

# Spatial assessment of PM<sub>10</sub> and ozone concentrations in Europe (2005)

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# Contents

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<b>Acknowledgements .....</b>	<b>4</b>
<b>Executive summary .....</b>	<b>5</b>
<b>1 Introduction.....</b>	<b>9</b>
<b>2 Interpolation methodologies, supplementary data selection and interpolation uncertainty analysis.....</b>	<b>12</b>
2.1 Interpolation methodologies and supplementary data selection.....	13
2.2 Interpolation uncertainty analysis.....	13
2.3 Exceedance probability mapping.....	13
<b>3 PM<sub>10</sub> maps for 2005.....</b>	<b>15</b>
3.1 PM <sub>10</sub> annual average map .....	16
3.2 PM <sub>10</sub> 36th highest daily average map .....	16
3.3 Population exposure and health impacts .....	17
3.4 Health impact assessment.....	20
3.5 Uncertainties and probability of exceeding the limit values .....	22
<b>4 Ozone maps for 2005 .....</b>	<b>24</b>
4.1 Ozone health related indicators .....	24
4.1.1 Population exposure and health impacts.....	24
4.1.2 Uncertainties in calculating the probability of exceeding the target.....	26
4.2 Ozone vegetation indicators.....	31
4.2.1 Exposure of vegetation and forests .....	31
4.2.2 Uncertainties .....	35
<b>5 Recommended interpolation methods for regular updating.....</b>	<b>37</b>
<b>References .....</b>	<b>39</b>
<b>Annex 1 Input data .....</b>	<b>42</b>
A1.1 Introduction.....	42
A1.2 Measured air quality data.....	42
A1.3 Unified Emeq model output.....	42
A1.4 Lotos-euros model output .....	43
A1.5 Altitude .....	43
A1.6 Meteorological parameters .....	43
A1.7 Population density.....	44
A1.8 Land cover.....	44
<b>Annex 2 Statistical formulas.....</b>	<b>45</b>
<b>Annex 3 Exceedance probability mapping .....</b>	<b>48</b>

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The EEA project manager was Anke Lükewille and the EEA-ETC/ACC task manager was Peter de Smet.

# Executive summary

High PM<sub>10</sub> and ozone concentrations can have harmful effects on human health and vegetation. The EU air quality legislation sets two legally binding limit values for PM<sub>10</sub> mass concentrations (EC, 1999). European citizens shall not be exposed to:

- (1) annual mean levels exceeding 40 µg PM<sub>10</sub> per cubic metre (m<sup>-3</sup>), and
- (2) PM<sub>10</sub> concentrations exceeding 50 µg m<sup>-3</sup> for more than 35 days per year (36th maximum daily average).

For protection of human health from the ground-level ozone pollution, the air quality legislation sets a target value (not legally binding; EC, 2002):

- an eight-hour average ozone concentration of 120 µg m<sup>-3</sup> shall not be exceeded by more than 25 days per year (26th highest maximum daily value).

This report presents particulate matter (PM<sub>10</sub>) and ground-level ozone concentration maps covering the whole of Europe. The interpolated maps are based on a combination of measurement and regional modelling results. Using measured concentrations as a primary source of information, the report summarizes the methodologies and the methodological choices taken in order to derive such maps.

The maps use monitoring data for 2005 as a basis, i.e. values reported by the EEA member countries in 2006 under the Exchange of Information Decision (EC, 1997).

To estimate people's exposure to PM<sub>10</sub> and ozone concentrations and possible health impacts, measurement data in denser populated areas have got a higher weight than those in less populated regions.

Additionally, the study considers the World Health Organization (WHO) human health indicator SOMO35 <sup>(1)</sup> and the vegetation-related indicator

AOT40 <sup>(2)</sup> for vegetation/crops (EC, 2002) or forests. Both metrics, the SOMO35 and the AOT40, are measures of the accumulated exposure of humans or vegetation to high ozone levels over a certain period.

## Particulate matter (PM<sub>10</sub>) mapping results

The interpolated PM<sub>10</sub> maps suggest that the number of Europeans exposed to annual mean concentrations of PM<sub>10</sub> above the annual limit value (40 µg m<sup>-3</sup>; EC, 1999) was more than 9 % of the total population in 2005. The estimated probability of exceeding this PM<sub>10</sub> limit value was higher than 75 % in the urban areas of the Balkan region. The probability is also high in southern Poland, the Czech Republic, Hungary and southern Spain.

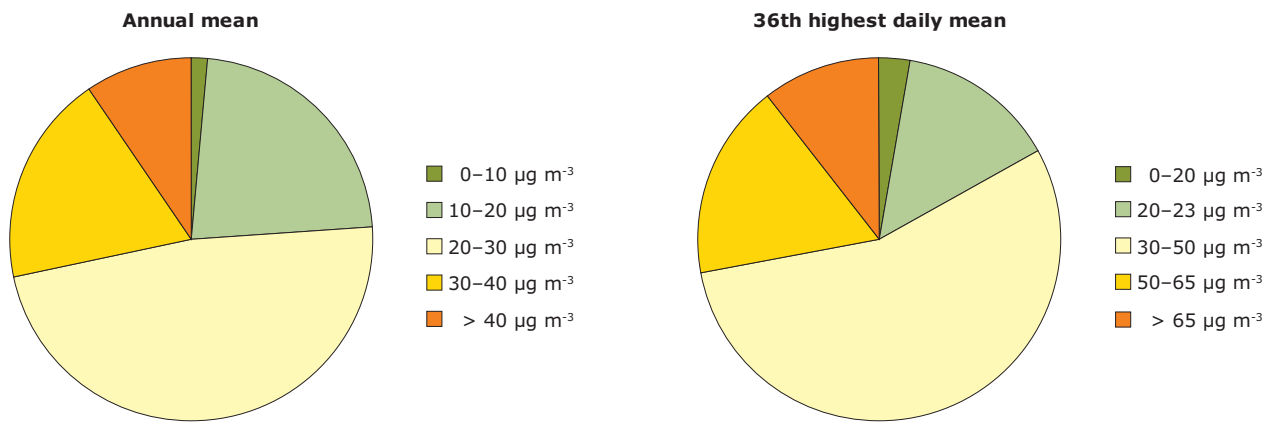
The results indicate that in 2005, about 28 % of the European population were exposed for more than 35 days to PM<sub>10</sub> concentrations of above 50 µg m<sup>-3</sup> (that is, the limit value; EC, 1999). The estimated probability of exceedance for the PM<sub>10</sub> daily limit value is considerable in large areas of the eastern European countries and in the entire Po Valley in Italy (higher than 75 % probability). The probability is also high in Spain, Portugal, Italy, Greece, some of the Balkan countries, Belgium, the Netherlands and Luxembourg, where it ranges from 25 to 50 %, with increased levels of 50 to 75 % in the more urbanized centres of these regions.

The number of premature deaths per million inhabitants attributable to PM<sub>10</sub> exposure (the EU-27 as a whole) was estimated to range from 510 to 1 150 cases per million, with a best estimate of 830 deaths per million (median). The observed range is mainly a result of the differences in PM<sub>10</sub> concentrations over Europe, and partly of the differences in age distributions and baseline mortalities. Measured particulate matter (PM) concentrations in Europe have not shown, in general, any downward tendencies over the period from 2000 to 2005. The number of 830 premature deaths per million inhabitants corresponds to about 373 000 premature deaths in the EU-25 countries

<sup>(1)</sup> SOMO35 (expressed in µg m<sup>-3</sup> times hours) is the accumulated ozone concentration in excess of 35 ppb (70 µg m<sup>-3</sup>). It is the sum of the differences between the maximum daily 8-hour concentration exceeding 70 µg m<sup>-3</sup> for each day in the calendar year.

<sup>(2)</sup> AOT40 (expressed in µg m<sup>-3</sup> times hours) stands for the sum of the differences between hourly concentrations greater than 80 µg m<sup>-3</sup> (= 40 ppb) and 80 µg m<sup>-3</sup> over a given period using only the 1-hour values measured between 8:00 and 20:00, Central European Time, each day. Target value for the protection of vegetation, AOT40, is calculated from the 1-hour values from May to July.

**Exposure of the European population to PM<sub>10</sub> concentrations, annual average (left) and 36th highest daily average (right), 2005**



in 2005. This estimate agrees well with the results of the CAFE Programme where 348 000 premature deaths caused by (only) anthropogenic primary PM and PM precursors emissions were estimated for the year 2000 <sup>(3)</sup>.

**Ground-level ozone mapping results**

The map of the 26th highest maximum daily value indicates that in 2005, 38 % of the European population were exposed to the ozone levels above the target value (120 µg m<sup>-3</sup>, 26th highest daily maximum 8-hour average). The estimated probability that the target value for this indicator shall be exceeded south of the geographic line Biarritz – Basel – Luxemburg – Hannover – Lodz is moderate to considerable (probability > 50 %; and in southern parts, it is > 75 %).

The impact on health, due to elevated ozone concentrations in the EU-27, seems to be lower than the PM<sub>10</sub> impact by an order of magnitude. According to the assessment presented in this report, it ranges from 75 deaths per million inhabitants (for the south-eastern and southern Europe) to less than 10 (for the northern and north-western European countries).

Furthermore, the results suggest a high accumulated exposure of crops and forests to ozone concentrations (AOT40). According to the interpolated maps, almost 50 % of all agricultural land was exposed to ozone levels exceeding the EU target value for 2010 (that of 18 000 µg m<sup>-3</sup> times hours), and almost 90 % was exposed to levels in

excess of the EU long-term objective of 6 000 µg m<sup>-3</sup> times hours (EC, 2002).

**Interpolation methodologies**

The report relies on the ground-based PM<sub>10</sub> and ozone measurements as the primary information, and on results from chemistry transport modelling (CTM) and other data – as the secondary, supplementary sources. This approach differs from the work in support of the development of the European Thematic Strategy on Air Pollution. The Thematic Strategy relies on modelling as its primary source of information, while using monitoring data to calibrate a chemical transport model (EMEP). These two approaches can be regarded as complementary.

The maps for rural and urban areas were created separately and subsequently merged, using the population density as a basis. This approach aims to provide an objective method for dealing with the differences found between the rural and urban interpolated concentration fields in most areas of Europe.

The resolution of the interpolated PM<sub>10</sub> and ozone maps, covering all of Europe, is 10 x 10 km<sup>2</sup>. They include rural background data as well as urban and suburban background monitoring data retrieved from the AirBase <sup>(4)</sup> database.

Different mapping procedures were developed and evaluated for the annual aggregated PM<sub>10</sub> indicators as well as ozone indicators mentioned above. Spatially resolved supplementary data were

<sup>(3)</sup> See for example: [http://ec.europa.eu/environment/air/pdf/sec\\_2005\\_1133.pdf](http://ec.europa.eu/environment/air/pdf/sec_2005_1133.pdf).

<sup>(4)</sup> <http://www.eea.europa.eu/themes/air/airbase>.



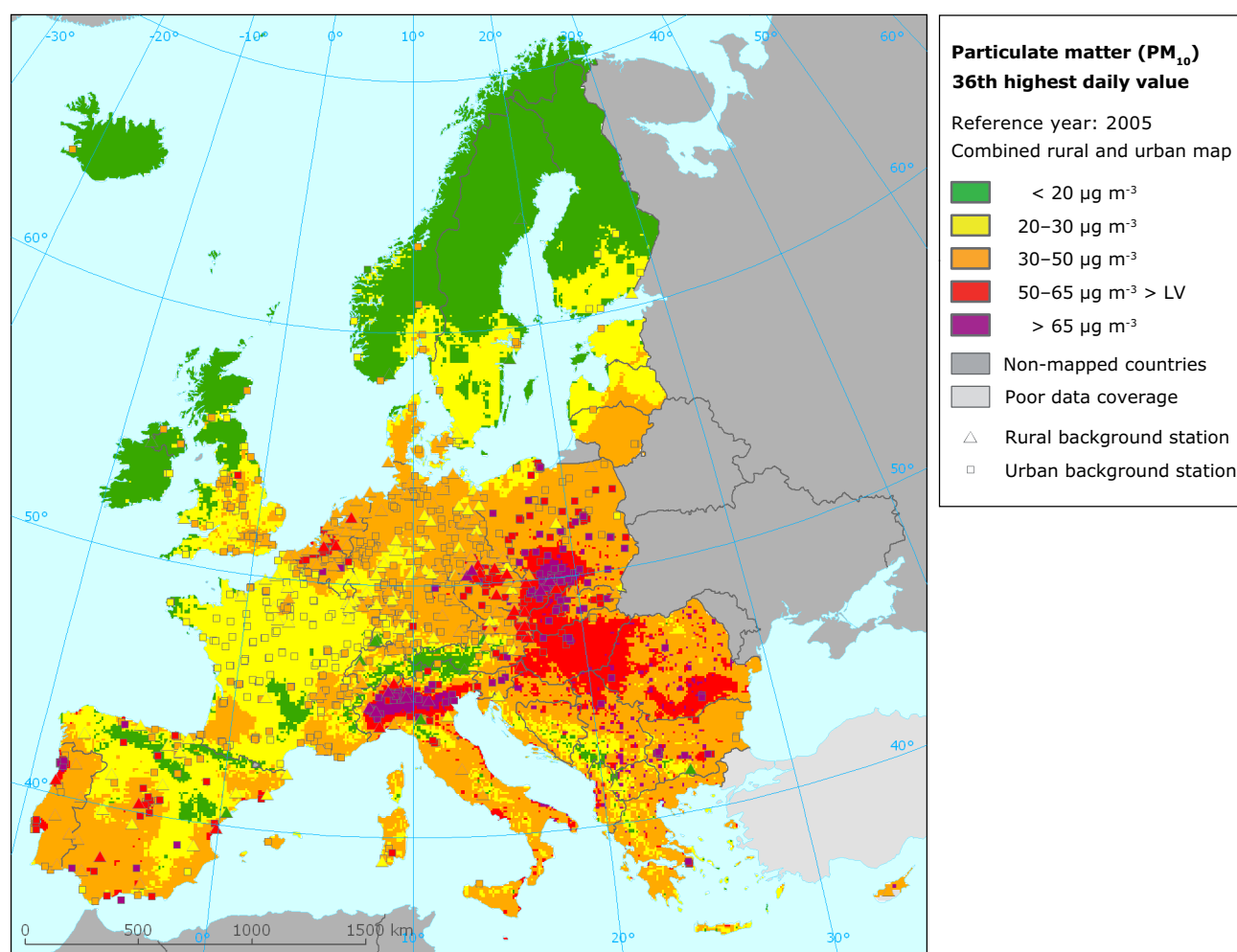
used to improve the regression and interpolation procedures and the spatial assessments (e.g. altitude, meteorological parameters, EMEP CTM and LOTOS-EUROS CTM outputs). The LOTOS-EUROS model performed best for  $PM_{10}$ , while the EMEP model performed best for ozone. Residual kriging following the use of a linear regression model was the best performing interpolation methodology for all indicators.

Uncertainty and variability related to the respective interpolation methodologies used were assessed, and the uncertainty maps were prepared. Based on the concentration and uncertainty maps, those showing the probability of exceedance were compiled.

The results obtained through basing the mapping methods on the annual data were compared to those where daily data were used for assimilation and interpolation (Denby *et al.*, 2008). The emphasis was on the statistical performance of the applied methods as well as on the methodologies available for determining uncertainties.

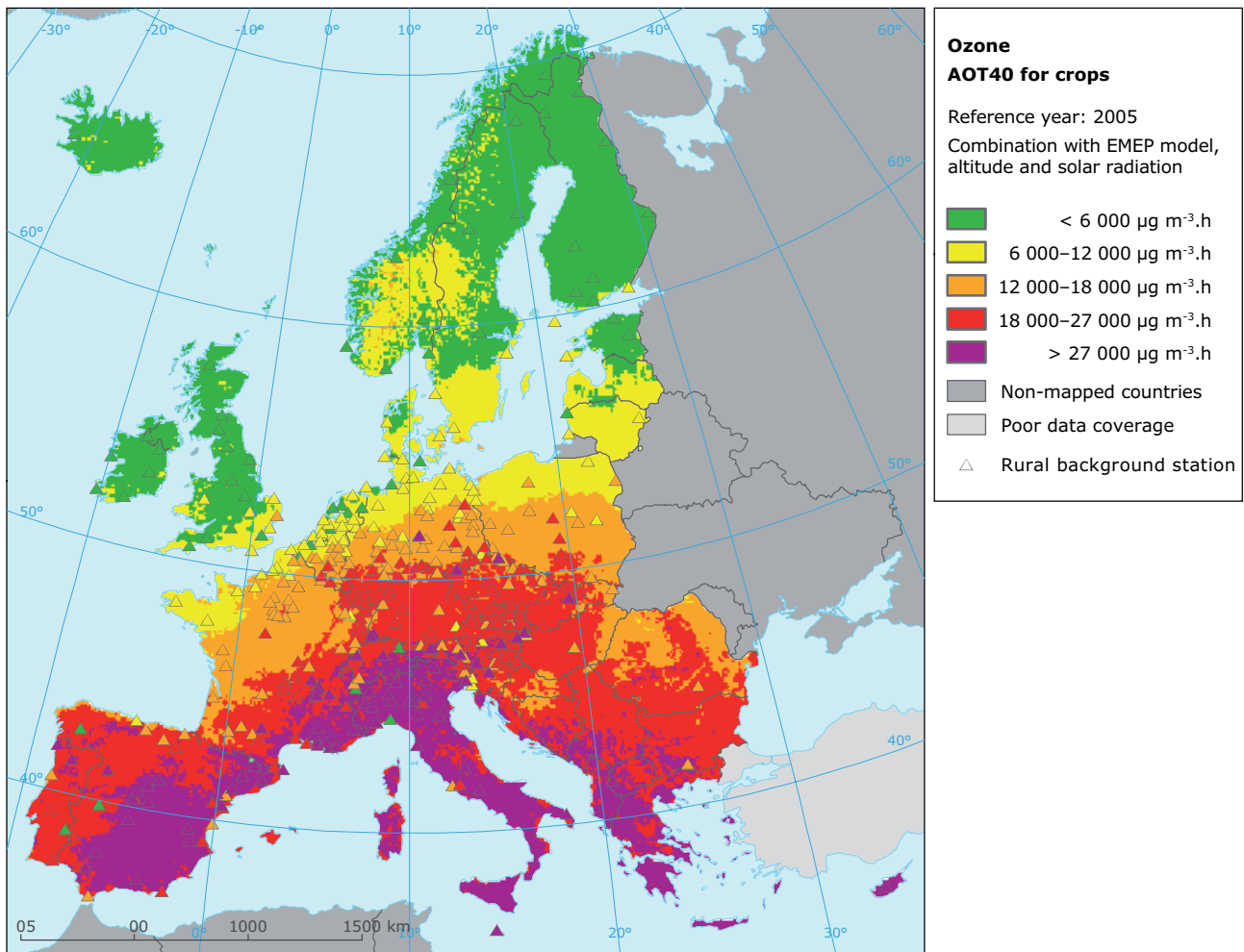
The approach used to assess the impact on human health followed, basically, the algorithms for the relative risk functions as seen when assessing the impact on health due to air pollution (Künzli *et al.*, 2000) and as used in other (model-based) assessment programmes and projects (IIASA, 2005). The report provides tables that show the estimated

### $PM_{10}$ – 36th maximum daily average value in $\mu g m^{-3}$ , 2005 – the limit value for the protection of human health is $50 \mu g m^{-3}$



**Note:** In comparison to its neighbouring countries, the  $PM_{10}$  concentration in France is relatively low. These apparently low levels are probably due to the  $PM_{10}$  values not being measured by using the recommended reference method and by applying a correction factor of (only) 1,0.

**Ozone AOT40 indicator for crops: the map shows concentrations in  $\mu\text{g m}^{-3}$  times hours, accumulated over the period from May to July 2005**



**Note:** Measurements at rural background stations were combined with EMEP model outputs and other supplementary data.

population at risk per country and in Europe as a whole. Similar impact tables were compiled for the vegetation-related indicators showing the areas at risk, i.e. subject to damage, change or yield reduction.

However, we must not forget that the interpolated  $10 \times 10 \text{ km}^2$  maps are only smooth estimates of the real pollution. They tend to overestimate the average concentrations and underestimate both

low and high values. Consequently, exposure of the population to both high and low concentrations is underestimated, while exposure to average concentrations is overestimated. Such bias can be significant, particularly in under-sampled areas.

Full technical details are given in two Technical Papers compiled by the European Topic Centre on Air and Climate Change (Horálek *et al.*, 2007 and Horálek *et al.*, 2008).

# 1 Introduction

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## General objectives

The new Directive on Ambient Air Quality and Cleaner Air for Europe (EC, 2008) requires that air quality should be assessed throughout the territory of each Member State, using the combined efforts of monitoring and modelling.

An EEA project was initiated with the objective to assess the spatial distribution of air pollution and its impacts, and to interpolate maps primarily based on air quality measurements as reported by European countries under the Exchange of Information Decision (EoI; EC, 1997, 2001). The aim was to provide the spatial information on air quality complementary to the model-based European-wide maps that were used for example in the CAFE Programme (IIASA, 2005) and City-Delta project (<http://aqm.jrc.it/citydelta/>). The reported measurement data, including the meta-data information on the single stations, are stored in the database AirBase that is hosted by the EEA and is accessible through the internet <http://www.eea.europa.eu/themes/air/airbase>.

The maps developed within the framework of this project provide **geospatially-referenced information on air quality-related status and impact indicators** that can be used in air pollution assessments. Typical status indicators are concentration levels, exceedances of thresholds and limit values as defined by the Air Quality Framework Directive and its daughter directives. Examples of impact indicators are population at risk, number of premature deaths attributable to ambient air pollution as well as ecosystems, vegetation or crops at risk. These impact indicators can in turn provide input contributing to the development of indicators and assessments used with respect to potential yield reductions as well as economic and ecological losses. However, such assessments are beyond the scope of this paper.

## General considerations concerning the spatial assessments of air quality

There exists a large variety of interpolation methods, developed for many different applications, which

use individual point measurement datasets and create spatially distributed fields based on this information (Denby *et al.*, 2005). Methods such as kriging, radial basic functions and inverse distance weighting have been used for interpolating air quality data.

An indispensable method to assess the impacts of air pollution and to evaluate compliance with various current EU Directives and UN ECE Protocols is the use of local point measurements. Due to the large range of temporal and spatial scales involved in the physic-chemical atmospheric cycles of some air pollutants, it is necessary to observe changes at all scales — from local to global. Density of air quality monitoring stations varies significantly throughout Europe and from pollutant to pollutant. Rural stations are assumed to be representative of regions within a radius of approximately 50 km <sup>(5)</sup> but the average distance between stations is much larger than this. Any spatially distributed analysis reliant on air quality data, e.g. ecosystem or health impact studies, will naturally require information on the status of air quality between the stations. Thus, there is a marked need for efficient, robust and reliable interpolation methods.

In addition to air quality measurements, in order to improve the interpolation it is also possible to introduce some supplementary data, with a better spatial coverage. Typically, such supplementary data should be either representative of the data to be interpolated, e.g. the use of PM<sub>10</sub> to represent the distribution of PM<sub>2.5</sub>, or should reflect correlations between the physical processes that lead to the spatial distribution of the data to be interpolated. Examples are elevation, levels of precipitation and temperature. Whatever supplementary data are chosen, in order to be applicable they must have a significant correlation with the data to be interpolated.

Numerical models simulating the atmospheric chemical composition provide also complex information on the spatial air quality for the region. They integrate information on emissions into the atmosphere and reflect the complex physical and chemical processes involved in the dispersion of air

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<sup>(5)</sup> For station representativeness see, e.g.: Interim report on representativeness and classification of air quality monitoring stations at <http://ec.europa.eu/environment/air/quality/legislation/assessment.htm>.

pollutants. The possibility of integrating modelling results and measurements from different platforms (*in situ* measurements, remote sensing, satellite data, etc.) with different spatial and temporal scales into a consistent analysis of the state of the atmosphere is provided by the assimilation procedures. However, the assumptions made in the model and a large number of uncertainties in model parameters and input values (such as emissions, meteorology) require that the model should be validated against measured/observed data. Different models used for regional scale assessments do not always provide the same results for the various pollutants. In general, when the models are evaluated at specific measurement points the results are considered less certain than the measured data. A possible solution for decreasing uncertainty in the resulting spatial fields may be through combination of modelling results and monitoring data in an optimised way.

### Comparison of three different interpolation methodologies

To carry out a proper evaluation of the improvements in interpolation results, we employed a type-wise poor usage analysis technique, pursuing different types of input data in three subsequent steps and including their involved uncertainties. As a result, we have formulated these three types of interpolation methodologies that we analysed and compared:

- (1) using monitoring data only (by geostatistical methods based on kriging);
- (2) combining monitoring data and chemical transport model (CTM) data (using a linear regression model followed by kriging of its residual);
- (3) combining monitoring data, CTM data and other supplementary data (using a multiple linear regression model followed by kriging of its residual).

The reason for comparing these types of methods was to examine their respective levels of performance and improvement of interpolation by adding additional information on top of the monitoring data. From the results, the statistically best or operationally preferred method was selected for the final step – the compilation of interpolated European maps of air quality.

Based on the results of such comparison, we arrived at a conclusion about the most robust and

preferred method for each indicator describing both rural and urban areas. Such preferred methods that we used for producing rural, urban and combined rural/urban maps (the year 2005) are presented in this report. In the most cases, we were selecting the above-mentioned type-3 methodology (see Chapter 5).

The Unified EMEP model has been used for the type 2 and type 3 analyses and the respective mapping exercises (Fagerli *et al.*, 2004). Further, a comparison has been made, using output from the Unified EMEP model and from another chemical transport and dispersion model, LOTOS-EUROS (Schaap *et al.*, 2008). Both applied CTM models give similar results, however, the LOTOS-EUROS model performs best for PM<sub>10</sub> <sup>(6)</sup>, while the EMEP model performs best for ozone.

### Uncertainty of the interpolations

The interpolation uncertainty of the preferred method for mapping has been assessed quantitatively. The analysis of interpolation uncertainty of the maps has been applied to all air quality indicators analysed in this report. The analyses are preliminary based on cross-validation. In cross-validation, the spatial interpolation for each measurement point is computed through the use of all available information except the information from that one point. The procedure is repeated for all points. The predicted and measured values are compared with the help of statistical indicators.

### Mapping probability of exceedance of threshold values

From a political point of view, a key question is to what extent the concentration of a certain air pollutant in certain areas is either above or below a limit, target or threshold value (as defined in the EU air quality legislation). However, there are some significant uncertainties associated with estimating pollution levels by using monitoring sites located at a certain spot in the area of a grid cell. It means that this question cannot be answered accurately for any particular location in the chosen grid cell of e.g. 10 x 10 km<sup>2</sup>. Therefore, it makes sense that the probability of values for limit, target or threshold being exceeded should be evaluated taking into account the uncertainties of interpolation.

In this report, maps showing the probability of exceeding limit or target values are presented on the

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<sup>(6)</sup> PM<sub>10</sub> is particulate matter with an aerodynamic diameter of up to 10 µm, i.e. the 'fine' (PM<sub>2.5</sub>) and 'coarse' (PM<sub>10</sub> minus PM<sub>2.5</sub>) PM fractions together.

basis of an approach described in the Annex 3. What is taken into account is only the uncertainty of the interpolation, not measurement uncertainties.

### Use of the mapping results

As summarized above, interpolation techniques have been applied for the compilation of detailed maps for air quality in Europe. The interpolations are based on a combination of air quality monitoring data, modelling results and other supplementary data. This approach complements the methods used for carrying out spatial analyses implemented for the European Thematic Strategy on Air (7). These analyses rely primarily on the results of chemical transport modelling and the data obtained was used in support of negotiations on emission reduction measures and for assessing of the feasibility of such measures.

The pan-European maps presented in this report are used as the basis for an assessment of risks to public health and to ecosystems that are related to air pollution. In addition to the benefit of mapping indicators of air pollution in order to inform the general public, the maps also contribute to the quality and relevance of the assessments of exposure to air pollution and its impacts in rural and urban areas across Europe.

Maps prepared within this task were used and presented in several EEA reports such as:

- (1) *Air pollution in Europe in 1990–2004* (EEA, 2007a);
- (2) *Europe's environment — The fourth assessment* (the Belgrade report, EEA, 2007b);
- (3) *EMEP particulate matter assessment report* (EMEP, 2007);
- (4) the EEA technical reports on air pollution by ozone in Europe in summer (EEA, 2003–EEA, 2007c).

The products of this work will be also made available to the general public via the EEA website and as part of the EEA data service.

### Background and outline of the report

This report summarises results of air quality mapping activities performed by the European Topic Centre on Air and Climate Change (ETC/ACC) for the European Environment Agency (EEA). It is mainly based on the document produced by Horálek *et al.*, in 2008 (Horálek *et al.*, 2008), which, in its turn, was based on or linked to several ETC/ACC technical papers (e.g. Horálek *et al.*, 2007 and Denby *et al.*, 2008). This report presents the maps drawn with the use of most recent (officially reported) data on PM<sub>10</sub> and ozone air quality (the year 2005). Unfortunately, the available data were insufficient to allow the mapping of PM<sub>2.5</sub> concentrations or indicators.

The ETC/ACC technical papers mentioned above provide details on the mapping methodologies developed and applied by ETC/ACC.

A recently presented ETC/ACC technical paper (Denby *et al.*, 2008) places emphasis on the use of daily statistical data i.e. when creating, on a daily basis, interpolated fields and combining these fields to derive indicators of air quality. The ETC/ACC technical paper prepared by Horálek *et al.* (2008) presents an uncertainty analysis of the interpolated maps.

Chapter 2 gives an overview of the methodologies used. Chapters 3 and 4 present and discuss the mapping results for PM<sub>10</sub> and ozone, respectively (8). Combined maps for each pollutant and each indicator are presented at the end of each of these chapters. Chapter 5 discusses how the preferred interpolation methodologies were identified and selected for the mapping of indicators for pollution and air quality in Europe. Annex 1 presents the sources of the input data and the detailed parameters used in the interpolations and mapping exercises. Annex 2 gives a brief overview of statistical formulae used. Annex 3 describes the methodologies for mapping the probability of threshold values being exceeded.

(7) European Thematic Strategy on Air, [http://ec.europa.eu/environment/air/index\\_en.htm](http://ec.europa.eu/environment/air/index_en.htm).

(8) Information on other pollutants (NO<sub>x</sub>, NO<sub>2</sub> and SO<sub>2</sub>) in relation to human health and ecosystem thresholds can be found in Horálek *et al.*, 2007.

## 2 Interpolation methodologies, supplementary data selection and interpolation uncertainty analysis

### 2.1 Interpolation methodologies and supplementary data selection

Air pollution measurements from ground stations constitute an accurate and reliable source of air quality information. As the number of measuring sites is limited, the information obtained from these measurements has to be generalized to improve the spatial coverage. There are various ways of compiling spatial maps based on measurements at a monitoring station. Spatial interpolation is the most straightforward approach. If spatial interpolation does not use any further information (except the coordinates and altitude of the measurement stations) in addition to the measurements, we refer to spatial *interpolation using primarily monitoring data*, i.e. type 1 of interpolation methods evaluated.

The second type of methods considered in this study consists in focusing on ground-based measurements as primary information and using output of chemistry transport models (CTMs) as a secondary, supplementary source. While some of the methods and data sources are similar, to some extent these two approaches may be regarded as complementary. CTMs integrate information on atmospheric emissions (as an independent source of information) and simulate the complex physical and chemical processes unfolding in the atmosphere. CTMs simulations have the advantage of providing spatial air quality information for the whole of Europe. However, they create sources of uncertainty others than those associated with measurements. Combining monitoring and dispersion modelling data represents the second type of the interpolation methods. This type presupposes creation of *linear regression models through the use of monitoring and modelling data which is then followed by interpolation of its residuals*. We have examined and compared the performance of the same method by using two different CTMs — the Unified EMEP model and the LOTOS-EUROS model.

Finally, the method of the third type is arrived at through an extension of the second type by the inclusion of supplementary parameters. These parameters are spatially resolved and show

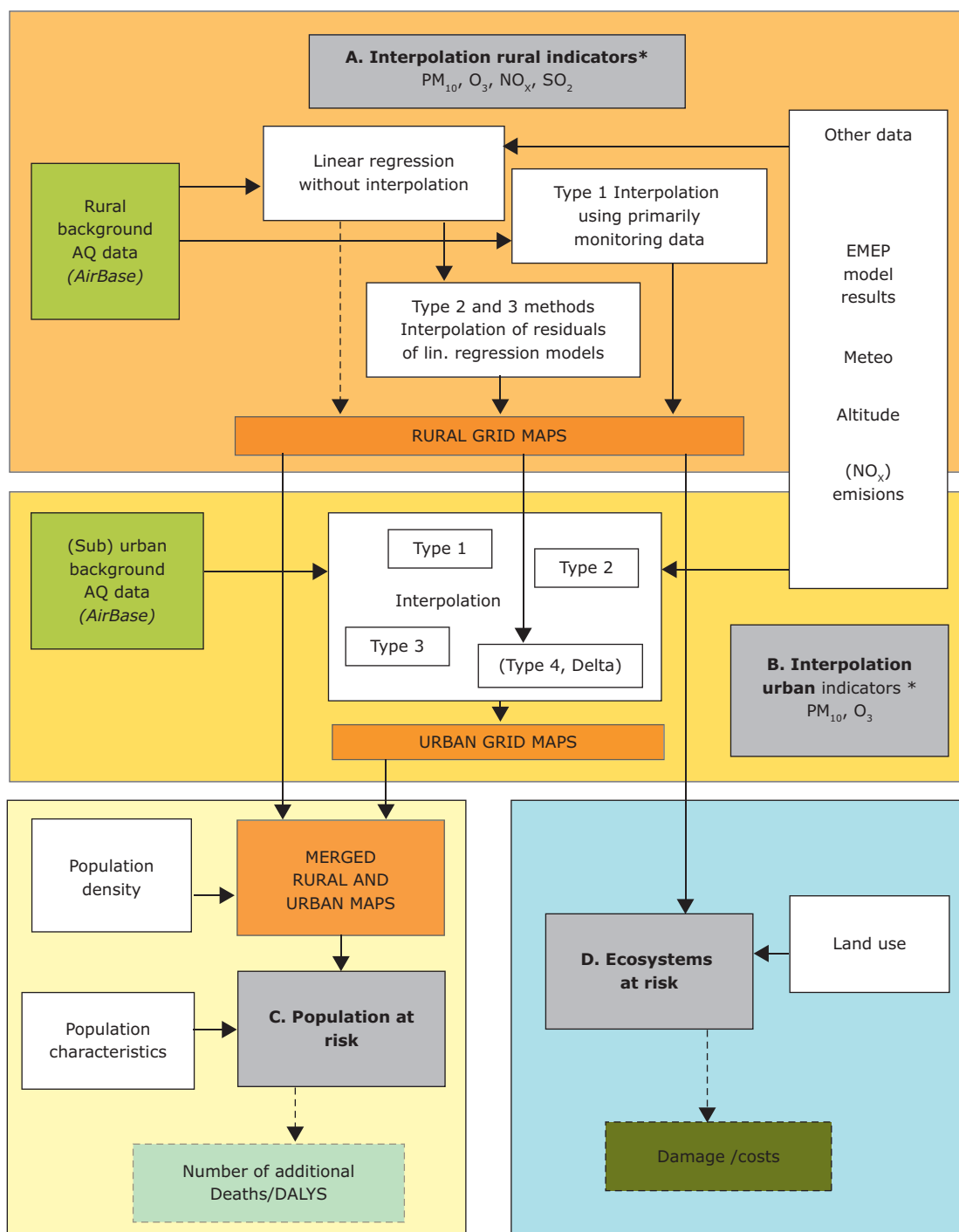
statistical correlation with the air quality data, thus providing more spatially resolved information about the whole territory in comparison to the pure air quality measurements. Examples of such supplementary data considered are meteorology, topography, population density and emissions. The linear regression analysis is primarily aiming at identifying and selecting the most descriptive supplementary parameters for inclusion in this third type. This type may be described as the multiple *linear regression models built with the use of monitoring, modelling and other supplementary data subsequently subjected to interpolation of their residuals*.

A detailed description of these methods is given in Annex 2. The description of such linear regression models and explanation of how they are used in order to select the most significantly correlating supplementary parameters for the interpolations can be found in Horálek *et al.*, (2007), Sections 2.4 and 2.5. The supplementary data sources considered in this report are described in more detail in Annex 1.

The three types of methodologies examined can be summarized by the following list (see also Annex 2).

- (1) Interpolation methods using primarily monitoring data are:
  - (a) ordinary kriging;
  - (b) lognormal kriging;
  - (c) ordinary co-kriging that uses the altitude of the measuring stations;
  - (d) lognormal co-kriging that uses the altitude of the measuring stations.
- (2) Linear regression models that use monitoring and chemical transport model data (1. EMEP; 2. LOTOS-EUROS) with a subsequent interpolation of its residuals through the use of ordinary kriging (residual kriging).
- (3) Linear regression models that use monitoring, chemical transport model data (EMEP) and other supplementary data with a subsequent interpolation of their residuals through the use of ordinary kriging (residual kriging).

**Figure 2.1 Process of building maps showing concentration of air quality indicators, exceedance of indicator concentrations and exposure estimates**



**Note:** \* indicators: pollutant concentration, but also derived statistics such as number of cases of exceeding limit or target values and exposure indicators like SOMO35<sup>(9)</sup> and AOT40<sup>(10)</sup>.

<sup>(9)</sup> SOMO35 (expressed in  $\mu\text{g m}^{-3}$  times hours) is the accumulated ozone concentration in excess of 35 ppb ( $70 \mu\text{g m}^{-3}$ ). It is the sum of the differences between the maximum daily 8-hour concentration exceeding  $70 \mu\text{g m}^{-3}$  for each day in the calendar year.

<sup>(10)</sup> AOT40 (expressed in  $\mu\text{g m}^{-3}$  times hours) stands for the sum of the differences between hourly concentrations greater than  $80 \mu\text{g m}^{-3}$  ( $= 40 \text{ ppb}$ ) and  $80 \mu\text{g m}^{-3}$  over a given period using only the 1 hour values measured between 8:00 and 20:00, Central European Time, each day. Target value for the protection of vegetation: AOT40, calculated from 1-hour values from May to July.

For all interpolation methods, i.e. different types of kriging, variogram parameters are estimated by the minimization of the cross-validation RMSE (root mean squared error).

For all three types of methods, the mutual comparison and uncertainty analyses are executed through cross-validation (for details, see Annex 2 and Horálek *et al.*, 2008) for all pollutants considered and for their indicators. Based on the cross-validation analysis and other criteria as described in Horálek *et al.* (2007), the selection was made for the preferred method.

The approaches described above are applied separately for the rural and for urban areas. The merging of the selected rural and urban maps into one combined map is done based on the aggregated 10 x 10 km<sup>2</sup> population density grid, applying the criterion as described in Annex 2.

Figure 2.1 presents a flow chart summarizing how the maps showing concentration and exceedance of air quality indicators are built and how we derive the exposure through the use of monitoring data, supplementary data, linear regressions and/or interpolation techniques.

To assess the impact of ozone on mortality, the UN-ECE Task Force on Health <sup>(11)</sup> recommends the adoption of a daily maximum 8-hour mean concentration as the principal benchmark. It should be followed with assessment over a full year. The Task Force stated that it was not possible to identify a threshold for the effects of ozone on mortality. However, in practice, a recommendation was made to the European CAFE Programme to use a cut-off for integrated assessment modelling at 35 ppb, and consider it to be a daily maximum 8-hour mean ozone concentration. For days when ozone concentration was higher than 35 ppb, only the increment exceeding 35 ppb has been used to calculate effects. No effects of ozone on health would then be calculated on days when it is less than 35 ppb, meaning that the exposure parameter is the sum of excess of daily maximum 8-h means

over the cut-off of 35 ppb calculated for all days per year. This parameter, the SOMO35 (sum of means over 35 ppb), is thus a measure of accumulated high exposure. Therefore, this metric was also considered in this study.

## 2.2 Interpolation uncertainty analysis

Four approaches to quantify interpolation uncertainty were applied in this project:

- (i) comparison of the maps created by the selected and alternative methods, using differences in these maps;
- (ii) cross-validation analysis, using statistical indicators and their scatter plots;
- (iii) non cross-validation scatter plots for comparison of measured and estimated values; and
- (iv) the creation of uncertainty maps additional to the interpolated maps produced with the preferred interpolation method.

Principle results of the uncertainty analysis presented for particular indicators in this report are based on the cross-validation analysis.

## 2.3 Exceedance probability mapping

Based on the concentration and uncertainty maps, there have been constructed the maps of the probability of exceedance (PoE) of a specific threshold value (e.g. limit or target value).

Maps showing the probability of exceeding limit or target values, calculated according to the procedure described in Annex 3, are also presented in the report. The probability of exceeding the limit or target value is estimated from the uncertainties in the interpolation methods and from the actual concentrations in each grid cell. The resulting maps provide guidance on further actions with respect to measures aimed at the implementation of abatement and the design of monitoring networks.

<sup>(11)</sup> UN ECE Task Force on Health, <http://www.unece.org/env/lrtap/WorkingGroups/wge/who.htm>.



### 3 PM<sub>10</sub> maps for 2005

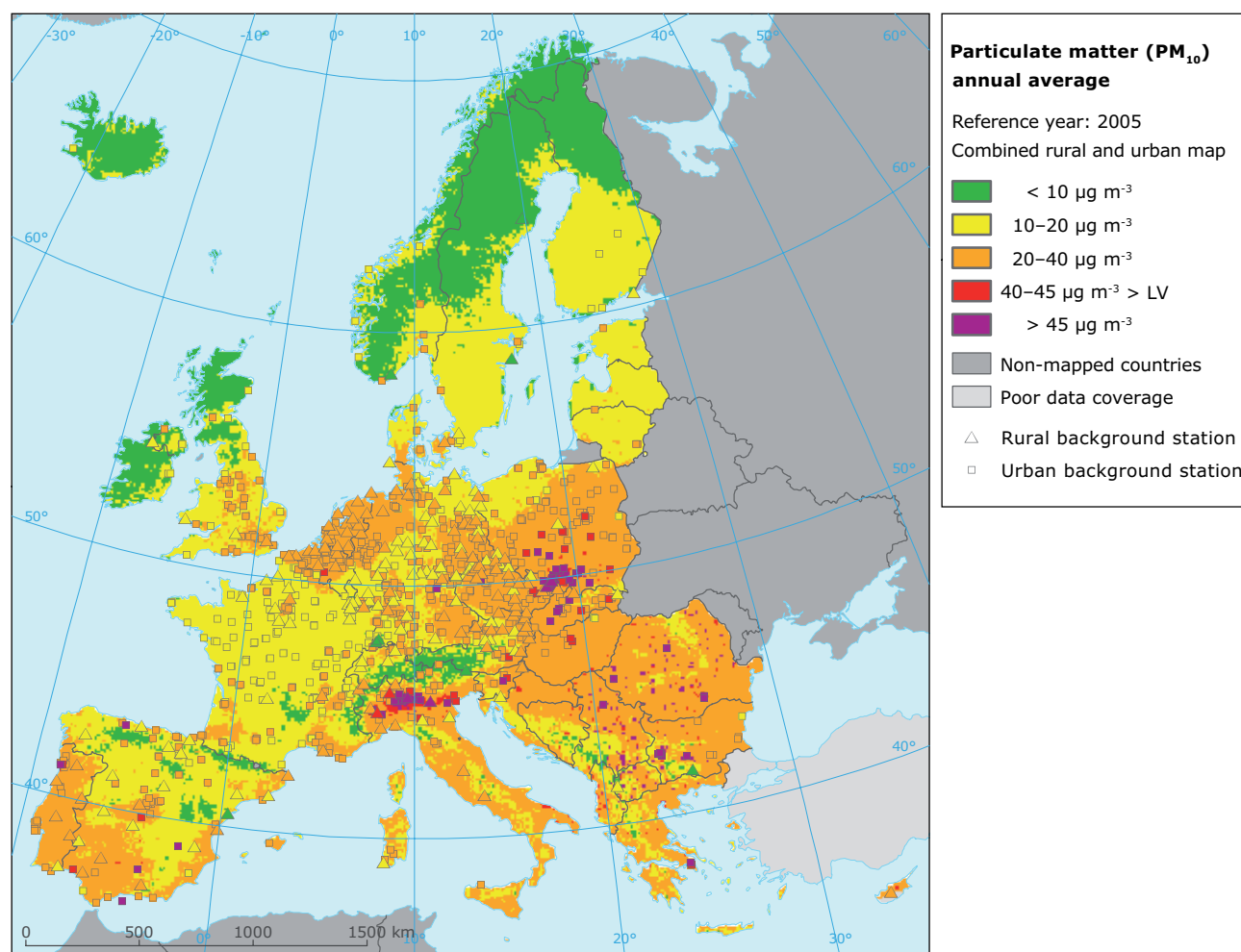
For PM<sub>10</sub>, the two health-related indicators considered are the annual average and 36th highest maximum daily average. The analyses and selection of the best and/or preferred interpolation method was carried out separately for the rural area and the urban ones (Table 7.1). Based on the selected method for each indicator, the rural and urban maps are then compiled. From these maps, a combined interpolated PM<sub>10</sub> indicator map is created, the one covering the whole of Europe and using, as criterion a population density relation. The process of preparing separate maps for rural and urban areas is described in detail in Annex 2.

#### 3.1 PM<sub>10</sub> annual average map

The combined interpolated map for the 2005 PM<sub>10</sub> annual averages (Map 3.1) is created by combining the rural and urban maps and using a 10 × 10 km<sup>2</sup> grid showing the aggregated population density field. It is done in accordance with the criteria described in Horálek *et al.* (2007).

The rural map is created by combining the annual averages of the measured PM<sub>10</sub> concentrations obtained from the rural background stations and the supplementary data from the EMEP model

**Map 3.1 Combined rural and urban concentration map of PM<sub>10</sub> — annual averages, the year 2005. Spatial interpolated concentration fields and the measured values at the single measurement station**



**Note:** Unit: µg m<sup>-3</sup>. The limit value is 40 µg m<sup>-3</sup>.

output, the altitude field, wind speed and surface solar radiation in a multiple linear regression model, followed by residual kriging. It should be noted, though, that the methods based on lognormal co-kriging (using altitude) give slightly better results. However, the method selected has been the one described, as it gives a better European coverage.

The 2005 urban map is created by combining the measured PM<sub>10</sub> annual averages obtained from the urban and suburban background stations with the EMEP model output only in a linear regression, followed by the interpolation of its residuals by ordinary kriging.

The areas and stations in the combined map where the limit value of 40 µg m<sup>-3</sup> is exceeded are coloured red and purple. Comparing this map with the 2004 concentration map (Horálek *et al.*, 2007), a similar spatial pattern is observed but the concentration

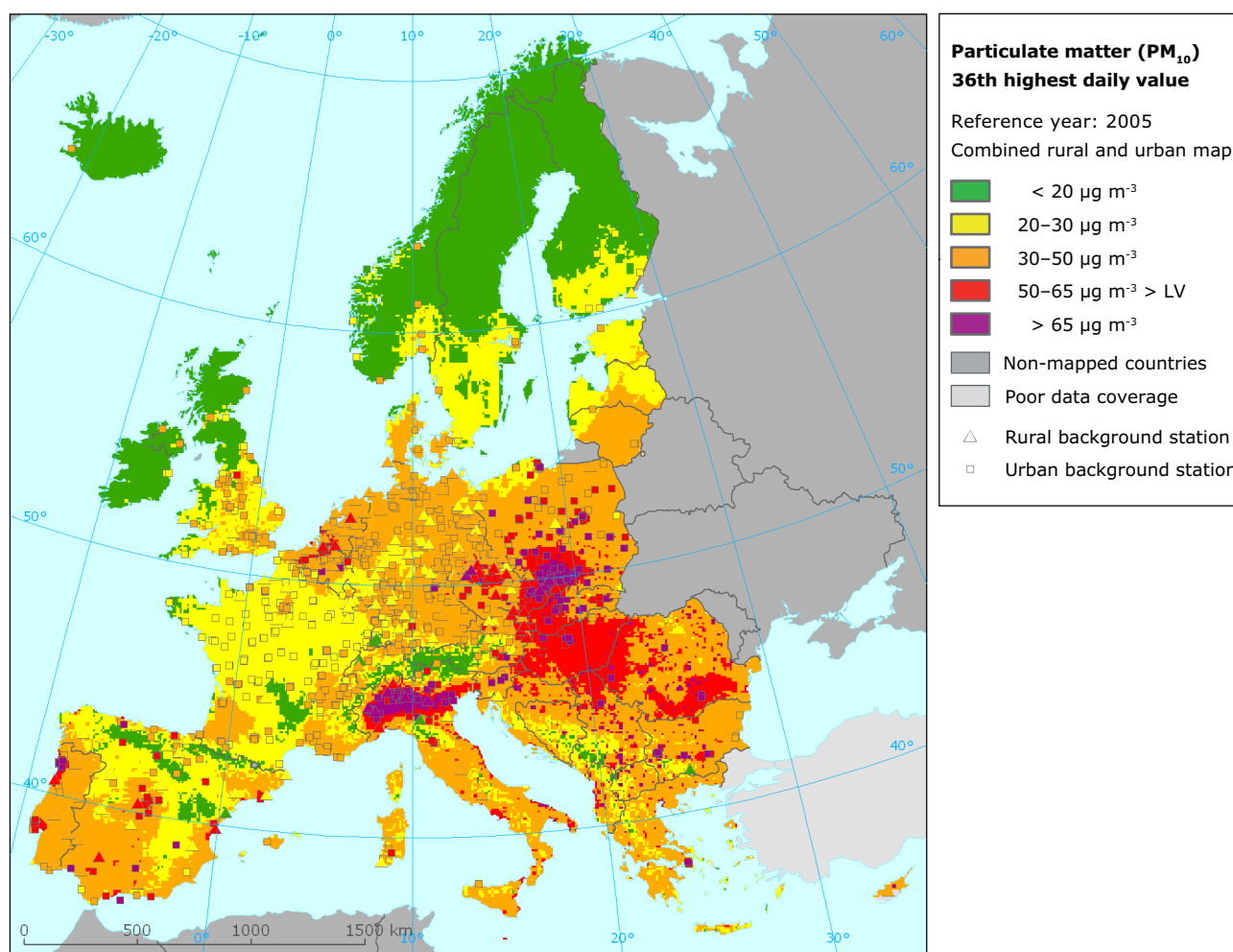
in 2005 tends to be slightly higher than in 2004. Similarly, a small increase can also be seen in the monitoring data viewed separately (Mol *et al.*, 2007).

### 3.2 PM<sub>10</sub> 36th highest daily average map

The combined map for the 2005 PM<sub>10</sub> 36th highest daily averages (Map 3.2) is created by the same interpolation methods as the annual mean map. Because of its good European coverage, this method is recommended as 'standard' in future applications for both indicators.

The areas and stations in the combined map where the limit value of 50 µg m<sup>-3</sup> is exceeded are coloured red and purple. Compared to the situation in 2004, the 2005 concentrations in central-eastern Europe and the Po valley seem to be higher than last year, while the levels observed along the Atlantic coast and the North Sea are lower.

**Map 3.2 Combined map of rural and urban concentration of PM<sub>10</sub> – 36th maximum daily average value, 2005**



**Note:** Unit: µg m<sup>-3</sup>. The limit value is 50 µg m<sup>-3</sup>.

In comparison to its neighbouring countries, the PM<sub>10</sub> concentrations in France are relatively low (both for annual and for 36th maximum daily average values). Probably, these low levels are biased — due to the fact that PM<sub>10</sub> monitoring data from France are not corrected, although the recommended reference method was not applied (mostly TEOM) (de Leeuw, 2005).

### 3.3 Population exposure and health impacts

The final concentration map of the annual PM<sub>10</sub> mean concentration (Map 3.1) shows increased concentrations in the urbanized areas in Europe.

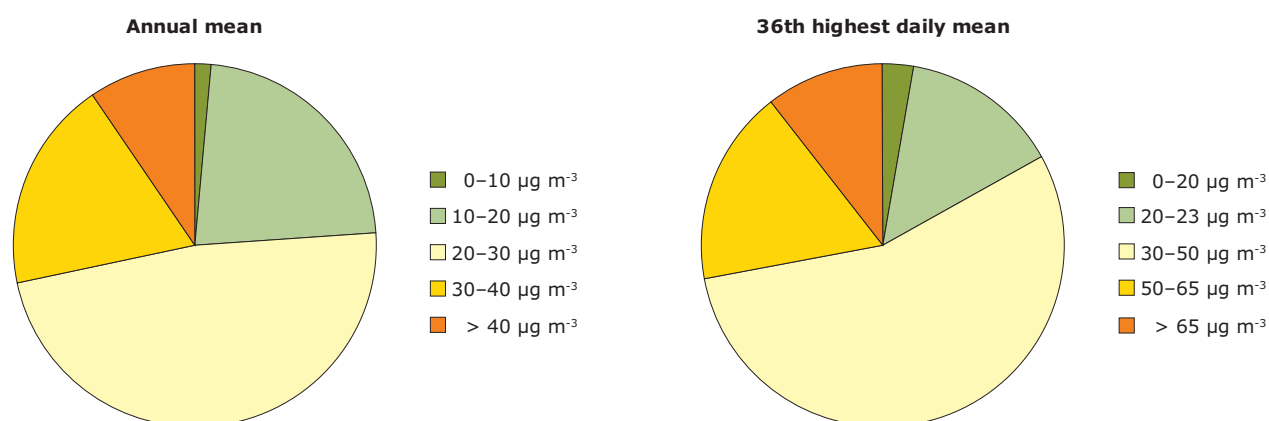
Table 3.1<sup>(12)</sup> and Figure 3.1 show the distribution of the population frequency for a limited number of exposure classes. Almost a quarter (23 %) of the European population is exposed to concentrations below the WHO Guideline value of 20 µg m<sup>-3</sup> (WHO, 2001). In 2005, about two thirds of the European population lived in areas where the PM<sub>10</sub> concentration is estimated to have been between 20 and 40 µg m<sup>-3</sup>. About 9 % of the population live in areas where the PM<sub>10</sub> annual limit value is exceeded. However, as the mapping methodology

tends to underestimate high values, in reality the figure of 9 % is probably higher<sup>(13)</sup>. The frequency distribution shows a large variability over Europe; in six countries (Albania, Bulgaria, Cyprus, the former Yugoslav Republic of Macedonia, Greece and Romania), it is estimated that more than a quarter of the population is exposed to concentrations above the limit value. In a number of countries in the north and north-west of Europe, the limit value does not seem to be exceeded at the 10 x 10 km<sup>2</sup> level applied in the mapping.

In general, we must not forget that the interpolated 10 x 10 km<sup>2</sup> maps are only smooth estimates of the real pollution. They tend to overestimate average concentrations and underestimate both low and high values. Consequently, the exposure of the population to both high and low concentrations is underestimated, while the exposure to average concentrations is overestimated. Such bias can be significant, particularly in under-sampled areas.

In comparison to the annual limit value, the daily limit value of PM<sub>10</sub> is exceeded over a much larger part of Europe (Figure 3.1; see also the map in Map 3.2 and Table 3.2). In 2005, more than 25 % of the European population were exposed to ambient concentrations of above 50 µg m<sup>-3</sup> for more than 35 days (limit value of daily PM<sub>10</sub> concentrations).

**Figure 3.1 Exposure of the European population to PM<sub>10</sub> concentrations, annual mean (left) and 36th highest daily mean (right), (reference year 2005)**



<sup>(12)</sup> When providing the tables showing the exposure of the population, all countries shown are split into two groups. The reason for doing it is as follows. The exposure was calculated with the help of different databases of population densities (see also Annex 1, A 1.7). Population density [inhbs km<sup>-2</sup>] is based on the data from the EC's Joint Research Centre describing the majority of countries. For the countries not included in this database (i.e. Andorra, Albania, Bosnia and Herzegovina, Cyprus, Iceland, Liechtenstein, the former Yugoslav Republic of Macedonia, Norway, Serbia and Montenegro, Switzerland, and Turkey), the population density was calculated on the basis of the data from an alternative source, the ORNL LandScan (2002) Global Population Dataset.

<sup>(13)</sup> Kriging in general 'smoothens' the interpolation field and the interpolated value is the average of a 10 x 10 km grid.

**Table 3.1 Population exposure and population weighted concentration – PM<sub>10</sub> annual average, 2005**

Country	% of population					Population-weighted conc. $\mu\text{g m}^{-3}$
	< 10 $\mu\text{g m}^{-3}$	10–20 $\mu\text{g m}^{-3}$	20–40 $\mu\text{g m}^{-3}$	40–45 $\mu\text{g m}^{-3}$	> 45 $\mu\text{g m}^{-3}$	
Austria	5.8	16.9	77.4	0	0	23.5
Belgium	0	2.8	97.2	0	0	28.9
Bulgaria	0.3	4.5	65.3	3.2	26.7	37.2
Croatia	0	4.4	87.5	8.1	0	30.6
Czech Rep.	0	1.5	88.8	2.9	6.8	31.5
Denmark	1.2	40.8	58.0	0	0	19.7
Estonia	2.4	83.7	14.0	0	0	16.3
Finland	7.9	92.1	0.0	0	0	13.3
France	0.5	60.9	38.5	0	0	19.1
Germany	0.1	22.7	77.2	0	0	22.1
Greece	0.1	7.2	48.7	35.4	8.5	34.7
Hungary	0	0	98.0	2	0	33.5
Ireland	40.0	60.0	0.0	0	0	11.4
Italy	0.6	5.1	71.8	10.1	12.4	32.7
Latvia	1.3	58.6	40.1	0	0	18.7
Liechtenstein	0	0	100	0	0	21.4
Lithuania	0	54.0	46.0	0	0	20.3
Luxembourg	0	100	0.0	0	0	18.4
Malta	0	0	100	0	0	36.5
Netherlands	0	0.2	99.8	0	0	29.1
Poland	0	8.1	75.6	4.1	12.2	30.5
Portugal	0	1.8	85.3	12.9	0.0	30.6
Romania	0	1.9	59.2	12.3	26.7	37.4
San Marino	0	0	100	0	0	24.9
Slovakia	0	2.0	92.6	5.4	0	31.4
Slovenia	0.1	11.5	88.4	0	0	27.5
Spain	0.6	15.8	82.7	0.8	0	27.4
Sweden	8.8	84.6	6.6	0	0	15.0
United Kingdom	2.3	30.5	67.2	0	0	20.8
Albania	1	13.5	39.5	32.9	12.9	33.8
Andorra	18	16.1	65.7	0.0	0.0	16.2
Bosnia and Herzegovina	0.6	17.7	60.7	20.9	0.0	30.1
Iceland	47.4	52.6	0.0	0	0	9.5
the former Yugoslav Republic of Macedonia,	1.2	9.9	25.5	5.2	58.2	42.7
Norway	15.6	53.2	31.1	0.0	0.0	16.5
Serbia and Montenegro	0	6	47	7	40	38.7
Switzerland	3	42.0	54.8	0.0	0	19.8
<b>Total</b>	<b>1.4</b>	<b>23.0</b>	<b>66.7</b>	<b>3.8</b>	<b>5.2</b>	<b>26.2</b>

**Note:** Countries for which air quality or population density data are missing (Cyprus, Turkey) have been excluded from calculations in this paper. To adjust for the differences in dense versus less populated areas, only weighted annual values measured at urban AirBase background stations in agglomerations were used in this report <sup>(14)</sup>.

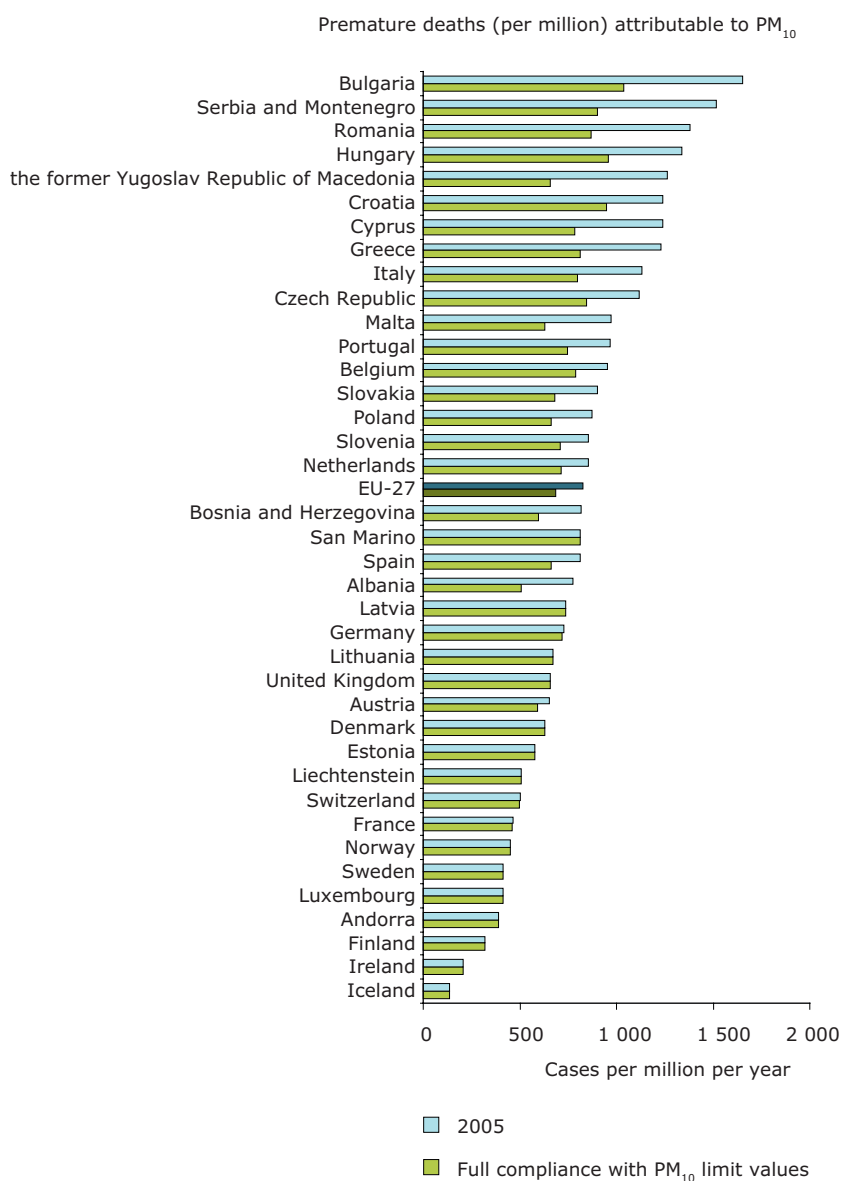
<sup>(14)</sup> Weighted concentration =  $\sigma(\text{population per grid cell times concentration per cell})/\sigma(\text{population per cell})$ .

**Table 3.2 Population exposure and population weighted concentration – PM<sub>10</sub>, 36th maximum daily average value, 2005**

Country	% of population					Population-weighted conc. $\mu\text{g m}^{-3}$
	< 20 $\mu\text{g m}^{-3}$	20–30 $\mu\text{g m}^{-3}$	30–50 $\mu\text{g m}^{-3}$	50–65 $\mu\text{g m}^{-3}$	> 65 $\mu\text{g m}^{-3}$	
Austria	5.4	7.4	54.9	30.0	2.3	42.3
Belgium	0	2.3	71.7	26.0	0	46.5
Bulgaria	0.1	2.1	54.6	9.5	33.7	62.7
Croatia	0.0	2.2	46.8	31.2	19.7	52.0
Czech Rep.	0	0.0	31.3	49.5	19.3	57.5
Denmark	2.9	15.2	81.9	0	0	32.7
Estonia	5.3	52.9	41.8	0	0	28.8
Finland	33.1	66.9	0	0	0	22.1
France	1.9	51.9	46.2	0	0	29.6
Germany	0.1	7.5	90.9	1.5	0	37.2
Greece	0.3	5.7	37.9	42.0	14.2	54.1
Hungary	0	0	9.1	66.0	24.8	59.1
Ireland	63.6	36.4	0	0	0	14.8
Italy	0.5	2.9	39.2	30.6	26.9	56.2
Latvia	1.8	33.6	64.6	0	0	33.6
Liechtenstein	0	0	100	0	0	36.2
Lithuania	1.2	9.3	89.4	0	0	36.8
Luxembourg	0	30.7	69.3	0	0	30.6
Malta	0	0	14.5	85.5	0	61.6
Netherlands	0	0	76.6	23.4	0	47.4
Poland	0.1	2.0	49.5	27.4	21.0	54.7
Portugal	0	0.3	44.6	37.9	17.3	51.7
Romania	0.0	0.3	29.6	29.3	40.8	63.9
San Marino	0	0	100	0	0	39.8
Slovakia	0	0.1	27.6	59.7	12.6	55.3
Slovenia	0.0	1.6	53.8	44.6	0	48.4
Spain	1.2	10.2	50.2	38.2	0.2	43.7
Sweden	25.8	50.7	23.5	0	0	24.0
United Kingdom	6.3	22.9	70.9	0	0	31.4
Albania	1.1	10.2	29.6	13.3	45.8	55.4
Andorra	18.3	16.1	65.7	0	0	28.7
Bosnia and Herzegovina	0.9	11.2	42.8	14.6	30.5	50.1
Iceland	100	0	0	0	0	9.6
the former Yugoslav Republic of Macedonia	1.2	5.2	28.9	0	64.7	71.4
Norway	31.6	32.5	35.9	0	0	26.1
Serbia and Montenegro	0.6	4.2	27.0	20.7	47.6	63.9
Switzerland	2.3	13.6	82.0	1.2	1.0	33.9
<b>Total</b>	<b>2.9</b>	<b>14.6</b>	<b>54.8</b>	<b>17.4</b>	<b>10.2</b>	<b>43.5</b>

**Note:** Countries for which air quality or population density data are missing (Cyprus, Turkey) have been excluded from calculations in this paper. To adjust for the differences in densely populated versus less populated areas, only weighted annual values measured at urban AirBase background stations in agglomerations were used in this report.

**Figure 3.4** Number of premature deaths per million inhabitants attributable to PM<sub>10</sub> exposure in the reference year 2005



**Note:** The 'no\_ex scenario' corresponds to the (hypothetical) situation that the daily limit value were not exceeded at European hot-spot locations.

### 3.4 Health impact assessment

In a health impact assessment, the number of premature deaths attributable to long-term exposure to PM<sub>10</sub> has been estimated. An update of country-specific demographic data has been taken from the UN Population Division (UN, 2006). The health impact assessment is performed according to standard population attributive principles (WHO, 2001). Use has been made of a relative risk of 4.3 % per 10 µg m<sup>-3</sup> PM<sub>10</sub> for total mortality (excluding violent deaths, adults of 30 years of age and older) (Künzli *et al.*, 2000).

The natural background PM<sub>10</sub> concentration for the whole of Europe is difficult to assess. Regional variations are high, e.g. due to the presence of the sea-salt spray in strips close to the coastline, PM re-suspension from soils or because of secondary aerosol formation from biogenic organics. In this study, scenarios applied were assuming natural background concentration across Europe as a constant of 5 µg PM<sub>10</sub> m<sup>-3</sup>. This value for the background concentration was subtracted from the interpolated concentrations.

The estimated number of premature deaths per million inhabitants attributable to PM<sub>10</sub> exposure is given in Figure 3.4. The observed range is partly caused by the differences in PM<sub>10</sub> concentration over Europe and partly — by the differences in age distributions and baseline mortalities. The uncertainties in the numbers caused by the uncertainties in the relative risk factor are relatively large: for the EU-27 as a whole, the number ranges from 510 to 1 150 deaths per million, with a statistically best estimate of 830 deaths per million inhabitants. The estimates are in good agreement with the results of the CAFE Programme. The figure of 830 premature deaths per million inhabitants correspond to about 373 000 premature deaths in the EU-25 countries in 2005, assuming a PM<sub>10</sub> natural background concentration of 5 µg m<sup>-3</sup>. The CAFE Programme results included estimation of 348 000 premature mortalities due to anthropogenic emissions of primary PM and PM precursors in the year 2000 <sup>(15)</sup>.

The results in Figure 3.4, labelled as 'no\_ex scenario' correspond to a sensitivity calculation in which it is assumed that the daily limit value (a daily mean of 50 µg m<sup>-3</sup> that may not be exceeded on more than 35 days per year) is exceeded nowhere in Europe, not even at hot-spot locations. This situation has been simulated by truncating the annual mean concentration calculated for each 10 x 10 km<sup>2</sup> grid cell to 25 µg m<sup>-3</sup> and re-estimating the number of premature deaths using the truncated concentration field. The rationale for the truncation value of 25 µg m<sup>-3</sup> is as follows. The monitoring data show that the daily limit value of PM<sub>10</sub> is equivalent to an annual average concentration of 31 µg m<sup>-3</sup> (see e.g. Buijsman *et al.*, 2005 and Stedman *et al.*, 2007). In our mapping exercise, the concentration in the 10 x 10 km<sup>2</sup> grid cell is assumed to be representative for the rural or urban background situations. On the average, the annual average concentration at a traffic hotspot is 20 to 25 % higher than at an urban background station (see e.g. Mol *et al.*, 2007). Therefore, with a background concentration of 25 µg m<sup>-3</sup>, exceeding the daily limit value at hotspots may largely be avoided.

In this 'no\_ex scenario', the reduction in the number of premature deaths is particularly significant in the central eastern European countries. For the EU-27, we estimate a reduction of about 17 % to be an average. However, even if the limit values are met in all the EU-27 countries, the health problem shall not

be solved: a substantial number of premature deaths can still be expected.

### 3.5 Uncertainties and probability of exceeding the limit values

#### Annual PM<sub>10</sub> averages

The analysis of uncertainties, presented in detail in Horálek *et al.* (2008), shows that the absolute mean interpolation uncertainty in the combined map is equal, in the rural areas (taken from the rural map), to 5.5 µg m<sup>-3</sup>. This refers to a relative interpolation uncertainty of about 26 % of the mean of the measurement-based PM<sub>10</sub> annual averages at all the rural background stations. For the urban areas (taken from the urban map), the standard uncertainty/error was 5.5 µg m<sup>-3</sup>, i.e. about 20 % at all urban and suburban background stations.

#### PM<sub>10</sub> 36th maximum daily average

The absolute mean interpolation uncertainty in the combined map is (for the rural areas in this map) 9.7 µg m<sup>-3</sup>. This refers to a relative interpolation uncertainty of about 26 % of the mean of the measurement-based PM<sub>10</sub> 36th maximum daily average at all rural background stations. For the urban areas in the combined map, uncertainties are 9.9 µg m<sup>-3</sup> and about 21 %, respectively, at all urban and suburban background stations (for details, see sections A.2.1.3 and A 2.2.3 in Horálek *et al.*, 2008).

#### Probability of exceeding the limit values

The maps showing the probability of exceeding the limit values (Maps 3.3 and 3.4) were computed according to the procedure described in Annex 3. The process was based on the use of concentration maps (Maps 3.1 and 3.2) and the uncertainty maps — along with the results of the interpolation uncertainty analysis (Chapter 2.2 and Horálek *et al.*, 2008). The uncertainty in the probability maps arises only from the uncertainties caused by the interpolation and innate spatial variability of concentrations within a grid cell. Neither uncertainties in the measurements or the supplementary data nor those caused by the combination of urban and rural maps are included.

After constructing the uncertainty map, it is possible to compile the probability map — using the concentration value given for each grid cell

<sup>(15)</sup> See for example, [http://ec.europa.eu/environment/air/pdf/sec\\_2005\\_1133.pdf](http://ec.europa.eu/environment/air/pdf/sec_2005_1133.pdf).

of the concentration map and its corresponding uncertainty value for this particular grid cell. The probability of exceeding the limit values for a particular grid cell can be estimated by taking into account the respective limit value and assuming a Gaussian distribution.

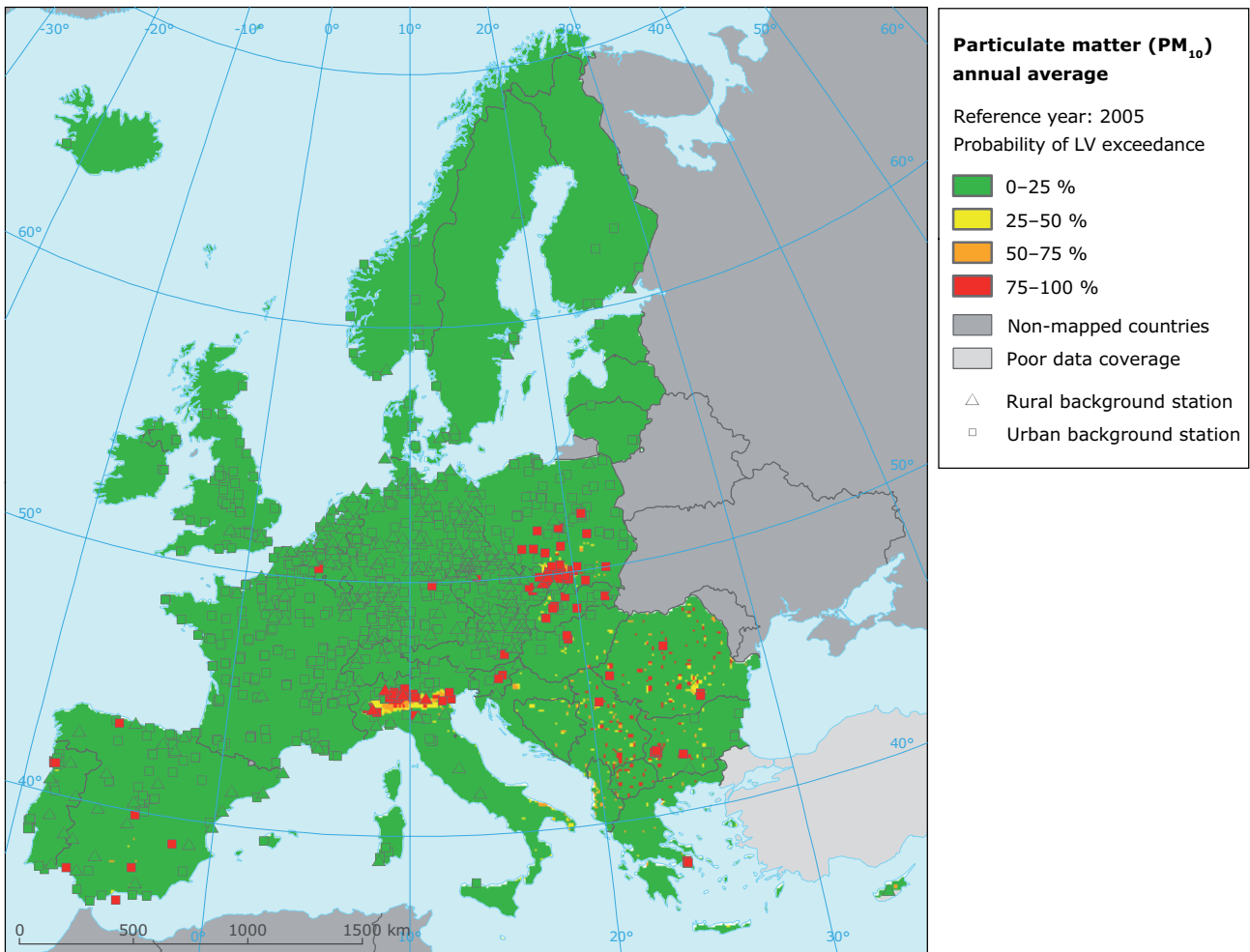
Map 3.3 and 3.4 show the map of estimated probability of exceeding the annual average for PM<sub>10</sub> and the 36th maximum daily mean, respectively. Areas where the probability of exceeding the limit value is above 75 % are marked in red; areas below the 25 % probability are marked in green.

Apart from the estimated probability of exceeding the limit value in a grid cell, the maps (Map 3.3 and 3.4) also show the single measurement stations (squares and triangles in Map 3.3 and 3.4), indicating cases of exceeding the limit value (marked red) or

non-exceeding it (marked green) at those stations in 2005.

The estimated probability of exceeding the annual averages for PM<sub>10</sub> (Map 3.3) is moderate to considerable in urban areas of the Balkan region (red grid cells, i.e. larger than 75 % probability). In the southern Poland, the Czech Republic, Hungary and southern Spain, the probability of exceeding these values is also relatively high. In the Balkan area, southern Poland and the Italian Po Valley, the whole regions show a probability of exceeding the limit as ranging from 25 to 50 %, or to even higher levels of 50 to 75 % and more in their centres. The analyses suggest that the likelihood of annually exceeding the limit for PM<sub>10</sub> is very strong in those regions. It is also clear that considerable improvements in air quality are needed — if we were to discuss reaching the target (low) levels in the future. The situation in

**Map 3.3** Map showing the probability of exceeding the limit value for the annual average of the PM<sub>10</sub> indicator (in µg m<sup>-3</sup>) on the European scale in 2005



**Note:** The spatial resolution is 10 x 10 km<sup>2</sup> (grid). Single stations with annual average measurement values above or below the limit value are marked red or green, respectively.



the countries of north-western and northern Europe is better, i.e. the probability of exceeding the limit is only  $\leq 25\%$ , indicating that policy targets have been or might be met in the background areas.

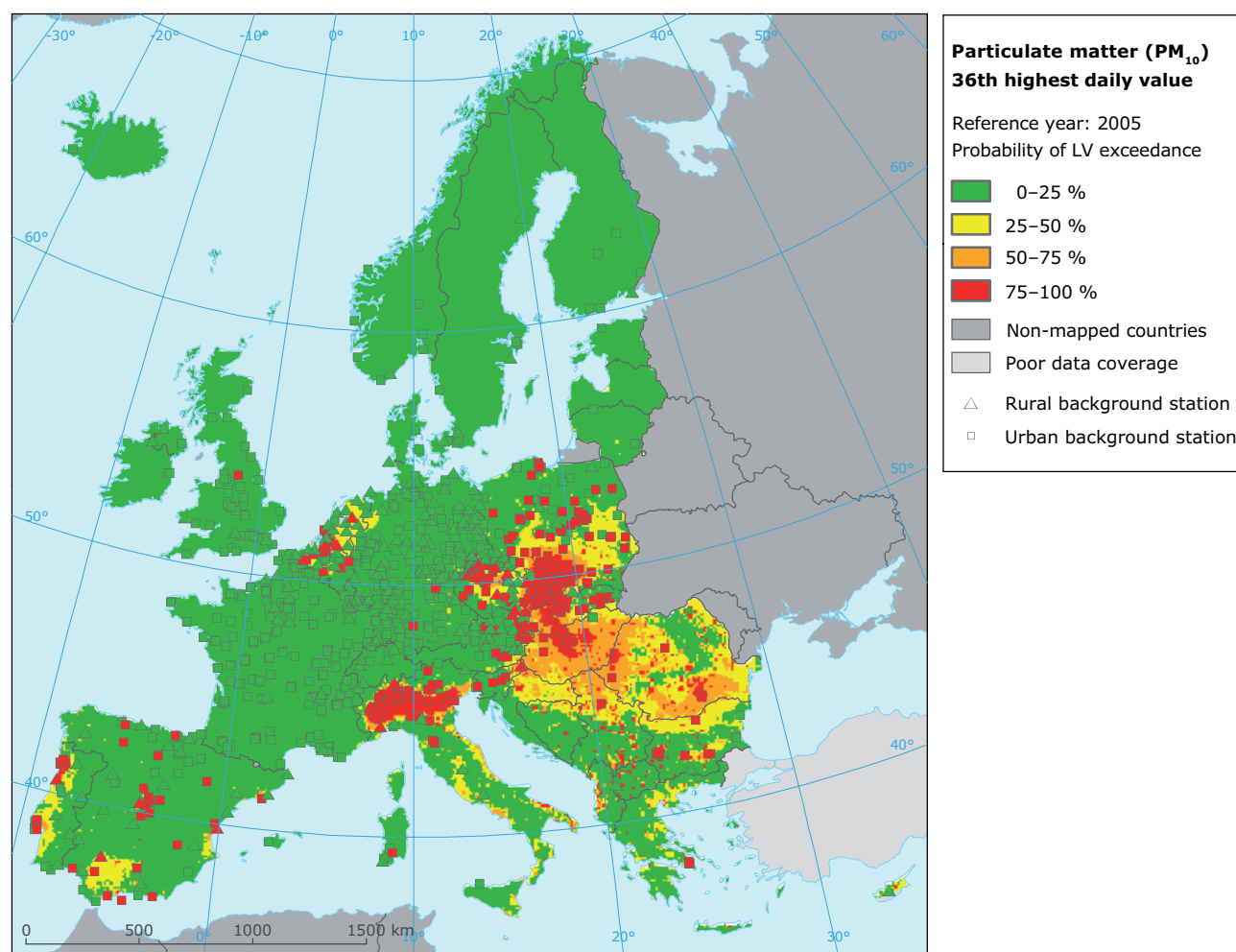
The 10 x 10 km<sup>2</sup> map does not give information on the possible cases of exceeding the limit values locally. The low percentages of such probability for France may be a result of the French networks not applying correction factors for PM<sub>10</sub> measurements.

The estimated probability of exceeding the PM<sub>10</sub> 36th maximum daily average (Map 3.4) is considerable in large areas of the eastern European countries and along the entire Po Valley (red areas indicating larger than 75 % probability of exceeding the values). The analyses suggest that the value for PM<sub>10</sub> is likely to be exceeded there on a daily basis and considerable improvements in air quality are

needed to prevent this overstepping of levels in the future. The estimated probability of exceeding the target values is lower in Spain, Portugal, Italy, Greece, some of the Balkan countries, Belgium, the Netherlands and Luxembourg where this probability ranges from 25 to 50 %, with increased levels of 50 to 75 % in the more urbanized centres of the regions. In the northwestern and northern European countries, the probability of exceeding the levels is lower, i.e.  $\leq 25\%$ , suggesting that for rural background areas the policy targets have been or might be reached.

The 10 x 10 km<sup>2</sup> map does not give information on possible local cases of exceeding this level. The low probability percentages for France may be explained by the fact that in the French networks did not apply correction factors for PM<sub>10</sub> measurements in 2005.

**Map 3.4** Map showing the probability of exceeding the limit value for the PM<sub>10</sub> indicator – 36th maximum daily mean (in  $\mu\text{g m}^{-3}$ ) – on the European scale in 2005



**Note:** The spatial resolution is 10 x 10 km<sup>2</sup> (grid). Single stations with annual averaged measurement values above or below the limit value are marked red or green, respectively.

## 4 Ozone maps for 2005

The ozone indicators addressing human health analysed in this report are the:

- (1) The average for 26th highest daily maximum 8-hour concentration (target value from Air Quality Framework Directive, 3rd Daughter Directive (or 'the Ozone' Directive; EC 2002); and
- (2) SOMO35, i.e., the accumulated ozone concentration in excess of 35 ppb ( $70 \mu\text{g m}^{-3}$ ).

The SOMO35 is the sum of the differences between the maximum daily 8-hour concentrations exceeding  $70 \mu\text{g m}^{-3}$  for each day in the calendar year.

The two indicators chosen for vegetation were:

- (1) AOT40 for crops, i.e. the accumulated exposure over a threshold of 40 ppb — or  $80 \mu\text{g m}^{-3}$ , over a period of 3 months (May–July);
- (2) AOT40 for forests, i.e. the accumulated exposure over a threshold of 40 ppb — or  $80 \mu\text{g m}^{-3}$ , over a period of 6 months (April–September).

The AOT40 is the sum of the differences between the hourly mean ozone concentration (in ppb or  $\mu\text{g m}^{-3}$ ) and  $80 \mu\text{g m}^{-3}$  when the concentration exceeds  $80 \mu\text{g m}^{-3}$  during daylight hours, accumulated over a period of three (for crops) or six (for forests) months.

For the health-related indicators, the most suitable and preferred methods for interpolation mapping were analyzed separately for rural and urban areas. For the vegetation-related indicators, only rural maps were considered for the selection of preferred mapping methods, assuming that no relevant sensitive vegetation can be found in the urban areas.

### 4.1 Ozone health related indicators

The combined interpolated map for both ozone health indicators, 26th highest daily maximum 8-hour average ozone concentrations and SOMO35, is presented in Map 4.1. Both maps have been compiled by combining the rural and urban maps.

The rural maps are created by combining the measurement data from rural background stations with the data from the EMEP model output, altitude and surface solar radiation in a multiple linear regression model, which was followed by the interpolation of its residuals by ordinary kriging. Although this method does not give the best statistical results, it has been preferred here because of its better geographical coverage and consistency with the ozone indicators aiming at protecting vegetation. All rural ozone maps are compiled by using the same method, which is also recommended for future application.

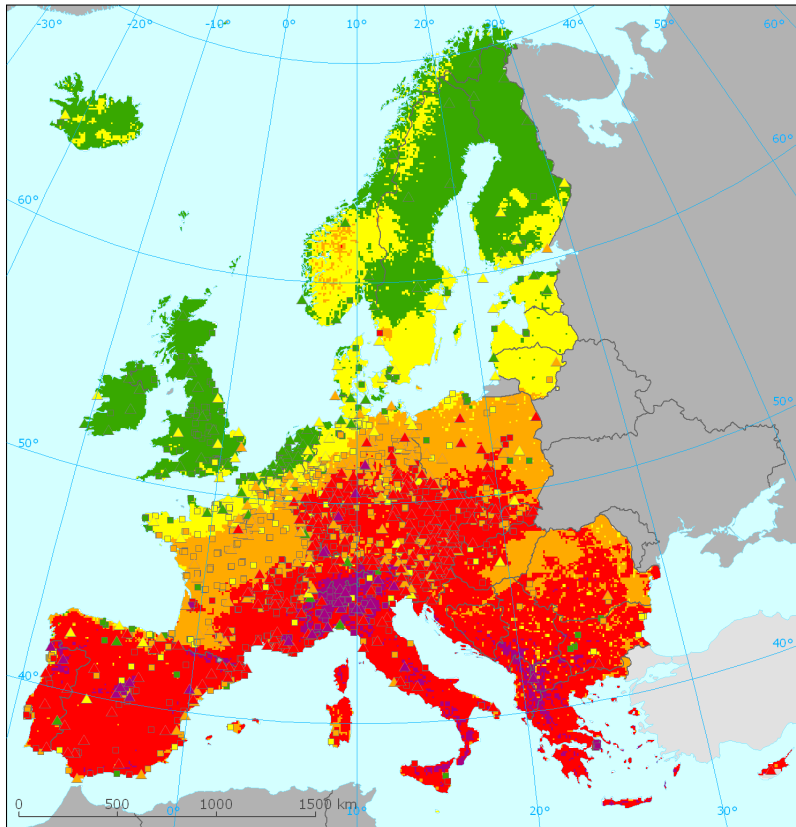
For both human health indicators, the urban maps have been created by combining the measurement data from the urban and suburban background stations with the data from the EMEP model output, wind speed and surface solar radiation in a linear regression model, which was followed by interpolation of its residuals by ordinary kriging. This method is recommended for future applications because of the better spatial coverage compared to other tested methods (ordinary kriging with EMEP model results only).

#### 4.1.1 Population exposure and health impacts

The map of the 26th highest maximum daily value (Map 4.1, top) shows somewhat higher values in the central and eastern parts of Europe and lower levels in the northern regions. The SOMO35 map (Map 4.1, bottom) suggests, in general, that the concentrations in 2005 were, in both magnitude and spatial distribution, similar to the concentrations in 2004. However, in 2005, there was an increase in SOMO35 values in southeastern Europe and the eastern Mediterranean. In the EU legislation, no limit or target values are set for SOMO35. Table 4.2 shows that values are varying significantly from country to country as well.

The population exposure in terms of both parameters is shown in Figure 4.2 (note that in contrast to Tables 4.1 and 4.2, Figure 4.2 includes all countries shown in Map 4.1). In 2005, 38 % of the European population was exposed to ozone levels above the target value ( $120 \mu\text{g m}^{-3}$ , 26th highest daily maximum 8-hour average; Figure 4.2). As Table 4.1 shows, this fraction

**Map 4.1 Combined maps of rural and urban concentration of the ozone health indicators – 26th highest daily maximum 8-hour value in  $\mu\text{g m}^{-3}$  (top) and SOM035 in  $\mu\text{g m}^{-3}$  times days (bottom), 2005**

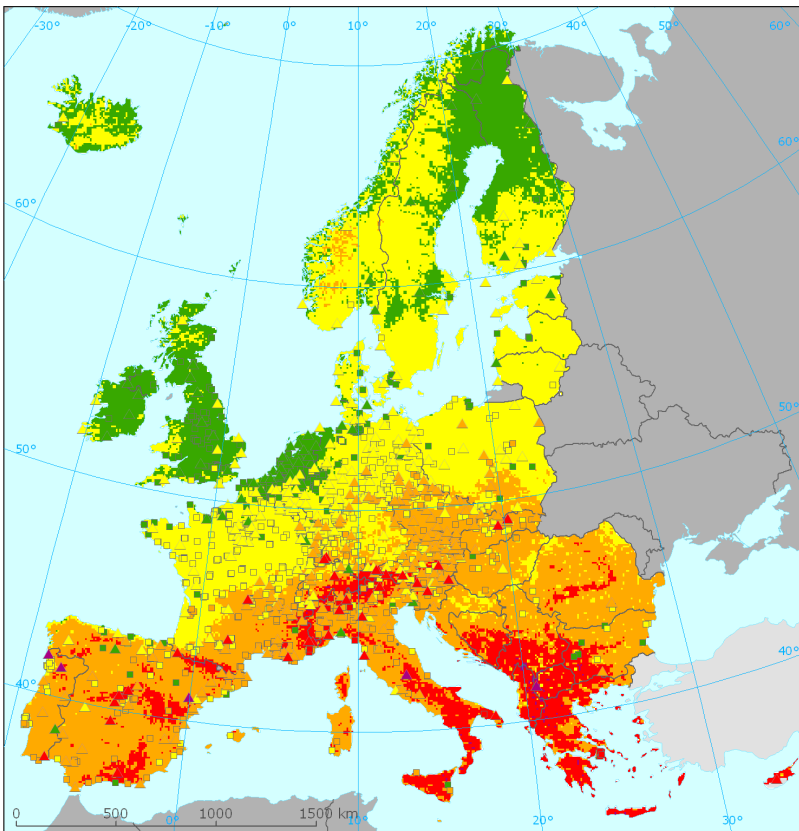


**Ozone – 26th highest maximum 8-hour daily value**

Reference year: 2005

Combined rural and urban map

- < 100  $\mu\text{g m}^{-3}$
- 100–110  $\mu\text{g m}^{-3}$
- 110–120  $\mu\text{g m}^{-3}$
- 120–140  $\mu\text{g m}^{-3}$
- > 140  $\mu\text{g m}^{-3}$
- Non-mapped countries
- Poor data coverage
- △ Rural background station
- Urban background station



**Ozone SOM035**

Reference year: 2005

Combined rural and urban map

- < 3 000  $\mu\text{g m}^{-3}.\text{d}$
- 3 000–6 000  $\mu\text{g m}^{-3}.\text{d}$
- 6 000–10 000  $\mu\text{g m}^{-3}.\text{d}$
- 10 000–15 000  $\mu\text{g m}^{-3}.\text{d}$
- > 15 000  $\mu\text{g m}^{-3}.\text{d}$
- Non-mapped countries
- Poor data coverage
- △ Rural background station
- Urban background station

varies strongly from country to country <sup>(16)</sup>. In the Mediterranean, more than half of the population lives in non-compliance areas with concentrations above the target value, while in northern Europe, the results suggest no exceedance at all.

As already stated for PM<sub>10</sub> (Chapter 3.3), we must not forget that the interpolated maps show only smooth estimates of the real pollution. They tend to overestimate average concentrations and underestimate both low and high values. Consequently, population exposure to both high and low concentration values is underestimated, exposure to average concentrations is overestimated. Such bias can be significant, particularly in under-sampled areas.

Following the recommendation of the WHO (WHO, 2001), the health impact is assessed as a relative risk of all-cause mortality of 1,003 (statistical confidence interval 1,001 to 1,004) for a 10 µg m<sup>-3</sup> increase in the daily maximum 8-hour mean ozone concentration. The estimated number of premature deaths attributable to ozone concentrations in 2005 is presented in Figure 4.2. Taking into account the difference in concentrations, the estimates are in correspondence with the estimates made for 2004. The impact on health from ozone seems to be an

order of magnitude lower than the impact from PM<sub>10</sub>.

#### 4.1.2 Uncertainties and probability of exceeding the target

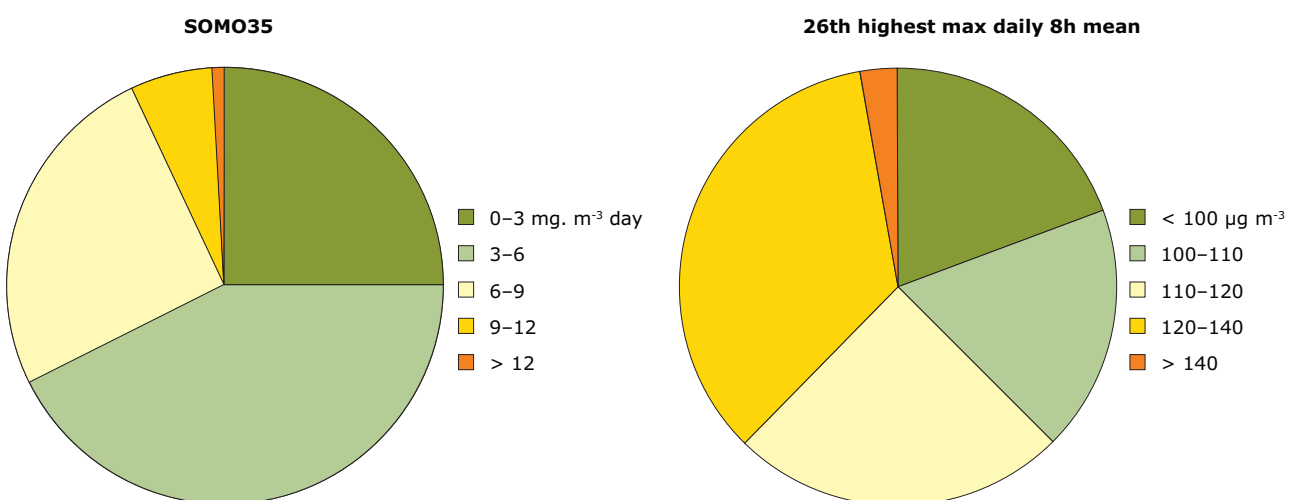
##### Twenty-sixth highest daily maximum 8-hour average ozone concentrations

The 26th highest daily maximum 8-hour average ozone concentrations for the rural areas in the combined map show an absolute mean interpolation uncertainty of 12.3 µg m<sup>-3</sup>. This refers to a relative mean interpolation uncertainty of about 10 %. For the urban map, the uncertainty is 10.0 µg m<sup>-3</sup>, i.e. about 9 % of the average of the measured indicator at all urban and suburban stations.

##### SOMO35

For SOMO35 in rural areas in the combined map the uncertainty is 2200 µg.m<sup>-3</sup>.days, i.e. 35.5 % of the average of SOMO35 values measured at all rural background stations. For the urban map the uncertainty is 1500 µg.m<sup>-3</sup> x days, i.e. about 32 % of the average of measured SOMO35 values at all urban and suburban stations.

**Figure 4.2 Exposure of the European population to ozone concentrations, SOMO35 (left) and 26th highest daily mean (right) – the reference year is 2005**



<sup>(16)</sup> The reason for splitting the countries in the following population exposure tables into two groups is that different databases of population densities were used to calculate exposure (see also Annex 1, A 1.7). Population density [inhbs km<sup>-2</sup>] is based on the data from the EC's Joint Research Centre for the majority of countries. For the countries that are not included in this database (i.e. Andorra, Albania, Bosnia and Herzegovina, Cyprus, Iceland, Liechtenstein, the former Yugoslav Republic of Macedonia, Norway, Serbia and Montenegro, Switzerland, and Turkey), the population density data used were from an alternative source, the ORNL LandScan (2002) Global Population Dataset.

**Table 4.1 Population exposure and population weighted concentration – ozone, 26th highest daily maximum 8-hour value, the year 2005**

Country	% of population					Population-weighted conc. $\mu\text{g m}^{-3}$
	< 100 $\mu\text{g m}^{-3}$	100–110 $\mu\text{g m}^{-3}$	110–120 $\mu\text{g m}^{-3}$	120–140 $\mu\text{g m}^{-3}$	> 140 $\mu\text{g m}^{-3}$	
Austria	0	2.4	18.4	78.6	0.6	122.6
Belgium	16.6	74.1	9.1	0.1	0	104.1
Bulgaria	14.3	24.5	18.2	42.3	0.6	115.0
Croatia	0	0	18.4	81.1	0.4	120.7
Czech Rep.	0	0	15.5	84.5	0	122.3
Denmark	48.0	51.3	0.8	0	0	97.0
Estonia	62.1	37.9	0	0	0	92.9
Finland	84.4	15.6	0	0	0	93.9
France	2.5	36.3	33.6	27.3	0.3	114.1
Germany	3.2	27.2	36.6	33.0	0	114.9
Greece	0	1.6	24.1	72.2	2.1	125.8
Hungary	0	0	34.7	65.3	0	120.0
Ireland	100	0	0	0	0	84.8
Italy	0	0.6	7.0	72.1	20.3	132.2
Latvia	48.1	51.9	0	0	0	92.3
Liechtenstein	0	100	0	0	0	106.6
Lithuania	24.5	73.8	1.7	0	0	103.2
Luxembourg	0	0	54.3	45.7	0	120.2
Malta	0	85.5	9.5	5.0	0	107.0
Netherlands	82.4	17.6	0	0	0	92.8
Poland	2.6	11.8	72.0	13.5	0	114.9
Portugal	1.1	14.6	33.6	48.4	2.3	119.0
Romania	1.4	24.8	47.6	26.1	0.1	115.0
San Marino	0	0	0	100	0	134.8
Slovakia	0	0	18.0	81.9	0.0	122.4
Slovenia	0	0	13.3	86.7	0.0	123.6
Spain	4.1	17.1	22.5	55.9	0.3	117.9
Sweden	57.4	39.4	3.2	0	0	95.5
United Kingdom	98.4	1.6	0	0	0	87.2
Andorra	20.36	18.1	24.6	34.3	2.6	130.6
Albania	0.0	2.7	45.9	39.4	12.0	125.1
Bosnia and Herzegovina	0	0	40.5	59.0	0.6	122.2
Serbia and Montenegro	0.0	19.6	31.653	47.108	1.651	119.3
Switzerland	0	4.2	16.1	74.2	5.5	123.4
Iceland	96.0	4.0	0.0	0	0	90.9
the former Yugoslav Republic of Macedonia	0	40.2	23.3	29.6	7.0	120.0
Norway	74.058	25.3	0.7	0.0	0.0	98.2
<b>Total</b>	<b>20.1</b>	<b>17.8</b>	<b>24.4</b>	<b>35.0</b>	<b>2.6</b>	<b>112.7</b>

**Note:** Countries for which air quality or population density data are missing (Cyprus and Turkey) have been excluded from calculations shown in this paper. To adjust for the differences between densely populated versus less populated areas, this report made use only of weighted annual values measured at urban AirBase background stations in agglomerations <sup>(17)</sup>.

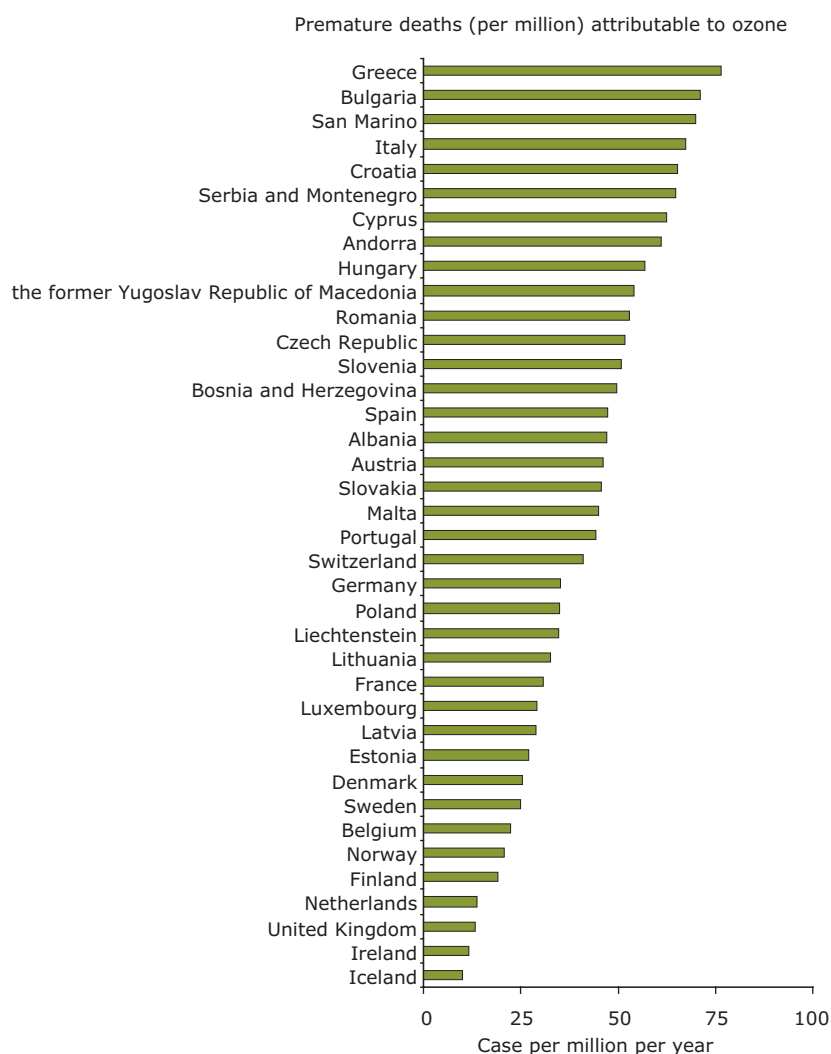
<sup>(17)</sup> According to the AQ Directives agglomeration 'shall mean a zone that is a conurbation with a population in excess of 250 000 inhabitants or, where the population is 250 000 inhabitants or less, with a given population density per km<sup>2</sup> to be established in the Member States.'

**Table 4.2 Population exposure and population weighted concentration – ozone, SOMO35, the year 2005**

Country	% of population					Population-weighted SOMO35 $\mu\text{g m}^{-3}\cdot\text{d}$
	< 3 000 $\mu\text{g m}^{-3}\cdot\text{d}$	3 000–6 000 $\mu\text{g m}^{-3}\cdot\text{d}$	6 000–10 000 $\mu\text{g m}^{-3}\cdot\text{d}$	10 000–15 000 $\mu\text{g m}^{-3}\cdot\text{d}$	> 15 000 $\mu\text{g m}^{-3}\cdot\text{d}$	
Austria	0	33.9	62.8	3.4	0	6 576
Belgium	71.1	28.9	0	0	0	2 787
Bulgaria	13.7	27.5	49.2	9.6	0.0	6 669
Croatia	0	41.1	56.1	2.8	0	6 667
Czech Rep.	0	47.5	52.5	0	0	6 087
Denmark	43.5	56.5	0	0	0	3 019
Estonia	53.5	46.5	0	0	0	2 722
Finland	63.5	36.5	0	0	0	2 580
France	16.0	61.9	21.5	0.5	0.0	4 756
Germany	24.6	70.0	5.4	0.0	0	4 164
Greece	0	13.6	58.4	27.8	0.1	9 062
Hungary	0	41.4	58.6	0	0	5 965
Ireland	97.8	2.2	0	0	0	1 852
Italy	0	4.0	83.8	12.2	0.0	8 134
Latvia	46.0	54.0	0	0	0	2 739
Liechtenstein	0	100	0	0	0	5 699
Lithuania	6.8	93.2	0	0	0	3 790
Luxembourg	0	100	0	0	0	4 796
Malta	0	0	95.0	5.0	0	7 140
Netherlands	99.5	0.5	0	0	0	1 920
Poland	2.2	87.4	10.4	0	0	5 037
Portugal	1.1	54.5	43.6	0.8	0	5 824
Romania	0	48.6	50.2	1.1	0	6 062
San Marino	0	0	100	0	0	8 612
Slovakia	0	17.5	82.2	0.3	0	6 622
Slovenia	0	24.3	75.5	0.3	0	6 669
Spain	4.6	29.2	63.9	2.2	0.0	6 514
Sweden	47.3	52.7	0.0	0	0	3 083
United Kingdom	97.8	2.2	0	0	0	1 634
Albania	0	21.8	42.7	33.7	1.7	8 563
Andorra	0	0.0	65.7	34.3	0	9 023
Bosnia and Herzegovina	0	40.2	44.5	15.3	0	7 490
Iceland	94.5	5.5	0	0	0	1 887
the former Yugoslav Republic of Macedonia	0	51.6	14.7	32.3	1.4	7 738
Norway	72.5	27.0	0.5	0	0	2 697
Serbia and Montenegro	0	43.6	41.6	14.8	0.0	6 978
Switzerland	0	61.0	34.4	4.6	0.0	6 150
<b>Total</b>	<b>25.9</b>	<b>40.4</b>	<b>30.4</b>	<b>3.3</b>	<b>0.0</b>	<b>5025</b>

**Note:** Countries for which the data for air quality or population density are missing (Cyprus and Turkey) have been excluded from calculations in this paper. To adjust for the differences in densely populated versus less populated areas, this report made use only of weighted annual values measured at urban AirBase background stations in agglomerations.

**Figure 4.2 Number of premature deaths per million inhabitants attributable to ozone exposure, reference year 2005**



### Probability of exceeding the target value

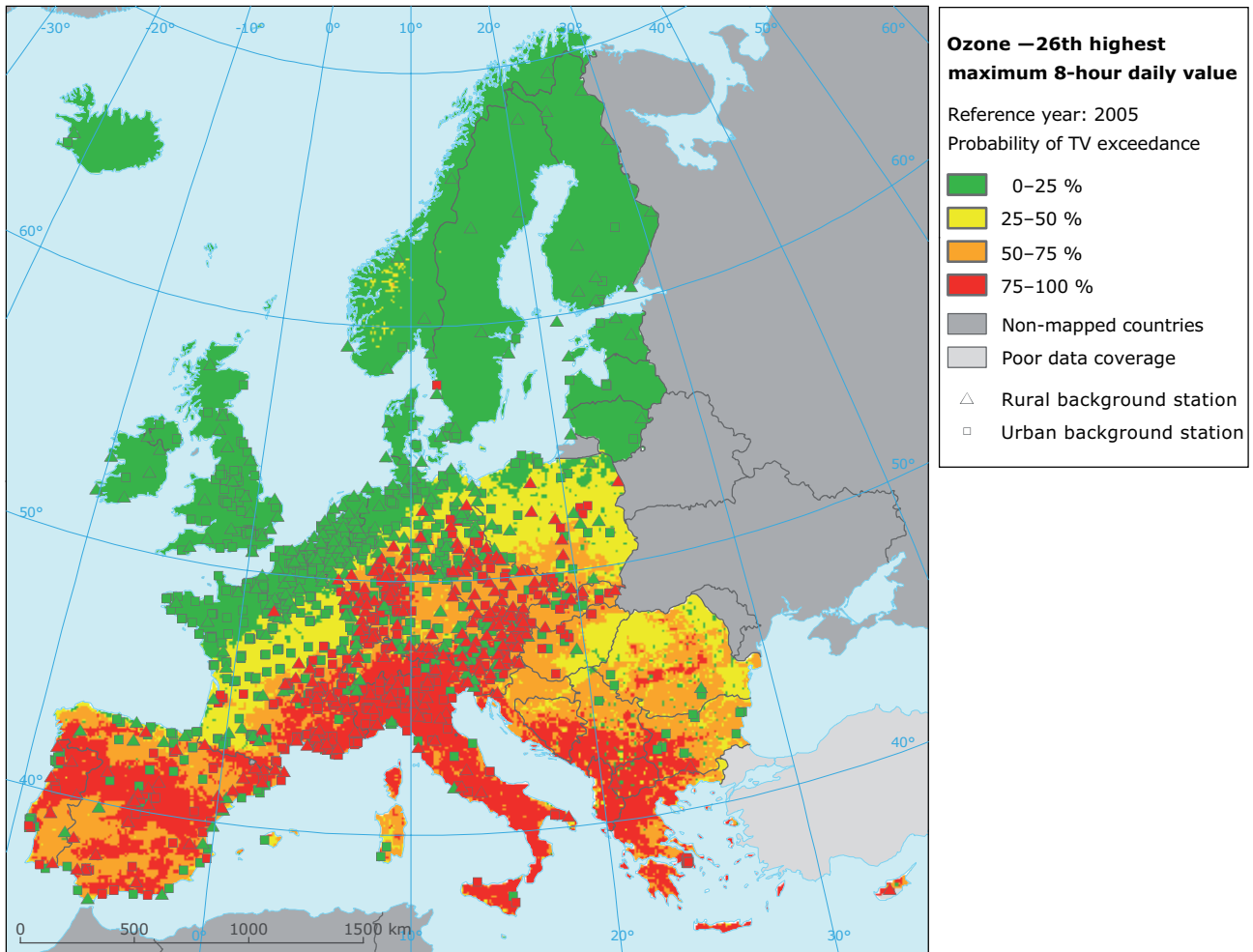
The map showing the probability of exceeding the target value (26th highest daily maximum 8-hour average ozone concentrations) is presented in Map 4.2. Areas where the probability of exceeding the limit value is above 75 % are marked red; areas below 25 % are marked green. The red colour indicates areas where it is very likely that target values shall be exceeded. This can be due to two reasons — to show:

- (1) areas with high concentrations close to or already above the target value;
- (2) areas with relatively low concentrations but high uncertainty levels related to these concentrations.

Vice versa, in the areas marked green, concentrations exceeding the target value are not very likely to occur.

The estimated probability of exceeding the target value for the ozone 26th highest daily maximum 8-hour average is moderate to considerable south of the line Biarritz – Basel – Luxembourg – Hannover – Lodz (orange grid cells show probability > 50 % and red cells > 75 %). The probability of exceeding the target values is also obvious from the changes in the altitude relief map used as supplementary information for mapping. The results suggest that for these southern European regions the target value will not be easily met.

**Map 4.2** Map with the probability (in %) of exceeding the target value for ozone indicator, 26th highest daily maximum 8-hour average values (in  $\mu\text{g m}^{-3}$ ) on the European scale in 2005



**Note:** The spatial resolution is  $10 \times 10 \text{ km}^2$  (grid). Single stations with annual averaged measurement values above or below the target value are marked red or green, respectively.

North of the demarcation line mentioned above, probability that the target values may be exceeded is much lower (25–50 %), indicating that those values might be easily met. Probability levels lower than 25 % can be found north of the boundary running La Rochelle – Rostock – Vilnius. Exceptions are some locations with higher altitudes in southern Norway (25–50 % probability of exceeding the limit). The results suggest that the target level has already been met or will be met over wider areas. The  $10 \times 10 \text{ km}^2$  map does not give information on possible cases of exceeding the targets locally.

What is remarkable is that the number of rural background stations marked red is relatively frequent (squares in Map 4.2), with measurements above the target value in grid cells showing a

probability of exceeding the limit as 25–50 %. This indicates that in rural areas local factors (emissions, meteorology, topography, etc.) not covered by the resolution of the interpolation might also play an important role. On the other hand, a relatively big number of urban and suburban background stations marked green, i.e. with measurements below the target value, are located in areas with a probability of exceeding the norms being 50–75 % (Map 4.2). This indicates that local quality of urban air is probably not always adequately covered by the resolution of the interpolation. The interpolated concentration field is, ultimately, a spatially smoothed representation of the background stations in the mapping domain, not necessarily reflecting the status of higher resolution of the local air quality.



## 4.2 Ozone vegetation indicators

The interpolated maps for both vegetation indicators (AOT40 for crops and AOT40 for forests) are presented in Map 4.3. It concerns only rural maps as it is assumed that there is no relevant vegetation in urban areas.

For both indicators, the maps have been created by combining measurement data from the rural background stations and the EMEP model output, altitude field and surface solar radiation in a linear regression model, and following this with the interpolation of its residuals by ordinary kriging (method of type 3, see Chapter 2). This interpolation method is recommended for future applications because of its consistency.

### 4.2.1 Exposure of vegetation and forests

The Ozone Directive (EC, 2002) defines a target value and a long-term objective for the protection of vegetation: AOT40, calculated from 1-hour values (daylight hours only, defined as the period between 8:00 and 20:00 CET) from May to July. The AOT40 target value for 2010 is 18 000  $\mu\text{g m}^{-3}$  times hours (.h), the long-term objective being 6 000  $\mu\text{g m}^{-3}$  .h. The term *vegetation* is not further defined in the Ozone Directive; comparing the given definitions with those in the Mapping Manual (UNECE, 2004) allows to suggest that we have to interpret the term *vegetation* in the Ozone Directive as agricultural crops. The exposure of *agricultural crops* has been evaluated here on a basis of the AOT40 for vegetation as defined in the Ozone Directive.

In addition, exposure of *forests* has been estimated on the basis of the relevant definition in the Mapping Manual: critical level of 10 000  $\mu\text{g m}^{-3}$  .h (corresponding to 5 ppm .h), accumulation over the full vegetation period, April 1– September 30.

### Agricultural crops

The rural map for ozone (AOT40 for vegetation) is given in Map 4.3. This map has been combined with the land cover CLC2000 map. Following a procedure similar to the one described in Horálek *et al.*, (2007), the exposure of agricultural areas has been calculated at the country-level. The agricultural areas were defined as the CORINE Land Cover level-1 class 2 *Agricultural areas* (encompassing the level-2 classes 2.1 *Arable land*; 2.2 *Permanent crops*; 2.3 *Pastures and 2.4 Heterogeneous agricultural areas*). Table 4.3 gives the absolute and relative agricultural

area for each country and for four European regions where the target value and long-term objective for ozone are exceeded. The table also presents the frequency distribution of the agricultural area per country over the classes of exposure.

Table 4.3 shows that about 50 % of all agricultural land is exposed to ozone the amounts of which are exceeding the target value of 18 000  $\mu\text{g m}^{-3}$  .h, and about 90 % – to levels in excess of the long-term objective of 6 000  $\mu\text{g m}^{-3}$  .h. In southern countries, the target values are exceeded over about 95 % of the area. In northern Europe, the ozone levels are below the target value for nearly 100 % of the agricultural area.

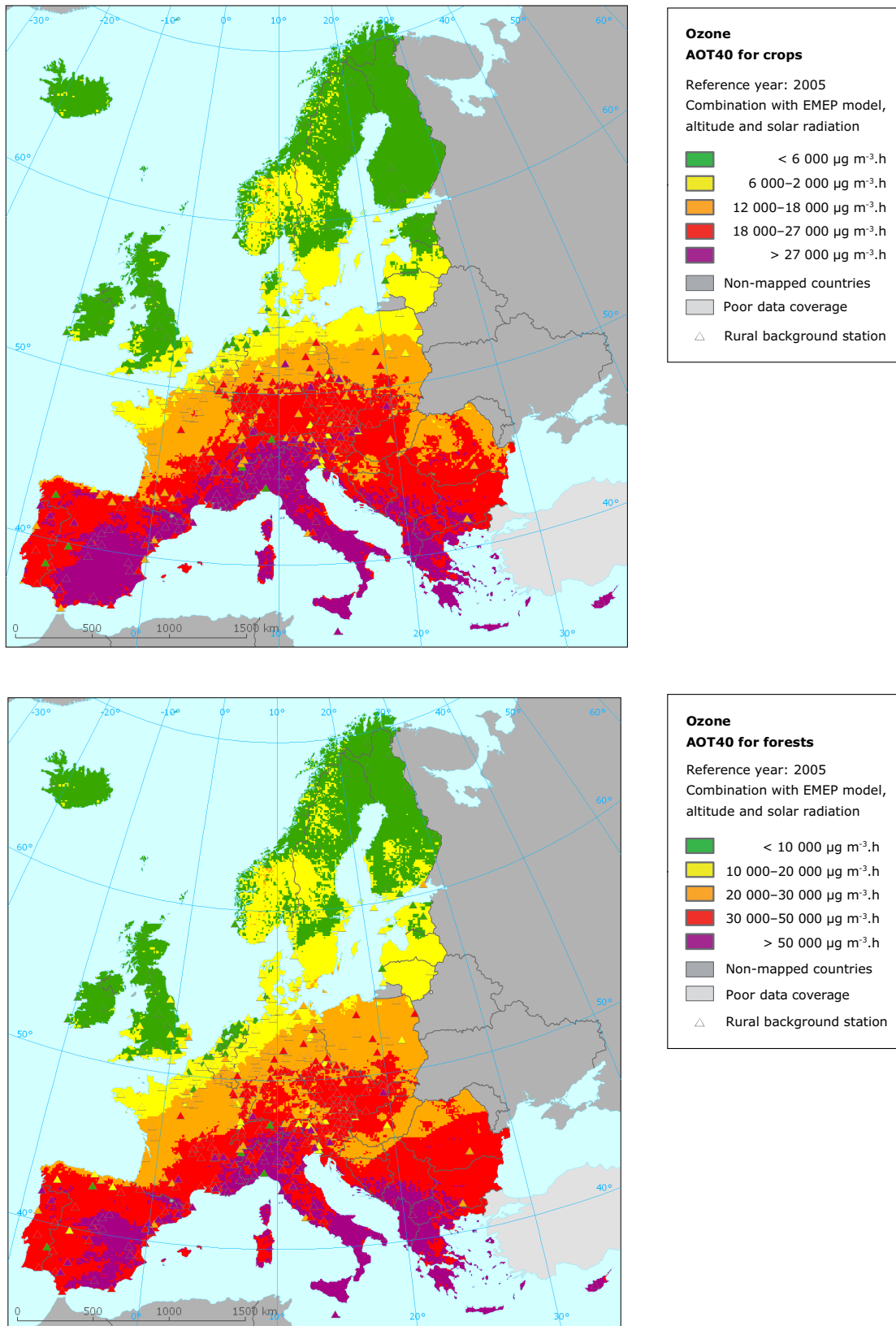
### Forests

The Ozone Directive provides neither a target value nor a long-term objective for the protection of forests. However, its Annex III – which defines the types of information to be submitted to the European Commission – mentions a level of 20 000  $\mu\text{g m}^{-3}$  .h. Following a procedure similar to the one described in detail in Horálek *et al.*, (2007), this level (indicated as 'reporting value' or *RV*) has been used as a reference point – in combination with the critical level (*CL*) of 10 000  $\mu\text{g m}^{-3}$  .h, as defined in the Mapping Manual (UNECE, 2004). The forest areas are defined as the Land Cover level-2 classes 3.1 *Forests* and 3.2 *Scrub and/or herbaceous vegetation associations* as the two out of the three of level-1 class 3. *Forests and semi-natural areas*, and is different from what has been used (level-2 class 3.1 only) in Horálek *et al.*, (2007).

The rural ozone map (AOT40 for forests) is given in Map 4.3. The gradients in this map are very similar to those in the AOT40 for the vegetation map where concentrations are increasing from north to south. Table 4.4 describes the forest area where the critical level for ozone is exceeded. Similar to the finding of the CAFE Programme, in many countries, except in the United Kingdom and some of the northern countries, all forest areas were exposed to levels above the critical level (*CL*). In 2005, the reporting level (*RV*) is exceeded in about 60 % of the European forested area. The frequency distribution of the forest exposure in single countries is given in Table 4.4 as well.

It is clear that in Northern Europe the reporting level of 20  $\text{mg m}^{-3}$  .h is not exceeded at all, in central and eastern Europe, cases of exceeding the level are observed almost everywhere and in southern Europe everywhere.

**Map 4.3** Maps of rural concentration of ozone vegetation indicators – AOT40 for crops (top) and AOT40 for forests (bottom), 2005



**Note:** Units:  $\mu\text{g m}^{-3}\times\text{hours}$ . The AOT40 target value (for crops only) given in the Ozone Directive is 18 000  $\mu\text{g m}^{-3}\cdot\text{hours}$  with the long-term objective being 6 000  $\mu\text{g m}^{-3}\cdot\text{hours}$  (EC, 2002).

**Table 4.3 Exposure of agricultural area and levels of exceeding target values (Long Term Objective, LTO, and Target Value, TV) for ozone, AOT40 for crops, 2005**

Country	Agricultural area					2005 percent [%]				
	Total	above LTO (6 mg m <sup>-3</sup> .h)		above TV (18 mg m <sup>-3</sup> .h)		< 6 000	6 000– 12 000	12 000– 18 000	18 000– 27 000	> 27 000
	km <sup>2</sup>	km <sup>2</sup>	%	km <sup>2</sup>	%	µg m <sup>-3</sup> .h	µg m <sup>-3</sup> .h	µg.m <sup>-3</sup> .h	µg m <sup>-3</sup> .h	µg m <sup>-3</sup> .h
Albania	7 109	7 109	100	7 109	100	0	0	0	32,3	67,7
Austria	27 450	27 450	100	27 069	99	0	0	1,4	93,6	5,0
Belgium	17 623	17 500	99	1 129	6	0,7	45,7	47,2	6,4	0
Bosnia and Herzegovina	19 251	19 251	100	15 026	78	0	0	21,9	64,5	13,5
Bulgaria	57 208	57 208	100	56 635	99	0	0	1,0	95,4	3,6
Croatia	23 745	23 745	100	17 602	74	0	0	25,9	64,1	10,1
Cyprus	4 088	4 088	100	4 088	100	0	0	0	3,1	96,9
Czech Republic	45 550	45 550	100	37 063	81	0	0	18,6	81,4	0
Denmark (ex. Faeroe Islands)	30 798	22 946	75	0	0	25,5	72,1	2,4	0	0
Estonia	14 418	962	7	0	0	93,3	6,7	0	0	0
Finland	28 582	837	3	0	0	97,1	2,9	0	0	0
France	327 337	327 337	100	110 461	34	0	16,2	50,1	28,1	5,6
the former Yugoslav Republic of Macedonia	9 515	9 515	100	9 515	100	0	0	0	66,6	33,4
Germany	212 360	210 699	99	72 072	34	0,8	28,7	36,5	33,9	0
Greece	48 918	48 918	100	48 918	100	0	0	0	33,3	66,7
Hungary	63 054	63 054	100	47 441	75	0	0	24,8	75,2	0
Ireland	45 312	1 552	3	0	0	96,6	3,4	0	0	0
Italy	153 591	153 591	100	153 193	100	0	0	0,3	14,7	85,0
Latvia	28 053	19 733	70	0	0	29,7	70,3	0	0	0
Liechtenstein	42	42	100	42	100	0	0	0,0	100	0
Lithuania	39 656	37 554	95	0	0	5,3	94,7	0,0	0	0
Luxembourg	1 410	1 410	100	1 347	96	0	0	4,4	95,6	0
Malta	91	91	100	91	100	0	0	0	0	100
Netherlands	24 347	22 391	92	0	0	8,0	89,2	2,8	0	0
Poland	199 623	199 623	100	12 031	6	0	32,1	61,8	6,0	0,0
Portugal	42 351	42 351	100	41 799	99	0	0	1,3	95,3	3,4
Romania	134314	134314	100	66 285	49	0	0,7	50,0	49,3	0,0
San Marino	44	44	100	44	100	0	0	0	0	100
Slovakia	24 248	24 248	100	18 534	76	0	0	23,6	76,4	0,1
Slovenia	7 133	7 133	100	6 176	87	0	0	13,4	84,1	2,5
Spain	251 487	251 487	100	248 108	99	0	0	1,3	44,7	54,0
Sweden	37 737	18 174	48	0	0	14,8	85,2	0	0	0
United Kingdom	137 813	33 353	24	0	0	75,8	24,2	0,0	0	0
<b>Total</b>	<b>2 064 257</b>	<b>1 833 258</b>	<b>89</b>	<b>1 001 777</b>	<b>49</b>	<b>10,4</b>	<b>16,0</b>	<b>24,1</b>	<b>32,7</b>	<b>16,8</b>

**Note:** Unit: mg m<sup>-3</sup>.hours.

Countries not included because of the lack of land cover data: Andorra, Iceland, Norway, Serbia and Montenegro, Switzerland and Turkey.

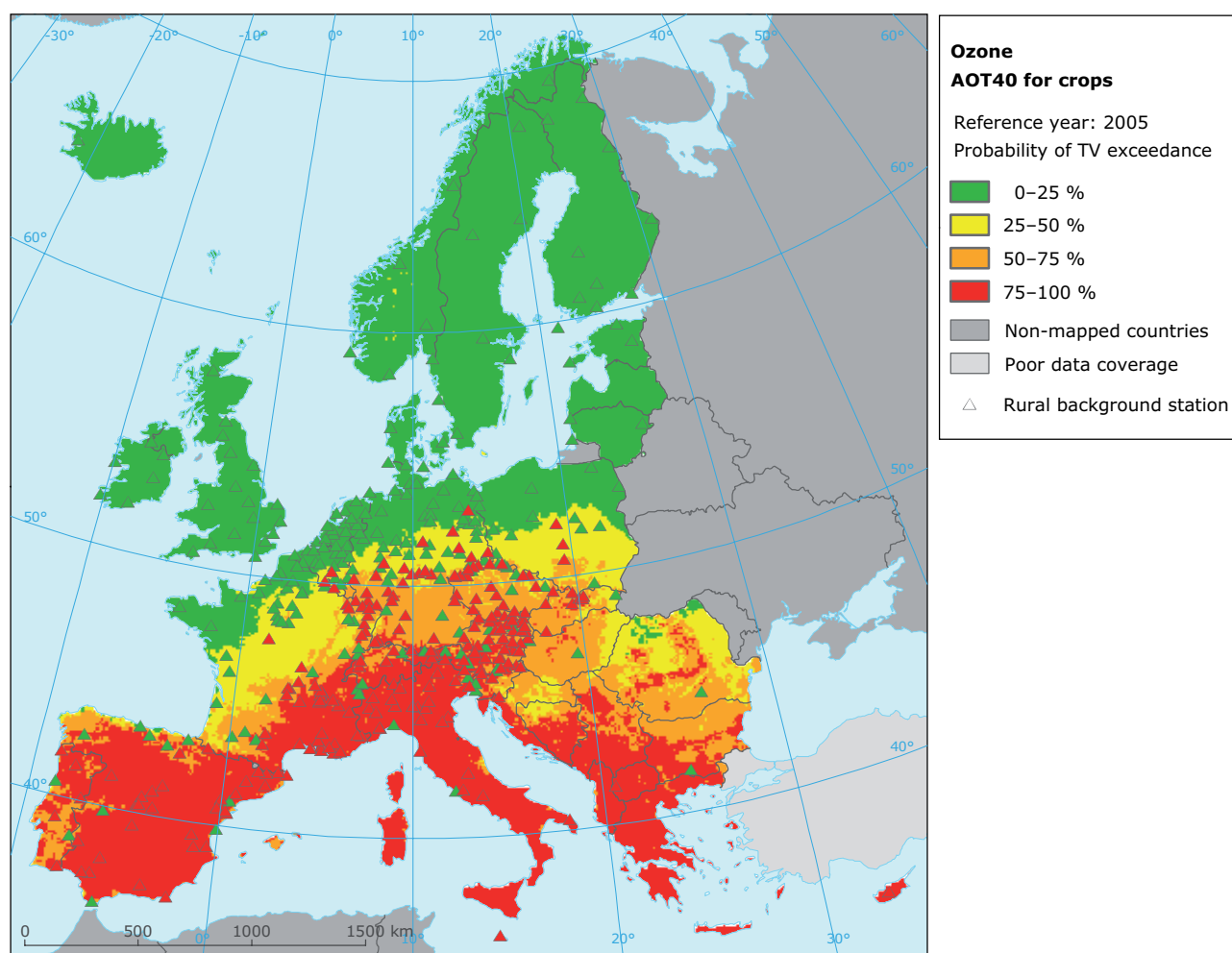
**Table 4.4 Exposure of forest areas and presence of cases where limites are exceeded (Critical level and Reporting Level) – ozone, AOT40 forests, the year 2005**

Country	Area of forests											
	Total			above CL (10 mg.m <sup>-3</sup> .h)			above RV (20 mg.m <sup>-3</sup> .h)			2005 percent [%]		
	km <sup>2</sup>	km <sup>2</sup>	%	km <sup>2</sup>	%	< 10 000 µg.m <sup>-3</sup> .h	10 000– 20 000 µg.m <sup>-3</sup> .h	20 000– 30 000 µg.m <sup>-3</sup> .h	30 000– 50 000 µg.m <sup>-3</sup> .h	> 50 000 µg.m <sup>-3</sup> .h		
Albania	7 738	7 738	100	7 738	100	0	0	0	0.5	99.5		
Austria	37 608	37 608	100	37 608	100	0	0	12.7	85.5	1.8		
Belgium	6 100	6 094	100	4 535	74	0.1	25.6	74.3	0	0		
Bosnia- Herzegovina	22 815	22 815	100	22 815	100	0	0	7.2	74.9	17.9		
Bulgaria	34 660	34 660	100	34 660	100	0	0	0	79.6	20.4		
Croatia	19 762	19 762	100	19 762	100	0	0	38.3	53.2	8.5		
Cyprus	1 498	1 498	100	1 498	100	0	0	0	1.7	98.3		
Czech Republic	25 501	25 501	100	25 501	100	0	0	11.2	88.8	0		
Denmark (ex. Faeroe Islands)	3 408	3 349	98	202	6	1.8	92.3	5.9	0	0		
Estonia	20 317	16 010	79	0	0	21.2	78.8	0	0	0		
Finland	191 690	28 748	15	0	0	85.0	15.0	0	0	0		
France	144 521	144 514	100	133 827	93	0.0	7.4	44.8	37.4	10.4		
the former Yugoslav Republic of Macedonia	8 619	8 619	100	8 619	100	0	0	0	2.8	97.2		
Germany	103 589	103 542	100	91 770	89	0.0	11.4	56.4	32.2	0		
Greece	22 978	22 978	100	22 978	100	0	0	0	21.1	78.9		
Hungary	17 331	17 331	100	17 331	100	0	0	35.2	64.8	0		
Ireland	2 892	142	5	0	0	95.1	4.9	0	0	0		
Italy	78 497	78 497	100	78 497	100	0	0	0.2	32.9	66.9		
Latvia	26 512	25 052	94	0	0	5.5	94.5	0	0	0		
Liechtenstein	63	63	100	63	100	0	0	41.1	58.9	0		
Lithuania	18 468	18 468	100	55	0	0	99.7	0.3	0	0		
Luxembourg	904	904	100	904	100	0	0	100	0	0		
Malta	2	2	100	2	100	0	0	0	0	100		
Netherlands	3 074	2 475	81	0	0	19.5	80.5	0	0	0		
Poland	91 182	91 182	100	87 305	96	0	4.3	84.1	11.7	0.0		
Portugal	24 190	24 190	100	24 190	100	0	0	0	98.3	1.7		
Romania	69 660	69 660	100	69 660	100	0	0	25.4	73.9	0.7		
San Marino	6	6	100	6	100	0	0	0	86.5	13.5		
Slovakia	19 248	19 248	100	19 248	100	0	0	12.7	87.2	0.1		
Slovenia	11 469	11 469	100	11 469	100	0	0	20.5	79.4	0.2		
Spain	91 489	91 489	100	91 463	100	0	0.0	6.9	59.2	33.9		
Sweden	248 597	111 768	45	567	0	55.0	44.7	0.2	0	0		
United Kingdom	19 158	3 598	19	0	0	81.2	18.8	0	0	0		
<b>Total</b>	<b>1 373 545</b>	<b>1 048 978</b>	<b>76</b>	<b>81 2271</b>	<b>59</b>	<b>23.6</b>	<b>17.2</b>	<b>18.8</b>	<b>29.5</b>	<b>10.8</b>		

**Note:** Unit: mg m<sup>-3</sup> x h.

Countries not included because of the lack of land cover data: Andorra, Iceland, Norway, Serbia and Montenegro, Switzerland and Turkey.

**Map 4.4** Probability (in %) of exceeding the target value for ozone vegetation indicator AOT40 for crops on the European scale in 2005, on the 10 x 10 km<sup>2</sup> grid resolution



**Note:** Stations with annual averaged measurement values above the target value are marked red; station measurements below the limit value are marked green.

#### 4.2.2 *Uncertainties and probability of exceeding the target*

The uncertainty of the absolute and relative mean interpolation of AOT40 for the crops map, expressed by the RMSE from the cross-validation, is  $7\,700 \mu\text{g m}^{-3} \cdot \text{h}$ , i.e. about 41 % of the average of the AOT40 crops values measured at all stations. For the map of AOT40 for forests, it is  $12\,500 \mu\text{g m}^{-3} \cdot \text{h}$ , i.e. about 42 % of the average of the AOT40 forest values measured at all stations.

The probability map showing exceedance of the target value is presented in Map 4.4 with the same probability legend as in Map 4.2.

As no ozone limit or target values for forests are defined in the Ozone Directive, no probability map has been prepared for the AOT40 for forests.

The estimated probability of exceeding the target value for the ozone vegetation indicator AOT40 for crops is moderate to considerable south of the line Biarritz – Basel – Luxemburg – Krakow (orange grid cells indicate probability > 50 %, and red cells > 75 %). In this area, the probability of exceeding the target value is also illustrated in the changes in the altitude relief map used as a supplementary source of information. The results suggest that for these southern European regions the target value will not be easily met.

North of the above mentioned line, the probability that target values may be exceeded is lower, about 25–50 %, thus suggesting that the target values might be met easier in these areas. Probability of exceeding the target lower than 25 % can be found north of the line La Rochelle – Hannover – Polish/Belarusian border. This indicates that target values are already met over a wide area or might be met in the future. The 10 x 10 km<sup>2</sup> map does not give information on possible cases of exceeding the targets locally.

It is remarkable how relatively frequent is the number of rural background stations marked

green (triangles in Map 4.2) where measurements below the target value in grid cells indicate a probability of exceeding the target being 50 %. There are also some stations marked red in areas with a probability of 25–50 %. This indicates that in rural areas local factors (emissions, meteorology, topography etc.) not covered by the resolution of the interpolation might also play an important role. The interpolated concentration field is, ultimately, a spatially smoothed representation of the background stations in the mapping domain, not necessarily reflecting the status of the local air quality at a higher resolution.

## 5 Recommended interpolation methods for regular updating

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Chapters 3 and 4 describe the process of mapping the air quality monitoring data for the year 2005 (years 2000 to 2004 were analysed in the earlier studies). The maps have been compiled on the basis of respectively preferred interpolation methods, basically weighting the method with the highest statistical score against the one giving the best pan-European spatial coverage. The analyses include an assessment of the interpolation uncertainty and the probability of exceeding limit, target values or (long-term) objectives as defined by the EU Air Quality Directives. Further on, the chapters present some examples of assessing the impact produced on health and vegetation in the year 2005.

The selection of an interpolation method used as the preferred method was based on several criteria. Taking the best statistical fit as the starting point, other criteria were also considered. The main criteria were correspondence with (proximity to) the best performers for other indicators of the same pollutant, extent of coverage of the European mapping domain, the best performer for the 2004-data, and subsequently practical and pragmatic reasons, like continuity of the indicator updating over the (past) years and the facilitating trend analyses. A method scoring higher on these additional (mainly) non-statistical criteria and not displaying a significantly worse statistical fit was selected as the preferred method. Hence, an acceptable choice of 'best' methods was underpinned by an expert judgement.

Table 7 provides an overview of the preferred interpolation methods used for the main air pollutants (data for 2005). For each of the entities, clear recommendations are given as to which methods should be applied for regular use in EEA assessments and indicators, especially if such assessments and indicators are produced and

updated on a regular basis. In addition, there is also given the motivation for the choice, as based on the selection criteria.

In principle, it is recommended to follow the creation of a linear regression model with residual kriging. This method provides the best consistent interpolation results for several years, based on the root mean squared error (RMSE) from cross-validation. The supplementary data used include EMEP modelling results, information on altitude and meteorological parameters (for details, see Table 7). Although kriging based on monitoring data only gives similar results for some indicators, the former method is preferred because it allows mapping of the entire European territory.

Although SO<sub>2</sub> and NO<sub>x</sub> mapping is not discussed here, the 'best' methods are summarized, for completeness, in Table 7 based on the work of Horálek *et al.* (2008). For SO<sub>2</sub> non-compliance with the limit values set for natural vegetation occurs only at few locations. Due to the meteorological variability and other factors, it might well be that in some years the limit values are exceeded over wider areas. However, it is unlikely that this is the case in more than 1–2 % of the total area. Therefore, it is recommended these indicators should not be mapped on a regular basis. Cases of exceeding the NO<sub>x</sub> limit values are more of a local (urban) hotspot than a regional problem.

The maps for PM<sub>2.5</sub> have not been compiled — because the number of relevant measuring stations continues to be too small to deliver reliable results. Relating the available measurement data to both EMEP model results and to PM<sub>10</sub> monitoring data gives poor fit, as concluded in a PM<sub>10</sub>–PM<sub>2.5</sub> assessment feasibility study (Horálek *et al.*, 2008).

**Table 7.1 Summary table on best or preferred interpolation methods**

Substance	Urban/ rural	Indicator	Preferred method recommended for regular mapping	Reasons for selected preferred method
PM <sub>10</sub>	Rural	Annual average <hr/> 36th maximum daily average	Residual kriging after multiple linear regression model using EMEP model output, altitude, solar radiation and wind speed.	Best RMSE results given by lognormal cokriging but 2nd best solution preferred because of better coverage of areas without measurements, consistency with earlier work and its performance close to best results.
	Urban	Annual average <hr/> 36th maximum daily average	Residual kriging after linear regression model using EMEP model.	Best RMSE results. <hr/> Best RMSE results.
Ozone	Rural	26th highest maximum 8-hour running average	Residual kriging after multiple linear regression model using EMEP model output, altitude and solar radiation.	Best RMSE results given by ordinary cokriging but the 2nd best is preferred: close to best, better coverage of area without stations, overall (close to) best performer at all other O <sub>3</sub> indicators (both rural and urban) and therefore considered to be robust.
		SOMO35		Over the years, appears to be a robust method.  Best RMSE results given by ordinary cokriging in 2005, but close 2nd best is preferred: used last year – see above and below.
		AOT40 crops		Best RMSE results given by ordinary cokriging but 2nd best is preferred: close to best, better area coverage of the area without stations than ordinary cokriging, preferred and used at all other O <sub>3</sub> indicators and appears to be robust.
	AOT40 forests	Best RMSE results, in line with AOT40crops' preferred performer for AOT40 crops, arguments above.		
	Urban	26th highest maximum 8-hour running average <hr/> SOMO35	Residual kriging after multiple linear regression model using EMEP model output, wind speed and solar radiation.	Best RMSE results, methodological consistency with all other ozone indicator maps (both rural and urban). <hr/> Best RMSE results.
SO <sub>2</sub> <sup>(18)</sup>	Rural	Annual average	Residual kriging after linear regression model using EMEP model output.	Best RMSE results. Appears to be a robust method.
		Winter average		Best RMSE results. In line with annual average results: methodology appears consistent and robust.
NO <sub>x</sub>	Rural	Annual average	Residual kriging after linear regression model using EMEP, altitude, wind speed and solar radiation.	Best RMSE results — addition of meteorological parameters appears to improve performance significantly.

<sup>(18)</sup> Little to no evidence of exceeding the limit values in Europe. Therefore, it is recommended not to map these indicators on a routine basis.



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# Annex 1 Input data

## A1.1 Introduction

The input data used depend on the mapping methodology applied. Chapter 4 of Horálek *et al.*, (2007) provides a complete overview of sources and specifications of the input data. For clarity and readability of this paper, we have provided here the full list of the data used. The interpolation methods of type 1 use measured air pollution data — together with the coordinates and altitude of the measurement stations. The advanced mapping methods also require the use of supplementary parameters, such as output from the dispersion models, altitude data covering the whole study area, meteorological parameters and population density. For vegetation and forest exposure the land cover data are used.

## A1.2 Measured air quality data

The air quality data were extracted from the European monitoring database AirBase, supplemented by several rural EMEP stations that are not reported on AirBase. The data used were only from stations classified by AirBase and/or EMEP as *rural*, *suburban* and *urban background* stations. *Industrial* and *traffic* station types are not considered, as they represent local scale concentration levels not applicable to the mapping resolution employed. The following components and their indicators were considered:

PM <sub>10</sub>	– annual average [ $\mu\text{g m}^{-3}$ ], the year 2005; – 36th maximum daily average value [ $\mu\text{g m}^{-3}$ ], the year 2005.
Ozone	– 26th highest daily maximum 8-hour average value [ $\mu\text{g m}^{-3}$ ], the year 2005; – SOMO35 [ $\mu\text{g m}^{-3} \cdot \text{day}$ ], the year 2005; – AOT40 for crops [ $\mu\text{g m}^{-3} \cdot \text{hours}$ ], the year 2005; – AOT40 for forests [ $\mu\text{g m}^{-3} \cdot \text{hours}$ ], the year 2005.
SO <sub>2</sub>	– annual average [ $\mu\text{g m}^{-3}$ ], the year 2005; – winter average [ $\mu\text{g m}^{-3}$ ], the winter 2004–2005.
NO <sub>x</sub>	– annual average [ $\mu\text{g m}^{-3}$ ], the year 2005;
NO <sub>2</sub>	– annual average [ $\mu\text{g m}^{-3}$ ], the year 2005 (for purposes of NO <sub>x</sub> mapping only);
NO	– annual average [ $\mu\text{g m}^{-3}$ ], the year 2005 (for purposes of NO <sub>x</sub> mapping only).

SOMO35 is the annual sum of maximum daily 8-hour concentrations above 35 ppb (i.e.  $70 \mu\text{g m}^{-3}$ ). Winter average is the average over the six months from October to March.

In case of components affecting human health (i.e. PM<sub>10</sub>, the ozone parameters of 26th highest daily maximum 8-hour average value and SOMO35), data considered are the data from *rural*, *urban* and *suburban* background stations. In case of components affecting vegetation (SO<sub>2</sub>, NO<sub>x</sub> and both AOT40 parameters for ozone), only *rural* background stations are considered.

Only the stations with annual data coverage of at least 75 % are used. The stations from French overseas areas (departments) have been excluded. Additionally, one Greek ozone station (GR0110R) with highly questionable data has been excluded from the analysis.

Table A1.1. shows the number of the measurement stations selected for the individual pollutants and their respective indicators.

In addition to the AirBase data, eight additional rural PM<sub>10</sub> stations from the EMEP database have been used to reach a more extended spatial coverage by measurement data.

## A1.3 Unified EMEP model output

The well-established European chemistry transport model we used is the photochemical version of the Unified EMEP model (revision rv2\_5\_beta2), which is an Eulerian model with a resolution of  $50 \times 50 \text{ km}^2$ . The disaggregation to the  $10 \times 10 \text{ km}^2$  grid cells is done as described in Section 4.4 of Horálek *et al.*, (2007). Output from this model (2005 data extracted in October 2007) is used for the same parameter set as the set of measurement parameters in Section A.1.2:

PM <sub>10</sub>	– annual average [ $\mu\text{g m}^{-3}$ ], year 2005; – 36th maximum daily average value [ $\mu\text{g m}^{-3}$ ], the year 2005.
Ozone	– 26th highest daily maximum 8-hour average value [ $\mu\text{g m}^{-3}$ ], the year 2005; – SOMO35 [ $\mu\text{g m}^{-3} \cdot \text{day}$ ], the year 2005; – AOT40 for crops [ $\mu\text{g m}^{-3} \cdot \text{hours}$ ], the year 2005;

**Table A1.1 Number of the stations selected for the individual indicators and areas. For rural areas – the rural background stations, and for urban areas – the urban and suburban background stations are used**

	PM <sub>10</sub>		Ozone		SO <sub>2</sub>		NO <sub>x</sub> direct	NO and NO <sub>2</sub>	NO <sub>2</sub> only	No <sub>x</sub> direct and derived
	Annual	36th max.	SOMO35	26th highest	Annual	Winter	Annual	Annual	Annual	Annual
	Average	Daily mean		Max. Daily 8h	Average	Average	Average	Average	Average	Average
Rural	214	206	440	442	283	271	132	+ 126	+ 67	= 325
Urban	800	800	843	841	-	-	-	-	-	-

- SO<sub>2</sub>
- AOT40 for forests [ $\mu\text{g m}^{-3}\cdot\text{hours}$ ], the year 2005.
  - annual average [ $\mu\text{g m}^{-3}$ ], the year 2005;
  - winter average [ $\mu\text{g m}^{-3}$ ], the season 2004/2005.
- NO<sub>x</sub>
- annual average [ $\mu\text{g m}^{-3}$ ], the year 2005.

The model is described by Simpson *et al.*, (2003) and Fagerli *et al.*, (2004). The model results are based on the emissions for the relevant year (Vestreng *et al.*, 2007) and actual meteorological data (from PARLAM-PS, i.e. special dedicated 2000 version of HIRLAM numerical weather prediction model, with parallel architecture (see Sandnes, Lenschow and Tsyro, 2000).

#### A1.4 LOTOS-EUROS model output

As a comparable air chemistry transport model for interpolated air quality mapping of the health-related pollutant indicators, the following 2005 data output of LOTOS-EUROS model (Schaap *et al.*, 2007) was used:

- PM<sub>10</sub>
- annual average [ $\mu\text{g m}^{-3}$ ], the year 2005;
  - 36th maximum daily average value [ $\mu\text{g m}^{-3}$ ], the year 2005.
- Ozone
- 26th highest daily maximum 8-hour average value [ $\mu\text{g m}^{-3}$ ], the year 2005;
  - SOMO35 [ $\mu\text{g m}^{-3}\cdot\text{day}$ ], the year 2005.

The data were extracted on 17–19 October 2007 by TNO in net CDF. The pollutant parameters extracted are PM<sub>10</sub> daily averages and ozone hourly averages in a grid resolution of 25 x 25 km<sup>2</sup> for the entire modelling domain, which is somewhat less extended than that of EMEP. The same disaggregation as with the EMEP data has been applied to meet the 10 x 10 km<sup>2</sup> grid interpolation resolution.

#### A1.5 Altitude

The station altitude from AirBase (or EMEP) is only considered in this study at the level of interpolation cokriging techniques when using primarily monitoring data (methodology type 1).

For the methodologies of type 3, when using the altitudes in their linear regression model as supplementary information, we used the altitude data field covering all Europe (in meters) of GTOPO30, original grid resolution of 30 x 30 arcsec. For details, see Horálek *et al.*, (2007).

#### A1.6 Meteorological parameters

Actual meteorological surface layer parameters are extracted from the Meteorological Archival and Retrieval System (MARS) of the ECMWF (European Centre for Medium-range Weather Forecasts). The currently used derived parameters are extracted from the ECMWF variables and specified in detail in Horálek *et al.*, (2007), Section 4.5. Those are:

- Wind speed
- annual average [ $\text{m s}^{-1}$ ], the year 2005;
- Surface solar radiation
- annual average [ $\text{MWs m}^{-2}$ ], the year 2005;
- Temperature (in 2 meters)
- annual average [ $^{\circ}\text{C}$ ], the year 2005;
- Relative humidity
- annual average [%], the year 2005.

We also tested the use of surface pressure (according the recommendation of Horálek *et al.*, 2007) but finally decided to ignore this parameter, because of almost no improvement of the results in the case of its use as a pollutant, except NO<sub>x</sub> (see Section A 2.1.1).

### A1.7 Population density

Population density [inhbs km<sup>-2</sup>] is based on JRC data for the majority of countries (Source: EEA, pop01c00v3int, official version August 2006; Owner: JRC). For the countries not included in this database (i.e. for Andorra, Albania, Bosnia and Herzegovina, Cyprus, Iceland, Liechtenstein, the former Yugoslav Republic of Macedonia, Norway, Serbia and Montenegro, Switzerland, and Turkey), we used population density data from an alternative source, the ORNL LandScan (2002) Global Population Dataset. However, these data were not available for the southern part of Cyprus. (see Horálek *et al.*, (2007), Section 4.9 for the detailed specification and the aggregation executed on the populations density data).

As mentioned in Horálek *et al.*, (2007), preliminary comparisons between the ORNL LandScan and the JRC datasets for countries covered by both datasets demonstrated significant differences between these two databases. Thus, we compared the aggregated data for the individual countries with the official UN population data (<http://www.un.org/popin/data.html>) for these countries. This comparison showed good agreement of JRC and UN data but

underestimation of ORNL data. Based on this comparison, the multiplied factor of 1.65 was applied for all ORNL data.

For the health impact assessment performed according to standard population attributive principles (WHO, 2001), an update of country-specific demographic data has been taken from the UN population Division (UN, 2006).

### A1.8 Land cover

The use was made of input data from CORINE Land Cover 2000 (CLC2000) – grid 250 x 250 m, version 8/2005 version 2, (Source and owner: EEA, lceugr250\_00). The countries missing in this database are Iceland, Norway, Switzerland, Serbia and Montenegro, and Turkey.

In an effort to reduce the time needed for calculations on large data quantity involved with the 250 x 250 m<sup>2</sup> grid resolution, an aggregation to a 500 x 500 m<sup>2</sup> grid resolution is performed first – before the mapping and table extraction of the cases of exceeding the targets takes place. The ultimate map and table results are not influenced by this resolution aggregation.

## Annex 2 Statistical formulas

### A2.1 Interpolation methods

Kriging is used in a different form. Kriging is a statistical interpolation method (for detailed description, see Cressie, 1993), which makes use of the assumption that the spatial variance of the value being interpolated can be described as a function of distance. In other words, the further away a point is from a measurement, the larger the uncertainty. Kriging exploits this assumption, which is described by the variogram and its parameters — nugget, sill and range (see below), by trying to minimise the mean square prediction error at the interpolation point, i.e. the most likely value at that point, given the surrounding measurements. Kriging enables to compute not only the estimation (resp. concentration) maps, but also the uncertainty maps.

#### Variogram parameters

The basic parameters of the variogram are called nugget, sill and range (see Figure A2.1).

Sill is the value at which the spatial variability does not change with distance (plateau); range is the distance at which the spatial variability does not change. The range gives information about the size of the search window, as it is not interesting to account for those points where spatial variance is not related to distance. If the range is large, the long-range variation dominates; if small — then the short distances dominate the variation. Nugget is the y-intercept, which represents the spatially uncorrelated noise and errors, since at zero distance we would expect no variability. The difference sill-nugget is sometimes called partial sill.

The empirical variogram is computed on the basis of measured data (for details, see Horálek, 2008).

#### Interpolation using monitoring data

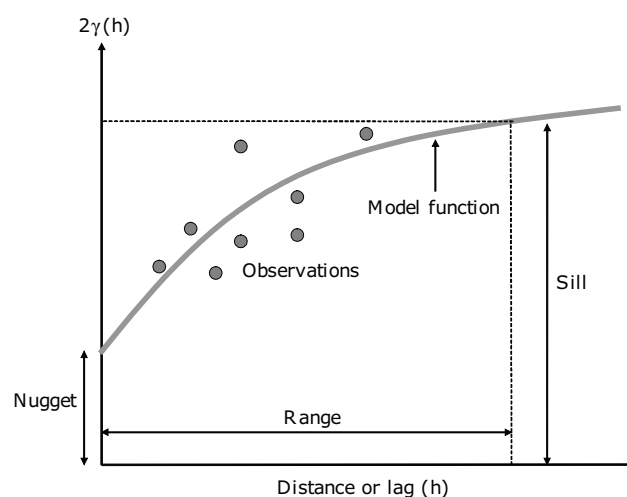
##### Ordinary kriging

Ordinary kriging is the most commonly used geostatistical method. It considers the basic statistical model

$$Z(s) = \mu + \eta(s) + \varepsilon(s) \quad (\text{A2.1})$$

where  $\mu$  represents the constant mean structure of the air quality field;

Figure A2.1 Diagram showing the important parameters that describe the variogram,  $2\gamma(h)$ , used in kriging



$\eta(s)$  is the (zero-mean)<sup>(\*)</sup> stochastic part; its statistical structure is described by a variogram;

$\varepsilon(s)$  is the measurement error or noise (zero-mean).

<sup>(\*)</sup> zero-mean is sliding all data in a profile so that their average is zero.

Interpolation is carried out according to the relation

$$\hat{Z}(s_0) = \sum_{i=1}^n \lambda_i Z(s_i), \quad \sum_{i=1}^n \lambda_i = 1 \quad (\text{A2.2})$$

where  $\lambda_1, \dots, \lambda_n$  are the weights assumed on the basis of a variogram in order to minimize the mean-square-error.

##### Ordinary cokriging

Ordinary cokriging makes use, in addition to primary measured data, of the supplementary quantities, for example altitude and temperature. The values of these quantities are considered only at the measuring sites (contrary to the methods presented below, where the complete parameter field is considered).

Interpolation is carried out according to the relation

$$\hat{Z}(s_0) = \sum_{i=1}^n \lambda_i Z(s_i) + \sum_{j=1}^m \sum_{i=1}^n \eta_{ij} Y_j(s_i) \quad (\text{A2.3})$$

where  $\lambda_i$  and  $\eta_{ij}$  are the weights assumed on the basis of a variogram and crossvariograms;

$Y_j(s_i)$  are the values of supplementary quantities ( $j = 1, \dots, m$ ) in the  $i$ -th point, with  $i = 1, \dots, n$ .

*Lognormal kriging and lognormal cokriging*

Lognormal kriging and cokriging can be used in a case where the considered quantity (e.g. measured concentrations) has a lognormal distribution (i.e. if the values gained by logarithmic transformations show a Gauss normal distribution).

This is similar to ordinary kriging and cokriging performed after logarithmic transformation. The interpolated field is back-transformed by exponentiation  $\exp(Z + \sigma^2/2)$ , where  $Z$  is the interpolated field and  $\sigma^2$  is the kriging error (Cressie, 1993).

**Linear regression models plus interpolation of their residuals (residual kriging)**

This method combines the linear regression models and kriging. The following statistical model is considered:

$$Z(s) = \mu(s) + \eta(s) + \varepsilon(s) \quad (\text{A2.4})$$

where  $\mu(s)$  represents the fixed part (which models mean concentration using regression models);

$\eta(s)$  is the (zero-mean) stochastic part; its statistical structure is described by a variogram;

$\varepsilon(s)$  is the measurement error or noise (zero-mean).

The method used is the spatial interpolation of the residuals of a linear regression model. Here interpolation is carried out according to the relation:

$$\hat{Z}(s_0) = c + a_1 \cdot X_1(s_0) + a_2 \cdot X_2(s_0) + \dots + \eta(s_0) \quad (\text{A2.5})$$

where  $\hat{Z}(s_0)$  is the estimated value of the air pollution parameter at the point  $s_0$ ;

$X_1(s_0), X_2(s_0), \dots$  are the individual supplementary quantities at the point  $s_0$ ;

$c, a_1, a_2,$  are the parameters of the linear regression model calculated at the points of measurement;

$\eta(s)$  is the spatial interpolation of the residuals of the linear regression model at the points of measurement.

Different linear regression models use different supplementary data, for example apart from the output from a dispersion model, they can include altitude or various meteorological parameters. The dispersion model can be used alone or in combination with other parameters. The spatial interpolation of residuals is carried out using ordinary kriging.

**A2.2 Uncertainty analysis: cross-validation**

The main indicator used in cross-validation is root mean squared error (RMSE):

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (Z(s_i) - \hat{Z}(s_i))^2} \quad (\text{A2.6})$$

where  $Z(s_i)$  is the measured concentration at the  $i$ -th point,  $i = 1, \dots, N$ ;

$\hat{Z}(s_i)$  is the estimated concentration at the  $i$ -th point arrived at using other information without measuring the concentration at the  $i$ -th point;

$N$  is the number of the measuring points.

RMSE should be as small as possible.

**A2.3 Merging criteria of rural and urban maps into combined concentration maps**

The Pan-European population density grid is used for merging the rural and urban maps into one combined air quality indicator map. Both the rural



and the urban maps are created for the whole of Europe. The population density grid helps to determine the part of the area for which the respective map should be used.

For areas with population density less than the defined value of  $\alpha_1$ , the rural map is applied, and for areas with population density grids greater than the defined value  $\alpha_2$ , the urban map is applied. For areas with population density within the interval  $(\alpha_1, \alpha_2)$ , the following relation is applied

$$\hat{Z}(s) = \frac{\alpha_2 - \alpha(s)}{\alpha_2 - \alpha_1} \cdot R(s) + \frac{\alpha(s) - \alpha_1}{\alpha_2 - \alpha_1} \cdot U(s) \quad (\text{A2.13})$$

where  $\hat{Z}(s)$  is the resulting value of concentration at the point  $s$ ;

$R(s)$  is the concentration at the point  $s$  for the rural map;

$U(s)$  is the concentration at the point  $s$  for the urban map;

$\alpha(s)$  is the density of population at the point  $s$ .

The separate mapping of rural and urban areas and their subsequent merging is based on the presumption that at locations not too far away from each other, rural air pollution levels are lower (in case of  $\text{PM}_{10}$ ), or higher (in case of ozone) than levels of urban air pollution. This holds in general. However, it is not the case for several small areas. (It is mainly caused by irregular distribution of measuring stations within the network, especially by the lack of rural stations). This supposed inconsistency is corrected through the use of the following approach. An auxiliary field is computed on the basis of the data from all background stations, both rural and (sub)-urban. In the areas where the rural map shows higher levels of air pollution (in case of  $\text{PM}_{10}$ ) or lower levels (in case of ozone) than those on the urban map, both rural and urban maps are modified according to the auxiliary field computed from all stations.

## Annex 3 Exceedance probability mapping

The maps with the probability of exceedance (PoE) of a specific threshold value (e.g. limit or target value), exceedance maps are constructed using the concentration and uncertainty maps:

$$PoE(x) = 1 - \Phi\left(\frac{LV - C_c(x)}{\delta_c(x)}\right) \quad (A3.1)$$

where  $PoE(x)$  is the probability of exceeding the limit value ( $LV$ ) in the grid cell  $x$ ;

$\Phi()$  is the cumulative distribution function of the normal distribution;

$LV$  is the limit value of the relevant indicator;

$C_c(x)$  is the estimated combined concentration value in the grid cell  $x$ ;

$\delta_c(x)$  is the combined standard error of the estimation in the grid cell  $x$ .

For the probability map of the combined (rural and urban) map, the standard error is calculated from the standard errors of the composing rural and urban maps:

$$\delta_c = \sqrt{A^2 \cdot \delta_r^2 + (1-A)^2 \cdot \delta_u^2 + 2A \cdot (1-A) \cdot \delta_r \cdot \delta_u \cdot r_{ru}} \quad (A3.2)$$

where  $\delta_c$  is the combined uncertainty (standard deviation) in the grid cell;

$A$  is the weight factor based on population density for the rural grid cells (see Annex 2);

$\delta_r$  and  $\delta_u$  are the uncertainties in the corresponding rural resp. urban grid cell;

$r_{ru}$  is the correlation coefficient of the rural and urban concentration fields.

In the case of the perfect correlation, the equation becomes:

$$\delta_c = A \cdot \delta_r + (1-A) \cdot \delta_u \quad (A3.3)$$

In the case of no correlation, it is:

$$\delta_c = \sqrt{A^2 \cdot \delta_r^2 + (1-A)^2 \cdot \delta_u^2} \quad (A3.4)$$

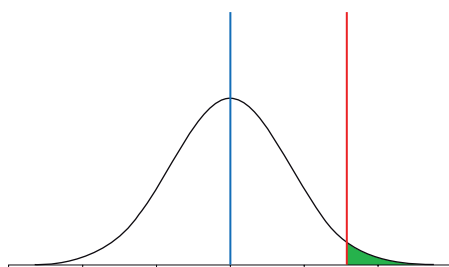
In the areas with a population density less than 100 inhabitants per km<sup>2</sup>, the weight factor  $A = 1$ , meaning the concentration and uncertainty of the rural map is assigned to the corresponding grid cell of the final map. In the areas with more than 500 inhbs.km<sup>2</sup>, the weight factor  $A = 0$ , meaning the concentration and uncertainty of the rural map is assigned to the corresponding grid cell of the final map. In the areas with a population density within the range of 100–500 inhbs.km<sup>2</sup>, the combined concentration  $C_c$  is derived according to the equation A2.13 and its related combined standard error uncertainty  $\delta_c$  — according to the equation A3.2.

To get an impression of the size of the area per population density 'class', the number of 10 x 10 km<sup>2</sup> grid cells are counted. From the total number of 50 918 cells the terrestrial mapping domain consists of, there are 40 942 cells classified as rural (< 100 inhbs km<sup>2</sup>),

1 831 cells — as urban (> 500 inhbs km<sup>2</sup>), and 8 145 cells as combined rural and urban (100–500 inhbs km<sup>2</sup>).

The probability of exceeding the targets is calculated based on residual kriging (assuming the Gaussian distribution of the residuals after linear regression).

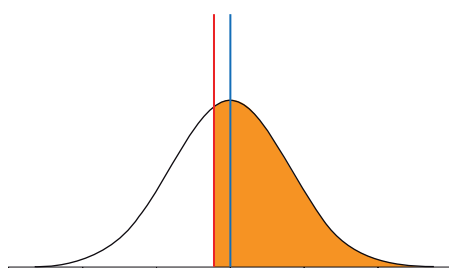
In the probability maps, the relations between the predicted value in a cell and the limit value (or target value) are grouped into four cases illustrated below.



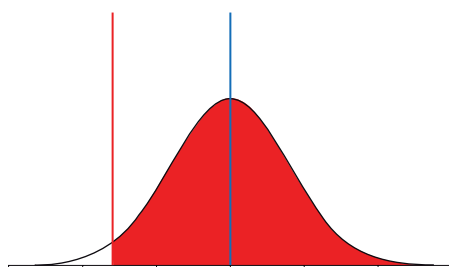
- (1) The graph shows the predicted concentration value in the grid cell (blue line) and the normal distribution centred around the grid cell value. The limit value is given by the red line. In this situation, the grid value is much lower than the limit value. However, when considering the uncertainties in the concentration value, exceeding the targets is unlikely but there is still a small chance of a limit/target value exceedance. When the total probability of exceedance is less than 25 % the cells are coloured green on the probability maps.



- (2) In this situation, the concentration value is slightly lower than the limit value. Limits/targets are not exceeded but when considering the uncertainty ranges, exceeding the limit value is possible. The chance of exceeding the limit/target value is up to 50 % when concentration and limit value are equal. In the map, the areas where the probability of exceedance is between 25 and 50 % are coloured yellow.



- (3) Here the concentration is larger than the limit/target value: such a situation is defined as an exceedance but as the graph shows that compliance with the limit value is possible. This kind of situations, where the probability of exceedance is between 50 and 75 %, is indicated by an orange colour.



- (4) Here the concentration is much larger than the limit value. Exceeding the limit value is most likely. In the probability maps, areas where the chance of exceeding the limit/target value is more than 75 % are coloured red.

The four probability classes can be defined in terms of the standard error  $\sigma(x)$  and predicted concentration value  $C$  in the grid:

- (1) if the limit value is more than  $C + 0.675 \sigma(x)$ , then the PoE is less than 25 %;

- (2) if the limit value is between  $C$  and  $C + 0.675 \sigma(x)$ , then the PoE is between 25 and 50 %;

- (3) if the limit value is between  $C - 0.675 \sigma(x)$  and  $C$ , then the PoE is between 50 and 75 %

- (4) if the limit value is less than  $C - 0.675 \sigma(x)$ , then the PoE is more than 75 %.

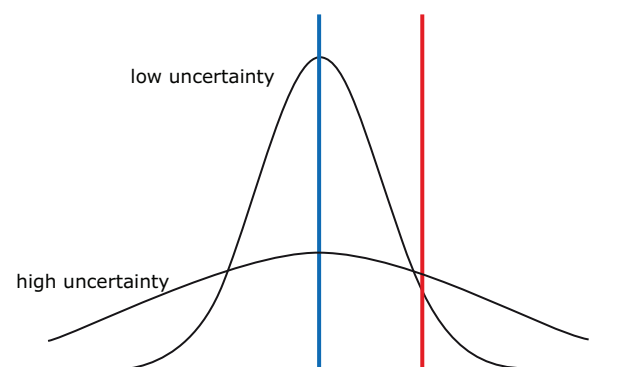
The difference between the predicted value and the limit value, and the level of interpolation uncertainty plays a combined role in the level of probability of exceeding the limits. This is illustrated in Figure A3.1. Here the distribution around the predicted value is given for two cases — with a low and high uncertainty.

With uncertainty increasing (that is when standard error of the predicted value in the grid cell is increasing), the curve broadens. The predicted value is below the limit value; the probability of exceeding the limits is given by the area on the right-hand side of the red line of the limit value. Although the difference between predicted value and limit value in both the high and low uncertainty cases is the same, it is clear that in the high uncertainty case the probability of exceeding limits is much higher than in the low uncertainty case. For the situation sketched in Figure A3.1., the grid cell may even fall in a different probability class: in the low uncertainty, the cell would be green, in the high uncertainty case, the cell would be yellow.

Next to the estimation of the probability of exceeding the limit value, the values presented in the probability maps are the real measured values at the stations: stations with the measured values above the limit or target values are marked red, whereas the station measurements below the limit value are marked green. (Neither orange nor yellow is applied to the stations, since that classification is related to the *interpolation uncertainty* only, not the measurement uncertainty at the stations).

The probability map may serve as a guide to further actions with respect to implementing of the abatement measures and to monitoring the network design. In regions with a high uncertainty in the indicator value, when the probability map indicates that exceeding the limits is most unlikely, there might be no need to reduce this uncertainty (e.g. by establishing additional stations). On the other hand,

**Figure A3.1** Distribution around the predicted value (blue) given for two cases with a low and high uncertainty. Red is the limit or target value



there will be areas where the concentration is below the limit value but the probability map indicates that limits are likely to be exceeded. Although these areas are ostensibly in compliance, one may conclude that, in order to avoid non-compliance in a different year, abatement measure should be considered.

Both the uncertainty and probability maps, presented in this report, account only for the interpolation error. Other sources of uncertainty (e.g. measuring errors, representativeness, and model uncertainty) have not been included. Therefore, it should be stressed that these maps are only estimates.

The exceedance probability maps showing the probability of exceeding a limit/target value indicate that the relationship between the actual value, the interpolated value and the associated uncertainty is much more complex than is often assumed. This calls for a careful consideration when selecting the particular output indicator to be presented and the way of presenting it, especially in view of the message to be communicated.

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