to ultrafine particles and cardiovascular and respiratory effects.

With respect to the chemical composition, the role of specific elements like elemental carbon, metals but also of container components like diesel exhaust, particles from tyre and break wear has been investigated. During the 13th meeting of the Task Force on Health (UN/ECE, 2010) the Netherlands presented a systematic analysis of available evidence to assess if black smoke[8], measured by reflectometric methods, could be a health-relevant indicator of air pollution from combustion sources additional to PM_{10} or $PM_{2.5}$ mass concentration. Preliminary results confirmed that such an indicator could be a sensitive health- and policy-relevant exposure metric to assess the health effects of traffic-related air pollution and to assess the effectiveness of traffic-related policy measures. However, the specificity of black smoke for health effects attributable to PM needs to be further evaluated.
Therefore the review of the AQ Directive could consider: The need for defining limit or target values for specific components in PM, for example combustion aerosol, or black carbon.

Black carbon forms an important fraction of PM; exposure to black carbon will lead to health impacts (see above). Black carbon is furthermore a so-called short-lived climate forcer (that is, a warming agent having a relative short atmospheric lifetime ranging from days to weeks). Reduction of black carbon emissions is presently being considered in the revision of the UN/ECE Gothenburg protocol.

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[8] Black smoke consists of fine solid particles suspended in air, which mainly arise from the incomplete burning of fossil fuels or bio fuels. Black smoke is measured by its blackening effect on filters. As this blackening effect depends on the aerosol composition, black smoke is a rather ill-defined pollutant: similar black smoke concentrations may have widely different aerosol composition. Black carbon (BC) is a better measure for particles produced by incomplete combustion.

Therefore the review of the air quality legislation could consider: The need for defining limit or target values for black carbon in ambient air and/or including measures that support or prioritise the reduction of black carbon emissions..

Carbon monoxide (CO) has a direct impact on human health and plays a key role in atmospheric chemistry as it, together with methane, regulates the budget of the hydroxyl-radical. During the photochemical oxidation of CO and methane, ozone and CO_2 are formed. CO is therefore a secondary greenhouse gas and in addition, it has indirectly an impact on human health via its contribution to ozone formation.
Therefore the review of the air quality legislation could consider: Should CO as precursor of CO_2, ozone, and methane be included in the scope of the revised EU air legislation because they are, as precursor of CO_2 and O_3, involved in air pollution health effects and climate change?

A number of Member States have set more strict limit or target values at the national or regional level for the pollutants listed in the Air Quality Directive and the 4th Daughter Directive (see Annex A). Several Member States have set standards for other air pollutants. Next to limit or target values in ambient air, limit or target values have been set for deposition.
Therefore the review of the AQ Directive could consider: Which lessons could be learned by reviewing the argumentations of Member States that have implemented more strict values than those set in the EU legislation?

Protection of ecosystems

For the protection of ecosystems the EU has only defined critical levels for the direct exposure to SO_2 and NO_x. Not included in the AQ Directive are critical levels for ammonia (NH3), and the issue of acidification and eutrophication of European ecosystems is not addressed. The critical loads of acidity and particularly of nutrient nitrogen are still exceeded in large parts of sensitive ecosystem areas in Europe (CCE, 2011). As acidification is the combined effect of the deposition of sulphur oxides and reduced as well as oxidised nitrogen compounds to ecosystems it can not a priori be stated whether the current critical levels set for SO_2 and NO_x are sufficient to protect ecosystems.

In the preparation of the CAFE Strategy and the NEC Directive, reductions in acidifying or eutrophying deposition loads played a key role in setting the ceilings for national air pollutant emissions. The NEC Directive sets an interim environmental objective for 2010: reducing areas where critical loads for acidification are exceeded by at least 50% in each grid cell for which critical load exceedances are computed compared with the 1990 situation. Calculations for 2010, assuming a full implementation of current policies, show in 84% of the grid cells a reduction in exceeded area of more than 50% (e.g. EEA, 2010). In spite of considerable improvements, the NEC objective has not been met. No environmental objective for reducing nutrient nitrogen loads alone has been defined; only emission ceilings for NO_x and NH_3 have been set. The area of sensitive ecosystems at risk of exceedances of critical loads of nutrient nitrogen (eutrophication) has only slightly decreased over the last decades (e.g. EEA 2010).
Therefore the review of the AQ Directive could consider: More emphasis might be put on the issue of eutrophication and acidification (and links to changes in biodiversity inter alia in Natura 2000 areas). Particularly, as the Air Policy Review by 2013 will address both in parallel, the revision of the AQ and NEC Directives and the respective ex post and impact assessments.

The adverse effects of ground-level ozone on vegetation, both agricultural crops and (semi)-natural vegetation are well documented. Initially average concentrations have been used to assess the risk associated with ozone for vegetation. In the 1990s it was shown that the integrated exposure over a certain threshold is better correlated with the effects than the concentration itself. The AOT40 concept (ozone concentrations above a threshold of 40 ppb accumulated during the growing period

of vegetation[9]) was introduced. However, since around the year 2000 it is becoming increasingly evident that the actual uptake of ozone particularly by plant leaves, i.e. the internal dose, rather than the external exposure should be considered in the risk assessment. From the first estimates (Emberson et al. 2000) onwards it has become clear that the distribution of the so-called ozone flux (that is, the actual amount of ozone taken up by vegetation) is much more uniform over Europe than AOT40 exposure (Figure 4.2). Using the EMEP model both AOT40 (left) and ozone flux (right) have been calculated for deciduous trees (Karlsson, 2009). For the AOT40 indicator, there is a difference in modelled exposure between Scandinavia and the Mediterranean area of about a factor of 10. This is supported by measured monitoring data (de Smet et al. 2009). When the exposure is based on ozone flux, the north-south gradient is about a factor of 2-3 (Figure 4.1).
Based on the recommendations of the Working Group on Effects the Executive Body of the CLRTAP has decided (UN/ECE 2009) to include the flux-based approach the integrated assessment modeling activities, especially in work done for the revision of the Gothenburg Protocol.
Therefore the review of the AQ Directive could consider: Should the AOT40 standard be replaced by the flux parameter? In favour of this is the improved risk assessment using the flux parameter; disadvantage is that the direct monitoring of ozone fluxes is difficult (and expensive). Alternatively, compliance checking of a flux standard could be based on the results of chemical transport models. The use of models in exceedance assessments of the health related limit values is already in place in a number of Member States.

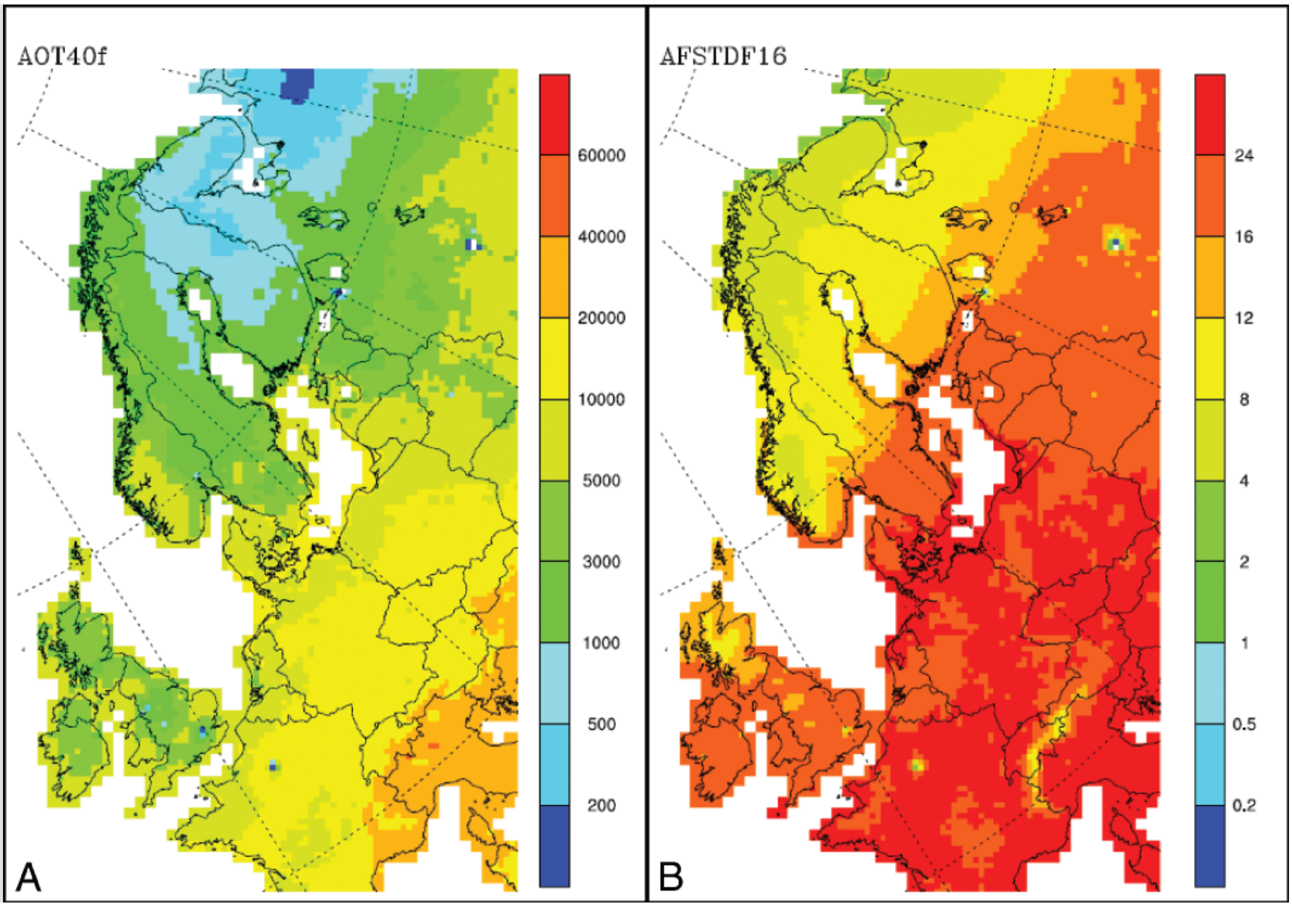

Figure 4.2. Comparison between AOT40 (April-September, in ppb.h) and flux (in mmol.m^{-2}) distributions over Europe. (source: Karlsson et al, 2009)

[9] For agricultural crops a three-month growing period (May - July) is adopted; for natural vegetation (forests) a 6-month period (April - September) is used.

Exposure assessment

In the assessment of exposure to air pollution the classification and representativeness of the monitoring station is an important factor. This information is generally provided by the local network managers. Analyses made by the Topic Centre (see e.g. Horalek et al 2009) suggest that not in all cases similar classification criteria have been used. For a better comparability between countries it is recommended to provide guidance on station classification and to develop tools for validation of station classification.

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Annex A. Additional limit or target values set by Member States

In May 2011 the ETC/ACM sent out an e-mail inquiry requesting the contact person responsible for the annual reporting on ambient air quality (AQ questionnaire, 2004/461/EC) additional information on limit or target value set by national or regional authorities:

Could you inform us on the situation in your country:

• have for the pollutants listed in the 4th Daugther Directive and the Air Quality Directive more strict limit/target values been set in your country (or will so in the near future); if so, please specify;

• have limit/target values been set for other pollutants than those listed in the AQ Directive?

Answers have been received from all Member States (except France) and also from Switzerland and Norway.

The results are summarized below. The summary table provides a qualitative overview of the pollutants for which the countries have set more strict values.

(a) Sulphur dioxide, SO_2

Austria: limit value of 120 µg/m^3 for daily mean concentration, not to be exceeded. For half-hourly mean a limit value of 200 µg/m^3 is set; exceedance is allowed up to three times per day and up to 48 times per year up to a level of 350 µg/m^3.

Switzerland: air quality standards are 30 µg/m^3 (annual average level), 100 µg/m^3 (95-oercentile of the half-hourly means) and 100 µg/m^3 (daily average, may be exceeded only once per year).

United Kingdom: a 15-minute mean of 266 µg/m^3 may not be exceeded more than 35 times a year.

Hungary: the hourly and daily limit values are 250 and 50 µg/m^3, respectively. 24 (hourly LV) and 3 (daily LV) exceedances are permitted each year.

Norway: a "national target"[10] is 90 µg/m^3 for daily mean levels, (no allowed exceedances)

Poland: no exceedances are allowed for the hourly LV (350 µg/m^3) and daily LV (125 µg/m^3)

Sweden: limit values are 200 and 100 µg/m^3 for averaging periods of 1h and 24, respectively. The permitted number of exceedances each year is 175 (hourly limit value) and 7 (daily limit values)

(b) Nitrogen dioxide, NO_2

Austria: limit value of 200 µg/m^3 for half-hourly mean. An annual limit value of 30 µg/m^3 enters into force by 1-1-2012; for the period 2010-2011 a Margin of Tolerance of 5 µg/m^3 is defined

Switzerland: air quality standards are 30 µg/m^3 (annual average level), 100 µg/m^3 (95-oercentile of the half-hourly means) and 80 µg/m^3 (daily average, may be exceeded only once per year).

Hungary: hourly and daily limit values are 100 and 85 µg/m^3, respectively. 18 exceedances of the hourly LV are permitted each year.

Norway: a "national target"[8] is 150 µg/m^3 for hourly mean levels, (max. 8 exceedances permitted)

Poland: no exceedances are allowed for the hourly LV (200 µg/m^3); the annual limit value is set to 35 µg/m^3

Sweden: limit values are 90 and 60 µg/m^3 for averaging periods of 1h and 24, respectively. The permitted number of exceedances each year is 175 (hourly limit value) and 7 (daily limit values)

(c) PM_{10}

Austria: not more than 25 exceedances of the daily limit value of 50 µg/m^3 are allowed.

Switzerland: air quality standards are 20 µg/m^3 (annual average level) and 50 µg/m^3 (daily average level, may be exceeded only once per year).

Estonia: the annual limit value is 20 µg/m^3

United Kingdom: Scotland has introduced stricter limit values: not more than 7 exceedances of the daily limit value of 50 µg/m^3 are allowed; the annual limit value is 18 µg/m^3

[10] Norway has set "national targets" for 2010 that are not mandatory binding.

Norway: a "national target"[8] has been set for daily means: 50 µg/m[3,] not to be exceeded more than 7 exceedances of the daily limit value of are allowed

(d) PM$_{2.5}$

Finland: the limit value of 25 µg/m^3 has to be met since 1-1-2010

United Kingdom: Scotland has introduced a stricter limit value: 12 µg/m^3 to be achieved by 2020.

(e) Lead

United Kingdom: a limit value for the annual mean of 0.25 µg/m^3 has to be met since 31 December 2008.

Hungary: limit value for annual mean is 0.3 µg/m^3

(f) Benzene

United Kingdom: in Scotland and Northern Ireland a running annual mean may not exceed 3.25 µg/m^3

Hungary: an hourly limit value of 10 µg/m^3 has been set.

Norway: a "national target"[8] is 2 µg/m^3 for annual mean levels (urban background, no allowed exceedances)

Poland: annual limit value of 4 µg/m^3

(g) Carbon monoxide, CO

Switzerland: air quality standard is 8 mg/m^3 (daily average, may be exceeded only once per year)

Hungary: limit values of 10, 5 and 3 mg/m3 have been set for hourly mean, maximum daily 8 hour mean , and annual mean, respectively.

Poland: limit value of 5 mg/m^3 for the maximum daily 8 hour mean.

(h) Ozone, O3

Switzerland: the 98-percentile of half-hourly average levels in a month may not exceed 100 µg/m^3, an hourly average level of 120 µg/m^3 may be exceeded only o once per year.

United Kingdom: a level of 100 µg/m^3 (daily maximum of running 8-hour mean) may not be exceeded more than 10 times a year.

(i) Other pollutants

Austria: limit values have been set for deposition of dust, lead and cadmium;

Switzerland: for cadmium in PM_{10} the standard is 1.5 ng/m^3 as annual mean; deposition standards have been set for total dust, lead, cadmium, zinc and thallium.

Belgium: Flanders has set regions limit or target values: for cadmium a limit value of 30 ng/m^3 has been set next to the EU target value of 5 ng/m^3. Limit values have been set for vinylchloride and hydrogen fluoride. Limit values have been set for the deposition of lead, cadmium and thallium. Target values have been set for acidifying and eutrifying deposition.

Bulgaria: for some specific pollutants like hydrogen sulphide (H_2S), ammonia (NH_3), sulphuric acid (H_2SO_4), hydrogen chloride (HCl) national regulations has been made.

Spain: objectives (for half-hourly eman and daily mean levels) have been set molecular chlorine, hydrogen chloride, fluorine compounds, hydrohen sulphide and carbon sulphide.

Finland: non-binding national air quality guide values substantially lower than the EU LV/TV, have been set for PM_{10}, CO, NO_2, and SO_2; Guide values have also been set for total suspended particulates and total reduced sulphur.

United Kingdom: for polycyclic aromatic hydrocarbons the limit value for annual mean is 0.25 ng/m^3 B(a)P. Additionally a limit value has been set for 1,3-butadiene.

Hungary: Limit values (annual mean) of 10, 5 and 25 ng/m3 have been set for arsenic, cadmium, and nickel, respectively. For B(a)P daily and annual limit values of 1 and 0.12 ng/m3 have been set..

Further limit values have been set for chromium, beryllium, 1,3-butadiene, dioxins, tetrachloretylene, trichloroethylene, vinyl chloride and asbestos. For a large number of chemical "planning target values" which are used during the permitting procedures are defined.

Lithuania: limit values have been set for 363 substances.

Summary table indicating for which pollutants additional limit or target values have been set by European countries. See text above for further discussion.

Country	SO_2	NO_2	PM_{10}	$PM_{2.5}$	Pb	ben zene	CO	O_3	others
Austria	(a)	(b)	(c)	--	--	--	--	--	(i)
Belgium	--	--	--	--	--	--	--	--	(i)
Bulgaria	--	--	--	--	--	--	--	--	(i))
Switzerland	(a)	(b)	(c)				(g)		(i)
Cyprus	--	--	--	--	--	--	--	--	--
Czech Republic	--	--	--	(d)	--	--	--	--	--
Germany	--	--	--	--	--	--	--	--	--
Denmark	--	--	--	--	--	--	--	--	--
Estonia	--	--	(c)	--	--	--	--	--	--
Spain	--	--	--	--	--	--	--	--	(i)
Finland	--	--	--	(d)	--	--	--	--	(i)
France					No information received				
United Kingdom (1)	--		(c)						
Greece	--	--	--	--	--	--	--	--	--
Hungary	(a)	(b)	--	--	(e)	(f)	(g)	--	(i)
Ireland	--	--	--	--	--	--	--	--	--
Italy	--	--	--	--	--	--	--	--	--
Lithuania	--	--	--	--	--	--	--	--	(i)
Luxembourg	--	--	--	--	--	--	--	--	--
Latvia	--	--	--	--	--	--	--	--	--
Malta	--	--	--	--	--	--	--	--	--
Netherlands	--	--	--	--	--	--	--	--	--
Norway									
Poland	(a)	(b)	--	--	--	--	(g)	--	--
Portugal	--	--	--	--	--	--	--	--	--
Romania	--	--	--	--	--	--	--	--	--
Sweden	(a)	(b)	--	--	--	--	--	--	--
Slovania	--	--	--	--	--	--	--	--	--
Slovakia	--	--	--	--	--	--	--	--	--

(1) For Gibraltar no additional limit or target values have been set.

Annex B. Number of operational monitoring stations in EU-27,

Table B.1 Concentration (in μg/m³ unless indicated) per station type averaged over all operational stations in EU-27 , period 2009. In case the limit or target value is expressed as a maximum allowable number of exceedances (N_{exc}) of a specified threshold value per year, the $(N_{exc}+1)^{th}$ highest value is given.

	SO_2-health		SO_2- vegetation		NO_2-health		NOx-vegetation	PM10		PM2.5
type	hourly	daily	year	winter	hourly	year	year	daily	year	year
Rural	23	11	2.8	2.8	58	11		33	19	12
Urban	40	19			98	24		46	27	17
Traffic	35	17			136	41		47	29	17
other	60	24			79	19		40	24	17
type	O3 8h max	O3 AOT40 (μg/m^3).h	CO maximum (mg/m^3)	Pb year	benzene	arsenic (ng/m^3)	cadmium (ng/m^3)	nickel (ng/m^3)	B(a)P (ng/m^3)	
Rural	115	13600	1.4	0.12	0.79	0.58	1.2	1.6	0.35	
Urban	110		2.4	0.026	1.4	1.2	1.2	2.9	2.4	
Traffic	99		2.7	0.023	1.9	0.97	0.68	3.1	0.79	
other	109		2.2	0.056	1.3	2.6	2.7	6.9	1.2	

Table B.2 Number of operational stations per stations type as used to prepare the distance-to-target graphs .

	SO_2-health		SO_2- vegetation		NO_2-health		NOx-vegetation	PM_{10}		$PM_{2.5}$
type	hourly	daily	year	year	hourly	year	year	daily	year	year
Rural	230	243	245	167	359	373		276	279	94
Urban	668	702			1119	1167		1026	1028	293
Traffic	342	343			792	800		717	723	124
other	470	471			475	477		377	379	60
total	1710	1759			2745	2817		2396	2409	571
type	O3 8h max	O3 AOT40	CO max	Pb year	benzene	arsenic	cadmium	nickel	B(a)P	
Rural	482	486	57	85	29	87	90	89	51	
Urban	1001		366	231	191	210	235	222	219	
Traffic	263		547	129	233	121	130	127	110	
other	272		168	128	108	112	126	119	50	
total	2018		1138	573	561	530	581	557	430	