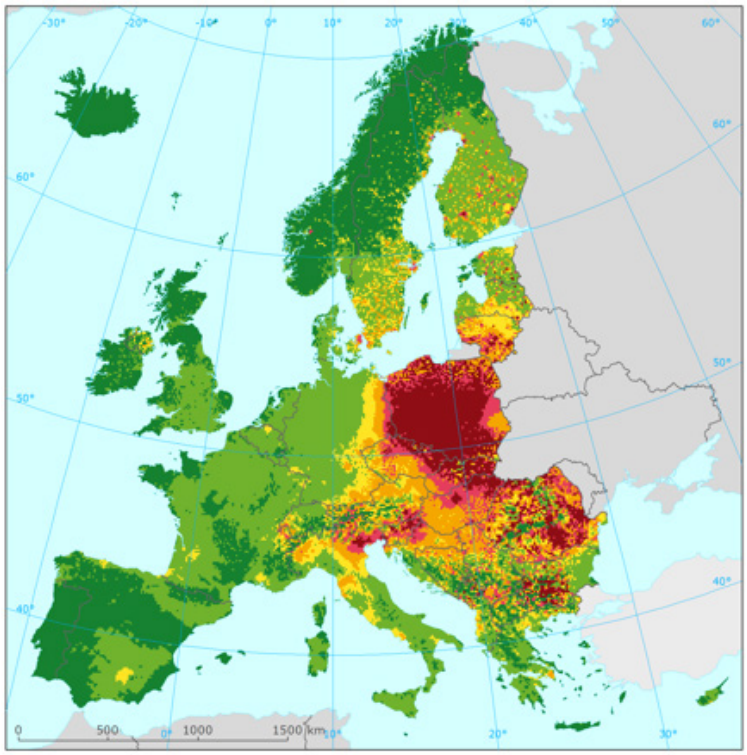
Mapping ambient concentrations of benzo(a)pyrene in Europe

Population exposure and health effects for 2012



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Cristina Guerreiro, Jan Horálek,
Frank de Leeuw, and Florian Couvidat

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Front page picture:
Estimated population-weighted concentration of benzo(a)pyrene in Europe in 2012

Author affiliation:

Cristina Guerreiro, Norwegian Institute of Air Research (NILU), Kjeller, Norway
Jan Horálek, Czech Hydrometeorological Institute (CHMI), Prague, Czech Republic
Frank de Leeuw, National Institute for Public Health and the Environment (RIVM), Bilthoven, The Netherlands
Florian Couvidat, National Institute for Industrial Environment and Risk (INERIS), France

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ETC/ACM Technical Paper 2014/6
European Topic Centre on Air Pollution and Climate Change Mitigation
PO Box 1
3720 BA Bilthoven
The Netherlands
Phone +31 30 2748562
Fax +31 30 2744433
Email etcacm@rivm.nl
Website http://acm.eionet.europa.eu/

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1 Executive summary

This study estimated current (2012) BaP concentration levels, population exposure and potential health impacts, and analysed the main emission sources of BaP. Current BaP concentration levels across Europe were estimated using the ETC/ACM mapping methodology, which combines monitoring data with modelling data and other supplementary data (e.g. altitude and meteorology). The estimated concentration maps have been compared to the EU target value. Furthermore, the European population exposure to BaP background ambient air concentrations and subsequent health impact were estimated.

Ambient air concentrations of BaP are high in large areas of Europe, mostly due to emissions from the domestic combustion of coal and wood. About one third of the BaP measurement stations in Europe are in exceedance of the target value (1 ng/m^3 annual average, to be met by 2013), mostly in urban and suburban areas[1]. Exceedances are most predominant in Central and Eastern Europe, but occur also in other European regions. About 20 % of the European population was exposed to BaP annual mean concentrations above the target value and only about 12 % of the European population live in areas with concentrations under the estimated reference level of 0.12 ng.m^{-3}. This reference value is the concentration for which a lifetime exposure corresponds to a risk of 1.10-5.

In the health impact assessment, the BaP effects have been estimated for lung cancer incidence using the exposure–response function recommended by WHO. The estimated number of lung cancer incidences in Europe due to BaP exposure was 550 (95% CI: 180-940) and 600 (95% CI: 200-1030), depending on the model selected in the interpolation. However, a sensitivity calculation showed that these results might be systematically underestimated; accounting for this bias the estimates are 630-700 new incidences. The largest health impacts can be found in the Eastern European countries, with about 50% of the incidences occurring in Poland and Romania.

Health impacts of exposure to PAHs in Europe are likely larger than this study's estimated lung cancer incidence due to exposure to BaP, for the following reasons:

- PAHs have several health impacts. In addition to lung cancer, there are other health impacts such as increased incidence of skin and bladder cancer, genotoxicity and mutagenicity. Exposure to PAHs affects children's' cognitive development and it is also linked to cardiovascular morbidity and mortality[2].
- BaP is a marker for total exposure to carcinogenic PAHs, but it only contributes to part of the total carcinogenic potential of PAHs;
- Only intake via inhalation is considered in this study. Airborne PAHs are deposited on soil and water and may be bioaccumulated in the food chain. Humans are therefore also exposed to airborn PAHs through consumption of food and water;
- Concentrations and exposure to BaPs is somewhat underestimated in this study, mainly due to the lack of measurement data.

[1] 36 % of all urban and sub-urban stations registered exceedance, while 14 % of all the rural stations were in exceedance of the target value in 2012.

[2] Although at present the effects of PAH exposure cannot be easily separated from those of particles.

2 Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are a class of complex organic chemicals of increasing concern for their occurrence in the environment and effects. The term polycyclic aromatic hydrocarbons (PAHs) commonly refers to a large class of organic compounds that contain mainly carbon and hydrogen and are comprised of two or more fused aromatic rings (also called benzene rings). The smaller PAHs with 2 to 4 rings are volatile and are found in the gas phase to a higher degree than the 5- to 7-ring PAHs, which occur mainly or entirely as particles (PM2.5 and PM10) (Possanzini et al., 2004). PAHs with low vapour pressures emitted from combustion or other high temperature sources (e.g. benzo(a)pyrene - BaP) are typically associated with particulate matter of small size (<1μm) (Sheu et al., 1997).

PAHs associated with particulate matter can be transported over large distances before being removed by wet deposition, dry depositions and chemical transformation. Dry deposition of benzo(a)pyrene (BaP) adsorbed to atmospheric aerosols accounts for most of the removal;. wet deposition is less significant by a factor of 3-5 (ATSDR, 1995). The predominant modes of chemical transformation of particulate PAH such as BaP are the reactions between PAH adsorbed on the particle surfaces and oxidants like OH, NO_2, O_3, NO_3 and SO_3 and photo-oxidation of PAH irradiated under solar radiation, which produces a variety of oxidized derivatives such as quinones, ketones, or acids (Kamens et al, 1990). Furthermore, the reaction of PAH with atmospheric oxidants may form Nitrated PAHs (or Nitro-PAHs), which are an important category of derivations of PAHs. They are of special interest because they are direct-acting mutagens and carcinogens and are, thus, considered to have far greater toxicity than unsubstituted PAHs (Byeong-Kyu Lee, 2010; Atkinson and Arey, 1994). Alebic-Juretic et al. (1990) found degradation of PAH on particle surfaces by ozone to be an important pathway for their removal from the atmosphere.

PAHs are considered among the most dangerous air pollutants due to their carcinogenic and mutagenic character. They possess high carcinogenic potential to animals and humans and are bio-accumulated in the food chain. Furthermore, they can be transported over long distances in the atmosphere resulting in a widespread distribution on the continental scale. Under certain atmospheric conditions and due to their low vapour pressure and large molecular weight, PAHs are believed to contribute to the fine particulate matter toxic potential (Dejmek et al., 2000; Binkova and Sram, 2004; Ohura et al., 2004; Hertz-Picciotto et al., 2007; Rubes et al., 2007; Soucy et al., 2007; Sram et al., 2011; 2013).

Due to their toxic and ecotoxic characteristics PAHs pose a threat to humans and the environment, and the international community has therefore implemented policies to reduce their emissions. The Protocol to the UN-ECE Convention on Long-range Transboundary Air Pollution (CLRTAP) on persistent organic pollutants (POPs[3]) (UNECE, 1998) obliges the parties to report PAH emissions[4] and has as objective to control, reduce or eliminate discharges, emissions and losses of POPs, including PAHs.

[3] "Persistent organic pollutants" (POPs) are organic substances that: (i) possess toxic characteristics; (ii) are persistent; (iii) bioaccumulate; (iv) are prone to long-range transboundary atmospheric transport and deposition; and (v) are likely to cause significant adverse human health or environmental effects near to and distant from their sources.

[4] Annex III of the Protocol specified four indicator PAHs for emission inventories: benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene.

BaP has five aromatic rings and is the most widely investigated PAH as a marker for the carcinogenic risk of PAHs in ambient air. Around 90% of BaP in ambient air is adsorbed onto aerosols and around 10% is in the gas phase. It has been classified as carcinogenic to humans (IARC, 2012). The European directive (EU, 2004) sets a target value for ambient air concentration of BaP, as a marker for the carcinogenic risk of PAHs in ambient air, in order to avoid, prevent or reduce harmful effects of PAHs on human health and the environment as a whole. The target value for BaP (measured in PM10) was set to 1 $ng.m^{-3}$ as an annual mean, to be met by 2013.

Environmental decisions must frequently be made without sufficient data on exposure. In particular, data for toxic air pollutant exposures like PAHs are usually scarcer in geographic and temporal coverage than data for other regulated pollutants such as nitrogen dioxide (NO_2), ozone (O_3) and particulate matter (PM). The present work attempts to improve the exposure estimate to ambient air BaP concentrations by combining the available measurement data to modelling results from two different chemical transport models.

The current work estimates the European population exposure to BaP concentrations both in urban and rural areas by combining measurements and modelled data for 2012. On that basis, it estimates the health impacts of BaP in Europe.

The information in this report is expected to support European, national and local environmental authorities in their evaluation of BaP pollution levels and their impacts on health, in order to develop adequate air pollution and climate policies.

3 Sources

Depending on their formation mechanism, PAHs may be classified into pyrogenic (from incomplete combustion or pyrolysis of organic material at elevated temperatures), petrogenic (from the transformation of biogenic organic materials such as fossil fuels at moderate temperatures), diagenic (from the transformation of organic material in soils and sediments), and biogenic (synthesized by organisms) (Neff, 2002). The first two kinds are clearly predominant in the environment, with the most important sources being anthropogenic from incomplete combustion of organic fuels (residual oil, wood, coal, gasoline and diesel). Natural sources of PAHs, with much less incidence than anthropogenic ones, are forest fires and volcanic eruptions (Ravindra et al., 2008).

Under the LRTAP Convention, parties are invited to report emissions data for PAHs and BaP, among other pollutants. Twenty EU countries[5] reported officially their BaP emissions for 2012 to EEA under the LRTAP convention[6]. For modelling purposes, EMEP estimates BaP emissions for their European modelling domain (see Figure 3.3) based on the officially reported emissions to the extent possible and doing some corrections and/or gap-filling (EMEP/CEIP, 2014).

The total BaP emission of these 20 countries that reported BaP emissions in the EU28 was 180 tonnes in 2012 (EEA, 2014a). On the other hand, the total estimated BaP emissions by EMEP for modelling purposes for EU28 in 2012 was 241 tonnes[7] (EMEP/CEIP, 2014). This considerable difference corresponds to the gap filling estimation done by EMEP in order to obtain a more complete emission inventory for modelling purposes. Table 3.1 shows the differences per country between the officially reported emissions of BaP for 2012 and the estimate for modelling purposes.

Emission data for modelling is based on a combination of official reported data supplemented with expert estimates for missing data and/or for data of low quality. Not reported emissions are gap-filled (by EMEP's interpolation routine) in order to create complete sectorial gridded emissions for the whole of the EMEP area so that EMEP is capable of performing dispersion modelling.

[5] The 20 countries that reported BaP emissions for 2012 and included in this total are: Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, France, Germany, Hungary, Ireland, Latvia, Lithuania, Malta, the Netherlands, Poland, Romania, Slovakia, Slovenia, Sweden, and the United Kingdom.

[6] It is important to note that there is at present no legislation requiring reporting of BaP emissions by the Member States.

[7] Meaning that a total emission of 60 tonnes was estimated for 2012 for the remaining 8 countries that did not report: Austria, Belgium, Finland, Greece, Italy, Luxembourg, Portugal and Spain. These 8 countries are estimated to account for 25 % of the total EU28 emissions of BaP in 2012.

Table 3.1: BaP emissions in 2012 in the EU28 countries: officially reported and estimated for modelling, unit: tonnes/year.

	Emissions (T/yr)			Emissions (T/yr)	
countries	for models	officially reported	countries	for models	officially reported
AT	2,26	-	HU	9,23	9,19
BE	12,06	-	IE	1,23	1,23
BG	8,95	8,95	IT	18,97	-
CY	0,29	0,29	LT	3,90	3,90
CZ	5,55	5,55	LU	0,26	-
DE	31,60	31,60	LV	4,23	4,23
DK	2,46	2,46	MT	0,02	0,02
EE	4,48	4,48	NL	1,20	1,20
ES	13,57	-	PL	43,51	43,51
FI	5,15	-	PT	0,02	-
FR	5,04	5,04	RO	37,76	37,76
GB	3,51	3,51	SE	4,40	4,40
GR	8,27	-	SI	3,83	3,83
HR	3,41	3,41	SK	5,37	5,37
Total EU28				**240,52**	**179,92**

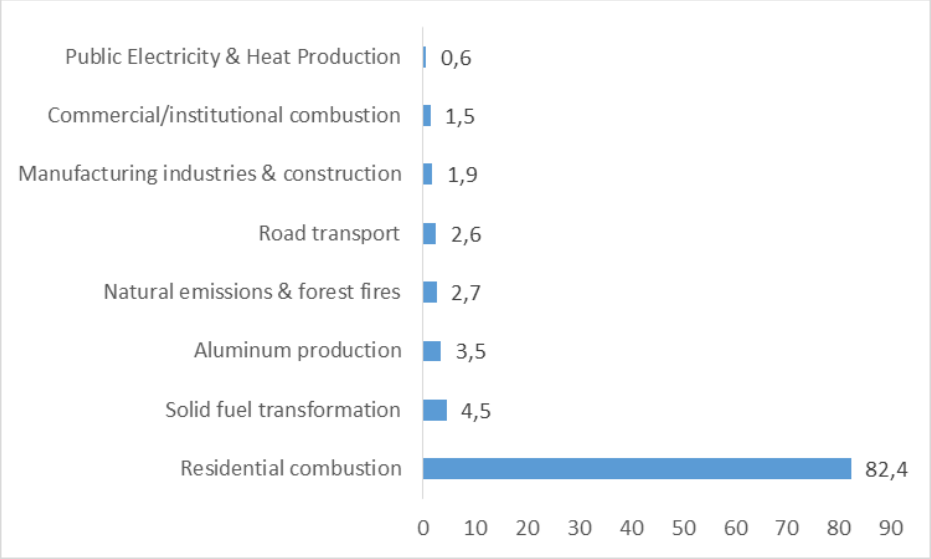


Figure 3.1: Officially reported emissions of BaP in 2012 (20 countries of EU28) by the main sectors and in % of total (source: EEA, 2014a).

Figure 3.1 shows the contribution (in %) of the main emission sectors to BaP emissions in the 20 countries of the EU28 that reported emissions in 2012. Residential combustion is by far the most important sector, contributing to 82 % of the total BaP emissions, mainly from wood- and coal-burning. Other sources are solid fuels transformation, aluminium production, natural emissions, and road traffic.

The development in the officially reported emissions of BaP from 2003 to 2012 is shown in Figure 3.2 for the same 20 countries and for Luxembourg, which reported emissions in a few years. The residential combustion sector has been the dominating emission sector over the period and the BaP

reported emissions for this sector have increased by 25 % between 2003 and 2012. This increase may be partially due to an increase in the use of solid fuels (e.g. wood, other bio-fuels, and coal) for domestic heating, due to either government incentives to increase the use of renewable fuels (in the case of wood and other bio-fuels), or to increasing costs of other energy sources and in response to economic hardship.

As further explained in Annex 1, no firm conclusions can be given on the temporal changes in total BaP emissions nor for the emission in the residential combustion sector in the last 10 years. Fig 2.2 on the official reported data suggest a 25% increase, however in the same period the EMEP-gapfilled emission data decreased with about 12%. Another reason for the increase in these reported emissions may be that in recent years emission inventories are more complete than in earlier years. In short, these changes in reported emissions are highly uncertain.

On the other hand, in the previous decade, i.e. from 1990 to 2000, it is clear that emissions decreased in several European countries. Reductions were especially great in Germany, the UK, and Hungary. In total the reporting countries reduced their emissions by 62% from 1990 to 2000 (EEA, 2014c).

EMEP's analysis on emission and concentration trends in BaP goes back to 1990. EMEP (2013) concludes that the reduction of PAH pollution levels was more significant during the 1990s. In particular, BaP air concentrations in the EU countries dropped by 38% from 1990 to 2000. However, after 2000 the decreasing trend was almost levelled off and during the recent years BaP air concentrations in more than a half of the EU countries tended to increase following the growth of their emissions (EMEP, 2013). Furthermore, EMEP (2013) concludes that for about 70% of the EMEP countries the contribution of transboundary transport to deposition of BaP over their territory exceeds the contribution of national emission sources.

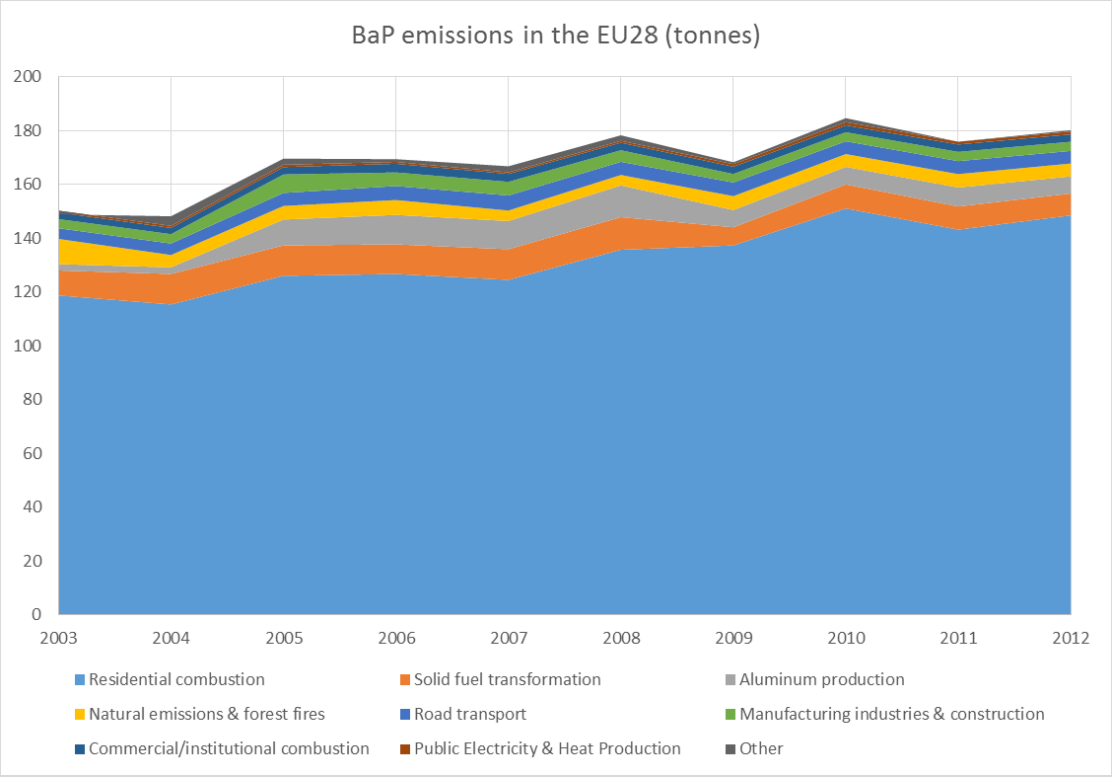


Figure 3.2: Development of BaP emissions in 21 countries in the EU28 from 2003 to 2012 in total and for the main emission sectors (source: EEA, 2014a).

The emission data used for dispersion modelling was the estimated emissions of BaP for use in models (EMEP/CEIP, 2014), for both EMEP and CHIMERE models. Considering this data, the main countries contributing to the total EU28 emissions in 2012 were Poland (18 %), Romania (16 %), Germany (13 %), Italy (8 %), Spain (6 %) and Belgium (5 %). In total, these 6 countries account for 66 % of the estimated total EU28 BaP emission in 2012. Figure 3.3 shows the EMEP emissions distribution over Europe, for use in models.

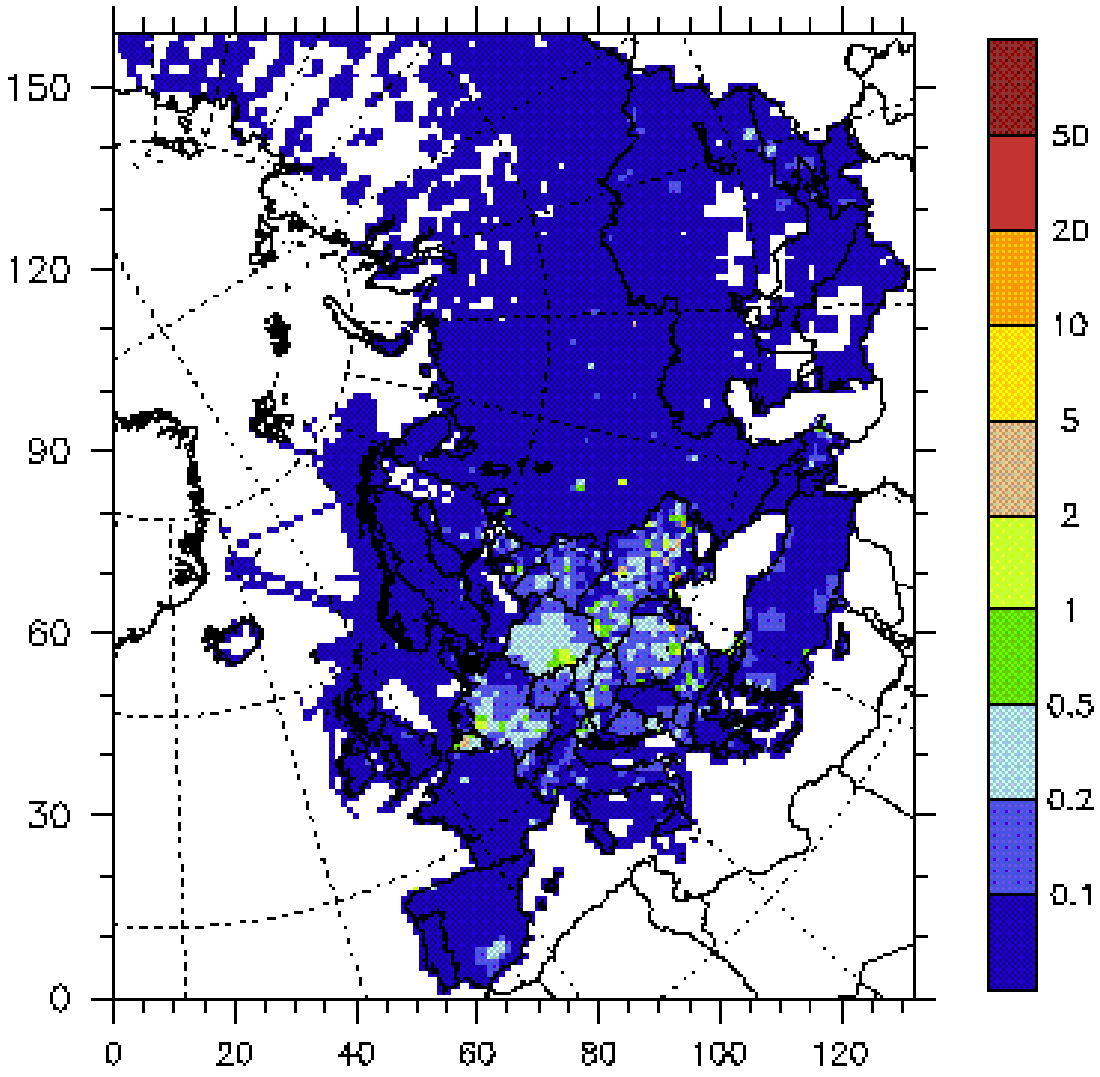


Figure 3.3: EMEP emissions distribution over Europe in 2012, for use in models (EMEP/CEIP, 2014). Unit: Mg/year.

4 General background on health effects of PAHs

The carcinogenicity of PAH in humans seems to be beyond dispute. Associations between soot, coal tar and pitch with human cancer have been reported since the end of 18th century (Dipple, 1985). In a series of monographs (see, for example, IARC, 1987), the International Agency for Research on Cancer has evaluated a number of PAH-containing materials or mixtures and occupational situations in which exposure to PAH occurs. Many of the studied materials/exposure were classified as group 1 (carcinogenic to man) or Group 2A, 2B (probably/possibly carcinogenic). In the latest review of human carcinogens, IARC (2012) concluded: "The strong and extensive experimental evidence for the carcinogenicity of BaP in many animal species, supported by the consistent and coherent mechanistic evidence from experimental and human studies provide biological plausibility to support the overall classification of BaP as a human carcinogen (Group 1)." Moreover, ambient air pollution as a whole, as well as PM as a separate component of air pollution mixture, have been classified recently as carcinogenic (IARC, 2013).

In ambient air, a number of individual PAH can be found (see, for example, Brown et al, 2013; Garrido et al., 2014). The carcinogenic potency varies widely over the PAH. Although BaP is not the most abundant pollutant, its carcinogenic potency is amongst the highest (Boström et al., 2002; Collins et al., 1998; Amarillo et al., 2014). BaP alone will underestimate the carcinogenic potential of ambient PAH mixtures since co-occurring substances are also carcinogenic. Given that the relative contributions of more potent PAHs, such as dibenzo[a, l]pyrene (Pufelete et al., 2004), in ambient air have not been adequately evaluated and there are only limited data on their presence and formation in ambient air, it is possible that their relative contribution to the carcinogenic activity of a total PAH mixture is far greater than that of BaP, and suggests further work is required to investigate the potential role of high potency PAHs in air pollution related lung cancer (Okona-Mensah et al., 2005). Nevertheless, in view of its carcinogenic potency and its abundance in the PAH mixture, BaP is widely used as an indicator.

The air quality directive (EU, 2004) sets a target value (TV) for annual mean BaP concentrations in ambient air equal to 1 ng.m^{-3}. This TV is very high compared to an estimated reference level of 0.12 ng.m^{-3}, assuming WHO unit risk for lung cancer, and an acceptable risk of additional lifetime cancer risk of approximately 1 x 10^{-5} (de Leeuw and Ruyssenaars, 2011). WHO (2013) proposes that the acceptability of the level of risk associated with the current EU target value (1 x 10^{-4}) should be reviewed and discussed.

PAHs can be absorbed through the respiratory tract, both through inhalation of tobacco smoke and ambient air, gastrointestinal tract (diet is the main route of exposure to PAHs in the general population), and skin. Most studies to date have not considered all routes of exposure (IARC, 2010; Reid et al., 2005). After absorption into the human body, PAHs may be altered into substances that can damage the genetic material of cells and initiate the development of cancer, although individual PAHs have different capacities to damage cells tissues.

Long-term exposure to PAH has been associated, in addition to lung cancer, to other health endpoints such as increased incidence of skin and bladder cancer. PAH are genotoxic (Topinka et al., 2011); prenatal exposure to PAH affects children's' cognitive development at the age of 5 with potential implications for school performance (Edwards et al., 2010). Exposure to PAH in early-life (age 3-5) might play a role in ADHD behaviour problems (Perera et al., 2014). The high concentrations found in occupational situations might lead to fatal ischemic heart diseases (Burstyn et al., 2005). WHO (2013) has found new evidence linking PAH exposure to cardiovascular morbidity and mortality, although at present the effects of PAH exposure cannot be easily separated from those of particles.

5 BaP concentration map for Europe

In order to estimate the population exposure to BaP and its associated health effects in Europe, it is necessary to have the best possible knowledge on the status of the BaP concentrations across Europe. Since the geographical coverage of BaP measurements is insufficient for preparing a European concentration map, the chosen approach is to combine the existent measurement data with modelling results.

5.1 Methodology

The methodology used for the creation of the BaP concentration map over Europe is in principle the same as documented in Horálek et al. (2014a) and previously used to create concentration maps for PM10, PM2.5, O_3, NO_2 .

The mapping method used is a linear regression model followed by kriging of the residuals produced from that model (residual kriging), as explained further in Section 5.1.1. In the linear regression model, the measured data are taken as a dependent variable, while the output concentration data from a chemical transport model and other supplementary data (e.g. altitude, meteorology, population density) are used as independent variables.

The maps are constructed for the rural and urban background areas separately, which are later merged into one combined air quality concentration map using population density information. This map is used for the population exposure estimates. For presentational purposes of European map, a spatial aggregation to 10x10 km^2 resolution is sufficient and as such applied in this paper. More details are given in Section 5.1.2.

5.1.1 Interpolation

The interpolation method used is a linear regression model followed by interpolation (kriging) of the residuals produced from that model (residual kriging). Interpolation is carried out according to the relation:

$$\hat{Z}(s_0) = c + a_1.X_1(s_0) + a_2.X_2(s_0) + ... + a_n.X_n(s_0) + \eta(s_0) \qquad (4.1)$$

Where:

$\hat{Z}(s_0)$	is the estimated value of the air pollutant concentration at the point s_o,
$X_1(s_0), X_2(s_0), ..., X_n(s_0)$	are the individual supplementary variables at the point s_o,
$c, a_1, a_2, ..., a_n$	are the parameters of the linear regression model calculated based on the data at the points of measurement,
$\eta(s_0)$	is the spatial interpolation of the residuals of the linear regression model at the point s_o calculated based on the residuals at the points of measurement; the interpolation method used is ordinary kriging.

Generally, for different air pollutants and area types (rural, urban), different supplementary data are used. For some pollutants (e.g. PM10, PM2.5), prior to linear regression and interpolation, a logarithmic transformation of measurement and model data is used. After the interpolation, a back-transformation is applied. For details, see Horálek et al. (2007) and De Smet et al. (2011).

In this study, the supplementary data for use in the mapping has been explored and selected separately for the rural and the urban background areas. In addition, the use of interpolation with or without the use of the logarithmical transformation has been tested.

5.1.2 Merging of rural and urban background maps

The BaP annual mean concentration map is constructed for the rural and urban areas separately on a grid of 10x10 km^2 resolution. The rural map is based on rural background stations and the urban map on urban and suburban background stations. The reason for the separate mapping of rural and urban areas is the assumption that BaP concentrations in general are higher in urban than in the rural areas. However, in some areas the assumption is not valid. Thus, in the areas where the estimated urban map value is lower than the estimated rural map value, both the rural and the urban background maps are adapted with the use of the joint rural/urban map. (Such map is constructed based on both rural and urban/suburban background stations.) Subsequently to this, the adapted rural and urban background maps are merged into one combined air quality concentration map using a European-wide population density grid at 1x1 km^2 resolution. For the 1x1 km^2 grid cells with a population density less than a defined value of α_1, we select the rural map value and for grid cells with a population density greater than a defined value α_2, we select the urban map value. For areas with population density within the interval (α_1, α_2) a weighting function of α_1 and α_2 is applied. For details, see Horálek et al. (2007) and De Smet et al. (2011).

Summarising, the separate rural and urban background maps are constructed at a resolution of 10x10 km; their merging however takes place on basis of the 1x1 km^2 resolution population density grid, resulting in a final combined pollutant concentration map on this 1x1 km^2 resolution grid, which is aggregated to a 10x10 km^2.

5.1.3 Calculation of population exposure

Population exposure for Europe as a whole is calculated from the BaP annual mean concentration map and population density data, both at 1x1 km^2 resolution. It is expressed by the percentage of total European population living in areas with BaP air concentrations within specific intervals, including below and above the BaP TV (1 $ng.m^{-3}$). In addition, the European-wide exposure is calculated as the population-weighted concentration, i.e. the average concentration per inhabitant. In addition to the population-weighted concentration for the whole Europe, also the population-weighted concentration for each 10x10 km^2 grid was calculated, which enables to present the population-weighted concentration map in 10x10 km^2 resolution.

The population-weighted concentration was calculated according to the equation:

$$\hat{c} = \frac{\sum_{i=1}^{N} c_i p_i}{\sum_{i=1}^{N} p_i} \qquad (4.2)$$

where $\hat{c}$ is the population-weighted average concentration in the whole Europe or in the given 10x10 km^2 grid cell,
p_i is the population in the i^{th} 1x1 km^2 grid cell,
c_i is the concentration in the i^{th} 1x1 km^2 grid cell,
N is the number of grid cells in Europe as a whole or in the given 10x10 km^2 grid cell.

5.1.4 Uncertainty analysis of concentration map

The uncertainty estimation of the European concentration map is based on the one hand on cross-validation and on the other hand on the interpolation standard error map, calculated based on kriging theory.

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 in the available set. The advantage of this cross-validation technique is that it enables evaluation of the quality of the predicted values at locations without measurements, as long as they are within the area covered by the measurements.

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

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(\hat{Z}(s_i) - Z(s_i))^2} \qquad (4.3)$$

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

where $Z(s_i)$ is the measured concentration[8] at the i^{th} point, $i = 1, ..., N$,

$\hat{Z}(s_i)$ is the estimated concentration[8] at the i^{th} point using other information, without the measured concentration at the i^{th} point,

N is the number of the measuring points.

In addition to the RMSE expressed in the absolute units, one could express this uncertainty in relative terms by relating the RMSE to the mean air pollution concentration measured at all stations:

$$RRMSE = \frac{RMSE}{\overline{Z}}.100 \qquad (4.4)$$

where $RRMSE$ is the relative RMSE, expressed in percent,

$\overline{Z}$ is the arithmetic average of the measured concentrations[8] $Z(s_1), ..., Z(s_N)$.

RMSE and RRMSE should be as small as possible, and bias should be as close to zero as possible.

The standard error map is calculated based on the spatial statistics theory, see Cressie (1993). The standard error of the combined (rural and urban background) map is calculated from the standard errors of the separate rural and urban background maps, as in De Smet et al. (2011).

[8] i.e. all measured BaP annual mean concentration(s).

5.2 Data used

5.2.1 Measurement air quality data

Monitoring data for BaP concentrations was extracted from the European monitoring database AirBase (Mol and Van Hooydonk, 2012). Only data from stations classified as the type *background* for the areas *rural*, *suburban* and *urban* are used. *Industrial* and *traffic* station types are not considered, as they represent local scale concentration levels not applicable at the mapping resolution employed. The following indicators of BaP concentrations in ambient air (with the Airbase component number, cp_number) were considered:

Benzo(a)pyrene, 2012 annual average ($ng.m^{-3}$)
- BaP in PM2.5, aerosol (cp_number = 1029)
- BaP in PM10, aerosol (cp_number = 5029)
- BaP in PM10, air+aerosol (cp_number = 5129)
- BaP, air+aerosol (cp_number = 6015)

Despite the fact that the European directive (EU, 2004) prescribes that the BaP concentration measurements should be made in the PM10 fraction, available data for all the species listed above were considered in this study. The justification for this is that the focus of this study is on the estimation of exposure and its health effects and this is independent on which size fraction the BaP concentrations were measured in. As mentioned in Chapter 1, most of the BaP is present in PM2.5 and not in the coarser fraction of PM10, and the gaseous fraction of the total BaP is quite small. This may introduce some systematic differences in the measured data, but on the other hand, the inclusion of additional measured data decreases of uncertainties in the interpolated concentration maps.

Only the stations inside the EEA map extent *Map_1c* (EEA, 2011) are used, so one station located in the French overseas departments was excluded. To reach a more extended spatial coverage AirBase data was supplemented with data from:

a) three Slovak urban background stations (SHMI, 2013), and
b) two French stations[9] (provided by INERIS).

Measurements from stations with data coverage of at least 14 percent valid daily measurements (24 hours samples) a year were used, i.e. a minimum of 51 days a year with valid data in order to maximise the use of the available measurement data, already scarce in large areas of Europe. This requirement for the data coverage is very low and allows an increase in the uncertainty of the measurement data used, compared to other pollutants. A data coverage of 14% corresponds to the minimum time coverage for indicative measurements laid down in Annex IV of Directive 2004/107/EC (EU, 2004). EC (2001) recommends a sampling frequency of one day every third day of the year, allowing an evenly distributed data sampling and a data coverage of 43%. Furthermore, it says that individual measurements can be expected to have an uncertainty of about +/- 50% at the 95% confidence level. When individual measurements are used for generating annual means, the likely expanded uncertainty arising from reducing daily to once in every 6th day sampling would be between 25 and 30% falling to ~10% when every third day is sampled (EC, 2001).

As stated above, sampling must be spread evenly over the weekdays and the year, in order to secure that the annual average calculated from the available data is representative of the real value. The available measurement data was therefore checked to see if it is randomly distributed across the year, i.e. the reported number of valid measurements has been compared with the expected number of measurements per day of the week and per month (de Leeuw, 2012). Some deviations from the regular distribution of the data were observed. Two stations with greatest deviation in the data

[9] With the station codes: FR01020 and FR24009.

sampling distribution[10] were excluded, namely the Spanish stations ES0006R and ES0007R. For the future, it is recommended to examine the issue of the data coverage and sampling distribution more in depth, in order to secure a lower uncertainty in the measured annual average.

In total, 84 rural background and 289 urban/suburban background stations were used. Figure A2.1 (Annex 2) shows the rural and urban/suburban background measurement stations used for the BaP annual mean concentration map for 2012, including their measurement concentration range.

5.2.2 Chemical transport model data

Modelled BaP annual mean concentrations for 2012 were used to produce the BaP concentration maps. Results from two different chemical transport models (CTMs) were used for comparison: the EMEP MSC-E model[11] (EMEP, 2014) and the CHIMERE model (Menut et al., 2013; http://www.lmd.polytechnique.fr/chimere). In both cases, the model concentration data is imported into ArcGIS as a point shape file, subsequently converted into a 100 x 100 m^2 resolution raster grid, and spatially aggregated into the reference EEA 10 x 10 km^2 grid.

Emissions for 2012 with spatial resolution 50 x 50 km^2 provided by EMEP/CEIP (2014) were used for both model runs. The models were driven by ECMWF meteorology for 2012.

A. EMEP MSC-E model output (original resolution circa 50 x 50 km^2)

EMEP MSC-E POP model is a three-dimensional Eulerian multi-compartment chemistry transport model (Gusev et al., 2005). The model's output covers completely the mapping domain (i.e. the area of the EEA member and cooperating countries within the map extent Map_1c, EEA, 2011). The model was run by EMEP MSC-East in its own grid specification in circa 50 x 50 km^2 resolution. EMEP (2014) provides details on the EMEP modelling, but it presents concentration fields calculated for 2011 emissions, which we did not use.

The chemical transport model EMEP MSCE-POP simulates the photodegradation of BaP adsorbed to particulate matter. It uses the calculated value of half-life obtained in Chen et al. (2001) on the basis of Quantitative Structure-Property Relationship studies (QSPRs) on the direct photolysis of BaP in atmospheric aerosol. Its numerical value agrees with the interval of experimentally determined values of BaP half-life due to photodegradation process (Gusev et al., 2006). In addition, the model takes into account the degradation of BaP in the gaseous phase due to reactions with hydroxyl radicals (OH); all other reactions are neglected (Gusev et al., 2005).

B. CHIMERE model output (original resolution 0.25°x0.25°, i.e. circa 20x30 km)

Air concentrations of BaP for 2012 were simulated with the air quality model CHIMERE[12] (Menut et al, 2013) over Europe with a resolution of 0.25°. Chimere was modified to take into account the partitioning of BaP between the gas phase and the particle phase according to its saturation vapor pressure taken from Gusev et al. (2005). The degradation of BaP in the gaseous phase due to reactions with hydroxyl radicals (OH) is also taken into account.

The modelling domain is <-14.5°W/34.5°E> <35.2°N/69.5°N>, i.e. the mapped area of the EEA member and cooperating countries, without Iceland, northern Norway and southern Cyprus. A short description of the model setting is given in Annex 4.

[10] In these two cases, the monitoring data cover only two or three consecutive months.
[11] The results were directly provided by EMEP MSC-E using 2012 emissions, the report EMEP (2014) shows preliminary results for 2012 using 2011 emissions.
[12] http://www.lmd.polytechnique.fr/chimere/

5.2.3 Other supplementary data

A. Altitude (original resolution 30x30 arc-seconds)

The altitude data field (in meters above sea level) is taken from GTOPO30[13], which is a global digital elevation model (DEM) with a horizontal grid spacing of approximately 1. For details, see Horálek et al. (2007).

B. Meteorology parameters (original resolution 0.25°x0.25°)

Wind speed (annual average, in $m.s^{-1}$), surface solar radiation (annual average of daily sum, in $MWs.m^{-2}$), temperature (annual average, in °K, consequently converted to °C), and relative humidity (annual average, in %) were used. The hourly and daily data for 2012 were extracted from the Meteorological Archival and Retrieval System (MARS) ERA-interim reanalyses of ECMWF[14]. For details, see Horálek et al. (2007).

C. Population density

Population density (in $inhabitants.km^{-2}$, census 2001) for the majority of countries is based on data provided by the European Commissions (EC) Joint Research Centre (JRC). The original resolution is 100x100 m^2. For countries and regions lacking JRC data, we use ORNL population data in the 1x1 km^2 resolution. For details, see Horálek et al. (2014a).

[13] https://lta.cr.usgs.gov/GTOPO30
[14] http://www.ecmwf.int/en/research/climate-reanalysis/browse-reanalysis-datasets

6 BaP map creation including analysis

6.1 Selection of supplementary data and kriging type

For the BaP map creation, the best variant of the mapping method had to be selected. At first, we selected the set of supplementary data, separately for the rural and background areas. Secondly, we examined the need of a logarithmical transformation of measurement and model (CTM) data (Section 5.1.1).

The first step is to execute the linear regression analysis, for the urban and the rural background stations separately. The aim is to identify useful supplementary data for their eventual use in the residual kriging. For the selection of the useful supplementary data, the backward elimination is used and confirmed by automatic stepwise regression (see Horálek et al.,2007). Both the variants with and without the logarithmical transformation are examined.

The results of the regression analysis can be seen in Table 6.1. R^2 and adjusted R^2 should be as close to one as possible, standard errors should be as small as possible. (However, the standard errors for the linear regression models with and without logarithmical transformation are not mutually comparable, due to the logarithmical transformation.) The choice made in this section is indicative for the best linear regression result. However, this choice might not necessarily provide the best residual kriging result.

Table 6.1 Statistical indicator values of the selected linear regression models indicating the correlation between observed and calculated annual mean BaP concentrations (2012), using the specified supplementary data.

linear regression model	rural areas					
	using EMEP			using CHIMERE		
	R^2	adj. R^2	std. err.	R^2	adj. R^2	std. err.
EMEP/CHIMERE	0,20	0,19	0,76	0,27	0,26	0,73
EMEP/CHIMERE, log. transformed	0,20	0,19	1,29	0,32	0,31	1,20
EMEP/CHIMERE, altitude, log. transformed	0,23	0,21	1,28	0,35	0,33	1,17
EMEP/CHI., altitude, wind speed, log. transf.	0,38	0,35	1,16	0,42	0,40	1,12
linear regression model	urban background areas					
	using EMEP			using CHIMERE		
	R^2	adj. R^2	std. err.	R^2	adj. R^2	std. err.
EMEP/CHIMERE	0,25	0,25	2,97	0,18	0,18	3,11
EMEP/CHIMERE, temperature	0,30	0,29	2,88	0,26	0,25	2,96
EMEP/CHIMERE, log. transformed	0,34	0,34	1,19	0,27	0,27	1,25
EMEP/CHIMERE, temperature, log. transf.	0,44	0,44	1,10	0,45	0,45	1,09
EMEP/CHI., temp., popul. density, log. transf.	0,51	0,50	1,04	0,53	0,52	1,02

The selected linear regression models shown in Table 6.1 and Table 6.2 are explained here under.
For rural areas:

1) **EMEP/CHIMERE** is the linear regression model using the BaP measurements in rural background stations as dependent variable and the CTM results as independent variable;
2) **EMEP/CHIMERE, log. transformed** is the linear regression model using the BaP measurements as dependent variable and the CTM results as independent variable, where both the measurements and the CTM results have been previously logarithmic transformed;
3) **EMEP/CHIMERE, altitude, log. transformed** is the same as in 2), with altitude as an independent variable, but altitude has not been logarithmic transformed;
4) **EMEP/CHI., altitude, wind speed, log. transf.** is the same as in 2), with altitude and wind speed as an independent variables which have not been logarithmic transformed.

For urban background areas:

I. **EMEP/CHIMERE** is the linear regression model using the BaP measurements in urban/suburban background stations as dependent variable and the CTM results as independent variable;
II. **EMEP/CHIMERE, temperature** is the linear regression model using the BaP measurements in urban/suburban background stations as dependent variable and both the model results and temperature as independent variables;
III. **EMEP/CHIMERE, log. transf**. is the linear regression model using the BaP measurements in urban/suburban background stations as dependent variable and the model results as independent variable, where both the measurements and the CTM results have been logarithmic transformed;
IV. **EMEP/CHIMERE, temp., log. tr.** is the same as in III, with temperature as an independent variable, but temperature has not been logarithmic transformed;
V. **EMEP/CHIMERE, temp., pop.d., log. tr**. is the same as in III, with temperature and population density as an independent variables, where temperature has not been logarithmic transformed and population density has previously been logarithmic transformed.

For the best variants, the spatial interpolation was executed and mutually compared, see Table 6.2. The comparison is performed using cross-validation, see Section 5.1.4. The main indicator is RMSE and the additional ones are bias and linear regression parameters from the cross-validation scatterplot.

RMSE should be as small as possible, bias should be as close to zero as possible, R^2 and slope should be as close to one as possible, and intercept should be as close to zero as possible. The best results are marked in Table 6.2 by dark green, the second best by light green.

Table 6.2 Comparison of different variants of spatial interpolation showing RMSE, bias, R^2 and linear regression from the cross-validation scatter plots of BaP annual mean predicted values, 2012. Unit: ng.m^{-3}, except R^2.

spatial interpolation variant	rural areas							
	using EMEP				using CHIMERE			
	RMSE	bias	R^2	regr. eq.	RMSE	bias	R^2	regr. eq.
EMEP/CHIMERE	0,70	0,03	0,33	y=0.384x+0.33	0,69	0,03	0,36	y=0.433x+0.31
EMEP/CHIMERE, log. transf.	0,67	0,13	0,44	y=0.568x+0.34	0,62	0,08	0,49	y=0.587x+0.29
EMEP/CHI., alt., wind sp., log. tr.	0,64	0,08	0,44	y=0.477x+0.33	0,60	0,06	0,51	y=0.509x+0.31
spatial interpolation variant	urban background areas							
	using EMEP				using CHIMERE			
	RMSE	bias	R^2	regr. eq.	RMSE	bias	R^2	regr. eq.
EMEP/CHIMERE	1,98	0,05	0,67	y=0.723x+0.79	2,11	0,09	0,63	y=0.708x+0.88
EMEP/CHIMERE, temperature	1,97	0,03	0,67	y=0.733x+0.73	2,11	0,07	0,63	y=0.716x+0.83
EMEP/CHIMERE, log. transf.	1,89	-0,01	0,70	y=0.734x+0.69	2,07	0,02	0,64	y=0.723x+0.77
EMEP/CHIMERE, temp., log. tr.	1,84	0,04	0,71	y=0.756x+0.69	2,02	0,08	0,66	y=0.746x+0.76
EMEP/CHI., temp., pop.d., log tr.	2,11	0,22	0,68	y=0.863x+0.58	1,97	0,15	0,70	y=0.836x+0.59

Based on the analysis, we selected:

- For the rural map: 4) **EMEP/CHIMERE, altitude, wind speed, log. tr.**, i.e. EMEP or CHIMERE model, with logarithmic transformation, altitude and wind speed.
- For the urban background map: IV) **EMEP/CHIMERE, temp., log. tr**., i.e. EMEP or CHIMERE model, with logarithmic transformation, and temperature.

In both cases, the logarithmical transformation is applied to the measurement and CTM data (see Section 5.1.1). In the analysis, EMEP and CHIMERE models give mostly quite similar results; (e.g., the same supplementary data are selected in both cases).

At the urban background areas, the best performing linear regression model is different for the EMEP and CHIMERE models. When using EMEP data, the linear regression model IV is the best performing variant, while when using CHIMERE data the linear regression model V is the best performing variant. Ultimately, the variant IV (i.e. without population density) was selected for urban background mapping both for EMEP and CHIMERE. This choice was done for two reasons: 1) for consistency (i.e. the same variant is selected for both EMEP and CHIMERE), 2) the negative dependency of the population density (i.e. increasing BaP with the decreasing population density) for which we do not have a clear explanation. This negative dependency might be caused by differences in residential heating behaviour between countries and between rural and urban areas. Temperature is used in the chosen model IV. In the multiple linear regression, with increasing temperature, BaP is decreasing. The main reason for this relation is that domestic heating is the main source of BaP emissions, as discussed in Chapter 3.The supplementary data used for the mapping in the rural areas is similar to the ones used in PM10 mapping (Horálek et al, 2014a).

6.2 Spatial interpolation

Based on the supplementary parameters and the kriging variant selected in Section 4.1, the concentration map of BaP was constructed using both EMEP and CHIMERE model outputs. As previously explained, the maps were created separately for the rural and urban background areas, and subsequently combined using population density, see Sections 5.1.1 and 5.1.2.

Table 6.3 presents the estimated parameters of the linear regression models (c, a_1, a_2,... in equation 4.1) and of the residual kriging (*nugget, sill, range*), and it includes the statistical indicators of both the regression and the kriging. The adjusted R^2 and standard error are indicators for the fit of the

regression relationship, where the adjusted R^2 should be as close to 1 as possible and the standard error should be as small as possible. RMSE and bias are the cross-validation indicators, showing the quality of the resulting map.

Table 6.3: ***Parameters of the linear regression models and of the ordinary kriging variograms (nugget, sill, range) and their statistics of BaP annual average for 2012 in rural and urban areas, using EMEP (left) and CHIMERE (right) model results.***

linear regr. model + ord. kriging of its residuals	using EMEP		using CHIMERE	
	rural areas	urban b. areas	rural areas	urban b. areas
	coeff.	coeff.	coeff.	coeff.
c (constant)	1,95	2,81	1,45	3,29
a1 (log. EMEP model 2012)	0,456	0,72		
a1 (log. CHIMERE model 2012)			0,606	0,79
a2 (altitude GTOPO)	-0,00163		-0,00141	
a3 (wind speed 2012)	-0,599		-0,439	
a4 (temperature 2012)		-0,19		-0,26
adjusted R^2	**0,35**	**0,44**	**0,40**	**0,45**
standard error [ng.m^{-3}]	**1,16**	**1,10**	**1,12**	**1,09**
nugget	0,62	0,58	0,07	0,08
sill	1,29	1,16	0,69	0,71
range [km]	950	900	250	250
RMSE [ng.m^{-3}]	**0,64**	**1,84**	**0,60**	**2,02**
RRMSE [%]	**129,0%**	**68,9%**	**120,9%**	**75,4%**
bias (MPE) [ng.m^{-3}]	**0,08**	**0,04**	**0,06**	**0,08**

The rural and urban background maps were constructed at 10x10 km^2 resolution and combined using a population density grid at 1x1 km^2 resolution, as explained in Section 5.1.2. The combined BaP concentration map has a 10x10 km^2 resolution.

Figure 6.1 presents the combined final maps for the 2012 BaP annual mean for Europe in a 10x10 km^2 grid resolution, using EMEP and CHIMERE model outputs. Turkey was excluded due to lack of air quality data. Red and purple areas and stations exceed the TV of 1 ng.m^{-3}.

It should be noted that the concentration levels presented in the maps are related to the aggregated 10x10 km^2 grid. Note that for estimating population exposure and health impacts a 1x1 km^2 resolution is used to account for the variability within a 10x10 km^2 grid cell. Although the estimated concentrations at the 1x1 km^2 resolution are highly uncertain (and therefore not shown), part of the correlation between concentration and population density (for example, small towns not resolved in the 10x10 km^2 grid) are accounted for. For better visualising the actual urban concentration levels at the actual urbanised areas, i.e. without the influence of the dominating pattern of extended rural areas, a separate urban background map is presented in Annex 3, Figure A3.1. In this map, the non-urban areas are masked.

Figure A5.1 in Annex 5 presents the difference between the interpolated maps created using EMEP and CHIMERE output data. This map can also be compared with the map showing the differences between just the EMEP and CHIMERE model outputs, see Figure A5.2 in Annex 5. The major differences are observed in the areas with large differences between the CTM outputs and similarly with low density of measurement stations (e.g. Romania). Contrary to that, large differences between the CTM outputs in the areas with high density of measurement stations (e.g. Benelux) do not lead into large differences of the model outputs. The reason is that the spatial interpolated mapping results are primarily driven by the measurement results, see Horálek et al. (2014b).

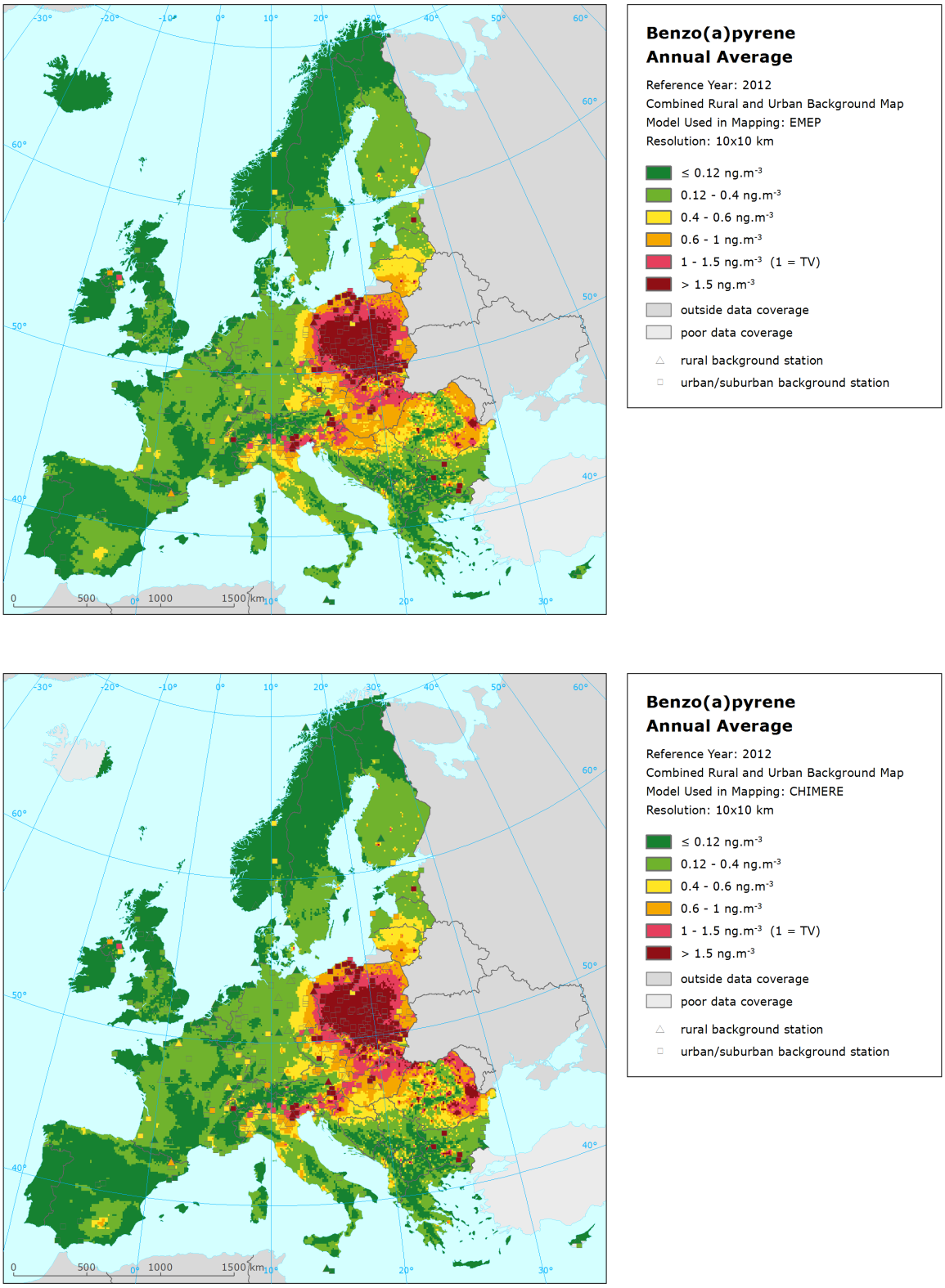


Figure 6.1 ***Spatial interpolated concentration field of annual mean BaP in 2012 using EMEP (top) and CHIMERE (bottom) models and the measured values at measurement points.***

6.3 Uncertainties

The air quality directive (EU, 2004) sets a TV for annual mean BaP concentrations equal to 1 ng.m^{-3}. In addition, the upper and lower threshold values (UAT and LAT) of 0.6 and 0.4 ng.m^{-3} are established for the assessment of concentrations and exposure. These thresholds are quite high compared to an estimated reference level of 0.12 ng.m^{-3}, corresponding to an additional lifetime cancer risk of 1 x 10^{-5}. The same directive does not require monitoring for levels under the LAT (0.4 ng.m^{-3}). This means that over large parts of Europe BaP concentrations are not measured or the measurement density is very low, increasing the uncertainties in a population exposure estimate.

Uncertainty estimated by cross-validation

Using RMSE as the most common indicator, the *absolute mean uncertainty* of the final combined map at areas 'in between' the station measurements can be expressed in ng.m^{-3}, see Table 6.3. RMSE for rural areas is 0.64 ng.m^{-3} for the concentration map using the EMEP model and 0.60 ng.m^{-3} for the map using CHIMERE results. For urban background areas, with higher concentrations, the RMSE is also higher, i.e. 1.8 ng.m^{-3} for the map using EMEP and 2.0 ng.m^{-3} for the map using CHIMERE. If we express the uncertainty in relation to the mean BaP concentration of all the stations, we obtain a relative RMSE (RRMSE) of 129% (EMEP) and 122% (CHIMERE) for rural areas and of 69% (EMEP) and 75% (CHIMERE) for urban background areas. The mean measured annual value at the stations is 0.5 ng.m^{-3} for rural background stations and 2.7 ng.m^{-3} for urban/suburban background stations. Both absolute and relative uncertainty values are influenced by these levels. It can be stated that the quality of the map using CHIMERE is slightly better for rural areas, while the quality of the map using EMEP is slightly better for urban background areas.

Figure 6.2 shows the cross-validation scatter plots, for both rural and urban background areas. R^2 and the slope a (from the linear regression equation $y = a \cdot x + c$) should be as close to one as possible, the intercept c should be as close to zero as possible. The R^2 indicates that for the rural areas about 44 % (using EMEP model) or 51 % (using CHIMERE model) and for the urban background areas about 71 % (using EMEP model) or 66 % (using CHIMERE model) of the variability is attributable to the interpolation.

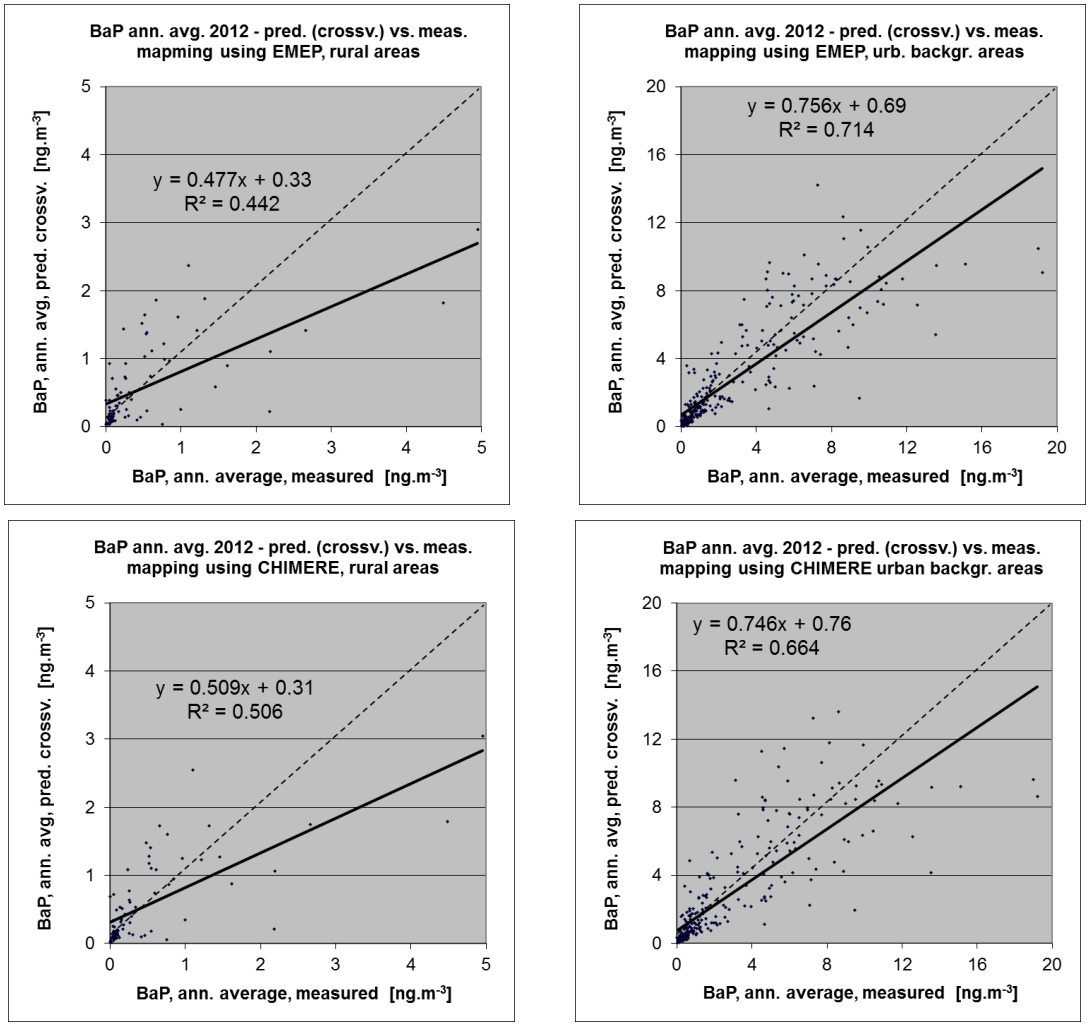
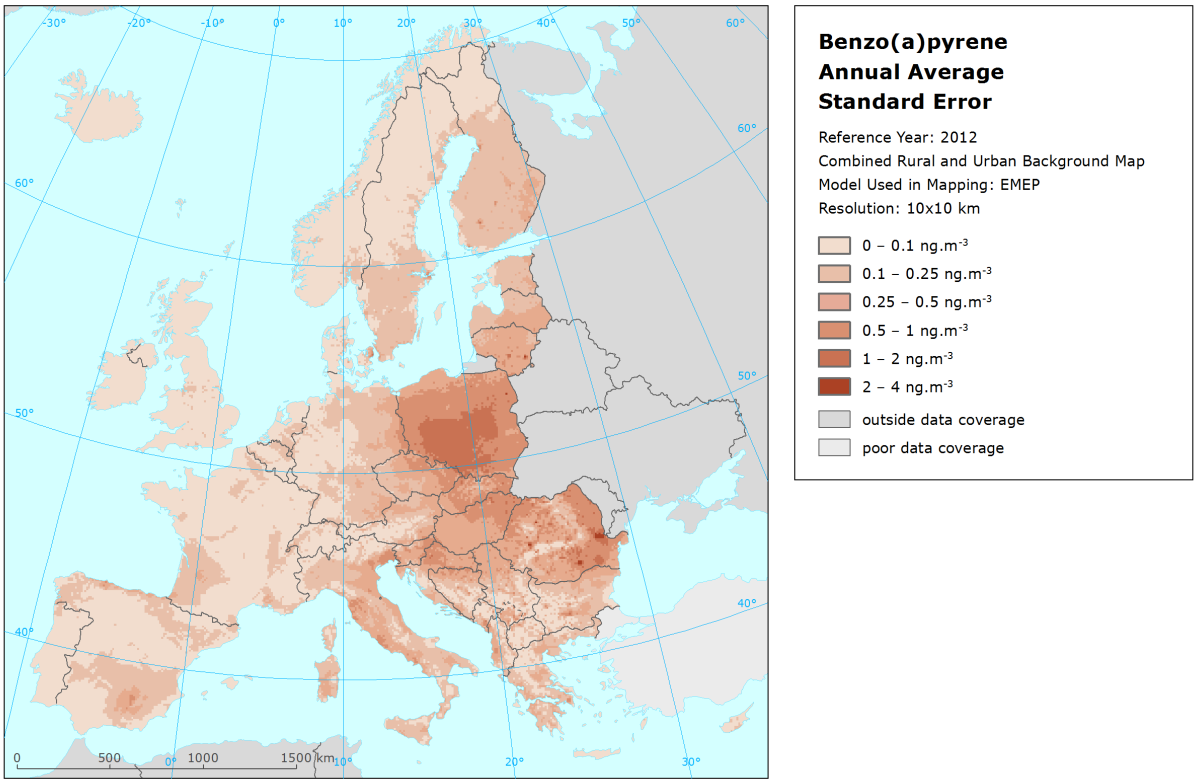


Figure 6.2 ***Correlation between cross-validation predicted values (y-axis) and measurements (x-axis) for the BaP annual average map for 2012 created using EMEP (upper) and CHIMERE (lower) for rural (left) and urban (right) areas.***

Uncertainty maps based on the kriging theory

In addition to the cross-validation analysis, the uncertainty can be expressed also based on the kriging theory. Figure 6.3 shows the uncertainty maps, using EMEP (top) and CHIMERE (bottom) model outputs, expressed as the interpolation standard error. These uncertainty maps are directly related to the concentration maps of the combined rural and urban background BaP annual average in 2012 presented in Figure 6.1



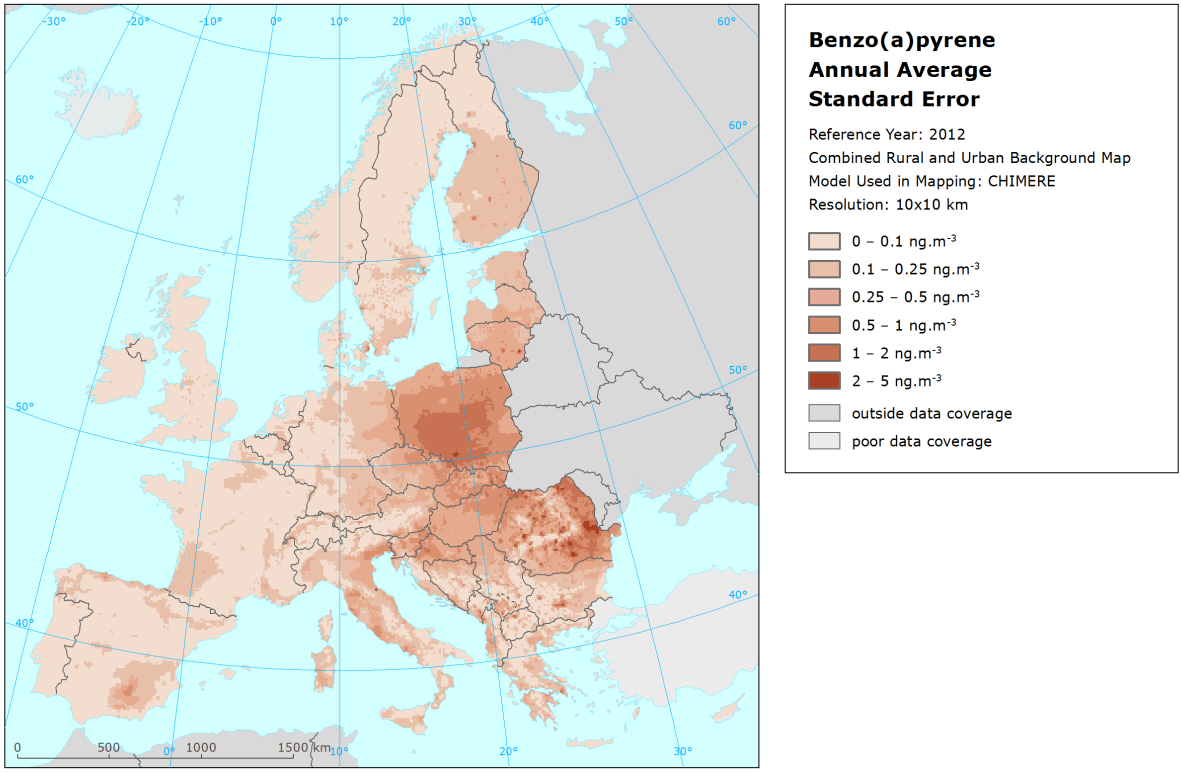


Figure 6.3: ***Uncertainty map showing interpolation standard error for concentration map of BaP annual average in 2012, using EMEP (top) and CHIMERE (bottom) models.***

The uncertainty is expressed in ng.m^{-3}, i.e. in the same units as the concentration maps (Figure 6.1). In general, the highest uncertainty is in the areas of the highest concentrations. Note that the uncertainty maps presented in Figure 6.3 are related to the 10x10 km^2 resolution.

Figure 6.4 shows the uncertainty maps, expressed as the relative standard error. The relative standard error is calculated by dividing the standard error (Figure 6.3) by the concentration (Figure 6.1) for each grid cell. It's resolution is also 10x10 km^2 and the unit relative numbers.

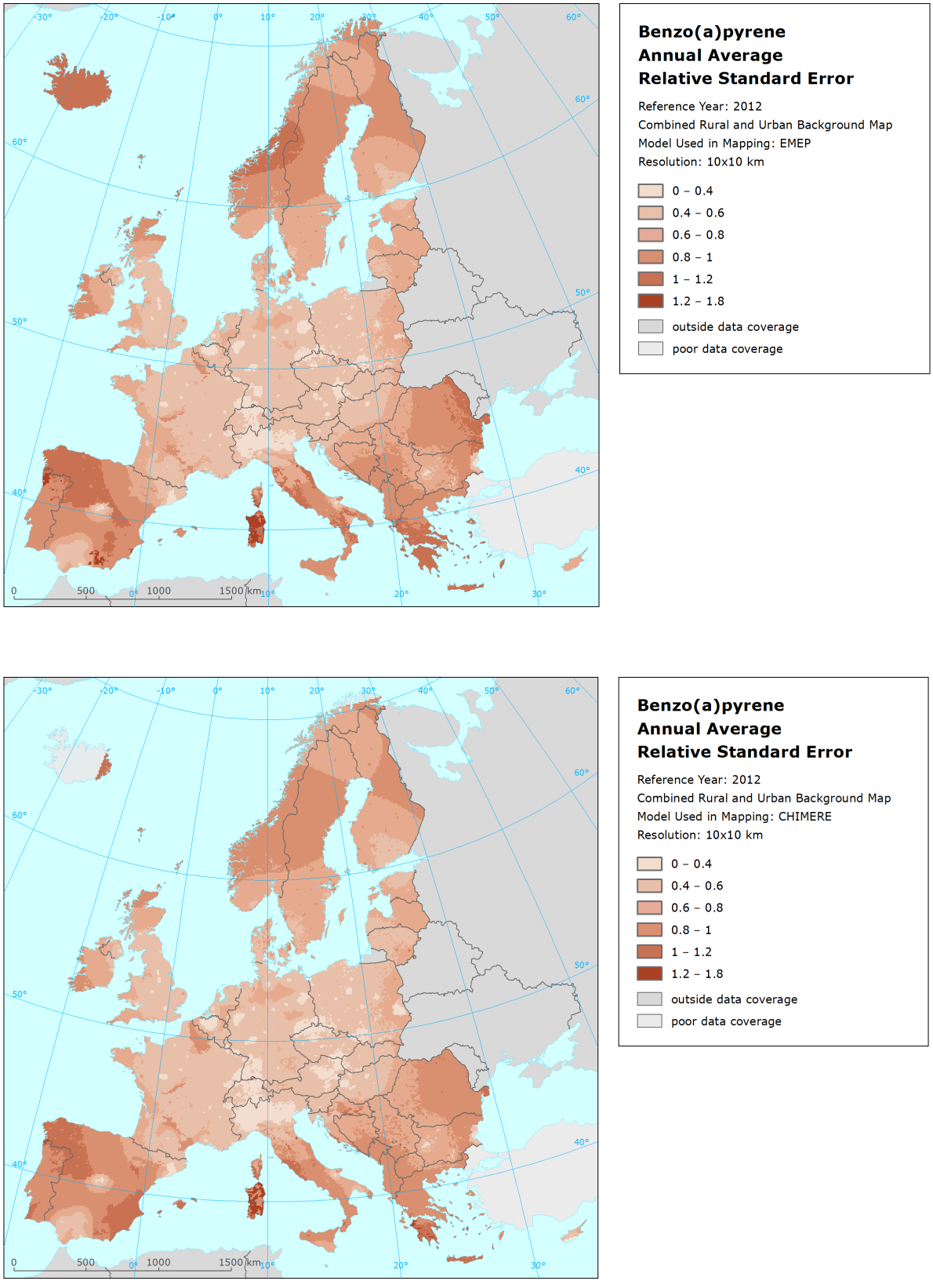
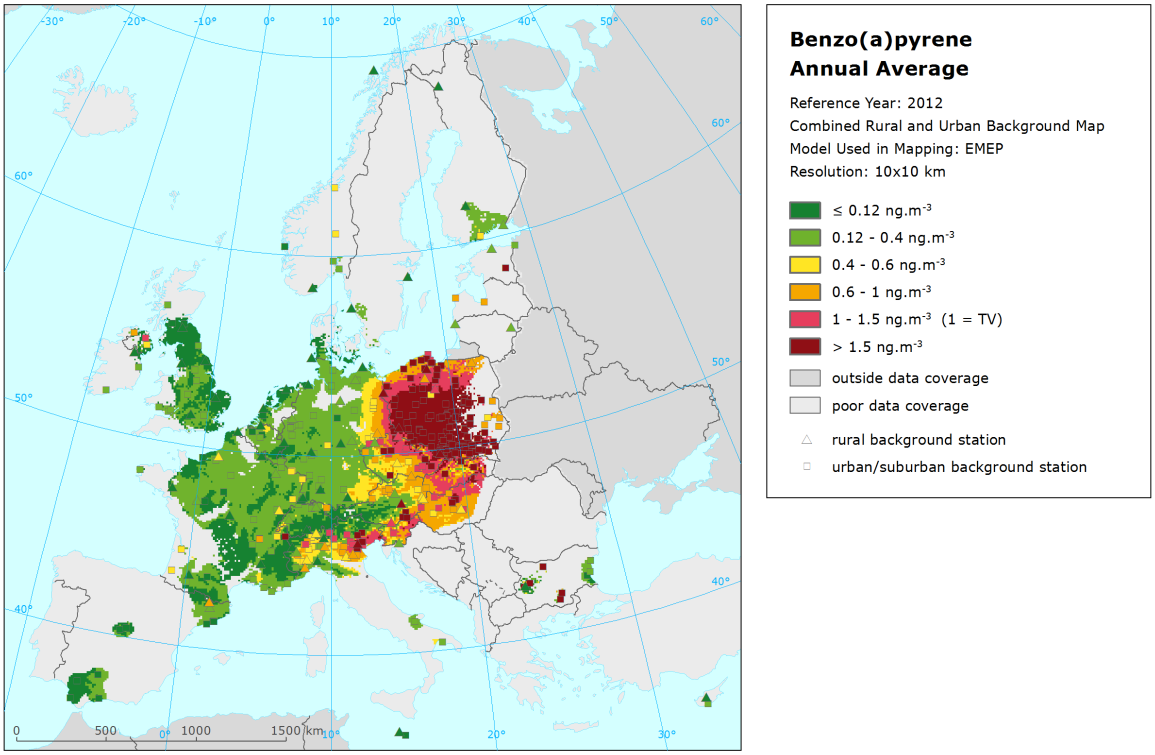


Figure 6.4 ***Uncertainty map showing interpolation relative standard error for concentration map of BaP annual average in 2012, using EMEP (top) and CHIMERE (bottom) models.***

Limiting the BaP concentration map to the countries with most of their territory within an area with acceptable relative standard error below 0.60 (see Directive EC 2004/107/EC, Annex IV, objectives for modelling), we obtain the concentration maps presented in Figure 6.5. It shows the spatial interpolated concentration field and the values in the measurement points of annual mean BaP concentrations in 2012 limited to areas with a relative standard error under 0.60.

Following such criterion, the following countries present a relative uncertainty too high to be included in the assessment: Portugal, Spain, Ireland, Iceland, Norway, Sweden, Finland, Romania, Bulgaria, Serbia, Croatia, Bosnia, Montenegro, Albania, Greece, FYR of Macedonia, and Malta. For Italy, the NUTS1 regions are taken into account: the regions "Centre", "South" and "Islands" are excluded, while the region "North" show quite low uncertainty and have therefore been included in the map. The relative uncertainty is mostly driven by the density of the measurement network. Thus, more measurements are necessary to reduce the uncertainty in the BaP concentration map, particularly in the areas with high uncertainty (light grey areas in Figure 6.5). BaP measurement stations are particularly necessary in the densely populated areas where high levels are expected, e.g. in Romania, Bulgaria, other Balkan states, and the Baltic States.



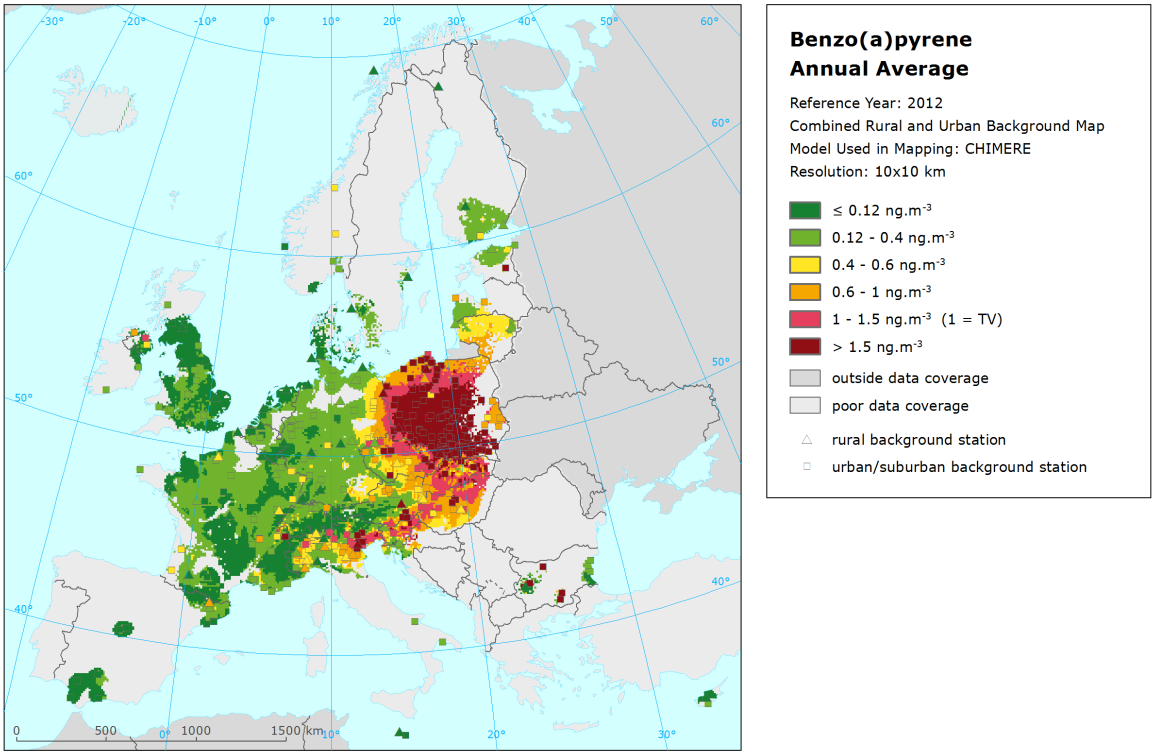


Figure 6.5 ***Spatial interpolated concentration field of annual mean BaP in 2012 limited to areas under 0.60, using EMEP (top) and CHIMERE (bottom) models, and the measured values at measurement points.***

7 Exposure

Exposure of individuals to PAHs occurs through inhalation, ingestion, and absorption through the skin. PAHs emitted to the air may contribute to human exposure by these three different pathways. PAHs in the air may be inhaled, or may be deposed and contaminate the soil, crops and plants, leading to low concentrations of PAHs in food and water. More substantial amounts of PAHs, including BaP, may be found in food, as a consequence of various methods of cooking, preservation and storage. As PAHs are formed in smouldering as well as flaming combustion, "burnt" food contains PAHs, as do smoked foods. Inhalation is the dominant pathway for BaP exposure for smokers, while for non-smokers the main route of exposure is through food (Kim et al., 2013; WHO 2000; USEPA 1994). The focus of this chapter is on the estimation of the European population exposure to ambient BaP concentrations.

The population exposure to BaP concentrations in Europe in 2012 was estimated based on the final combined concentration map (in 1x1 km^2 resolution) and the population density map, see Section 5.1.3.

Table 7.1 gives the population exposure frequency distribution for a limited number of Bap annual mean concentration ranges, as well as the population-weighted concentration for 2012. The population-weighted concentration of BaP for the whole Europe (i.e. the average BaP concentration which the average European inhabitant is exposed to) was about 0.9 $ng.m^{-3}$ in 2012.

Figure 7.1 shows the population-weighted concentration[15] for 2012, with the two different models outputs. It gives a better picture of the population exposure than the concentration maps presented in Figure 6.1, as it takes into account the population density. It shows that in Baltic countries (especially Lithuania) and some regions of Eastern Europe the population-weighted concentration is very high, i.e. above 1.5 $ng.m^{-3}$. Ten countries in Eastern Europe[16] have an average population-weighted concentration above 1 $ng.m^{-3}$ estimated using both models; these are Poland, Romania, Bulgaria, Hungary, the Czech Republic, Slovakia, Slovenia, Montenegro, Estonia, and Lithuania. People live close to the source, dominated by domestic heating (see Chapter 3) and therefore in areas of higher concentrations than where population density is lower.

About 20 % of the European population has been exposed to BaP annual mean concentrations above the TV of 1 $ng.m^{-3}$ in 2012 and only about 12 % of the European population live in areas with concentrations under the estimated reference level (RL) of 0.12 $ng.m^{-3}$ (Table 7.1). Both the percentage of inhabitants living in the areas above TV (20%) and the average population-weighted concentration in Europe (7.5 times higher than the WHO RL) are quite high. As mentioned, the reason for this is the fact that the most of the European population live in urban areas, in general more polluted than the rural areas. (See Annex 3, Figure A3.1).

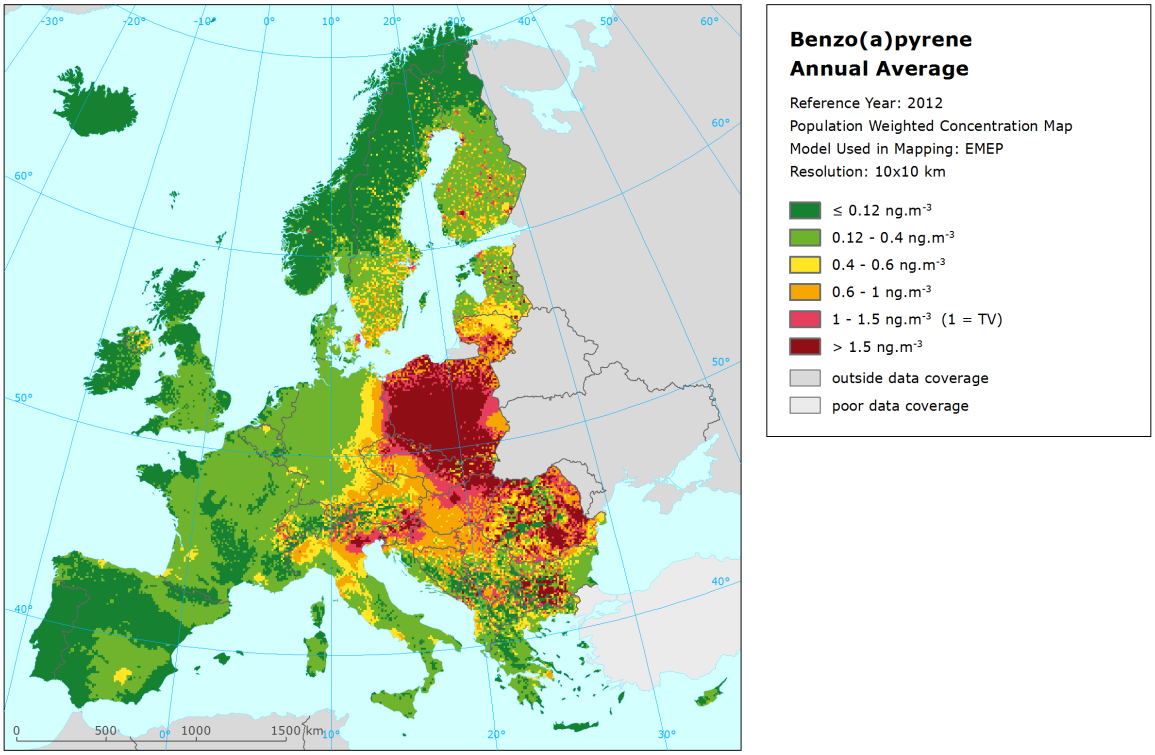
[15] The population weighted concentrations presented with a 10x10 km^2 resolution were calculated for each 10x10 km^2 gridcell by: 1) multiplying concentration with population at each 1x1 km^2 grid cell; 2) summing this product for the hundred gridcells (1x1 km^2) within the 10x10 km^2 gridcell; and 3) dividing this sum by the total population of the 10x10 km^2 gridcell. See equation 4.2.

[16] The average population-weighted concentration in the Former Yugoslavian Republic of Macedonia is above 1 $ng.m^{-3}$ when estimated using the CHIMERE model, but not using the EMEP model.

Table 7.1 Population exposure and population-weighted concentration for BaP annual mean in 2012, based on the interpolated concentration map using EMEP (top) and CHIMERE (bottom).

Map	Population [inhbs . 1000]	BaP, annual average, exposed population [%]						Population-weighted conc. [ng.m^{-3}]
		< TV				> TV		
		< 0.12 ng.m^{-3}	0.12 - 0.4 ng.m^{-3}	0.4 - 0.6 ng.m^{-3}	0.6 - 1 ng.m^{-3}	1 - 1.5 ng.m^{-3}	> 1.5 ng.m^{-3}	
using EMEP	516 730	11.7	46.7	10.4	10.7	6.8	13.6	0.84
		79.5				20.5		
using CHIMERE	516 169	12.6	46.2	9.8	11.0	6.1	14.3	0.92
		79.6				20.4		

Note: The population covered is different for the two models, as their modelling domain differs slightly. For the map using CHIMERE, Iceland, northern Norway and southern Cyprus is not included in the calculation, as it's outside the model domain. Turkey is not included in the calculation due to lacking air quality data.



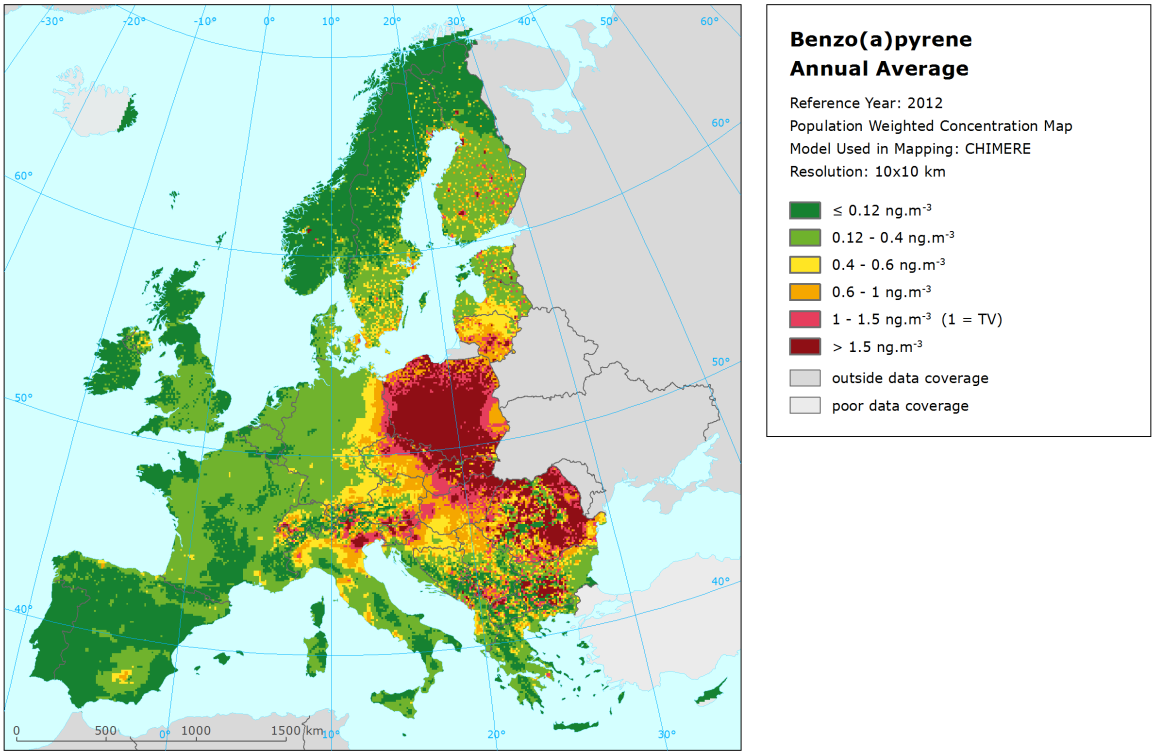


Figure 7.1: ***Population-weighted concentration field of annual mean BaP in 2012, using EMEP (top) and CHIMERE (bottom) models.***

8 Estimation of health effects of BaP in Europe

In this health impact assessment, the BaP effects have been estimated for lung cancer. Other health endpoints have not been selected either because of a lack of an adequate dose-response relation or because the relation was derived for occupational conditions with typical concentrations one or two orders of magnitude higher than under ambient conditions. By this restriction the total burden of disease attributable to the exposure to ambient BaP will evidently be underestimated.

For lung cancer we use here the exposure –response function as recommended by the WHO Air Quality Guidelines (WHO, 1987), a value which has not been changed in later revisions of the guidelines. Based on epidemiological data from studies in coke-oven workers, a unit risk for BaP as indicator air constituent for PAH is estimated to be 8.7 x 10^{-5} per ng.m^{-3} (WHO 1987, 2000). The unit risk is defined as "the additional lifetime cancer risk occurring in a hypothetical population in which all individuals are exposed continuously from birth throughout their lifetimes to a concentration of 1 ng.m^{-3} in air". This might be interpreted as follows: if unit risk = 8.7 × 10^{-5} per ng.m^{-3}, 8.7 excess cancer cases are expected to develop per hundred thousand people if exposed daily for a lifetime to 1 ng.m^{-3}. This unit risk value falls within the range of estimated unit risks as summarized by Boström et al (2002): 2.3 – 43 x 10^{-5} per ng.m^{-3} with a median value of 8.7 x 10^{-5} per ng.m^{-3}. From this we estimate a 95% confidence interval of 2.9 – 15 x 10^{-5} per ng.m^{-3}.

The number of lung cancer incidences (N) is estimated as

$$N = C \cdot Pop \cdot (UR/L) \qquad (7.1)$$

Where:
C is the ambient air concentration of the pollutant
Pop is the exposed population,
UR is the unit risk,
L is the lifetime exposure, set to 70 year.

Concentration data are taken from the interpolated concentration fields as described in Section 5.1.3. The calculations are performed on the 1x1 km^2 grids; in this way we account for the correlation between concentration level and population density (Liu et al., 2014). Regional totals are obtained by summation over all grid cells in a region.

Using the concentration data from both CTMs, the number of lung cancer incidences within the whole model domain has been estimated as 550 (95% CI: 180-940)[17]and 600 (95% CI: 200-1030) for the EMEP and CHIMERE model, respectively. In line with the concentration and population exposure maps shown in Figure 6.1, the largest health impacts can be found in the eastern European countries. About 50% of the incidences would occur in Poland and Romania.

[17] Note that the 95% confidence interval is based on the uncertainties in the unit risks. It does not include the uncertainties in the concentration maps.

Interpolated maps tend to smooth concentrations, masking areas of higher concentrations that are smaller than the grid cell size. The concentration maps show that the concentrations in urban areas – and therewith the health impacts – are probably underestimated. In addition, in large parts of Europe the BaP concentration is below the lower assessment threshold (LAT) of 0.4 ng.m^{-3} and monitoring is not required by the air quality directive (EC, 2004). In these regions the interpolated concentrations are highly uncertain and most probably underestimated, as models provide smoothed background concentrations. In a sensitivity analysis we tried to compensate for this. In each grid cell the concentration has been modified according to:

- when the concentration is below the LAT it is set to two-third of the LAT (C_{min}=0.27 ng.m^{-3});
- the inverse of the regression lines found in the cross-validation (Figure 6.2) is used to correct for the underestimation: $C_{cor} = (1/a)\ C_{int} - (b/a)$ where a and b are slope and intercept of the regression lines in Figure 6.2 and C_{int} is the original interpolated concentration;
- the concentration in the grid cell is replaced by the maximum value of C_{int}, C_{min} and C_{cor}.
- a small fraction of BaP (< 10%) is in the gaseous phase. This fraction differs over Europe depending on temperature and aerosol loading and is difficult to quantify. It is not included in the sensitivity run.

Rerunning the impact assessment for the modified concentration fields, as explained above, results in a nearly 20% increase in the number of incidences: to 630 (EMEP) and 700 (CHIMERE) for 2012.

Ambient particulate matter contributes to lung cancer incidence in Europe (Hamra et al., 2014). Using a relative risk of 1.09 (95% CI: 1.04, 1.14) resulted from a meta-analysis (Hamra et al., 2014) and an interpolated PM2.5 exposure map (Horálek et al., 2014a) we estimate 34600 new case of lung cancer in Europe in 2012. As most of the BaP in ambient air is associated to PM2.5, this estimate includes also the impact of BaP exposure on increased lung cancer incidence.

Although relatively small compared to PM, the estimates given above are likely an underestimation of the health impacts of ambient BaP mainly due to:

- concentrations are underestimated mainly due to the lack of measurement data;
- only intake via inhalation is considered, indirect intake via deposition and uptake in the food chain might play a role in certain regions;
- other health endpoints have not been included.

9 Summary and conclusions

PAHs are considered among the most dangerous air pollutants due to their carcinogenic and mutagenic character. They can be transported over long distances in the atmosphere resulting in a widespread distribution on the continental scale, and they bio-accumulate in the food chain. The European directive (EU, 2004) sets a target value for ambient air concentration of BaP, as a marker for the carcinogenic risk of PAHs in ambient air, in order to avoid, prevent or reduce harmful effects of PAHs on human health and the environment as a whole. The target value for BaP (measured in PM10) was set to 1 $ng.m^{-3}$ as an annual mean, to be met by 2013. This target value is quite high compared to an estimated reference level[18] of 0.12 $ng.m^{-3}$.

The present work summarised the main emission sources of BaP and discussed the development in emissions over the last decade in Europe. Residential combustion is by far the most important sector, contributing with 82 % (2012) of the total BaP emissions, mainly from wood- and coal-burning. Other sources are solid fuels transformation, aluminium production, natural emissions, and road traffic. The assessment of the Europe-wide BaP emission trend is very uncertain. Changes in emissions over the last 10 years vary considerably from country to country and the analysis is very uncertain due to the incompleteness of BaP emission inventories, especially in earlier years. In addition, seven[19] out of the EU28 countries have never reported their BaP emissions, as there is at present no legislation requiring reporting of BaP emissions by the Member States.

This study estimated current BaP concentration levels, population exposure and potential health impacts. These estimates were done by combining the best available information from chemical transport models (CTMs) and observations through the use of spatial interpolation methods. The work was based on the current ETC/ACM mapping methodology to estimate regional and urban background pollution levels in Europe and their uncertainties. Based on the estimated concentration maps compliance with the EU TV was assessed. Furthermore, the European population exposure to BaP background ambient air concentrations and subsequent health impact were estimated.

The current spatial interpolation method used at ETC/ACM is residual kriging. This is a linear regression method followed by kriging of the residuals. The linear regression uses measured air quality data (mostly from the AirBase database) as a dependent variable, while results from a CTM and supplementary data such as altitude and meteorology are introduced as independent variables. In this study model results from both the EMEP MSC-E and CHIMERE models have been used and analysed. The spatial interpolation was carried out separately for rural and urban background maps. The rural map is based on rural background stations, while the urban background map is based on urban and suburban background stations. Residual kriging results are largely driven by observations. On the other hand, the modelled concentrations and the chosen auxiliary variables also have an impact in the spatial interpolated results, especially where measurement data is scarce. The quality of these air quality concentration maps relies primarily on the quality of the input data used, namely the quality of the observations, modelled results and auxiliary information introduced in the spatial interpolation method.

The spatial interpolated concentration fields, with a spatial resolution of 10x10 km^2, show quite similar results when comparing the use of EMEP and CHIMERE model results for 2012. Both

[18] Corresponding to an additional lifetime cancer risk of 1×10^{-5}.
[19] Austria, Belgium, Finland, Greece, Italy, Portugal, and Spain.

concentration maps show high 2012 annual mean concentrations of BaP in Eastern Europe, especially Poland and the Czech Republic, as well as in Northern Italy.

The analysis of the uncertainties of the interpolated map shows that in several regions in Europe the concentration estimate is quite uncertain. In large parts of Europe the relative standard error exceeds the 60% as required in the European directive (EU, 2004). The high uncertainty is mostly due to the low density of the measurement network. The air quality directive (EU, 2004) sets a lower assessment threshold (LAT) of 0.4 $ng.m^{-3}$ for the assessment of concentrations and exposure and does not require monitoring for levels under it. This threshold is quite high compared to an estimated reference level of 0.12 $ng.m^{-3}$. Thus, in large parts of Europe BaP concentrations are not measured or the measurement density is very low, increasing the uncertainties in the interpolated concentration map and population exposure estimate. In addition, there is lack of measurement stations also in areas where high concentrations are expected, e.g. in Romania, Bulgaria, other Balkan states, and the Baltic States. It is particularly important to establish stations in densely populated areas where higher concentrations are expected in order to decrease the uncertainty in the concentration map and population exposure estimates.

The population exposure frequency distribution was estimated for a few BaP annual mean concentration intervals, as well as the population-weighted concentration (i.e. the average BaP concentration which the average European inhabitant is exposed to) for 2012. The population-weighted concentration of BaP averaged over the whole Europe was about 0.9 $ng.m^{-3}$. About 20 % of the European population was exposed to BaP annual mean concentrations above the target value of 1 $ng.m^{-3}$ in 2012 and only about 12 % of the European population live in areas with concentrations under the estimated reference level of 0.12 $ng.m^{-3}$. The estimated exposure map for Europe shows that the population-weighted concentration of BaP is very high and above 1.5 $ng.m^{-3}$ in large regions of Eastern Europe, including the Baltic countries (especially Lithuania). Both the percentage of inhabitants living in the areas above TV (20%) and the average population-weighted concentration in Europe (7.5 times higher than the WHO RL) are quite high. People live close to the source, dominated by domestic heating, and therefore in areas with higher concentrations than where population density is lower.

In the health impact assessment, the BaP effects have been estimated for lung cancer incidence using the exposure –response function recommended by WHO. Using the concentration data from both models, the number of lung cancer incidences within the whole model area has been estimated as 550 (95% CI: 180-940) and 600 (95% CI: 200-1030) for the EMEP and CHIMERE model, respectively. Taking into account the inherent underestimation of concentrations and exposure due to the mapping procedure, leads to correcting the number of incidences to 630 (EMEP) and 700 (CHIMERE) for 2012. The largest health impacts can be found in the eastern European countries. About 50% of the incidences occur in Poland and Romania. These results are most probably underestimated, and a sensitivity test indicates the number of incidents may be underestimated by 20% or more.

In addition, it is important to bear in mind that BaP only represents part of the overall carcinogenicity amongst the group of PAH compounds for which data are available (WHO, 2010; Holland et al., 2001; Pufelete et al., 2004). The mutagenic and carcinogenic potency varies widely over the PAH. Although BaP is not the most abundant pollutant, its carcinogenic potency is amongst the highest. Never the less, BaP alone will underestimate the carcinogenic potential of ambient PAH mixtures since co-occurring PAHs are also carcinogenic. The relative contributions of more potent PAHs (e.g. dibenzo[a, l]pyrene) to the carcinogenic activity of a total PAH mixture may be far greater than that of BaP, but further research is needed to quantify it (Okona-Mensah et al., 2005).

The findings from this study suggest that the health impacts from exposure to BaP in Europe are considerably smaller than for exposure to PM2.5. Never the less, BaP is an important indicator for PAHs and is likely among the four most important pollutants regulated by the European Air Quality Directives (EU, 2004 and EU, 2008) (together with PM, O_3 and NO_2).

BaP is a good indicator of domestic combustion emissions, as more than 80% of total BaP emissions in Europe come from this sector. Thus, BaP is a very useful indicator for air quality management targeting emissions from the increasingly important domestic combustion sector. Domestic combustion is also a very important contributor to PM2.5 primary emissions (EEA, 2014b). Nonetheless, since PM2.5 also has other important sources, BaP is probably a better indicator to test the result of implemented measures and policies designed to mitigate emissions from domestic combustion.

Large uncertainties are associated with the current assessment of health impacts from BaP or PAHs in general. Two main improvements are needed in order to reduce the uncertainties related to mapping BaP concentrations and exposure in Europe and support air quality management: 1) improve the quality and completeness of the PAH and BaP emission inventories; 2) improve the monitoring network over Europe for BaP concentrations, especially in areas with expected higher concentrations (e.g. in the Balkan and Baltic countries) and with higher population density. The results from the CTMs are very sensitive to the input emission data, and measurement data is crucial for understanding the behaviour of BaP in the atmosphere, as well as to test and validate the CTMs. Furthermore, the methodology applied here to produce concentration and exposure maps of BaP would be greatly improved by the increase in availability of BaP measurements, especially in areas of low measurement density and regions with expected concentrations above LAT.

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Annex 1 Development in BaP emissions from 2003 to 2012

The comparison of the differences between the EMEP estimated emissions for modelling and the officially reported emissions over the last 10 years shows that the gap between the two have decreased in the last 10 years. Table A1.1 shows the total EU28 BaP emissions for the officially reported emissions, for the difference between the EMEP estimate for modelling and the officially reported emissions, as well as this difference in percentage of the total officially reported emissions for each year. It shows that while the officially reported emissions in 2003 where less than half (48%) of EMEP estimated emissions for modelling, in 2012 the officially reported emissions were 75 % of the total estimated for modelling. Thus, the difference in percentage of the total officially reported emissions was reduced from 107 % in 2003 to 34 % in 2012. On the other hand, the number of not reporting countries have not changed much. Nine countries did not report BaP emissions between 2003 and 2007, eight in 2008, 2009, 2011 and 2012, and seven failed to report in 2010. The only two countries that reported only a few out of the 10 years were Luxembourg and Malta, both with very low emissions. Consequently, the change in completeness of the officially reported emissions over the years is within the reporting countries and not due to the fact that more countries reported their BaP emissions.

The seven countries that did not report their emissions in any of the 10 years were Austria, Belgium, Spain, Finland, Greece, Italy and Portugal.

Table A1.1: Differences in BaP total emissions for the EU28 countries between officially reported and estimated by EMEP for modelling in the period 2003 to 2012.

	EU28 BaP emissions (T/yr)			
Year	Officially reported	EMEP model emis.	Difference	% difference
2003	132,6	274,6	142,0	107
2004	147,9	278,4	130,5	88
2005	169,3	278,5	109,1	64
2006	169,1	274,3	105,2	62
2007	166,4	271,1	104,7	63
2008	178,3	283,5	105,2	59
2009	168,2	281,7	113,5	67
2010	184,7	309,0	124,3	67
2011	175,9	269,7	93,8	53
2012	179,9	240,5	60,6	34

The analysis of the BaP emission trends shows that the development differs considerably across Europe. The trends were calculated using the Mann-Kendall test estimated Sen's slope and the result of Mann-Kendall's test (Gilbert, 1987) for testing the presence of the trend. In total, and as Table A1.1 shows, the emissions estimated for modelling purposes have decreased from 2003 to 2012 in the EU28. However, any trend analysis should be interpreted with much caution. The trend methodology

assumes that the data are not correlated; for gap-filled data, e.g. when based on inter- or extrapolation, this will not be the case. In addition, improving statistics (on emission factors, fuel usages) will also induce an artificial trend.

On the other hand, the officially reported emissions increased considerably in total over the last 10 years, especially due to the increase reported by Germany and Poland. It is unclear if this increase is real, due to an increase in completeness of the emission inventories over the period, or any other reason. Particularly Germany, Poland and Romania have reported a considerable increase in their emissions between 2003 and 2012, while EMEP estimates indicate a decrease in the period. These three countries together account for more than half of the total officially reported BaP emissions in the EU28 in the period, and explain the narrowing gap between the reported and estimated emissions previously discussed. In contrast, emissions estimated for modelling for Hungary and Bulgaria increased more substantially in the EMEP estimates than the officially reported data.

Annex 2 Input data for the BaP concentration map

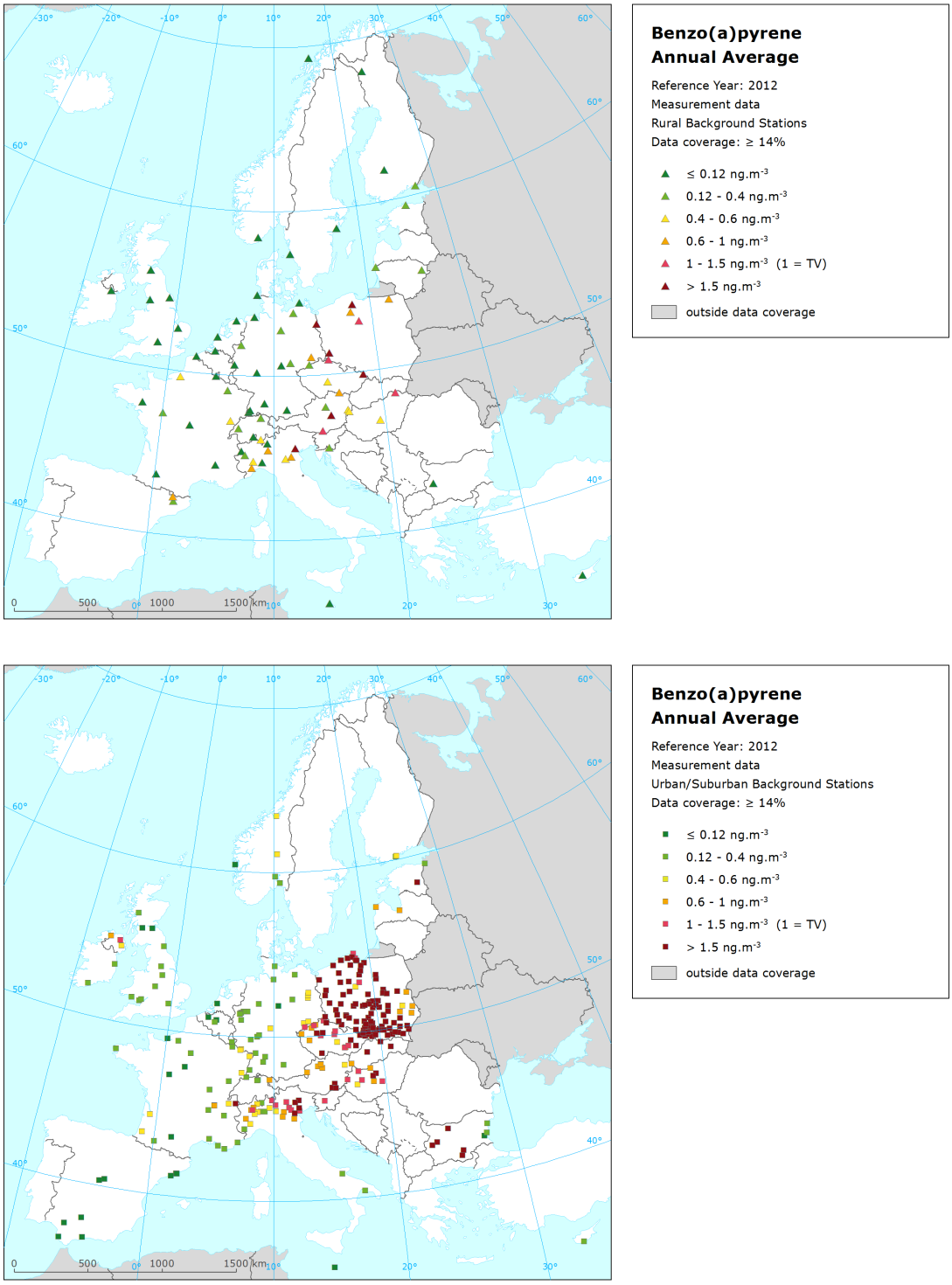

Figure A2.1 Measurement air quality data from rural (top) and urban/suburban (bottom) background stations. BaP, annual average, 2012. Units: ng.m^{-3}.

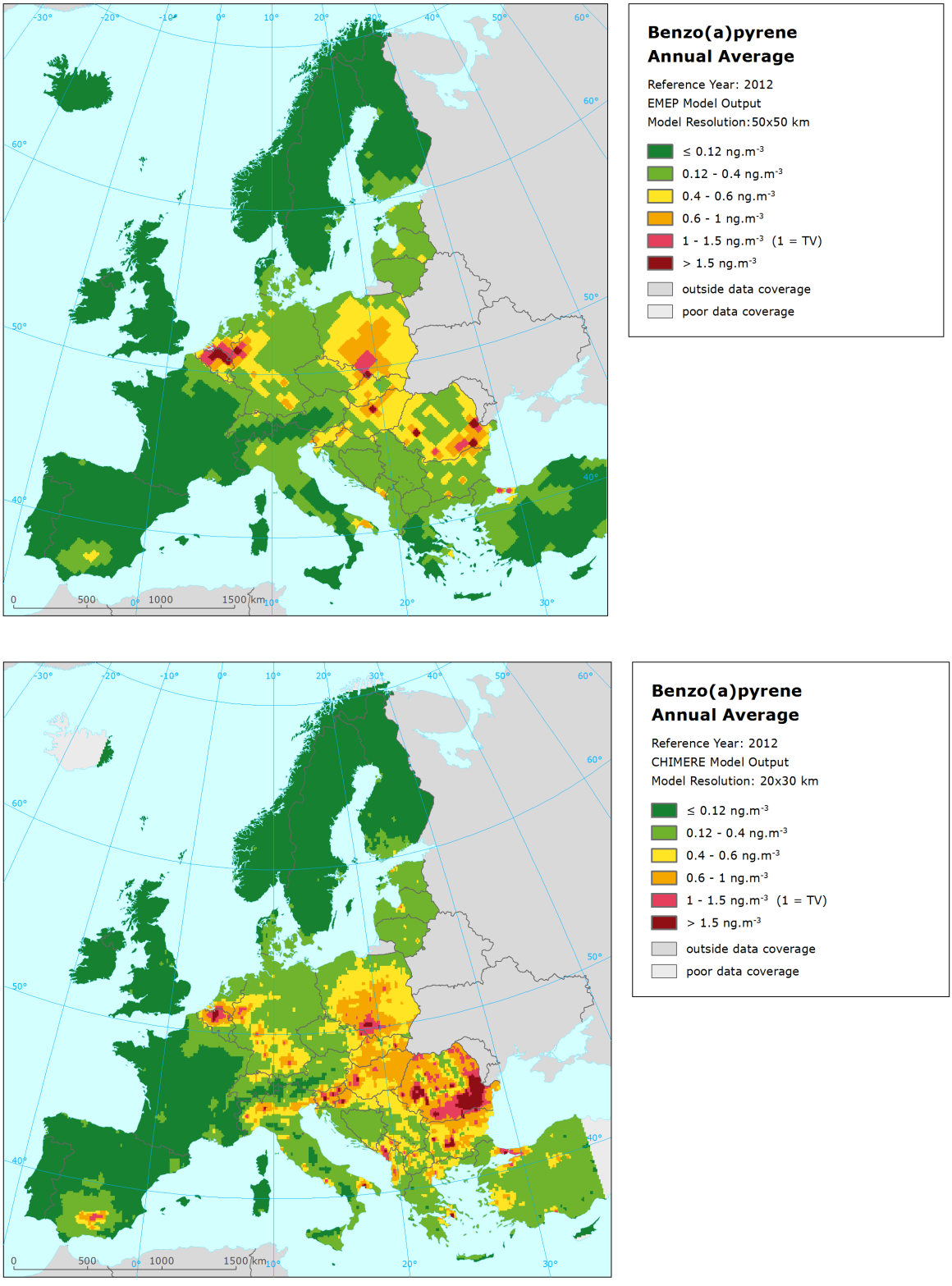

Figure A2.2 Output of chemical transport model EMEP (top) and CHIMERE (bottom). BaP annual average in 2012. Units: ng.m^{-3}.

Annex 3 Urban background maps

(applicable for urban areas only)

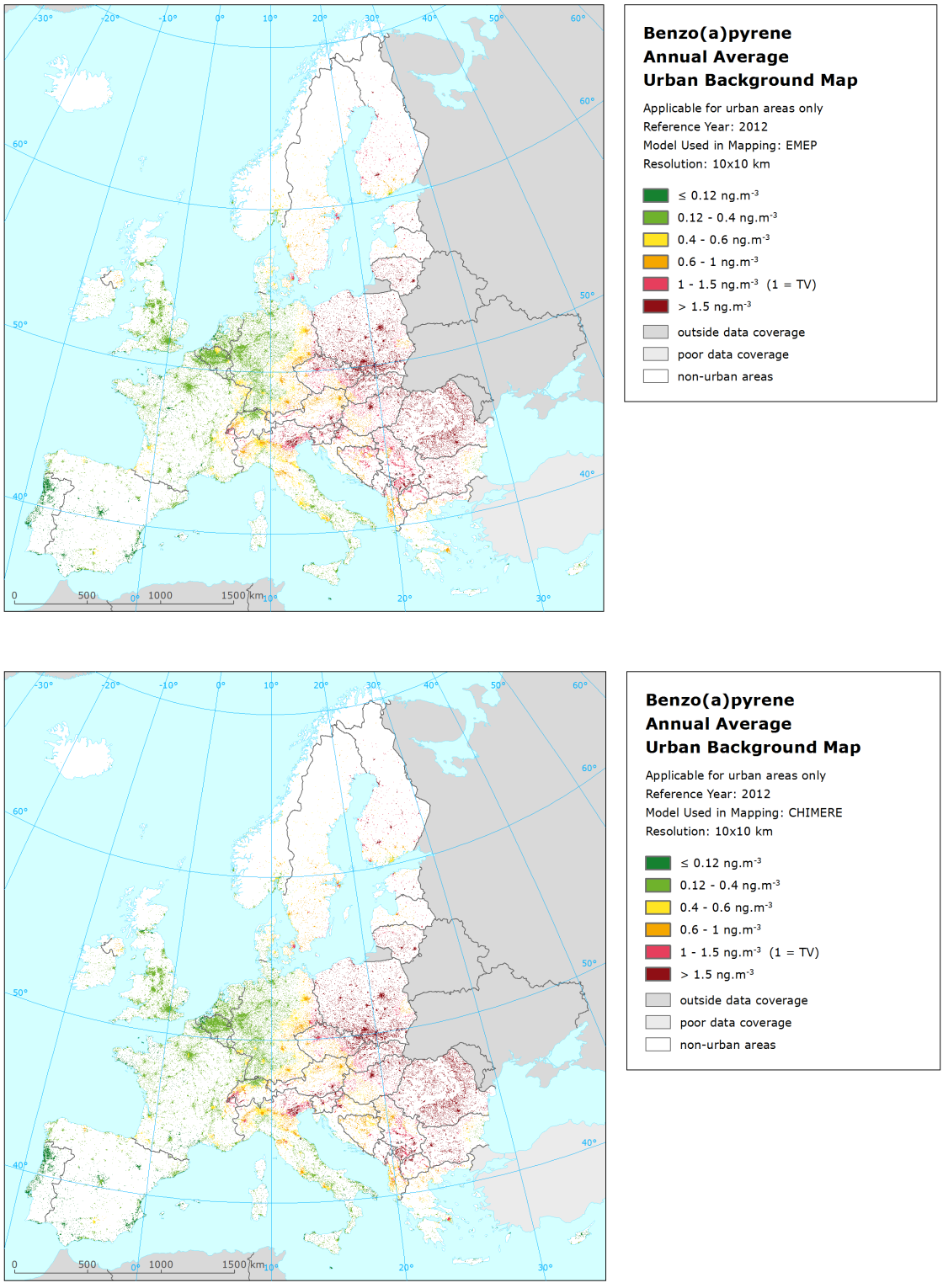

Figure A3.1 Urban background map of annual mean BaP, 2012. Spatial interpolated concentration field using EMEP (top) and CHIMERE (bottom) models and measurements at (sub)urban background stations.

Annex 4 Modeling BaP with the CHIMERE model - Technical note

Air concentrations of BaP for 2012 were simulated with the air quality model CHIMERE[20] over Europe with a resolution of 0.25°.

METHOD

Model Setting:
The 0.5° inventory emissions of EMEP (Vestreng et al., 2007 ; Vestreng, 2003) was used for BaP emissions. Data with a 0.25°x0.25° resolution from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used for meteorology. A climatology of MACC for aerosol and gas species are used for boundary conditions (boundary conditions for BaP are set at 0 $ng.m^{-3}$ due to lack of information).

Model description:
Gas-phase chemistry is simulated with the MELCHIOR mechanism. A sectional approach is used to represent particles over 8 sections (diameter from 40 nm to 10 nm). The evolution of particles follows Warren (1986). The formation of organic aerosol in CHIMERE is described in Bessagnet et al. (2008).

The partitioning of BaP between the gas phase and the organic phase of particles is computed according to its saturation vapor pressure (9.34×10^{-7} Pa according to Gusev et al., 2005). BaP should be almost entirely present into particles with at most 7% of BaP in the gas phase for very clean area (less than 1 $\mu g/m^3$ of organic aerosols).

Due to OH radicals in the atmosphere, BaP is degraded by photo-oxidation according to a first order reaction.

$$BaP + OH \rightarrow \text{Degradation Products}$$

A reaction rate constant of 5.00×10^{-11} $molecules^{-1}$ cm^3 s^{-1} is used based on Gusev et al. (2005).

[20] http://www.lmd.polytechnique.fr/chimere/

Annex 5 Differences in EMEP and CHIMERE concentration maps

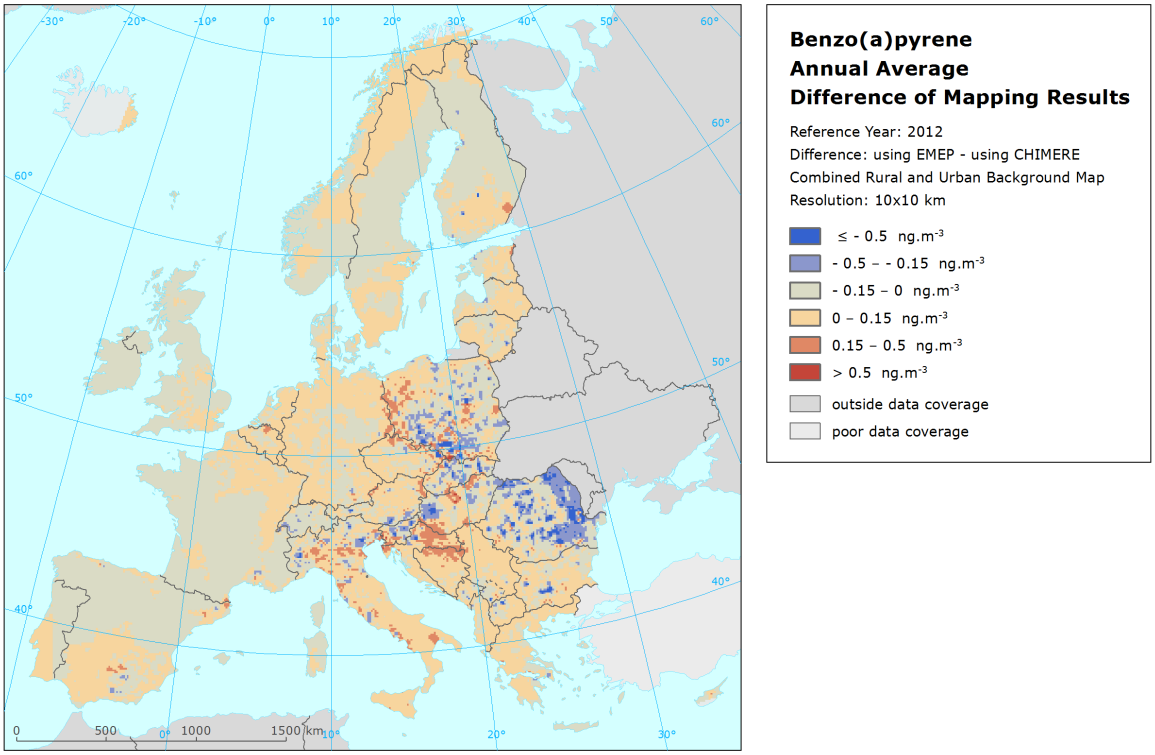

Figure A5.1: Difference of the mapping results (i.e. combined rural and urban background maps) of BaP annual average for 2012 using EMEP and CHIMERE models. Units: ng.m^{-3}.

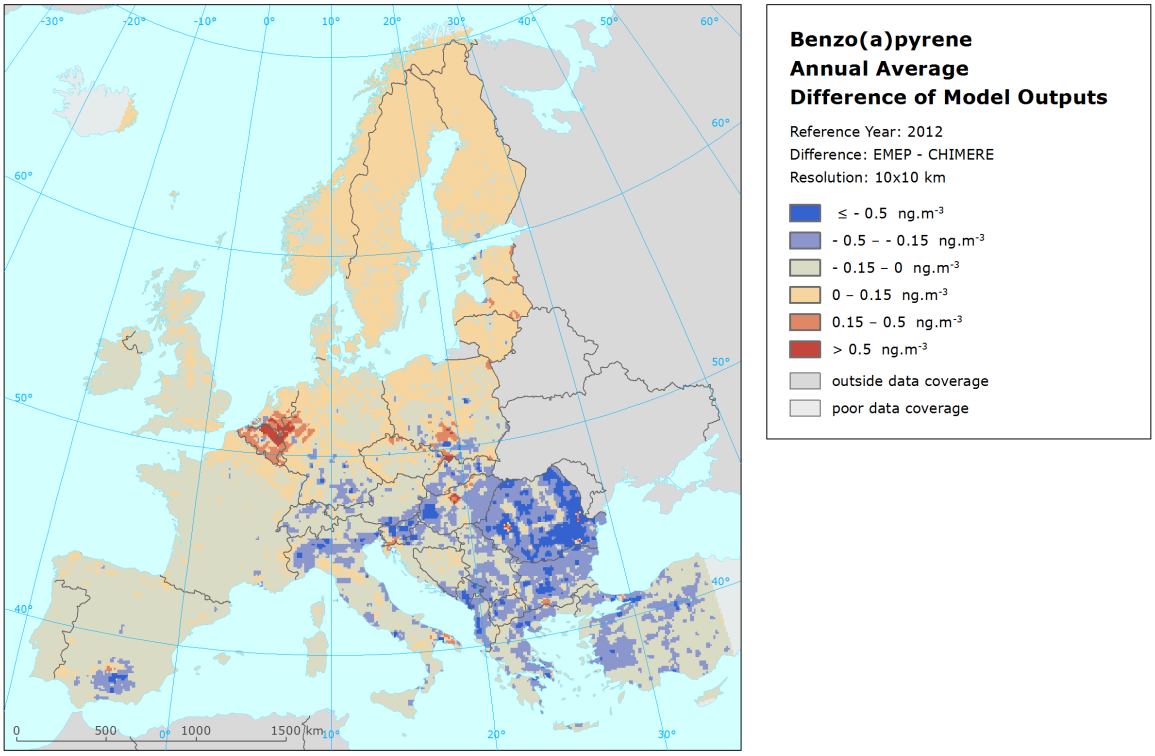

Figure A5.2: ***Difference of the EMEP and CHIMERE model outputs. BaP, annual average, 2012. Units: ng.m^{-3}.***