European air quality maps of ozone and PM<sub>10</sub> for 2007 and their uncertainty analysis



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#### Front page picture:

The ozone levels for the health indicators  $26^{th}$  highest daily maximum 8-hour value in  $\mu g.m^{-3}$  for both the rural and urban areas, combined into one final map for the year 2007. Its target value is 120  $\mu g.m^{-3}$ . (Figure 5.1 of this paper).

Note the considerable decrease in area where the ozone levels are above its target value, compared to those of 2006 (ETC/ACC Technical Paper 2008/8 cover figure), and being back to levels of somewhat below of those of 2005 (ETC/ACC Technical Paper 2007/7, cover figure).

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# 1 Introduction

This paper provides an update of European air quality concentration maps, their exceedance probability and population exposure estimates based on interpolation of annual statistics of the 2007 observational data reported by EEA Member countries in 2008. The paper presents the mapping results and includes an uncertainty analysis of the interpolated maps, building upon methodological developments of earlier reports from Horálek et al. (2007, 2008) and De Smet et al. (2009).

The products of this work are primarily intended to be used for the assessments and evaluations of European air quality by the EEA and its ETC/ACC, and for (interactive visual) public information purposes through the EEA website. Others may use the products under the restriction of source reference to this document equivalent to the format used at the reference section of this paper for the ETC/ACC Technical Papers.

We consider the two main pollutants of interest,  $PM_{10}$  and ozone, as most relevant pollutants for an annual update. Mapping of  $PM_{2.5}$  is a theme dealt with in a separate ETC/ACC Technical Paper (De Leeuw and Horálek, 2009).

The mapping methods applied are the same as recommended and described in Horálek et al. (2007, 2008) and De Smet et al. (2009). It includes creating at a European scale, using a 10x10 km grid resolution, separately a rural interpolated map based on rural background measurements and an urban map based on urban and suburban background station observations. Then subsequently derive a final combined map by merging them together, using a weighting function based on the population density. Next to air quality observational data, other supplementary data such as output from (currently) the Unified EMEP chemical transport model, altitude and meteorological parameters are used in the mapping procedure. Depending on their level of contribution in improving the interpolation calculations, we apply different sets of supplementary parameters for each pollutant indicator type and each area type, following the recommendations of Horálek et al. (2008).

Next to annual indicator maps, we present the population exposure to  $PM_{10}$  and ozone and the exposure of vegetation to ozone.

For all the maps, we include a quantitative estimate of their interpolation uncertainty, using cross-validation parameters and scatter-plots. In addition, the paper contains the maps with probability estimates of limit/target value exceedances.

Chapter 2 describes briefly the applied methodology. Chapter 3 documents the input data. Chapters 4 and 5 present the calculations, the mapping, the exposure estimates and the uncertainty results for  $PM_{10}$  and ozone respectively. Chapter 6 summarizes the overall conclusions.

# 2 Used methodology

### 2.1 Mapping method

In previous technical papers prepared by the ETC/ACC (Technical Papers 2008/8, 2007/7, 2006/6, 2005/8 and 2005/7) methodological details on spatial interpolations and their uncertainties have been discussed. No changes have been made in the methodology. Here a brief summary will be presented.

The mapping method used is the linear regression model followed by the kriging of its residuals (residual kriging). The interpolation is carried out according to the relation:

$$\hat{Z}(s_0) = c + a_1 \cdot X_1(s_0) + a_2 \cdot X_2(s_0) + \dots + a_n \cdot X_n(s_0) + \eta(s_0)$$
(2.1)

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

 $X_1(s_0), X_2(s_0), ..., X_n(s_0)$  are the *n* number of individual supplementary variables at the point  $s_o$ *c*,  $a_1, a_2, ..., a_n$  are the *n* selected parameters of the linear regression model calculated at the points of measurement,

 $\eta(s_0)$  is the spatial interpolation of the residuals of the linear regression model at the points of measurement.

The spatial interpolation of residuals is carried out using ordinary kriging based on variogram estimated using spherical function (with parameters: *nugget, sill, range*). For different pollutants and the area types (rural, urban), different supplementary data are used, depending on their contribution to improving the fit of the regression.

The maps are constructed separately for the rural and urban areas. The merging of the rural and urban maps into one combined air quality indicator map is done using a European-wide population density grid. For areas with population density less than the defined value of  $\alpha_1$ , the rural map is applied, and for areas with a population density 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 weighting function of  $\alpha_1$  and  $\alpha_2$  is applied (see Horálek et al., 2005, 2007 and 2008).

The separate mapping of rural and urban areas and their subsequent merging is based on the presumption that in neighbouring locations, rural air pollution levels are lower (in case of PM<sub>10</sub>), or higher (in case of ozone) than urban air pollution. This holds in general for most areas. However, it is not the case for several limited areas, of which the extent is between 5.4% and 8.8% of the total area in 2007, depending on the pollutant and indicator. For these cases, a modification in the concentrations is computed on the basis of all background stations, both rural and (sub)urban, for the given pollutant resulting in an auxiliary field. For the areas where the rural field shows higher levels of air pollution in case of PM<sub>10</sub>, or lower levels than the urban field in case of ozone, both rural and urban maps are modified according to the auxiliary field computed from all stations. The final merging of the resulting urban and rural maps is carried out by the application of the methodology described in Section 3.3 of Horálek et al. (2007), which uses the population density field. The value of the parameters  $\alpha$  1 and  $\alpha$  2 in its equation 3.2 are set again as  $\alpha$  1 = 100 inhbs.km<sup>-2</sup> and  $\alpha$  2 = 500 inhbs.km<sup>-2</sup> on the basis of the analysis presented in Horálek et al. (2005).

For detailed description of the used methods, see Horálek et al. (2007).

### 2.2 Calculation of population and vegetation exposure

Population exposure for individual countries and for Europe as a whole is calculated from the air quality maps and population density data, both at a 10x10 km resolution. For each concentration class, the total population per country as well as European-wide is determined. In addition, the population exposure per country and European-wide we expressed as the population-weighted concentration, i.e. the average concentration per inhabitant, according to:

$$\hat{c} = \frac{\sum_{i=1}^{N} c_i p_i}{\sum_{i=1}^{N} p_i}$$
(2.2)

where  $\hat{c}$  is the population weighted average concentration in the country or in the whole Europe,

- $p_i$  is the population in the  $i^{th}$  grid cell,
- $c_i$  is the concentration in the  $i^{\text{th}}$  grid cell,
- *N* is the number of grid cells in the country or in Europe as a whole.

Vegetation exposure for individual countries and for Europe as a whole is calculated based on the air quality maps and land cover data, both in 2x2 km grid. For each concentration class, the total vegetation area per country as well as European-wide is determined.

### 2.3 Methods for uncertainty analysis

The uncertainty estimation of the European map is based on cross-validation. The cross-validation method computes the quality of the spatial interpolation for each measurement point from all available information except from the point in question, i.e. it withholds one data point and then makes a prediction at the spatial location of that point. This procedure is repeated for all measurement points of the available set. The predicted and measured values at these points are compared by drawing its scatter plot. With help of statistical indicators, one demonstrates objectively the quality of the predictions. This method is quite exact – no suppositions have to be fulfilled. The advantage of the nature of this cross-validation technique is that it enables to evaluate the quality of the predicted values at locations without measurements, as long as they are within the area covered by the measurements.

In addition, we make a simple comparison between the point measurements and interpolated values of the 10x10 km grid. The 10x10 km grid value is the averaged result of the interpolations for that 10x10 km area. The interpolated value within a grid cell will only approximate the predicted value(s) at the station(s) lying within that cell.

Another method to estimate uncertainties is based on a geostatistical theory: together with the prediction, the prediction standard error is computed at all the grid cells, which represents in fact the interpolation uncertainty map (see Cressie, 1993 for a detailed discussion). Based on the concentration and the uncertainty map the exceedance probability map is created.

### 2.3.1 Cross-validation

The results of cross-validation are described by the statistical indicators and scatter plots. The main indicator used is root mean squared error (RMSE) and additional is the mean prediction error (MPE):

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (\hat{Z}(s_i) - Z(s_i))^2}$$
(2.3)

$$MPE = \frac{1}{N} \sum_{i=1}^{N} (\hat{Z}(s_i) - Z(s_i))$$
(2.4)

where  $Z(s_i)$  is the measured concentration at the  $i^{\text{th}}$  point, i = 1, ..., N,

- $\hat{Z}(s_i)$  is the estimated concentration at the *i*<sup>th</sup> point using other information, without the measured concentration at the *i*<sup>th</sup> point,
- *N* is the number of the measuring points.

RMSE should be as small as possible, MPE should be as close to zero as possible.

### 2.3.2 Comparison of the point measured and interpolated grid values

The comparison of measured and predicted grid values is described by the linear regression equation and its parameter and statistics values.

### 2.3.3 Exceedance probability mapping

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

$$PoE(x) = 1 - \Phi(\frac{LV - C_c(x)}{\delta_c(x)})$$
(2.5)

where PoE(x) is the probability of limit/target value (LV/TV) exceedance in the grid cell x,

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

*LV* is the limit or target value of the relevant indicator,

 $C_c(x)$  is the interpolated concentration in the grid cell *x*,

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

The standard error of the probability map of the combined (rural and urban) map is calculated from the standard errors of the composing rural and urban maps, see Horálek et al. (2008).

# 3 Input data

Chapter 4 of Horálek et al. (2007) provides a complete overview of sources and specifications of the input data. The air quality, meteorological and, where possible, the supplementary data has been updated. No further changes in selecting and processing the input data have been made. For clarity and readability of this paper we reproduce here the list of the used data. The key data is the air quality measurements at the monitoring stations extracted from AirBase. The supplementary data cover the whole area. All supplementary data is converted into the reference EEA LAEA5210 projection on a 10x10 km grid resolution, except for the AOT40 maps for which the data were converted into a 2x2 km grid resolution to allow for accurate land cover exposure estimates.

## 3.1 Measured air quality data

The air quality data were extracted from the European monitoring database AirBase (Mol et al. 2009), supplemented by several rural EMEP stations which are not reported to AirBase. Only data from stations classified by AirBase and/or EMEP of the type *background* for the areas *rural*, *suburban* and *urban* are used. *Industrial* and *traffic* station types are not considered, since they represent local scale concentration levels not applicable at the mapping resolution employed. The following components and their indicators are considered:

- $PM_{10}$  annual average [µg.m<sup>-3</sup>], year 2007
  - $-36^{th}$  maximum daily average value [µg.m<sup>-3</sup>], year 2007
- Ozone  $-26^{\text{th}}$  highest daily maximum 8-hour average value [µg.m<sup>-3</sup>], year 2007
  - SOMO35 [µg.m<sup>-3</sup>.day], year 2007
  - AOT40 for crops [µg.m<sup>-3</sup>.hour], year 2007
  - AOT40 for forests [µg.m<sup>-3</sup>.hour], year 2007

SOMO35 is the annual sum of maximum daily 8-hour concentrations above 70  $\mu$ g.m<sup>-3</sup> (i.e. 35 ppb). AOT40 is the sum of the differences between hourly concentrations greater than 80  $\mu$ g.m<sup>-3</sup> (i.e. 40 ppb) and 80  $\mu$ g.m<sup>-3</sup>, using only values measured between 7:00 and 19:00 UTC, calculated over the three months from May to July (AOT40 for crops), respectively over the six months from April to September (AOT40 for forests). Note that the term *vegetation* as used in the ozone directive is not further defined. Comparing the definitions in the Mapping Manual (UNECE, 2004) and those in the ozone directive suggests 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 basis of the AOT40 for vegetation as defined in the ozone directive.

For the indicators relevant to human health (i.e.  $PM_{10}$ , and the ozone parameters 26<sup>th</sup> highest daily maximum 8-hour average value and SOMO35) data from *rural*, *urban* and *suburban* background stations are considered. For the indicators relevant to vegetation damage (both AOT40 parameters for ozone) only *rural* background stations are considered.

Only the stations with annual data coverage of at least 75 percent are used. We excluded the stations from French overseas areas (departments). Additionally, the ozone station IT1726A with highly questionable data has been excluded from the analysis.

In addition to the AirBase data, 12 additional rural background  $PM_{10}$  stations from the EMEP database have been used to reach a more extended spatial coverage by measurement data.

Table 3.1 shows the number of the measurement stations selected for the individual pollutants and their respective indicators.

Table 3.1 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.

	PN	110	ozone			
	annual	36 <sup>th</sup> daily	26 <sup>th</sup> highest	SOM035	AOT40	AOT40
(*)	average	maximum	daily max. 8h	3010033	for crops	for forests
rural	241	241	413	413	412	415
urban	875	875	866	866		

(\*) Including the stations of Svalbard, Azores, Madeira and Canary Islands, not presented in the map figures.

## 3.2 Unified EMEP model output

The established European chemical dispersion model used is the Unified EMEP model (revision rv3.07), which is a Eulerian model with a resolution of 50 x 50 km. This model provides information at a 50x50 km scale; the disaggregation to the 10x10km grid cells is done as described in Section 4.4 of Horálek et al. (2007). Output from this model (2007 data extracted in September 2009) is used for the same parameter set as the set of measurement parameters in Section 3.1:

- $\begin{array}{ll} PM_{10} & \mbox{ annual average } [\mu g.m^{-3}], \mbox{ year } 2007 \\ & \mbox{ 36}^{th} \mbox{ maximum daily average value } [\mu g.m^{-3}], \mbox{ year } 2007 \end{array}$
- Ozone  $-26^{\text{th}}$  highest daily maximum 8-hour average value [µg.m<sup>-3</sup>], year 2007
  - SOMO35 [µg.m<sup>-3</sup>.day], year 2007
  - AOT40 for crops [µg.m<sup>-3</sup>.hour], year 2007
  - AOT40 for forests [µg.m<sup>-3</sup>.hour], year 2007

The model is described by Simpson et al. (2003), Fagerli et al. (2004) and at the web site <u>http://www.emep.int/OpenSource/index.html</u>. The model results are based on the emissions for the relevant year (Mareckova et al. 2009) and actual meteorological data (from HIRLAM numerical weather prediction model, version 7.1.3). Benedictow et al. (2009) provides details on the EMEP modelling with the 2007 data.

## 3.3 Altitude

We used the altitude data field (in meters) of GTOPO30 that covers the European continent, with an original grid resolution of  $30 \times 30$  arcsec. This data we spatially aggregated into the  $10 \times 10$  km grid. For details, see Horálek et al. (2007).

## 3.4 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 derived parameters currently used and extracted from the ECMWF variables (details specified in Horálek et al. 2007, Section 4.5) are:

Wind speed	– annual average [m.s <sup>-1</sup> ], year 2007
Surface solar radiation	– annual average [MWs.m <sup>-2</sup> ], year 2007

## 3.5 Population density

Population density [inhbs.km<sup>-2</sup>], census 2001, is based on JRC data for the majority countries (JRC, 2008), (source EEA, popugrid01v4\_1grid, official version 4.1, 15 Jan. 2008, resolution 100x100 m).

For countries (Andorra, Albania, Bosnia-Herzegovina, Iceland, Liechtenstein, FYR of Macedonia, Montenegro, Norway, Serbia, Switzerland, and Turkey) and regions (Faroe Islands, Jersey, Guernsey, Man, Gibraltar, and northern part of Cyprus) not included in this database, we used as alternative source the ORNL LandScan (2002) Global Population Dataset, based on census counts of mid-2002 provided by the International Programs Center (IPC) of the United States Bureau of the Census (ORNL, 2002).

JRC data is spatially aggregated into EEA 10x10 km grid.

ORNL data is also spatially aggregated into EEA 10x10 km grid. The data is compared on the one hand with JRC data for countries covered by both population density data sources, and on the other hand with UN population data of individual countries based on 2000 round of national population censuses and of recent specialized surveys carried out in countries around the world (UN, 2007). Based on these comparisons, showing good agreement of JRC and UN data but underestimation of ORNL data, a multiplication factor 1.72 was applied for all ORNL data. Figure 3.1 presents this comparison between JRC and ORNL data based on the national population totals of the individual countries.



Figure 3.1 Correlation between JRC (y-axis) and the UN 2006 revision (x-axis, left), respectively ORNL recalculated (x-axis, right) national population totals. The ORNL data is recalculated by multiplication factor 1.5.

### 3.6 Land cover

The input data from CORINE Land Cover 2000 (CLC2000) – grid 100 x 100 m, version 12/2009 is used (EEA, 2009), (source and owner: EEA, lceugr100\_00). The countries missing in this database are Switzerland and Turkey.

# 4 PM<sub>10</sub> maps

For  $PM_{10}$  the two health-related indicators annual average and  $36^{th}$  highest maximum daily average are considered. The maps are created using the combination of rural and urban areas, as described in Chapter 2. All the maps we constructed in the EEA LAEA5210 10x10 km grid.

## 4.1 Annual average

### 4.1.1 Concentration map

The final interpolated map for the 2007  $PM_{10}$  annual averages is created by combining the rural and urban maps following the procedures described in Chapter 2 and in more details by Horálek et al. (2007). Both rural and urban maps were created by combining the annual averages from the measured  $PM_{10}$  concentrations with supplementary data in a linear regression model, followed by the interpolation of its residuals by ordinary kriging.

Supplementary data are used in accordance with the recommendations given by Horálek et al. (2008). The recommended supplementary data for rural areas are EMEP model output, altitude, wind speed, and surface solar radiation; for urban areas it is EMEP model output. (The relevant linear regression submodels are identified as P.Eawr and UP.E, respectively).

The estimated parameters of the linear regression models (*c*,  $a_1$ ,  $a_2$ ,...) and of the residual kriging (*nugget, sill, range*) are presented in Table 4.1, including the statistical indicators of both the regression and the kriging. The R<sup>2</sup> and standard error are indicators for the fit of the regression relation, where R<sup>2</sup> should be as close to 1 as possible and the standard error should be as small as possible. The values for the adjusted R<sup>2</sup> of 0.40 for the rural areas and 0.10 for the urban areas show a slightly better fit of the regression than observed for the years 2006 (0.29 and 0.03) and 2005 (0.28 and 0.06) (De Smet et al. 2009, Table 4.1; Horálek et al. 2008, Tables A.21 and A2.6). Over the years, the low values for urban areas indicate that the fit of the regression in urban areas is poor (Horálek et al. 2007, 2008, De Smet et al. 2009). RMSE and MPE are the cross-validation indicators, showing the quality of the resulting map; the MPE indicates to what extend the estimation is un-biased. More detailed analysis and comparison with results of 2005 and 2006 are given in Section 4.1.3.

The final map is presented in Figure 4.1. The areas and stations in the combined map where the limit value (LV) of 40  $\mu$ g.m<sup>-3</sup> is exceeded are coloured red and purple.

Table 4.1 Parameters of the linear regression models (Eq. 2.1) and of the ordinary kriging variograms (nugget,
sill, range) - and their statistics - of $PM_{10}$ indicator annual average for 2007 in the rural (left) and urban (right)
areas as used for the final combined map, i.e. rural linear regression model P.Eawr (left), resp. urban UP.E
(right) followed by interpolation on its regression residuals using ordinary kriging (OK, coded with 'a').

linear regr. model + OK on	rural areas (P.Eawr-a)	urban areas (UP.E-a)
its residuals	coeff.	coeff.
c (constant)	13.4	18.9
a1 (EMEP model 2007)	1.38	1.06
a2 (altitude GTOPO)	-0.0062	
a3 (wind speed 2007)	-2.49	
a4 (s. solar radiation 2007)	0.82	
adjusted R <sup>2</sup>	0.40	0.10
standard error [µg.m⁻³]	5.56	8.01
nugget	12	16
sill	29	46
range [km]	460	430
RMSE [µg.m <sup>-3</sup> ]	4.60	4.96
MPΕ [μg.m <sup>-3</sup> ]	0.18	0.06



Figure 4.1 Combined rural and urban concentration map of  $PM_{10}$  – annual average, year 2007. Spatial interpolated concentration field and the measured values in the measuring points. Units:  $\mu g.m^{-3}$ .

### 4.1.2 Population exposure

Table 4.2 gives the population frequency distribution for a limited number of exposure classes, as well as the population-weighted concentration (i.e. the average concentration per inhabitant) for individual countries and for Europe as a whole according to Equation 2.2.

Almost a quarter (24%) of the European population is exposed to annual average concentrations below 20  $\mu$ g.m<sup>-3</sup>, the WHO air quality guideline. More than two-third (70.5%) of the European population lived in 2007 in areas where the PM<sub>10</sub> concentration is estimated to be between 20 and 40  $\mu$ g.m<sup>-3</sup>. About 6% of the population lived in areas where the PM<sub>10</sub> annual limit value is exceeded, but not any country shows a population weighted concentration or a median above the LV. However, as the next section discusses the current mapping methodology tends to underestimate high values. Therefore the exceedance percentage will most likely be higher.

The frequency distribution shows a large variability over Europe; in four countries (Bulgaria, FYR of Macedonia, Romania, and Serbia) at least more than one fifth of the population is estimated to be exposed to concentrations above the limit value. Poland and Italy, who were in 2006 part of this group, show in 2007 some improvement with reduced numbers below this one fifth. In a number of countries in north and north-western Europe, the LV of 40  $\mu$ g.m<sup>-3</sup> seems not to be exceeded at the 10x10 km level applied in the mapping. When comparing 2007 with 2006 and 2005 we see that the population exposed to the low levels below 20  $\mu$ g.m<sup>-3</sup> dropped in 2006 temporarily to 20% compared to the 24% in 2005 and 2007. More remarkable is the tendency of a reduced population living above the limit

value from 9% in 2005, through 7.7% in 2006 to 5.7% in 2007. Vixseboxse and De Leeuw (2009) also observe a slight decrease in the reported number of zones in exceedance to PM10 annual average. To what extend this can be considered as a significant trend should be confirmed by longer time series and reduced interpolation uncertainties.

Considering the average for the whole of Europe, the overall population-weighted annual mean  $PM_{10}$  concentration is some 25 µg.m<sup>-3</sup>. That is about 1 µg.m<sup>-3</sup> lower as for 2005 (Horálek et al. 2008), and 2 µg.m<sup>-3</sup> lower as for 2006 (De Smet et al. 2009). The decrease of the population-weighted concentration in comparison with 2006 results is present in almost all countries. The major exception is France, here an increase of 4 µg.m<sup>-3</sup> is seen. The reason for this is that prior to 2007 France had not submitted to AirBase PM10 data corrected for possible volatilization losses. De Leeuw and Fiala (2009) made a sensitivity run with/without corrected French data. If the correction is applied to previous years, the 2007-increase in France disappears.

Country			2007 Percent [%]					Denvilation
		Population		< LV		>	LV	Population
			< 10	10 - 20	20 - 40	40 - 45	> 45	weighted conc.
		x1000	µg.m⁻³	µg.m⁻³	µg.m⁻³	µg.m⁻³	µg.m⁻³	μg.m <sup>-3</sup>
Austria	AL	8271	1.5	41.0	57.5	0	0	20.5
Belgium	BE	10579	0	6.4	93.6	0	0	24.6
Bulgaria	BG	7982	0	0.6	72.7	0.8	26.0	36.5
Croatia	HR	4464	0	0.8	99.2	0	0	28.6
Cyprus	CY	852	0	0	100	0	0	33.3
Czech Republic	CZ	10164	0	23.6	74.8	1.6	0	24.0
Denmark	DK	5415	0	63.8	36.2	0	0	19.7
Estonia	EE	1335	4.9	95.1	0	0	0	14.1
Finland	FI	5129	9.0	91.0	0	0	0	12.7
France	FR	58495	0	14.8	85.2	0	0	24.0
Germany	DE	82111	0	47.7	52.3	0	0	19.9
Greece	GR	10967	0	0.1	99.5	0.5	0	32.7
Hundary	ΗU	10128	0	0	100	0	0	26.9
Ireland	IE	3730	1.0	98.8	0.1	0	0	14.4
Italy	IT	56794	0.1	2.0	78.6	11.7	7.6	32.8
Latvia	i.v	2383	0.5	88.6	10.9	0	0	16.7
Liechtenstein		67	0	42.9	57.1	0	0	19.5
l ithuania	LV	3469	0	68.3	31.7	0	0	17.8
Luxembourg	LU	425	0	49.9	50.1	0	0	19.0
Malta	MT	395	0	0	100	0	0	27.0
Netherlands	NL	15729	0	0.2	99.8	0	0	25.8
Poland	PL	38223	0	18.4	70.6	8.4	2.6	26.8
Portugal	PT	9906	0.1	21.2	78.7	0	0	25.9
Romania	RO	22428	0	2.9	72.3	23.6	1.2	32.2
San Marino	SM	20	0	0	100	0	0	30.5
Slovakia	SK	5298	0	5.0	95.0	0	0	26.1
Slovenia	SI	2030	0	11.1	88.9	0	0	25.1
Spain	FS	38992	01	11.8	86.8	13	0	28.3
Sweden	SF	8887	9.9	85.7	4 4	0	0	14 0
United Kingdom	IJК	59029	0.9	32.4	66.7	0	0	21.1
Alhania	AI	3927	0.0	0.9	99.1	0	0	30.8
Andorra		61	30.6	0.0	69.4	0	0	16.3
Rosnia-Herzegovina	RA	4175	0	1.0	97.6	14	0	30.8
Iceland	IS	178	40.8	59.1	0	0	0	10.6
Macedonia EYR of	MK	2275	0	2.6	58 9	10.7	27.7	35.7
Montenearo	ME	713	0	10.9	89.1	0		29.4
Norway		3187	17.6	78.1	4 3	0	0	14.4
Serhia	RS	10736	0	0.7	4.0	22.1	23.2	36.9
Switzerland	СН	7238	15	30.4	68.1	0	23.2	20.3
Switzenand		7230	0.6	22.2	00.1	26	21	20.5
Total		516188	0.0	3.9	70.5	5.0	.7	25.3

Table 4.2 Population exposure and population weighted concentration –  $PM_{10}$ , annual average, year 2007

Note: In the lower pane countries for which the population numbers are based on ORNL population data with uncertain quality are AD, AL, BA, CH, IS, ME, MK, NO, RS. Turkey could not be included in the calculation due to lack of air quality or population density data.

### 4.1.3 Uncertainties

#### Uncertainty estimated by cross-validation

Using RMSE as the most common indicator, the *absolute mean uncertainty* of the maps in positions without measurement within the areas covered by measurements - i.e. excluding areas lacking monitoring stations such as the Balkan - can be expressed in  $\mu$ g.m<sup>-3</sup>. Table 4.1 shows that the absolute mean uncertainty of the combined map of PM<sub>10</sub> annual average expressed by RMSE is 4.6  $\mu$ g.m<sup>-3</sup> for the rural areas and 5.0  $\mu$ g.m<sup>-3</sup> for the urban areas. In 2006, the RMSE values were 5.8 and 6.1  $\mu$ g.m<sup>-3</sup> respectively and in 2005 for both 5.5  $\mu$ g.m<sup>-3</sup>, meaning the map for 2007 shows a somewhat lower level of absolute mean uncertainty than the maps for 2006 and 2005.

Alternatively, this uncertainty can be expressed in relative terms by relating the absolute RMSE uncertainty to the mean air pollution indicator value for all stations. This *relative mean uncertainty* of the map of PM<sub>10</sub> annual average is 23.5% for rural areas and 18.4% for urban areas. This is slightly lower than for the 2006 map (27% and 21%) and the 2005 map (25% and 20%). The lower uncertainties have probably their cause in a better fit of the linear regression ( $R^2$  was 0.10 – see Table 4.1 – whereas in the years 2006 and 2005 it was 0.03 and 0.06). These relative uncertainty values fulfil the data quality objectives for models as set in Annex I of the new air quality daughter directive EC (2008).

Figure 4.2 shows the cross-validation scatter plots, obtained according Section 2.3, for both the rural and urban areas. The  $R^2$  indicates that for the rural areas about 59% and for the urban areas about 66% of the variability is attributable to the interpolation. Corresponding values of the 2005 map (52% and 71%) and the 2006 map (52% and 69%), show a somewhat improved fit at the rural interpolations in 2007, but a somewhat reduced fit for the urban interpolations.



Figure 4.2 Correlation between cross-validation predicted values (y-axis) and measurements (x-axis) for the  $PM_{10}$  annual average for 2007 for rural (left) and urban (right) areas.  $R^2$  and the slope a (from the linear regression equation  $y = a \cdot x + c$ ) should be as close 1 as possible, the intercept c should be as close 0 as possible

The scatter plots indicate that in areas with high concentrations the interpolation methods tend to underestimate the levels. For example, in urban areas (Figure 4.2, right panel) an observed value of 50  $\mu$ g.m<sup>-3</sup> is estimated in the interpolation as about 43  $\mu$ g.m<sup>-3</sup>, about 15% too low. This underestimation at high values is natural to all spatial interpolations. It can be reduced by either using a higher number of the stations at improved spatial distribution, or introducing a closer regression by using other supplementary data.

### Comparison of point measurement values with the predicted grid value

Additional to the above point cross-validation, a simple comparison has been made between the point measurement and interpolated values averaged in a 10x10 km grid. This point-grid value comparison indicates to what extend the predicted value of a grid cell represents the corresponding measured values at stations located in that cell. The results of the point cross-validation compared to this pointgrid validation examination are summarised in Table 4.3. The table shows a better correlated relation between station measurements and the interpolated values of the corresponding grid cells for both rural and urban areas (i.e. higher R<sup>2</sup>, smaller intercept and the slope closer to 1) than it does at the point cross-validation predictions. That is because the simple comparison between points measurements and gridded interpolated values shows the uncertainty at the actual station locations (points) itself, while the cross-validation simulates the behaviour of the interpolation at positions without actual measurements within the area covered by measurements. The uncertainty at measurement locations is caused partly by the smoothing effect of the interpolation and partly by the spatial averaging of the values in the 10x10 km grid cells. The level of the smoothing effect leading to underestimation at high values is there smaller than it is at areas without measurement. For example, in urban areas the predicted interpolation gridded value will be about 44  $\mu$ g.m<sup>-3</sup> at the corresponding station point with the measured value of 50 µg.m<sup>-3</sup>, i.e. an underestimation of about 11%.

Table 4.3 Linear regression equation and coefficient of determination  $R^2$  from the scatter plots of (i) the predicted point values based on cross-validation and (ii) the aggregated predictions into 10x10 km grid cells versus the measured point values for  $PM_{10}$  indicator annual average for rural and urban areas of 2007.

	rural areas		urban areas		
	equation	R <sup>2</sup>	equation	R <sup>2</sup>	
i) cross-validation prediction	y = 0.618x + 7.66	0.588	y = 0.672x + 8.89	0.656	
ii) 10x10 km grid prediction	y = 0.741x + 5.16	0.814	y = 0.751x + 6.68	0.807	

### Probability of LV exceedance map

Next to the cross-validation analysis and the comparison of predicted grid values with the points of measurement, the map with the probability of exceeding the limit value at the level of 10x10 km grid has been constructed, using the concentration maps (Figure 4.1), the uncertainty map and the limit value (40 µg.m<sup>-3</sup> for the annual average). The probability of exceedance (PoE) map is presented in Figure 4.3. Areas with the probability of limit value exceedance above 75% are marked in red; areas below 25% are marked in green. The red areas indicate areas for which exceedance may occur very likely due to either high concentrations close to or already above the LV including such enclosed uncertainty that exceedance is very likely, or red areas indicate lower concentrations with such high uncertainty levels reaching above the LV that excess is very likely. Vice versa, in the green areas, it is not likely to have prediction values and/or such enclosed uncertainties that levels do reach above the LV.

Areas with 25-50%, resp. 50-75% probability of LV exceedance are marked in yellow and orange. The yellow colour indicates the areas with the estimated values below limit value for which there exists a reasonable chance of exceeding the limit. Contrary, the orange areas are above the limit value according to estimation, but with a chance of non-exceedance caused by the uncertainty of the estimation. The patterns in the spatial distribution of the different probability of exceedance classes over Europe differ somewhat from those of 2005 and 2006. In the south-eastern countries of Europe where relative little measurement stations are located, Romania, Bulgaria and Greece show some reduced extend and level of probability of exceedance at many existing yellow and orange spots (red shifted to orange, orange to yellow). Especially the capitals of the Romania and Bulgaria show clear reductions. The Iberian Peninsula shows in 2006 an elevated number of spots with increased probability of exceedances compared to the years 2005 and 2007 with their very a similar spatial pattern.

In the other areas where exceedances are observed, such as the Po Valley and the south of Poland, the probability of exceedances are also the highest, meaning that in these areas substantial reductions may be needed to reach non-exceedance levels in the future.



Figure 4.3 Map with the probability of the limit value exceedance for  $PM_{10}$  annual average for 2007, in  $\mu g.m^{-3}$  on the European scale on the 10 x 10 km grid resolution.-Interpolation uncertainty only is considered, not other sources of uncertainty.

# 4.2 36<sup>th</sup> highest daily average

### 4.2.1 Concentration map

Similar to the  $PM_{10}$  annual average map, the final interpolated map of  $36^{th}$  highest daily values is created by combining the rural and urban maps. Both rural and urban maps were created by combining the annual averages from the measured  $PM_{10}$  concentrations with supplementary data in a linear regression model, followed by the interpolation of its residuals by ordinary kriging.

The recommended supplementary data, given by Horálek et al. (2008), are the same as for annual average, i.e. EMEP model output, altitude, wind speed and surface solar radiation for rural areas and EMEP model output for urban areas. (The relevant linear regression submodels are identified as P.Eawr and UP.E respectively).

The estimated parameters of the linear regression models and of the residual kriging are presented in Table 4.4, including the statistical indicators for both the linear regression models and the residual kriging.

Table 4.4 Parameters of the linear regression models (Eq.2.1) and of the ordinary kriging variograms (nugget, sill, range) - and their statistics - of  $PM_{10}$  indicator  $36^{th}$  maximum daily mean for 2007 in the rural (left) and urban (right) areas as used for final mapping, i.e. rural linear regression model P.Eawr (left), resp. urban UP.E (right), followed by the interpolation on its regression residuals using ordinary kriging (OK, coded with 'a').

linear regr. model + OK on	rural areas (P.Eawr-a)	urban areas (UP.E-a)
its residuals	coeff.	coeff.
c (constant)	22.55	31.02
a1 (EMEP model 2006)	1.44	1.01
a2 (altitude GTOPO)	-0.0098	
a3 (wind speed 2006)	-4.64	
a4 (s. solar radiation 2006)	1.26	
adjusted R <sup>2</sup>	0.41	0.09
standard error [µg.m <sup>-3</sup> ]	9.73	14.55
nugget	40	65
sill	92	157
range [km]	460	280
RMSE [µg.m <sup>-3</sup> ]	7.99	9.07
MPE [μg.m <sup>-3</sup> ]	0.28	0.13

The regressions on the 2007 data have an adjusted  $R^2$  of 0.41 for the rural areas and 0.09 for the urban areas. This fit is better than in the year 2006 (and closer to 2005), where the adjusted  $R^2$  was 0.27 (0.29) for rural areas and 0.02 (0.06) for urban areas (De Smet et al. 2009, Horálek et al. 2008). RMSE and MPE are the cross-validation indicators for the quality of the resulting map. The RMSE analysis and comparison with 2006 results is discussed in more detail in Section 4.2.3.

The final map is presented in Figure 4.4. The areas and stations in the combined map where the limit value (LV) of 50  $\mu$ g.m<sup>-3</sup> is exceeded on more than 35 days are coloured red and purple.



Figure 4.4 Combined rural and urban concentration map of  $PM_{10} - 36^{th}$  maximum daily average values, year 2007. Units:  $\mu g.m^{-3}$ .

### 4.2.2 Population exposure

Table 4.5 gives the population frequency distribution for a limited number of exposure classes, as well as the population-weighted concentration, for individual countries and for Europe as a whole.

Table 4.5 Population exposure and population weighted concentration  $-PM_{10}$ ,  $36^{th}$  maximum daily average value, year 2007.

Country				20	07 Percent	[%]		Denulation
		Population		< LV		>	LV	Population-
			< 20	20 - 30	30 - 50	50 - 65	> 65	weighted conc.
		x1000	µg.m <sup>-3</sup>	µg.m <sup>-3</sup>	µg.m <sup>-3</sup>	µg.m⁻³	µg.m <sup>-3</sup>	μg.m <sup>-3</sup>
Austria	AL	8271	2.0	13.9	81.5	2.6	0	37.2
Belgium	BE	10579	0	2.7	93.2	4.1	0	43.0
Bulgaria	BG	7982	0	0.1	40.8	28.0	31.1	61.8
Croatia	HR	4464	0	0.1	66.2	33.7	0	47.7
Cyprus	CY	852	0	0	15.9	84.1	0	53.6
Czech Republic	CZ	10164	0	4.2	80.7	7.9	7.3	43.6
Denmark	DK	5415	0.4	45.9	53.7	0	0	31.3
Estonia	EE	1335	26.8	35.4	37.8	0	0	24.8
Finland	FI	5129	40.1	59.9	0.0	0	0	21.7
France	FR	58495	0.5	6.4	88.8	4.3	0	40.0
Germany	DE	82111	0.1	18.6	81.3	0	0	34.5
Greece	GR	10967	0	0	32.4	67.5	0	51.4
Hungary	ΗU	10128	0	0	73.0	27	0	45.6
Ireland	IE	3730	11	86.1	3.1	0	0	23.7
Italy	IT	56794	0.2	0.9	37.3	30.1	31.5	56.8
Latvia	LV	2383	2.2	54.2	43.6	0	0	29.5
Liechtenstein	LI	67	0	0	100	0	0	36.9
Lithuania	LV	3469	0	56.6	43.4	0	0	31.4
Luxembourg	LU	425	0	18.4	81.6	0	0	31.4
Malta	мт	395	0	0	100	0	0	42.6
Netherlands	NL	15729	0	0.3	99.7	0	0	41.8
Poland	PL	38223	0	8.2	56.5	21.9	13.4	47.0
Portugal	PT	9906	0.5	14.0	62.8	22.7	0	42.8
Romania	RO	22428	0	1.0	44.7	27.0	27.3	53.2
San Marino	SM	20	0	0	0.0	100	0	53.2
Slovakia	SK	5298	0	0.5	78.3	17.5	3.7	45.5
Slovenia	SI	2030	0	1.1	72.5	26.4	0	42.9
Spain	ES	38992	0.5	7.5	59.1	31.5	1.3	45.0
Sweden	SE	8887	31.5	63.2	5.3	0	0	22.9
United Kingdom	UK	59029	3.0	19.0	77.8	0.1	0	33.7
Albania	AL	3927	0	0.3	32.3	67.5	0	52.0
Andorra	AD	61	30.6	0	69.4	0	0	25.7
Bosnia-Herzegovina	ВА	4175	0	0	41.9	58.1	0	50.6
Iceland	IS	178	57.9	41.8	0.3	0	0	18.1
Macedonia, F.Y.R. of	ΜК	2275	0	0.6	33.8	37.9	27.7	54.2
Montenegro	ME	713	0	0.6	51.9	47.5	0	48.2
Norway	NO	3187	27.7	41.9	30.4	0	0	24.3
Serbia	RS	10736	0	0.1	29.3	33.4	37.3	58.2
Switzerland	СН	7238	1.5	7.4	90.4	0.8	0.0	37.9
Tatal	-	E46400	1.8	11.7	64.4	14.7	7.3	40.4
iotai		510188		78.0	-	22	2.0	42.4

Note: In the lower pane countries for which the population numbers are based on ORNL population data with uncertain quality are AD, AL, BA, CH, IS, ME, MK, NO, RS. Turkey could not be included in the calculation due to lack of air quality or population density data.

It is estimated that in 2007 about 22% of the total European population lived in areas where the  $36^{\text{th}}$  maximum daily mean of PM<sub>10</sub> exceeds the limit value of 50 µg.m<sup>-3</sup>. However, in Bulgaria, Cyprus, Greece, Italy, Romania, Albania, Bosnia-Herzegovina, FYR of Macedonia and Serbia both the populated weighted indicator concentration and the median were above the LV, implicating that in these countries the average concentration per inhabitant exceeded the LV and more than half of its population was exposed to concentrations exceeding the LV. In San Marino the complete population lived above the LV. As the interpolation methodology tends to underestimate high values, these numbers would most likely be even higher. The percentage of the total European population living in areas above the LV reduced in 2007 by about 6% compared to that of 2005 and 2006.

Such reduction is less obvious at the overall European population-weighted concentration of the  $36^{\text{th}}$  maximum daily mean, which is estimated for the year 2007 as about 42 µg.m<sup>-3</sup>. That is about 1.5 µg.m<sup>-3</sup> lower than in 2005 (Horálek et al. 2008) and about 3 µg.m<sup>-3</sup> lower than in 2006 (De Smet et al. 2009). The decrease of the population-weighted concentration in comparison with 2006 results is present in almost all countries. The major exception is France: an increase of about 7 µg.m<sup>-3</sup> is seen here. Starting in 2007 France submits corrected PM10 data to guarantee equivalence with the reference method. In a sensitivity run with/without corrected French data De Leeuw and Fiala (2009) conclude that if the correction is applied to previous years, the 2007-increase in France disappears.

Comparing the observed PM10 exceedances in 2007 for the indicator annual average (section 4.1.2) with 36th maximum daily average, one can conclude that the daily limit value is the most stringent of the two. Therefore, to comply with EU ambient air pollution legislation on PM10, countries best give preference to measures reducing PM10 concentrations to levels below the limit value for the 36th maximum daily mean.

### 4.2.3 Uncertainties

### Uncertainty estimated by cross-validation

At first, the cross-validation analysis is executed. In Table 4.4 the absolute mean uncertainty of the combined map of  $PM_{10}$  indicator  $36^{th}$  highest daily mean expressed by RMSE is 8.0 µg.m<sup>-3</sup> for the rural areas and 9.1 µg.m<sup>-3</sup> for the urban areas. For the 2005 and 2006 maps the RMSE values were 13.3 and 9.8 for rural areas, and 9.9 and 11.7 µg.m<sup>-3</sup> for urban areas respectively. It indicates that both rural and urban maps of 2007 show a lower absolute uncertainty than those of the 2005 and 2006 map.

This relative mean uncertainty of the map of  $PM_{10}$  indicator 36<sup>th</sup> highest daily mean is 23.5% for rural areas and 19.6% for urban areas. Compared to the 2005 and 2006 map with 26.3% and 26.6% for rural areas and 21.4% and 23.5% for urban areas, the relative uncertainty in the 2007 map is somewhat lower for both the rural and the urban areas.

Figure 4.5 shows the cross-validation scatter plots for both rural and urban areas. The  $R^2$  indicates that for the rural areas about 60% and for the urban areas about 65% of the variability is attributable to the interpolation. Corresponding values with those of the 2005 map (55% and 75%) and the 2006 maps (56% and 65%) show a somewhat improved fit for the rural areas in 2007, but a somewhat reduced fit for the urban areas.

The scatter plots indicate that in areas with high concentrations the interpolation methods tend to underestimate the levels. For example, in urban areas (Figure 4.5, right panel) an observed value of 70  $\mu$ g.m<sup>-3</sup> would be estimated in the interpolation as about 62  $\mu$ g.m<sup>-3</sup>, i.e. about 11% too low.



Figure 4.5 Correlation between cross-validation predicted values (y-axis) and measurements (x-axis) for the  $PM_{10}$  annual average for 2007 for rural (left) and urban (right) areas.  $R^2$  and the slope a (from the linear regression equation  $y = a \cdot x + c$ ) should be as close 1 as possible, the intercept c should be as close 0 as possible.

#### Comparison of point measurement values with the predicted grid value

Additional to the cross-validation, a simple comparison has been made between the point measurement and interpolated grid values. The results of the cross-validation compared to the gridded validation examination are summarised in Table 4.6. The uncertainty at measurement locations is caused partly by the smoothing effect of the interpolation and partly by the spatial averaging of the values in the 10x10 km grid cells. The level of the smoothing leading to underestimation at high values is here smaller than it is at areas without measurement: For example, in urban areas the predicted interpolation gridded value will be about 64  $\mu$ g.m<sup>-3</sup> at the corresponding station point with the measurement value of 70  $\mu$ g.m<sup>-3</sup>, i.e. an underestimation of about 9%.

Table 4.6 Linear regression equation and coefficient of determination  $R^2$  from the scatter plots of (i) the predicted point values based on cross-validation and (ii) the aggregation into 10x10 km grid cells versus the measured point values for PM<sub>10</sub> indicator  $36^{th}$  maximum daily mean for rural and urban areas in 2007.

	rural areas	S	urban areas		
	equation	R <sup>2</sup>	equation	R <sup>4</sup>	
i) cross-validation prediction	y = 0.623x + 13.07	0.597	y = 0.660x + 15.80	0.647	
ii) 10x10 km grid prediction	y = 0.736x + 9.10	0.809	y = 0.744x + 11.79	0.806	

#### Probability of LV exceedance map

Next to the cross-validation analysis and the comparison of predicted grid values with the points of measurement, the map with the probability of the limit value exceedance has been constructed, using the concentration maps (Figure 4.4), the uncertainty map and the limit value (LV), defined in the directive as  $50 \ \mu g.m^{-3}$  for the  $36^{th}$  highest daily mean. The probability map is presented in Figure 4.6. Areas with the probability of limit value exceedance above 75% are marked in red; areas below 25% are marked in green. Areas with 25-50%, resp. 50-75% probability of LV exceedance are marked in yellow and orange. Section 4.1.3 explains in more detail the significance of the colour classes in the map.

Comparing the 2005 and 2006 with the 2007 probability of exceedance (PoE), one can conclude that in 2006 and 2007 most of the Iberian Peninsula shows that the yellow contribution has turned into green, except at the largest cities and agglomerations. It means hardly any probability of exceedance resides, indicating that policy targets are or may be reached for these areas. Furthermore, several urbanised agglomerations, like Madrid, Barcelona and Lisbon, show a clear shift from orange to yellow, indicating a decreased likelihood of exceedance at these urbanised areas. As the interpolated maps refer to the rural or (sub)urban background situations only it cannot be excluded that exceedances of the limit values occur at *hotspot* or traffic situations. Lowering of probabilities of exceedances from 2005 through 2007 are also observed in the Black Triangle. Whereas in the Benelux, Denmark, north-eastern Poland, Latvia, and Norway the increase from 2005 to 2006 diminishes mostly again in 2007 to levels of 2005. The same happens in Greece but to a less extend. The increases observed in 2006 in Hungary, Romania, Bulgaria, Balkan areas, east-coast of Italy, and some coastal zones of Greece, where the PoE had gone up from yellow to orange has been dropped again in 2007 to levels around or even below 2005. In 2007, the extent of the yellow and orange areas is smaller then in 2005 except for Bulgaria, but the red areas seem to have concentrated and intensified at several more local spots that are known as larger agglomerations and  $PM_{10}$  emitting industrial areas. In other words, one observes a considerably reduced likelihood of exceedances in the many rural areas in South-East Europe, but with an increased likelihood at several urban and more industrialised areas. Most striking is the ongoing enlargement from 2005 through 2007 of the red area at the Po-valley, indicating the likelihood of exceedances increases here from moderate to very likely. It is connected to an extension of an increased moderate likelihood of LV exceedances for the western and most populated part of northern Italy, with a large likelihood at the urbanised regions of Rome and Naples. In these areas considerable emission reductions may be needed to reach non-exceedance levels in the future.



Figure 4.6 Map with the probability of the limit value exceedance for  $PM_{10}$  indicators  $36^{th}$  maximum daily mean, in  $\mu$ g.m<sup>-3</sup> on the European scale in 2007 on the 10 x 10 km grid resolution. Interpolation uncertainty only is considered, not other sources of uncertainty.

# 5 Ozone maps

For ozone two health-related indicators, the 26<sup>th</sup> highest daily maximum 8-hour running mean and the SOMO35, and two vegetation-related indicators, the AOT40 for crops and the AOT40 for forests, are considered. The maps of health-related indicators are created using the combination of rural and urban areas as described in Chapter 2 and are presented in the EEA LAEA5210 10x10 km grid. The maps of vegetation-related indicators are created for rural areas only and in a 2x2 km grid covering the same domain as the 10x10 km grid. This higher resolution is to serve the needs of the EEA Core Set Indicator 005 on ecosystem exposure to ozone.

## 5.1 26<sup>th</sup> highest daily maximum 8-hour average

## 5.1.1 Concentration map

T 11 5 1 D

C 1 1.

The interpolated map for 26<sup>th</sup> highest daily maximum 8-hour average is created by combining the rural and urban maps following the procedures described in Chapter 2 and in more detail in Horálek et al. (2007). Both rural and urban maps were created by combining the measured ozone concentrations with supplementary data in a linear regression model, followed by the interpolation of its residuals by ordinary kriging.

Supplementary data are used in accordance with the recommendations given by Horálek et al. (2008). The recommended supplementary data for rural areas are EMEP model output, altitude and surface solar radiation for rural areas, and EMEP model output, wind speed and surface solar radiation for urban areas. (The relevant linear regression submodels are identified as O.Ear and UO.Ewr respectively).

The estimated parameters of the linear regression models and of the residual kriging are presented in Table 5.1 including the statistical indicators of both the regression and the kriging. The fit of the regression relation expressed as the  $R^2$  is in 2007 with values of 0.51 for rural areas and 0.48 for urban areas slightly better than observed for the years 2006 (0.40 and 0.43) and 2005 (0.45 and 0.51) (De Smet et al. 2009, Table 5.1; Horálek et al. 2008, Tables A3.1 and A3.11). The numbers show that over the years the fit of the regressions are of the same order of magnitude at both the rural and the urban areas. RMSE and MPE are the cross-validation indicators, showing the quality of the resulting map. Section 5.1.3 discusses in more detail the RMSE analysis and comparison with results of 2005 and 2006.

Table 5.1 Parameters of the linear regression models (Eq. 2.1) and of the oralinary kriging variograms (hugget,
sill, range) - and their statistics - of ozone indicator 26 <sup>th</sup> highest daily maximum 8-hour mean for 2007 in the
rural (left) and urban (right) areas as used for final mapping i.e. linear regression model O.Ear (left), resp.
UO.Ewr (right) followed by the interpolation on its residuals using ordinary kriging (OK, coded 'a').

linear regr. model + OK on	rural areas (O.Ear-a)	urban areas (UO.Ewr-a)	
its residuals	coeff.	coeff.	
c (constant)	22.14	42.40	
a1 (EMEP model 2007)	0.64	0.63	
a2 (altitude GTOPO)	0.012		
a3 (wind speed 2007)		-3.89	
a4 (s. solar radiation 2007)	1.39	0.71	
adjusted R <sup>2</sup>	0.51	0.48	
standard error [µg.m⁻³]	11.34	11.04	
nugget	59	49	
sill	108	84	
range [km]	440	180	
RMSE [µg.m <sup>-3</sup> ]	8.79	8.92	
MPE [μg.m <sup>-3</sup> ]	0.02	0.04	

The final map is presented in Figure 5.1. The areas and stations in the combined map where the target value (TV) of 120  $\mu$ g.m<sup>-3</sup> is exceeded are coloured red and purple.



Figure 5.1 Combined rural and urban concentration map of ozone health indicators  $26^{th}$  highest daily maximum 8-hour value in  $\mu$ g.m<sup>-3</sup> for the year 2007. Its target value is  $120 \ \mu$ g.m<sup>-3</sup>.

## 5.1.2 Population exposure

Table 5.2 gives for 26<sup>th</sup> highest daily maximum 8-hour mean the population frequency distribution for a limited number of exposure classes, as well as the population-weighted concentration for individual countries and for Europe as a whole.

liour mean jor me yea	. 2007			200	07 Percent [	%]		
		Population		< TV	-	- > T	v	Population-
Country			< 100	100 - 110	110 - 120	120 - 140	× 140	weighted conc.
		¥1000	ua.m <sup>-3</sup>	ua.m <sup>-3</sup>	ua.m <sup>-3</sup>	ua.m <sup>-3</sup>	<sup>2</sup> 140	ua.m <sup>-3</sup>
Austria	Δ1	8271		<b>~3</b>	<b>rg</b>	95 3	0.2	124 Q
Rolaium		10570	50.0	30.5	14.5	05.5	0.2	00.1
Bulgaria		7082	- 59.0 7.0	39.5	1.0	54.0	02	99.1
Croatia	ЫО	1902	7.9	9.4	20.5	02.5	0.2	10.5
Citoalia		4404	0	0	7.3 EC E	92.5	0.2	127.0
Cyprus Czach Dopublic		10164	0	0	00.0	43.3 72.4	0	119.0
		10104	0	0	20.0	73.4	0	122.0
Denmark		5415	01.1	38.7	0.2	0	0	97.9
Estonia		1335	97.5	2.7	0	0	0	95.0
Finiand		5129	99.9	0.1	0	0	0	90.3
France	FR	58495	10.6	46.8	26.5	16.1	0	109.7
Germany	DE	82111	0.3	30.3	46.0	23.4	0	114.5
Greece	GR	10967	0	0.4	12.8	84.5	2	128.9
Hungary	HU	10128	0	0	5.9	94.1	0	126.6
Ireland	IE 	3730	100	0	0	0	0	85.0
Italy	IT	56794	0	2.6	18.1	51.8	27.4	131.1
Latvia	LV	2383	98.3	1.7	0	0	0	96.1
Liechtenstein	LI	67	0	0	0	100	0	121.5
Lithuania	LV	3469	84	16.0	0	0	0	98.1
Luxembourg	LU	425	0	0	100	0	0	112.5
Malta	MT	395	0	77	14.7	7.9	0	110.9
Netherlands	NL	15729	78.3	21.7	0	0	0	94.3
Poland	PL	38223	0.1	26.2	52.8	20.9	0	114.3
Portugal	PT	9906	0	38.4	55.2	6.4	0	112.4
Romania	RO	22428	0	16.3	31.1	52.6	0	119.1
San Marino	SM	20	0	0	0	100	0	134.7
Slovakia	SK	5298	0	0	22.2	77.8	0	124.0
Slovenia	SI	2030	0	0	0	97.1	2.9	128.4
Spain	ES	38992	2.1	19.0	52.2	26.7	0	115.9
Sweden	SE	8887	74.8	25.2	0	0	0	96.4
United Kingdom	UK	59029	99.8	0.2	0	0	0	83.8
Albania	AL	3927	0	2.1	15.2	62.7	20.0	129.6
Andorra	AD	61	0	0	69.4	30.6	0	123.6
Bosnia-Herzegovina	BA	4175	0	1.8	18.3	78.0	1.9	125.4
Iceland	IS	178	100	0	0	0	0	84.3
Macedonia, F.Y.R. of	MK	2275	0	0	56.0	27.0	17.0	125.0
Montenegro	ME	713	0	0	32.3	42.0	26	130.7
Norway	NO	3187	96.3	3.7	0.0	0	0	92.5
Serbia	RS	10736	0	1.2	27.7	67.4	3.8	124.8
Switzerland	СН	7238	0	1.9	39.5	55.5	3.2	120.8
Total		516199	22.1	18.0	26.4	30.1	3.5	112.1
IUlai		510100		66.5		33.	5	112.1

Table 5.2 Population exposure and population weighted concentration – ozone,  $26^{th}$  highest daily maximum 8-hour mean for the year 2007.

Note: In the lower pane countries for which the population numbers are based on ORNL population data with uncertain quality are AD, AL, BA, CH, IS, ME, MK, NO, RS. Turkey could not be included in the calculation due to lack of air quality or population density data.

It is estimated that in 2007 almost 34% of the European population lived in areas where the ozone target value (TV of 120  $\mu$ g.m<sup>-3</sup>) of the 26<sup>th</sup> highest daily maximum 8-hour mean is exceeded. For Bulgaria and Romania the average concentration per inhabitant (i.e. population weighted concentration) is just under the target value, however slightly more than half of its population is exposed to concentrations above the TV. In Austria, Croatia, Greece and Hungary more than 80%, about three quarter of the Czech and Slovak population, more than half of the Swiss, and all inhabitants of Lichtenstein are exposed to concentrations just above the target value. All Slovenians lived above the TV, with a few percent exposed to higher exceedances. In San Marino all inhabitants are exposed to higher concentrations above the TV. At several Balkan countries a significant number

of inhabitants had been exposed to reasonably high concentrations above the target value. Three quarter of the Italians lived above the TV, with even a quarter of the total population exposed to values considerably above the target value. In Andorra and FYR of Macedonia the average concentration per inhabitant was above the target value, but with less than half of the population exposed to concentrations above the target value. As the current mapping methodology tends to underestimate high values, the numbers will most likely be higher.

In 2005 38% and in 2006 55% of the total European population was exposed to ozone levels above the TV. We observe in 2007 a considerable decrease in the higher ozone levels above the TV. In general, the frequency distribution shows for 2007 a shift to increased percentages at lower class intervals compared to 2006 and 2005. For example, the Iberian Peninsula shows lower values in 2007 compared to 2005 and 2006. In several southern and central European countries the values in 2007 are quite the same or higher than in 2006 and 2005. The northern and western European countries show in 2007 a rather similar pattern as in 2005.

The overall European population-weighted ozone concentration in terms of the  $26^{th}$  highest daily maximum 8-hour mean is estimated for the year 2007 as 112 µg.m<sup>-3</sup>. That is the same as for the year 2005 (Horálek et al. 2008) and a decrease of about 8 µg.m<sup>-3</sup> compared to the 2006 results (De Smet et al. 2009).

### 5.1.3 Uncertainties

### Uncertainty estimated by cross-validation

The basic uncertainty analysis is given by cross-validation. Table 5.1 shows RMSE values of 8.8  $\mu$ g.m<sup>-3</sup> for the rural areas and 8.9  $\mu$ g.m<sup>-3</sup> for the urban areas of the combined map. For the 2005 and 2006 map the RMSE values were 12.3 and 11.2  $\mu$ g.m<sup>-3</sup> for rural areas, and 10.0 and 10.2  $\mu$ g.m<sup>-3</sup> for urban areas respectively (Horálek et al. 2008, Tables A3.3, A3.12; De Smet et al. 2009, Table 5.1). It indicates that the 2007 map has a lower absolute mean uncertainty at both the rural and urban areas compared to the 2005 and 2006 map.

The relative mean uncertainty of the 2007 ozone map is 7.5% for rural areas and 7.9% for urban areas. Compared to the 2005 and 2006 map with relative uncertainties of 10.3% and 8.9% for rural areas, and 8.9% and 8.4% for urban areas, the relative uncertainty in the 2007 map is as well somewhat lower for both the rural and the urban areas of 2005 and 2006.

Figure 5.2 shows the cross-validation scatter plots for both the rural and urban areas of the 2007 map. The  $R^2$ , an indicator for the interpolation correlation with the observations, shows that for the rural areas about 71% and for the urban areas about 66% of the variability is attributable to the interpolation. Corresponding values for the 2005 map (51% and 50%) and the 2006 map (49% and 53%), show an improved fit at both the rural and urban interpolations in 2007.

The scatter plots indicate that the high values are underestimated by the interpolation method. This will lead to an underestimation at areas with higher ozone values. For example, in rural areas (Figure 5.2, left panel) an observed value of 170  $\mu$ g.m<sup>-3</sup> is estimated in the interpolation as 155  $\mu$ g.m<sup>-3</sup>, which is 8% too low.



Figure 5.2 Correlation between cross-validation predicted values (y-axis) and measurements (x-axis) for the ozone indicator  $26^{th}$  highest daily maximum 8-hour mean for rural (left) and urban (right) areas in 2007.

### Comparison of point measurement values with the predicted grid value

Additional to the cross-validation, a simple comparison has been made between the point measurement and interpolated grid values. The results of the cross-validation compared to the gridded validation examination are summarised in Table 5.3. The uncertainty at measurement locations is caused partly by the smoothing effect of interpolation and partly by the spatial averaging of the values in the 10x10 km grid cells. The level of smoothing leading to underestimation at high values is here smaller than it is at areas without measurement. For example, at rural areas the predicted interpolation grid value will be about 159  $\mu$ g.m<sup>-3</sup> at the corresponding station point with the observed value of 170  $\mu$ g.m<sup>-3</sup>, i.e. an underestimation of about 6%.

Table 5.3 Linear regression equation and coefficient of determination  $R^2$  from the scatter plots of (i) the predicted point values based on cross-validation and (ii) aggregation into 10x10 km grid cells versus the measured point values for the ozone indicator  $26^{th}$  highest daily maximum 8-hour mean for rural and urban areas of 2007.

	rural areas	S	urban areas			
	equation	R	equation	R		
i) cross-validation prediction	y = 0.732x + 31.42	0.707	y = 0.684x + 35.68	0.658		
ii) 10x10 km grid prediction	y = 0.798x + 23.61	0.832	y = 0.769x + 26.15	0.813		

#### Probability of TV exceedance map

Next to the cross-validation analysis and the comparison of predicted grid values with the points of measurement, the map with the probability of the target value exceedance has been constructed, using the concentration maps (Figure 5.1), the uncertainty map and the target value (TV). The probability map is presented in Figure 5.3. Areas with the probability of limit value exceedance above 75% are marked in red; areas below 25% are marked in green. Areas with 25-50%, resp. 50-75% probability of TV exceedance are marked in yellow and orange. Section 4.1.3 explains in more details the significance of the colour classes in the map.

Comparing the years 2005 to 2007 one observes for the year 2006 a temporal increase in the PoE to levels above 50% and even above 75% at large parts of specifically central Europe. There the areas change from low (green), reasonable (yellow) and moderate (orange) into a considerable (red) likelihood of exceedance. In 2007 in the north-western part (France, Benelux, northern Germany, Scandinavia) levels are mainly back to just below those of 2005. In the eastern part (South Poland,

Slovakia, Hungary, Balkan region) they stay somewhat elevated above those of 2005. The South-East of Europe shows in 2006 a considerable reduction of the area with PoE above 50%, and in 2007 an increase is observed to levels well above of those of 2005 and over a considerable more extended area.

The central-southern countries in Europe (Italy, South-East France, Alpine zone) show unchanged high PoE (red). Whereas, the Iberian Peninsula shows a continued decrease in the levels of PoE, meaning a ongoing reduced likelihood of exceeding target values. The natural meteorologically induced variations from year to year, combined with methodological uncertainties and the limited number of years considered here do not allow for conclusions on any significant tendency. For that purpose, one would need longer time series and reduced uncertainties.



Figure 5.3 Map with the probability of the target value exceedance for ozone indicator  $26^{th}$  highest daily maximum 8-hour average values (in  $\mu g.m^{-3}$ ) on European scale in 2007,  $10 \times 10$  km grid resolution. Interpolation uncertainty only is considered, not other sources of uncertainty.

## 5.2 SOMO35

### 5.2.1 Concentration map

The interpolated map for SOMO35 is created by combining the rural and urban maps, according the criterion as described in Section 2 and in Horálek et al. (2007). Both rural and urban maps were created by combining the annual averages from the measured ozone concentrations with supplementary data in a linear regression model, followed by the interpolation of its residuals by ordinary kriging.

The recommended supplementary data, given by Horálek et al. (2008), are the same as for 26<sup>th</sup> highest daily maximum 8-hour mean, i.e. EMEP model output, altitude and surface solar radiation for rural

areas and EMEP model output, wind speed and surface solar radiation for urban areas. (The relevant linear regression submodels are identified as O.Ear, resp. UO.Ewr.)

The estimated parameters of the linear regression models and of the residual kriging are presented in Table 5.4, including the statistical indicators of both the regression and the kriging. The fit of the regression is expressed by  $R^2$  and standard error. For 2007, the adjusted  $R^2$  is for both the rural areas and the urban areas 0.58. This fit is somewhat better than observed for the years 2006 (0.42 for rural areas and 0.38 for urban areas) and 2005 (0.51 and 0.49) (De Smet et al. 2009, Table 5.4; Horálek et al. 2008, Tables A3.1 and A3.11). RMSE and MPE are the cross-validation indicators showing the quality of the resulting map. Section 5.2.3 discusses in more detail the RMSE analysis and comparison with results of 2005 and 2006.

Table 5.4 Parameters of the linear regression models (Eq. 2.1) and of the ordinary kriging variograms (nugget, sill, range) - and their statistics - of ozone indicator SOMO35 for 2007 in the rural (left) and urban (right) areas as used for final mapping, i.e. rural linear regression model O.Ear (left), resp. UO.Ewr (right) followed by the interpolation on its residuals using ordinary kriging (OK, coded with 'a').

linear regr. model + OK on	rural areas (O.Ear-a)	urban areas (UO.Ewr-a)
its residuals	coeff.	coeff.
c (constant)	-2121	-1338
a1 (EMEP model 2007)	0.52	0.62
a2 (altitude GTOPO)	2.42	
a3 (wind speed 2007)		n. sign.
a4 (s. solar radiation 2007)	326.63	144.21
adjusted R <sup>2</sup>	0.58	0.58
standard error  [µg.m⁻³.d]	1925	1419
nugget	2.7E+06	1.0E+06
sill	3.0E+06	1.5E+06
range [km]	440	200
RMSE [µg.m <sup>-3</sup> .d]	1801	1260
MPE [µg.m <sup>-3</sup> .d]	-28	6

The final map is presented in Figure 5.4. SOMO35 is not subject to one of the EU air quality directives and no limit or target values have been defined, which does not offers the possibility to create a map with the probability of exceedances.



Figure 5.4 Combined rural and urban concentration map of ozone indicators SOMO35 in  $\mu$ g.m<sup>-3</sup>.days for the year 2007.

### 5.2.2 Population exposure

Table 5.5 gives for SOMO35 the population frequency distribution for a limited number of exposure classes, as well as the population-weighted concentration for individual countries and for Europe as a whole.

Country	1			2	007 Percent	[%]		Population-
		Population		3000 -	6000 -	10000 -		weighted
			< 3000	6000	10000	15000	> 15000	conc.
		x1000	µg.m⁻³.d	µg.m <sup>-3</sup> .d	µg.m⁻³.d	µg.m⁻³.d	µg.m⁻³.d	µg.m⁻³.d
Austria	AL	8271	0	22.8	75.6	1.7	0	6514
Belaium	BE	10579	94.6	5.4	0	0	0	2247
Bulgaria	BG	7982	0	35.6	59.3	5.1	0	6943
Croatia	HR	4464	0	6.9	92.5	0.6	0	7203
Cyprus	CY	852	0	0	72.0	28.0	0	8423
Czech Republic	CZ	10164	0	75.4	24.6	0	0	5457
Denmark	DK	5415	63.7	36.3	0	0	0	2704
Estonia	EE	1335	98.7	1.3	0	0	0	2200
Finland	FI	5129	99.9	0.1	0	0	0	1640
France	FR	58495	38.3	48.4	13.1	0.2	0	3832
Germany	DE	82111	28.0	69.3	2.7	0	0	3861
Greece	GR	10967	0	0.5	83.6	15.9	0	8820
Hungary	ΗU	10128	0	4.9	95.1	0	0	6761
Ireland	IE	3730	99.8	0.2	0	0	0	1494
Italy	IT	56794	0	7.4	88.0	4.6	0	7890
Latvia	LV	2383	94.7	5.3	0	0	0	2344
Liechtenstein	LI	67	0	100	0	0	0	5444
Lithuania	LV	3469	63.4	36.6	0	0	0	2809
Luxembourg	LU	425	0	100	0	0	0	3557
Malta	MT	395	0	0	97.9	2.1	0	7480
Netherlands	NL	15729	97.9	2.1	0	0	0	1823
Poland	PL	38223	0.9	96.4	2.6	0	0	4394
Portugal	PT	9906	0	80.1	19.9	0	0	5090
Romania	RO	22428	0	40.5	59.1	0.4	0	6405
San Marino	SM	20	0	0	100	0	0	8450
Slovakia	SK	5298	0	17.9	82.1	0	0	6503
Slovenia	SI	2030	0	14	85.9	0.2	0	7194
Spain	ES	38992	2.4	33.0	64.4	0.3	0	6145
Sweden	SE	8887	88.3	11.7	0	0	0	2229
United Kingdom	UK	59029	99.5	0.5	0	0	0	1209
Albania	AL	3927	0	3.2	77.4	19.4	0	8260
Andorra	AD	61	0	0	69.4	30.6	0	8473
Bosnia-Herzegovina	BA	4175	0	18.6	70.7	10.7	0	7554
Iceland	IS	178	100	0.0	0	0	0	1276
Macedonia, F.Y.R. of	MK	2275	0	51.6	28.1	20.3	0.0	7384
Montenegro	ME	713	0	13.0	46.3	40.6	0	8854
Norway	NO	3187	91.1	8.9	0.0	0	0	1937
Serbia	RS	10736	0	26.2	69.8	4.0	0	7225
Switzerland	СН	7238	0	81.5	16.6	1.8	0.0	5397
Total		516199	31.0	36.4	31.1	1.5	0.0	4670
		510100	67	7.4		32.6		40/9

Table 5.5 Population exposure and population weighted concentration – ozone, SOMO35, year 2007.

Note: In the lower pane countries for which the population numbers are based on ORNL population data with uncertain quality are give (AD, AL, BA, CH, IS, ME, MK, NO, RS). Turkey could not be included in the calculations due to lack of air quality or population density data.

It is estimated that in 2007 about 33% of the European population lived in areas with SOMO35 values above 6000  $\mu$ g.m<sup>-3</sup>.d. In 2005 34% and in 2006 37% of the population was exposed to ozone levels above this SOMO35 level, indicating that in 2007 only a limited decrease of population exposed to the higher SOMO35 levels is observed. Comparing the frequency distribution of the years 2005, 2006 and 2007, a slight overall shift to increased lower intervals is observed. This shift over the years is not observed in the maps due to the specific fluctuations in the distribution at the different European regions.

The table shows that, compared to 2005 and 2006, there is in 2006 an overall increase in the number of inhabitants exposed to higher ozone concentrations, which reduces again in 2007 slightly below the level of 2005. In 2006 the exposure shows a clear shift from the lowest class and a small shift from the higher classes to the medium-range exposure classes ( $3000 - 10000 \ \mu g.m^{-3}.d$ ); with a peak at class 3000-6000  $\mu g.m^{-3}.d$ . In 2007 the percentage of population exposed to medium-range SOMO35 levels reduces again in favour of a distribution over the lower SOMO35 exposure classes.

Northern and north-western Europe show in 2006 a shift in exposures from a lower class interval to its neighbouring higher interval. In 2007 an opposite shift occurs to the lower neighbouring classes and for a more extended area than in 2005. The Iberian Peninsula and a considerable part of the Balkan and Greece show decreasing values from 2005 to 2006 going down from above 10 mg.m<sup>-3</sup>.d to values between 6 - 10 mg.m<sup>-3</sup>.d. This tendency is reflected quite well by the shifts of the total population exposure percentages per class interval when comparing the 2005 values with those of 2006. In 2007 a slight further drop in the exposure levels is noticed at the Iberian Peninsula, Southern France and Italy, but in the south-eastern part of Europe levels go up again.

The total European population-weighted ozone concentration in terms of SOMO35 was estimated as 4700  $\mu$ g.m<sup>-3</sup>.d. This is about 370  $\mu$ g.m<sup>-3</sup>.d less than in 2005 and 800  $\mu$ g.m<sup>-3</sup>.d less than in 2006 (Horálek et al. 2008; De Smet et al. 2009) and may indicate some overall decrease in exposure from 2005 to 2007.

### 5.2.3 Uncertainties

### Uncertainty estimated by cross-validation

The basic uncertainty analysis is given by cross-validation. In Table 5.4 the absolute mean uncertainty is 1801  $\mu$ g.m<sup>-3</sup>.d for the rural areas and 1260  $\mu$ g.m<sup>-3</sup>.d for the urban areas. For the 2005 and 2006 map the RMSE values were 2173 and 2077  $\mu$ g.m<sup>-3</sup>.d for the rural areas, and 1459 and 1472  $\mu$ g.m<sup>-3</sup>.d for the urban areas. This indicates the 2007 map has a clearly lower absolute uncertainty at both the rural and the urban areas.

The relative mean uncertainty of the 2007 map of SOMO35 is 33.3% for rural areas and 29.5% for urban areas. The uncertainty for rural areas is in 2005 with 35.5% and in 2006 with 31.6% of the same extent. For urban areas the relative mean uncertainty in 2005 is with 32% slightly higher than in 2006 and 2007, where both have 29.2%.

Figure 5.5 shows the cross-validation scatter plots for interpolated values at both the rural and urban areas. The  $R^2$  indicates that for the rural areas about 63% and for the urban areas about 67% of the variability is attributable to the interpolation. The corresponding values for the 2005 map (55% and 58%) and 2006 map (47% and 49%), illustrate a somewhat improved fit at both rural and urban areas in 2007.

The scatter plots show again that in areas with high concentrations the interpolation methods tend to underestimate predicted values. For example, in urban areas (Figure 5.5, right panel) an observed value of 10 000  $\mu$ g.m<sup>-3</sup>.d is estimated in the interpolation as 8200  $\mu$ g.m<sup>-3</sup>.d, which is 18% too low leading in general to high underestimations at high SOMO35 values.



Figure 5.5  $\overline{C}$  orrelation between cross-validation predicted values (y-axis) and measurements (x-axis) for the ozone indicator SOMO35 for rural (left) and urban (right) areas in 2007.

### Comparison of point measurement values with the predicted grid value

Additional to the point cross-validation, a simple comparison has been made between the point measurement and interpolated values averaged in a 10x10 km grid. The results of the cross-validation compared to the gridded validation are summarised in Table 5.6. The uncertainty at measurement locations is caused partly by the smoothing effect of the interpolation and partly by the spatial averaging of the values in the 10x10 km grid cells. The level of the smoothing effect leading to underestimation at high values is there smaller than it is at the areas without measurement. For example, at urban areas the predicted interpolation grid value will be about 8600  $\mu$ g.m<sup>-3</sup>.d at the corresponding station point with the observed value of 10 000  $\mu$ g.m<sup>-3</sup>.d, i.e. an underestimation of about 14%.

Table 5.6	Linear	r regressio	n equ	ation	and	coeffic	cient	of	determination	$R^2$	from	the	scatter	plots	of (i)	the
predicted	point 1	values bas	ed on	cross	-vali	dation	and	(ii)	aggregation	into	10x1	0 k	n grid	cells	versus	the
measured	point v	alues for th	ie ozor	ie indi	icato	r SOM	035	for i	rural and urba	n ar	eas.					

	rural areas	6	urban areas			
	equation	R <sup>2</sup>	equation	R <sup>2</sup>		
i) cross-validation prediction	y = 0.641x + 1921	0.634	y = 0.685x + 1358	0.668		
ii) 10x10 km grid prediction	y = 0.673x + 1752	0.692	y = 0.752x + 1068	0.739		

No limit or target value is set for the WHO recommended ozone health indicator SOMO35, therefore no probability of exceedance map has been prepared.

## 5.3 AOT40 for crops and for forests

The ecosystem based accumulative ozone indicators described in this section are specifically intended for insertion in the EEA Core Set of Indicator 005 (CSI005, <u>http://themes.eea.europa.eu/indicators</u>). For the estimation of the vegetation and forest land areas exposures to accumulated ozone the maps in this section are created on a 2x2 km grid instead 10x10 km as used at the other indicators in this paper. This resolution is selected as compromise between calculation time and accuracy in the impact analysis done for the ozone impact assessment of the CSI005, which uses results of this section. It serves a refinement of the exposure frequency distribution outcomes of the overlay with the 100x100 meter CLC2000 land cover classes.

### 5.3.1 Concentration maps

The interpolated maps for AOT40 for crops and AOT40 for forests are created for rural areas only, combining AOT40 data derived from station observations with supplementary data sources, as recommended in Horálek et al. (2008). The recommended supplementary data are here the same as for the human health related ozone indicators: EMEP model output, altitude and surface solar radiation. (The relevant linear regression submodel is identified as O.Ear.)

The estimated parameters of the linear regression models and of the residual kriging are presented in Table 5.7, including their statistical indicators of the regression and kriging. The fit of the regression is expressed by  $R^2$  and the standard error. For 2007 the adjusted  $R^2$  for the rural areas is 0.49 for the AOT40 for crops and 0.59 for AOT40 for forests. This fit is somewhat better than the one for crops and especially for forests in 2006 (0.45 and 0.47) but less for crops and somewhat better for forests compared to 2005 (0.53 and 0.52) (De Smet et al. 2009, Table 5.7; Horálek et al. 2008, Table A3.2). RMSE and MPE are the cross-validation indicators, showing the quality of the resulting map. Section 5.2.3 discusses in more detail the RMSE analysis and comparison with results of 2005 and 2006.

Table 5.7 Parameters of the linear regression models (Eq2.1) and of the ordinary kriging variograms (nugget, sill, range) - and their statistics - of ozone indicators AOT40 for crops (left) and for forests (right) for 2007 in the rural areas as used for final mapping, i.e. rural linear regression model O.Ear followed by the interpolation on its residuals using ordinary kriging (OK, coded with 'a').

linear regr. model + OK on	AOT40 for crops (O.Ear-a)	AOT40 for forests (O.Ear-a)
its residuals	coeff.	coeff.
c (constant)	-4108	-10729
a1 (EMEP model 2006)	1.10	0.53
a2 (altitude GTOPO)	5.38	13.37
a3 (s. solar radiation 2006)	914.3	1163.3
adjusted R <sup>2</sup>	0.49	0.59
standard error [µg.m⁻³]	6889	11228
nugget	2.0E+07	4.7E+07
sill	3.3E+07	8.5E+07
range [km]	400	170
RMSE [µg.m <sup>-3</sup> ]	5876	10190
MPE [µg.m <sup>-3</sup> ]	-35	-72

Figure 5.6 presents the final map of AOT40 for crops. The areas and stations in the map that exceed the target value (TV) of 18 mg.m<sup>-3</sup>.h are marked in red and purple. It concerns a map for rural areas, just based on rural background station observations, representing an indicator for vegetation exposure to ozone while assuming there is no relevant vegetation in the urban areas.

The same holds for the final rural map of AOT40 for forests as presented in Figure 5.7. However, for AOT40 for forests there is no TV defined.

Both maps showed throughout Europe for 2006 an overall increase in the levels of AOT40 with large areas in exceedance to the target value for crops (red and purple), even extending into the UK and Norway where 2005 showed low values of below 6 000  $\mu$ g.m<sup>-3</sup>.h. In 2007 at most of these areas the levels have been dropped again even somewhat below those of 2005, except for Italy, the Balkan and the remaining of south-eastern Europe, where the TV for crops continues to be exceeded everywhere.



Figure 5.6 Rural concentration map of ozone vegetation indicator AOT40 for crops for the year 2007. Units:  $\mu g.m^{-3}$ .hours.



*Figure 5.7 Rural concentration map of ozone vegetation indicator AOT40 for forests for the year 2007. Units:*  $\mu$ *g.m<sup>-3</sup>.hours.* 

### 5.3.2 Vegetation exposure

### Agricultural crops

The rural map with ozone indicator AOT40 for vegetation, i.e. agricultural crops, as given in Figure 5.6 has been combined with the land cover CLC2000 map. Following a similar procedure as described in Horálek et al. (2007) the exposure of agricultural areas, 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) has been calculated at the country-level. Table 5.8 gives the absolute and relative agricultural area for each country and for four European regions where the target value (TV) and long-term objective (LTO) for ozone are exceeded. The table presents the frequency distribution of the agricultural area per country over the exposure classes as well.

The table indicates the country grouping with corresponding colours of the region; *Northern Europe*: Sweden, Finland, Norway, Estonia, Lithuania, Latvia and Denmark. *North-western Europe*: United Kingdom, Ireland, Iceland, the Netherlands, Belgium, Luxembourg and France north of 45 degrees latitude. *Central and Eastern Europe*: Germany, Poland, Czech Republic, Slovakia, Hungary, Austria, Liechtenstein, Bulgaria and Romania. *Southern Europe*: Albania, Bosnia-Herzegovina, France south of 45 degrees latitude, Portugal, Spain, Italy, San Marino, Slovenia, Croatia, Greece, Cyprus, F.Y.R. of Macedonia, Montenegro, Serbia and Malta.

Table 5.8 illustrates that in 2007 about 36% of all European agricultural land was exposed to ozone exceeding the target value (TV) of 18 mg.m<sup>-3</sup>.h and about 78% was exposed to levels in excess of the long-term objective (LTO) of 6 mg.m<sup>-3</sup>.h. This is a substantial decrease in the total area with agricultural crops considered to suffer from adverse effects to ozone exposure compared to both 2006 and 2005, where about 70% and 49% of agricultural land was exposed to ozone levels in excess of the target value. Considering the long-term objective a considerable smaller area is in excess than in 2005 (89%) and 2006 (98%). Finland, Iceland, Ireland and the UK show in 2007 accumulated ozone levels not being in excess of any of the thresholds; Estonia and Norway show only a few percent of their forests exposed to levels in excess of the LTO as most stringent; for Sweden, Latvia and The Netherlands it is less than half of their agricultural area. For the remainder of the countries at least more than half of the agricultural is in excess of the LTO, with for many their complete agricultural area. In several countries, even their complete agricultural area experienced exposures above the target value as least stringent threshold.

In southern Europe, about 55% of the total agricultural area exceeds in 2007 the target value. This is a substantial reduced amount compared to 2005 (96%) and in 2006 (94%). In northern Europe, the ozone levels are below the target value for about 95% in 2006 and in both 2005 and 2007 no area is mapped in excess. In the north-western region the area exceeding the target value is almost 50% in 2006, which is more than four times larger than in 2005 (11%). However, in 2007 ozone levels have dropped such that only less than 1% of the area is still in excess. For the central and eastern region the total area where ozone exceeds the target value increases considerably from 2005 to 2006: from 44% to 77%. In 2007 it drops to an area of 50% of the total, being just above the level of 2005. Compared to 2005, the frequency distribution of agricultural area over the exposure classes shows for 2006 a clear shift towards higher exposures leading to an increased total area exceeded. In 2007 this shift diminishes again to a distribution very similar to that of 2005, but with a small increase in the area not exceeding the target value.

o <b>_</b> on <b>o</b> , iio i , o joi	<i>e. ops, ye</i>	Agricu	Itural Area	, 2007		Per	centage of	agricultura	l area, 2007	[%]
Country	tot. area	> LTO (6	mg.m⁻³.h)	> TV (18 mg	g.m <sup>-3</sup> .h)	< 6	6 - 12	12 - 18	18 - 27	> 27
	[km <sup>2</sup> ]	[km <sup>2</sup> ]	[%]	[km <sup>2</sup> ]	[%]	mg.m <sup>-3</sup> .h	mq.m <sup>-3</sup> .h	mg.m <sup>-3</sup> .h	mg.m <sup>-3</sup> .h	mq.m <sup>-3</sup> .h
Albania	7185	7185	100	7185	100	0	0	0	0	100
Austria	27461	27461	100	22473	81.8	0	0	18.2	81.3	0.6
Belgium	17652	9132	51.7	0	0	48.3	51.7	0	0	0
Bosnia-Herzegovina	19316	19316	100	19316	100	0	0	0	51.1	48.9
Bulgaria	57387	57387	100	57141	99.6	0	0	0.4	94.2	5.4
Croatia	24135	24135	100	24135	100	0	0	0	52.3	47.7
Cyprus	4290	4290	100	4290	100	0	0	0	33.2	66.8
Czech Republic	45550	45550	100	37818	83.0	0	0	17.0	83.0	0
Denmark	32248	28816	89.4	0	0	10.6	88.8	0.6	0	0
Estonia	14686	327	2.2	0	0	97.8	2.2	0	0	0
Finland	28832	2	0.0	0	0	100.0	0.0	0	0	0
France	328466	190135	57.9	11134	3.4	42.1	45.6	8.9	3.4	0.0
Germany	213504	207084	97.0	7607	3.6	3.0	40.7	52.8	3.6	0
Greece	51570	51570	100	50252	97.4	0	0	2.6	27.6	69.8
Hungary	63087	63087	100	63087	100	0	0	0	74.9	25.1
Iceland	2381	0	0	0	0	100	0	0	0	0
Ireland	46393	0	0	0	0	100	0	0	0	0
Italy	155657	155657	100	130691	84.0	0	2.2	13.9	38.9	45.1
Latvia	28293	4775	16.9	0	0	83.1	16.9	0	0	0
Liechtenstein	42	42	100	3	7.7	0	0	92.3	7.7	0
Lithuania	40039	27693	69.2	0	0	30.8	69.2	0	0	0
Luxembourg	1411	1411	100	0	0	0	100	0	0	0
Macedonia, FYR	9506	9506	100	9506	100	0	0	0	0	100
Malta	123	123	100	122	99.1	0	0	0.9	99.1	0
Monaco	1	1	100	0	92.3	0	0	7.7	92.3	0
Montenegro	2396	2396	100	2396	100	0	0	0	0	100
Netherlands	24903	6413	25.8	0	0	74.2	25.8	0	0	0
Norway	15388	5/3	3.7	0	0	96.3	3.7	0	0	0
Poland	200498	200498	100	42538	21.2	0	24.6	54.1	21.2	0
Portugal	42569	42130	99.0	0	0	1.0	98.1	0.9	0	0
Romania See Marine	134888	134888	100	130902	97.0	0	0	3.0	83.0	13.4
San Marino Sarbia	43	43	100	43	100	0	0	0		72.0
Selbia	40000	40000	100	40000	100	0	0	02	20.2	/ J.O 2 0
Slovania	24339 7127	24009 7107	100	24209 7107	100	0	0	0.3	90.9 70.7	3.0 27.3
Silvenia Snain	252313	250683	00 /	685/1	27.2	06	35.6	36.6	26.3	0.8
Sweden	38625	16881	99.4 13.7	00041	27.2	56.3	43.6	0.1	20.5	0.0
United Kingdom	141050	70		0	0	00.0 00 0	40.0 0.1	0.1	0	0
Total	2152765	1669239	77.5	769070	35.7	22.5	24.0	17.8	25.2	10.6
	2102700	1000200		100010	0011	2210	2.110		2012	1010
France N of 45N	260757	260672	100.0	191280	73.4	0.0	3.0	23.6	63.1	10.3
France S of 45N	67706	67706	100.0	64879	95.8	0.0	0.1	4.1	43.5	52.3
					- 5.0					- 1.0
Northern	198111	79068	39.9	0	0					
North-western	495473	147444	29.8	526	0.1					
Central & eastern	766757	760337	99.2	385829	50.3					
Southern	692424	682390	98.6	382716	55.3					
Total	2152765	1669239	77 5	769070	35.7					
10101	2102100	1000200		100010	00.1					

Table 5.8 Agricultural area exposure and exceedance (Long Term Objective, LTO, and Target Value, TV) for ozone, AOT40 for crops, year 2007.

Note: Countries not included due to lack of land cover data: Andorra, Switzerland, Turkey.

### Forests

The rural map with ozone indicator AOT40 for forests, as given in Figure 5.7 has been combined with the land cover CLC2000 map as well. Following a similar procedure as described in Horálek et al. (2007) the exposure of forest areas, defined as the Corine Land Cover level-2 class *3.1. Forests,* has been calculated at the country-level.

Table 5.9 gives for each country, four European regions and Europe as a whole the absolute and relative forest area where – let us call it – the *Reporting Value* (RV of 20 mg.m<sup>-3</sup>.h, as Annex III of the ozone directive defines it) in combination with the Critical Level (CL of 10 mg.m<sup>-3</sup>.h, as defined in the Mapping Manual) are exceeded. The table presents the frequency distribution of the forest area per country over the exposure classes as well. The reporting value of the ozone directive (RV) of 20 mg.m<sup>-</sup> <sup>3</sup>.h is exceeded in 2007 at almost 50% of the total European forest area, while in 2006 it was almost 70% and in 2005 about 60%. This means that the area of forest exposed to levels above the accumulated ozone reporting value initially increases in 2006 with 10% and than diminishes again in 2007 to a smaller area of even 10% below that of 2005. In 2005 three-quarter of the European forest area exceeded the critical level of 10 mg.m<sup>-3</sup>.h. In 2006 about all forested area the critical level was exceeded. This reduces again in 2007 to almost two-third of the total. Parallel to the agricultural thresholds, Finland, Iceland, Ireland and the UK show in 2007 accumulated ozone levels not exceeding any of the forest thresholds. Estonia and Latvia show only a few percent of their forest area exposed to levels exceeding the critical level as most stringent; for Sweden, Norway and The Netherlands it is less than half. For the remainder of the countries it is more then half of their forests, while for many even all their forests suffered exposure to levels in excess of the least stringent reporting value.

As in previous years, in 2007 the southern European region has AOT40 levels where about all forested areas exceed the critical level. Contrary to 2005 and 2006 where accumulated ozone concentrations were exceeded for all European forested areas, in 2007 a slight reduction resulted in some 94% exceedance. The central and eastern regions show over the three years a continued 100% exceedance of the critical levels. Whereas the area exceeding the reporting value shows a peak of 100% in 2006 followed by a reduction to about 85% in 2007, being some 10% lower than in 2005.

In the north-western region the area exceeding the critical level increases from 84% in 2005 to practically the whole area (98%) in 2006, and with an area increase above the reporting value from 69% in 2005 to 80% in 2006. In 2007 the critical level exceedance drops again to 78%, which is somewhat below that of 2005. Furthermore, the forest area exceeding the reporting value reduced quite prominently with more than 50% in 2006 to 28%, being more than 40% below that of 2005. Specifically in the northern region of Europe the area in exceedance peaks considerably in 2006: the area above the critical level enlarges from 40% in 2005 to even 100% in 2006 and reduces thereafter to just 12% in 2007. The reporting value peaks from no exceedance in 2005 to 23% in 2006 back to none in 2007. In comparison with 2005, the frequency distribution of the forested area over the exposure classes for 2006 shows a clear shift to higher exposures, specifically for the areas which had the lowest class values and values well above the reporting value in 2005. In 2007 an opposite shift occurs to the lower neighbouring classes and for a more extended area than in 2005. Especially the area with AOT40 levels below the exceedance thresholds increased.

<i>jei jei esis, jeu. 200</i>		Area c	of forests	, 2007		Pe	ercentage	of forest a	rea, 2007 [	%]
Country	tot. area	> CL (10 r	ng.m <sup>-3</sup> .h)	> <b>RV</b> (20	) mg.m <sup>-3</sup> .h)	< 10	10 - 20	20 - 30	30 - 50	> 50
	[km <sup>2</sup> ]	[km <sup>2</sup> ]	[%]	[km <sup>2</sup> ]	[%]	mg.m⁻³.h	mg.m⁻³.h	mg.m⁻³.h	mg.m⁻³.h	mg.m⁻³.h
Albania	7818	7818	100	7818	100	0	0	0	7.1	92.9
Austria	37608	37608	100	37608	100	0	0	7.6	90.0	2.4
Belgium	6104	5761	94.4	485	7.9	5.6	86.4	7.9	0	0
Bosnia-Herzegovina	22962	22962	100	22962	100	0	0	0	41.6	58.4
Bulgaria	34844	34844	100	34844	100	0	0	0	66.2	33.8
Croatia	20198	20198	100	20198	100	0	0	0	67.8	32.2
Cyprus	1552	1552	100	1552	100	0	0	0	1.0	99.0
Czech Republic	25484	25484	100	25484	100	0	0	19.6	80.4	0.0
Denmark	3694	3322	89.9	35	0.9	10.1	89.0	0.9	0	0
Estonia	20778	851	4.1	0	0	95.9	4.1	0	0	0
Finland	193325	44	0.0	0	0	100.0	0.0	0	0	0
France	144854	142413	98.3	73790	50.9	1.7	47.4	24.6	22.0	4.3
Germany	103821	103637	99.8	79888	76.9	0.2	22.9	58.5	18.4	0.0
Greece	23559	23559	100	23559	100	0	0	0	32.8	67.2
Hungary	17350	17350	100	17350	100	0	0	0	94.6	5.4
Iceland	314	0	0	0	0	100	0	0	0	0
Ireland	2913	1	0.0	0	0	100.0	0.0	0	0	0
Italy	78800	/8800	100	78800	100	0	0	1.6	42.8	55.6
Latvia	26960	1115	4.1	0	0	95.9	4.1	0	0	0
	63	63 11007	100	63	100	0		32.7	67.3	0
	18664	11687	62.6	0	0	37.4	62.6 05.0	0	0	0
Luxembourg	903	903	100	202	04.8 100	0	35.2	04.8	0	005
Malta	0030	0030	100	0030	100	0	0	0	0.5	99.5
Monaco	2 1	2 1	100	ے 1	100	0	0	0	100	0
Montenegro	5787	5787	100	5787	100	0	0	0	100	88.7
Netherlands	3100	1022	33.0	0	0	67 0	33.0	0	0	00.7
Norway	106324	31355	29.5	209	02	70.5	20.3	02	0	0
Poland	91851	91851	100	60007	65.3	0	34.7	42.4	22.9	00
Portugal	24320	24281	99.8	22165	91.1	0.2	87	79.9	11.3	0
Romania	69788	69788	100	69788	100	0	0	0.7	81.5	17.8
San Marino	6	6	100	6	100	0	0	0	32.0	68.0
Serbia	26687	26687	100	26687	100	0	0	0	34.8	65.2
Slovakia	19300	19300	100	19300	100	0	0	0	99.5	0.5
Slovenia	11475	11475	100	11475	100	0	0	0	69.7	30.3
Spain	91884	91881	100.0	86670	94 3	0.0	57	30.0	61.3	21
Sweden	249929	22907	9.2	27	0.0	90.8	9.2	0.0	0	0
United Kinadom	19660	148	0.8	0	0.0	99.2	0.8	0.0	0	0
Total	1521312	945090	62.1	735775	48.4	37.9	13.8	12.8	25.3	10.3
			•=			0.10			20.0	
France N of 45N	89507	87398	97.6	32978	36.8	24	60.8	27.8	85	0.5
	55247	55015	00.4	40912	72.7	2.4	25.7	10.4	12.0	10.4
FIGHCE S OF 45IN	55547	55015	99.4	40012	13.1	0.0	23.1	19.4	45.9	10.4
Northern	619674	71280	11.5	270	0.0					
North-western	122502	95232	77.7	34048	27.8					
Central & eastern	400108	399924	100.0	344331	86.1					
Southorn	370029	378654	00.0	357125	0/ 2					
	3/9028	3/0034	99.9	30/120	94.2					
Iotal	1521312	945090	62.1	735775	48.4					

Table 5.9 Forest area exposure and exceedance (critical level, CL, and reporting value, RV) for ozone, AOT40 for forests, year 2007.

Note: Countries not included due to lack of land cover data: Andorra, Switzerland, Turkey.

### 5.3.3 Uncertainties

### Uncertainty estimated by cross-validation

The absolute mean uncertainty of the map is given by the RMSE of cross-validation in  $\mu$ g.m<sup>-3</sup>.h. In Table 5.7 the absolute mean uncertainty is 5876  $\mu$ g.m<sup>-3</sup>.h for the AOT40 for crops and 10190  $\mu$ g.m<sup>-3</sup>.h for the AOT40 for forests. It indicates that the year 2007 has lower absolute mean uncertainties for the crops and forests than 2006 (7674 and 11990  $\mu$ g.m<sup>-3</sup>.h) and 2005 (7700 and 12500  $\mu$ g.m<sup>-3</sup>.h).

The relative mean uncertainty of the 2007 map of ozone indicator AOT40 for crops is about 40% and of the map of AOT40 for forests about 37%. These uncertainty values are somewhat higher than the uncertainties of the 2006 maps (30% for crops; 34% for forests), but slightly lower than for 2005 (41% for crops; 42% for forests).

Figure 5.9 shows the cross-validation scatter plots of the AOT40 for both crops and forests. The  $R^2$  indicates that for AOT40 for crops about 64% and for AOT40 for forests about 66% of the variability is attributable to the interpolation. The corresponding values for the 2005 maps (63% and 66% in (Horálek et al. 2008, Tables A3.5 and A3.6) and 2006 (53% and 49% in De Smet et al. 2009, Table 5.10), showing a higher level of performance for the interpolations in 2007, compared to 2006 and 2005.

The cross-validation scatter plots show again that in areas with higher accumulated ozone concentrations interpolation methods tend to seriously underestimate the predicted value. For example, in agricultural areas (Figure 5.9, left panel) an observed value of 30 000  $\mu$ g.m<sup>-3</sup>.h is estimated in the interpolation as about 25 000  $\mu$ g.m<sup>-3</sup>.h, i.e. an underestimation of about 16%. One can reduce this underestimation by extending the number of measurement stations and optimise the spatial distribution of those stations, specifically in areas with high values.



Figure 5.9 Correlation between cross-validation predicted values (y-axis) and measurements (x-axis) for the ozone indicators AOT40 for crops (left) and AOT40 for forests (right) for rural areas in 2007.

### Comparison of point measurement values with the predicted grid value

Additional to the cross-validation, a simple comparison has been made between the point measurement and interpolated values averaged in a 2 x 2 km grid. The results of the cross-validation compared to the gridded validation are summarised in Table 5.10. The table indicates at both receptors a better correlation between the station measurements and the averaged interpolation values of the corresponding grid cells (case ii) than it does at the point cross-validation predictions (case i) of Figure 5.9. Case ii) represents the uncertainty in the predicted gridded interpolation map. Whereas the cross-validation of case i) simulates the behaviour of the interpolation at point positions without actual measurements within the area covered by measurements. The uncertainty at measurement locations is

caused partly by the smoothing effect of interpolation and partly by the spatial averaging of the values in the 2x2 km grid cells. The level of the smoothing effect leading to underestimation at high values is there smaller than it is at the areas without measurement. For example, at agricultural areas the predicted interpolation grid value will be about 26 500  $\mu$ g.m<sup>-3</sup>.h at the corresponding station point with the observed value of 30 000  $\mu$ g.m<sup>-3</sup>.h, i.e. an underestimation of about 12%.

Table 5.10 Linear regression equation and coefficient of determination  $R^2$  from the scatter plots of (i) the predicted point values based on cross-validation and (ii) aggregation into 2x2 km grid cells versus the measured point values for  $PM_{10}$  indicator annual average for rural (left) and urban (right) areas of 2007.

	AOT40 for cr	ops	AOT40 for forests			
	equation	R <sup>2</sup>	equation	R <sup>2</sup>		
i) cross-validation prediction	y = 0.677x + 4777	0.634	y = 0.684x + 8647	0.664		
ii) 2x2 km grid prediction	y = 0.765x + 3538	0.795	y = 0.801x + 5554	0.864		

The AOT40 for crops with a target value of is 18 000  $\mu$ g.m<sup>-3</sup>.h would allow us to prepare a probability of exceedance map. However, we limited the preparations to the human health related indicators, thus not involving the accumulative ozone indicators used in the EEA CSI005, not demanding such probability of exceedance maps.

# 6 Conclusions

### Mapping and exposure results

This paper presents the interpolated maps for 2007 on the  $PM_{10}$  and ozone human health related air pollution indicators, together with their frequency distribution of the estimated population exposures and exceedances. It concerns the annual average and the 36<sup>th</sup> maximum daily mean for  $PM_{10}$  and the SOMO35 for ozone. Additional for ozone the interpolated maps on the ecosystem based indicators AOT40 for crops and AOT40 for forests are presented, including their frequency distribution of estimated land area exposures and exceedances. A similar mapping approach, primarily based on station observational data, has been used as in previous years (De Smet et al. (2009) and references cited therein).

### Human health PM10 indicators

Table 6.1 summarises for both *human health*  $PM_{10}$  *indicators* the average concentration the European inhabitant is exposed to, i.e. the population-weighted concentration, and the number of Europeans exposed to PM<sub>10</sub> concentrations above their limit values (LV) for the years 2005 to 2007.

Table 6.1 Percentage of the total European population exposed to  $PM_{10}$  concentrations above the limit values (LV) and the population-weighted concentration for the human health  $PM_{10}$  indicators annual average and 36th maximum daily average for 2005 to 2007.

PM <sub>10</sub>		2005	2006	2007
Annual average				
Population-weighted concentration	(µg.m⁻³)	26	27	25
Population exposed > LV (40 $\mu$ g.m <sup>-3</sup> )	(% of total)	9	8	6
36 <sup>th</sup> max. daily average				
Population-weighted concentration	(µg.m⁻³)	44	45	42
Population exposed > LV (50 µg.m <sup>-3</sup> )	(% of total)	28	28	22

The population exposed to annual mean concentrations of  $PM_{10}$  above the limit value of 40 µg.m<sup>-3</sup> is at least 6% of the total population in 2007. Furthermore, it is estimated that the European inhabitants living in the background (neither hot-spot nor industrial) areas – without regard whether urban or rural – are exposed on average to the annual mean  $PM_{10}$  concentration of 25 µg.m<sup>-3</sup>. In comparison with the previous two years, the number of people living in the areas above the LV, tends to reduce slightly. This trend is unlikely to be significant when taking into account the natural, meteorologically induced variations and the uncertainties involved in the interpolation. Longer time series and reduced uncertainties will be needed to make any conclusions on a possible trend.

In 2007 at least 22% of the European population lived in areas where the  $PM_{10}$  limit value of 50 µg.m<sup>-3</sup> for the 36<sup>th</sup> maximum daily mean is exceeded, being 6% lower than at its previous two years. The overall European population-weighted concentration of the 36<sup>th</sup> maximum daily mean for the background areas is estimated on about 42 µg.m<sup>-3</sup>. Compared to its previous two years one cannot conclude on some tendency, except that in 2007 the highest daily averages had lower concentrations probably leading to a population exposed to slightly lower concentrations.

Comparing the observed exceedances in 2007 for both PM10 indicators, one can conclude that the daily limit value is the most stringent. Subsequently, to comply to EU ambient air pollution legislation on PM10, countries giving preference to measures reducing PM10 concentrations in ambient air below the limit value for the 36th maximum daily mean, will most likely comply in addition to the annual limit value.

### Human health ozone indicators

Table 6.2 summarises for both *human health ozone indicators* the average concentration the European inhabitant is exposed to, i.e. the population-weighted concentration. Furthermore, the number of Europeans exposed to concentrations above the limit values of the 26<sup>th</sup> highest daily maximum 8-hour mean and above a level of 6 mg.m<sup>-3</sup>.d for the SOMO35 for the years 2005 to 2007 is presented.

Table 6.2 Percentage of the total European population exposed to ozone concentrations above the target value (TV) for the 26th highest daily maximum 8-hour average and an indicative chosen threshold for SOMO35, including their population-weighted concentrations for 2005 to 2007.

Ozone		2005	2006	2007
26 <sup>th</sup> highest daily max. 8-hr average				
Population-weighted concentration	(µg.m⁻³)	113	120	112
Population exposed > TV (120 mg.m <sup>-3</sup> .h)	(% of total)	38	55	34
SOMO35				
Population-weighted concentration	(µg.m <sup>-3</sup> .d)	5047	5485	4679
Population exposed > 6 mg.m <sup>-3</sup> .d	(% of total)	34	37	33

For ozone indicator  $26^{th}$  highest daily maximum 8-hour mean is estimated that at least 34% of the population lived in 2007 in areas above the ozone target value (TV) of 120 µg.m<sup>-3</sup>. The overall European population-weighted ozone concentration in terms of the  $26^{th}$  highest daily maximum 8-hour mean in the background areas is estimated as almost  $112 \mu g.m^{-3}$ . Compared to the previous two years one could conclude that 2006 is a year with elevated ozone concentrations leading to increased exposure levels compared to 2005 and 2007.

Similar tendency is observed at the SOMO35: in 2005 and 2007 one third of the population lived in areas where a level of 6 mg.m<sup>-3</sup>.d is exceeded being slightly lower than the estimated 37% in 2006. The population weighted SOMO35 concentrations show a similar pattern. The temporal increase occurs specifically in areas of northern and north-western Europe where the lowest SOMO35 levels are found. Some limited reductions in 2007 are found at the Iberian Peninsula.

### Agricultural and forest ozone indicators

Exposure indicators describing *agricultural and forest areas exposure to accumulated ozone* concentrations above defined thresholds are summarised in Table 6.3: the target value (LV) of 18 mg.m<sup>-3</sup>.h and the long-term objective (LTO) of 6 mg.m<sup>-3</sup>.h for the AOT40 for crops; the reporting value (RV) of 20 mg.m<sup>-3</sup>.h and the critical level (CL) of 10 mg.m<sup>-3</sup>.h for the AOT40 for forests.

Table 6.3 Percentages of the total European agricultural and forest area exposed to ozone concentrations above their thresholds: target value (TV) and long-term objective (LTO) for AOT40 for crops, and critical level (CL) and reporting value (RV) for AOT40 for forests for2005 to 2007.

Ozone		2005	2006	2007
AOT40 for crops				
Agricultural area % > TV (18 mg.m <sup>-3</sup> .h)	(% of total)	49	70	36
Agricultural area % > LTO (6 mg.m <sup>-3</sup> .h)	(% of total)	89	98	78
AOT40 for forests				
Forest area exposed > RV (20 mg.m <sup>-3</sup> .h)	(% of total)	59	69	48
Forest area exposed > CL (10 mg.m <sup>-3</sup> .h)	(% of total)	76	100	62

In 2007 about 36% of all agricultural land is exposed to accumulated ozone concentrations exceeding the target value and about 78% are exposed to levels in excess of the long-term objective. Compared to previous two years one could conclude that 2006 is a year with elevated ozone concentrations leading to increased exposure levels that reduced in 2007 to levels clearly below those of 2005.

For the ozone indicator AOT40 for forests the level of 20 mg.m<sup>-3</sup>.h is in 2007 exceeded in about 48% of the European forest area, which is clearly below those of previous two years. A similar course one

observes for the forest area exceeding the critical level. The elevated ozone accumulations in 2006 seem to be a one-off event.

The temporal pattern of the AOT40 for forests exceedances shows large similarity with those of the AOT40 for crops despite their different definitions.

The results in this report show that in general over Europe, and most significantly over northern and north-western Europe, 2006 was characterised by higher ozone levels than in 2005 and 2007: all indicators show an increase in 2006.

### Uncertainty results

Next to the creation of European wide interpolated air pollutant maps and exposure tables, the uncertainty of the presented maps is evaluated and maps with estimated probability of threshold exceedance are derived for the human health indicators. As exactly the same method and data sources has been applied over the years 2005 to 2007 a change in uncertainty is in principle related to the data content itself, thus out of our control. Denby et al. (2009) discusses a diversity of uncertainty factors potentially involved, including their possible levels of influence. The paper recommends options to reduce uncertainties systematically of which some are investigated currently and will be published about in forthcoming ETC/ACC Technical Papers and journal articles.

The relative uncertainty of rural  $PM_{10}$  maps of 2007 is about 23% for the annual average and 24% for the 36<sup>th</sup> maximum daily mean. For the urban  $PM_{10}$  map it is about 18% for the annual average and about 20% for the 36th maximum daily mean. The uncertainties are slightly lower than those of 2006 and 2005; this is given probably by the better fit of the linear regression with supplementary data in 2007 compared to its two previous years.

The relative uncertainty of ozone maps differs in 2007 for the distinguished indicators: for 26<sup>th</sup> highest daily maximum 8-hour average it is about 8% for both rural and urban areas; for SOMO35 it is about 33% for rural and 30% for urban areas. The relative uncertainty of the map for the AOT40 for crops is 40%, for the map of AOT40 for forests it is 37%.

The scatter plots of the interpolation results versus the measurements show that at both the  $PM_{10}$  and the ozone indicators at areas with high values a systematic underestimation of the predicted values occurs, leading to a considerable underestimation of the indicator values at locations without measurements. This effect is demonstrated most prominently at the ozone indicators. The underestimation would be reduced if an improved fit of the linear regression with (other) supplementary data would be reached. For example, in the near future more and more contribution from satellite imagery data and interpretation techniques is expected. Other options are extending the number of measurement stations and/or use additional mobile stations (in campaigns) which however are both expensive. Continued efforts in aiming for a more optimised spatial distribution of (such) stations, especially at areas with high air pollution and reduction of external uncertainties would certainly contribute in reducing uncertainties in the interpolations. For further reading on this subject we refer to Denby et al. (2009) and forthcoming ETC/ACC Technical Papers.

### Probability of exceedance

Maps with the probability of exceedance to its limit or target value have been prepared for the human health indicators of  $PM_{10}$  and ozone only. These probability maps are derived from combining the indicator map and its uncertainty map following the same method for the years 2005 to 2007. The differences in the maps between years depend fully on annual fluctuation in concentration levels, supplementary data and their involved uncertainties. Explaining their systematic relation is hard, since their direct causes are still a matter of study (Denby et al. 2009).

For the annual average  $PM_{10}$  the patterns in the spatial distribution of the different probability of exceedance (PoE) classes over Europe in 2007 has slightly reduced compared to 2006 and 2005.

The 36th maximum daily means of  $PM_{10}$  do show in 2007 a pattern that is relative close to that of 2005. The year 2006 had considerable increases in the PoE, leading to the conclusion that at some areas considerable systematic reductions in  $PM_{10}$  concentrations may be needed to reach non-exceedance levels in the future.

Interpreting 2007 with its previous years one can conclude that in 2006 the probability of exceedance (PoE) for the ozone increased in most parts of Europe. The central-southern and south-eastern countries in Europe show at many areas unchanged high PoE (red), where reaching compliance to non-exceedance may need considerable efforts. The Iberian Peninsula shows considerable large areas attaining in 2007 a considerable likelihood of non-exceedance. Forthcoming years may have to confirm whether one can speak of a significant positive trend.

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