3. CASE STUDY OF B(a)P POLLUTION IN SPAIN AND FRANCE

This chapter summarizes information on current progress in the case study of B(a)P pollution in the selected EMEP countries. Country-specific studies, performed by MSC-E in co-operation with national experts, represent an important activity that can contribute to further improvement of POP pollution assessment in the EMEP region [*Travnikov et al.*, 2018]. The B(a)P case study was initiated for Spain following the recommendation of the 2nd joint session of the Working Group on Effects and the Steering Body to EMEP. Objectives of the study include:

- Collection of detailed national scale information on emissions and monitoring data;

- Fine resolution modelling of B(a)P pollution levels using GLEMOS and CHIMERE model;

- Evaluation of modelling results against measurements of EMEP and national monitoring stations;

- Model simulations of B(a)P pollution using different scenarios of B(a)P emissions;

- Analysis of model predictions sensitivity to application of different parameterizations of most important processes affecting B(a)P long-range transport and removal from the atmosphere;

- Source-receptor and sector-specific modelling of B(a)P pollution on national scale.

The outcome of initial stage of the B(a)P case study is described in the previous POP Status Report [*Gusev et al.*, 2017]. This year evaluation of national scale B(a)P pollution levels in Spain is continued. In addition, similar work is initiated for France. Main emphasis at the current stage is given to the analysis of discrepancies between B(a)P modelling results and observed pollution levels taking place for Spain and France as well as for some other EMEP countries. This activity includes construction of experimental emission scenarios in order to evaluate sensitivity of model predictions to the uncertainties in the emissions data. Analysis of scenario modelling results can indicate areas of elevated uncertainties and characteristics of emissions that require further refinement. Particular attention is also paid to the effect of possible uncertainties in the applied modelling approach. Thus, different model parameterizations for the evaluation of B(a)P degradation and gas-particle partitioning are planned to be considered and tested. Model simulations and their analysis are performed in close cooperation with national experts in modelling of B(a)P pollution from Spain and France.

3.1. Model domains and emission data for modelling

Modelling of B(a)P pollution levels for this case study is carried out using nesting approach. The configuration of three modelling domains applied in the study is shown in Fig. 3.1. It consists of the coarse domain EU02 with spatial resolution 0.2°x0.2° and two nested domains FR005 and SP005 with spatial resolution 0.05°x0.05°. The year 2015 was selected as a reference year for model simulations.

Emissions for B(a)P modelling were prepared on the basis of the data from different sources. In particular, fine resolution gridded B(a)P emissions for Spain and France were provided by national experts of these countries. In case of Spain it was national inventory of PAH emission with spatial resolution 0.05°x0.05°. To obtain B(a)P emissions from the PAH emission data a scaling factor 0.14 was

used following the information from national experts of Spain, with reference to the EMEP/EEA Guidebook 2016 [*EMEP/EEA Guidebook*, 2016]. In case of France national inventory of B(a)P emission with spatial resolution 0.125°x0.0625° was applied. Both inventories for Spain and France were based on the SNAP sectoral distribution. For other EMEP countries, covered by the defined modelling domains, gridded data on annual B(a)P emissions with spatial resolution 0.1°x0.1° were obtained from CEIP. To compile consistent emission dataset for modelling, gridded data of CEIP, defined for the NFR sectors, were transformed into the SNAP sectors.
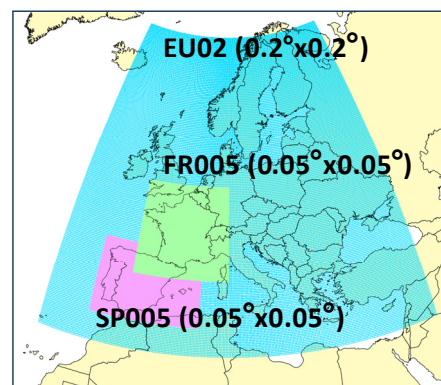

Fig. 3.1. *Domains for model simulations: EU02, FR005, and SP005.*

Spatial distribution of annual B(a)P emissions from anthropogenic sources for 2015within the three modelling domains is illustrated in Fig 3.2.
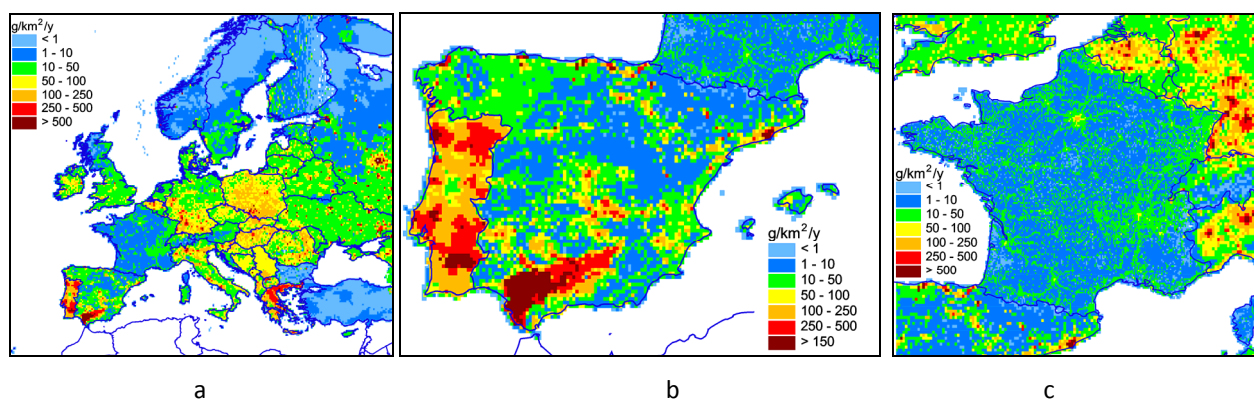

Fig. 3.2. *Spatial distribution of annual B(a)P emission fluxes in 2015 for the EU02 domain (a), SP005 domain (b), and FR005 domain (c), g/km^2/y*

3.2. Preliminary modelling results and their analysis

Preliminary model simulations of B(a)P pollution levels were performed for the coarse resolution domain EU02 using the GLEMOS model and the CHIMERE model [*Menut et al.*, 2013]. Both models have in general similar description of main processes governing B(a)P transport in the atmosphere, in particular, degradation in air, dry deposition of B(a)P in particulate phase, and scavenging with precipitation. At the same time, the models have different parameterizations of gas-particle partitioning. Besides, the CHIMERE model has no parameterization of air-surface gaseous exchange for B(a)P. Thus, these differences can noticeably affect model predictions of B(a)P pollution levels.

Figure 3.3 presents annual mean B(a)P air concentrations in the EU02 domain simulated by the two models for 2015. In general, both models predicted close pattern of B(a)P pollution levels in the European region. The highest B(a)P air concentrations (about 0.6 ng/m^3 and higher) were estimated for the countries of Central, Southern, and Eastern Europe (e.g. Poland, Slovakia, Hungary, Romania, Serbia,

Italy, and Greece), which in some areas exceeded the EU target level (1 ng/m^3). High levels of pollution were also predicted for southern Spain, Portugal, and some urban areas of Russia. B(a)P concentrations below the WHO reference level 0.12 ng/m^3 were estimated for Scandinavian countries, Denmark, Estonia, the UK, France, and northern Spain.

The spatial pattern of modelled B(a)P air concentrations can be compared with the outcome of mapping of ambient B(a)P concentrations in Europe [*Guerreiro et al.*, 2016]. Model estimates of B(a)P concentrations differ from the results of mapping in some areas of European region. In particular, higher B(a)P concentrations, in comparison to the predictions of the GLEMOS and CHIMERE models, were estimated by ETC/ACM mapping for Poland and France, and lower concentrations for Spain, Portugal, and Germany. The methodology applied for mapping was based on linear regression followed by kriging of the residuals and is largely driven by the observations. Therefore these differences may be explained by uncertainties of national emission inventories used for modelling as well as deficiencies of modelling approaches.
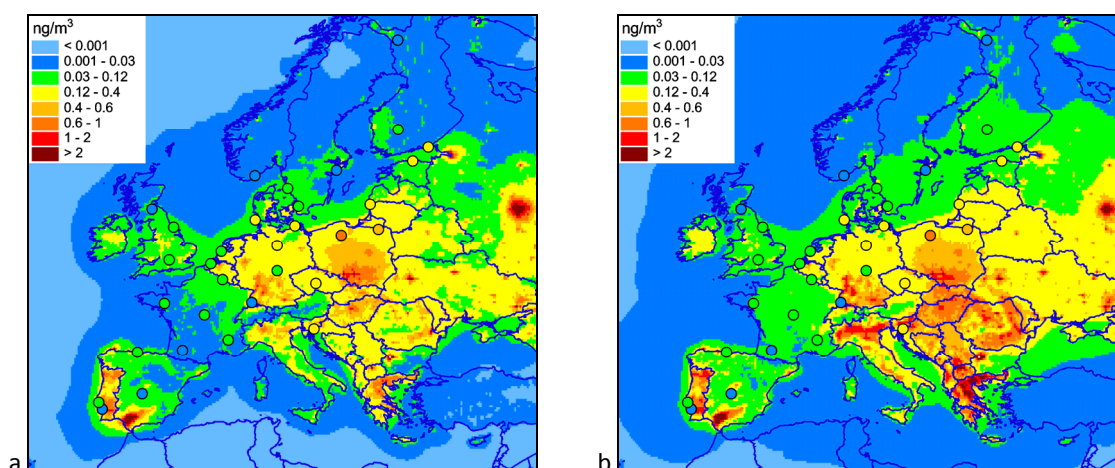

Fig. 3.3. *Annual mean B(a)P air concentrations (ng/m^3) in 2015 simulated by the GLEMOS (a) and CHIMERE (b) models for the EU02 domain 0.2°x0.2°; and B(a)P concentrations, observed at the EMEP monitoring sites, overlaid as coloured circles in the same scale as modelled values.*

Comparison of model predictions made by the two models showed that B(a)P concentrations simulated by CHIMERE were somewhat higher than the concentrations of GLEMOS. Lower levels of concentrations in the GLEMOS results can be attributed to the difference in the parameterizations applied for B(a)P modelling. As it was mentioned above, mechanisms of gas-particulate partitioning were different in the two models and parameterization of air-surface gaseous exchange was not included in the CHIMERE model.

Evaluation of model results against measurements was carried out using the data of EMEP monitoring network for 2015. Summary of statistical analysis of the agreement between modelled and observed B(a)P air concentrations is presented in Table 3.1. Annual mean measured B(a)P concentrations of 28 EMEP monitoring sites were selected for the comparison. As shown in the Table, statistical metrics, calculated for the GLEMOS and CHIMERE model simulation results, are quite comparable in terms of bias (NMB), correlation (R), and errors (RMSE). Both models tend to under-predict observed B(a)P air concentrations by 25-27%. Geographical pattern of observed B(a)P concentrations is generally

reproduced by the models, which is indicated by the spatial correlation coefficients equal to 0.6-0.7. However, for more than a half of monitoring sites (~60%) B(a)P concentrations, calculated by GLEMOS and CHIMERE, deviate from measurements more than a factor of 2.

Table 3.1. *Summary of model performance metrics, calculated for annual mean modelled B(a)P air concentrations of GLEMOS and CHIMERE and measurements of EMEP monitoring stations for 2015.*

Model	NMB[a] (%)	R[a]	RMSE[a] (ng/m^3)	F2[a] (%)	F3[a] (%)
GLEMOS	-27	0.61	0.13	43	68
CHIMERE	-25	0.69	0.12	36	82

[a] NMB is normalized mean bias; R is the spatial correlation between modelled and observed concentrations; RMSE is the root mean square error; F2 and F3 represent fractions of sites for which deviation between modelled and observed values are within a factor of 2 and 3, respectively.

In Figure 3.4 comparison of modelled and observed annual mean B(a)P air concentrations is shown for individual EMEP monitoring sites. The figure illustrates results of the comparison for the two groups, namely, for monitoring sites where deviations between the modelled and observed concentrations are within a factor of 2 (Fig. 3.4a), and where they are higher than a factor of 2 (Fig.3.4b). Model results for the first group of sites are characterized by high level of correlation with measurements (more than 0.9 for both models). The second group includes the sites for which overestimation or underestimation of measured values exceeded a factor of 2. In particular, the models tended to over-predict annual mean B(a)P concentrations observed in Belgium, the Netherlands, southern Germany, northern France, and Portugal. At the same time, B(a)P air concentrations in northern Germany, Estonia, Latvia, Finland, and Poland are underestimated by the models. Besides, the spatial correlation of modelled and measured concentrations for this group of sites is low.
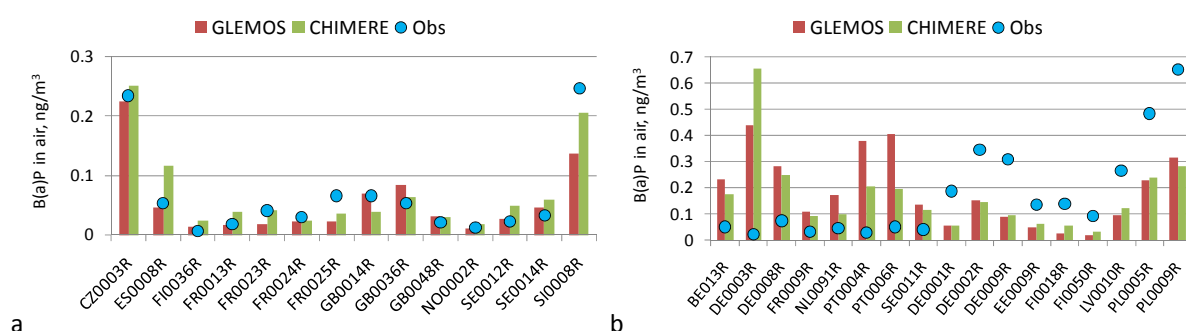

Fig. 3.4. *GLEMOS and CHIMERE modelling results for 2015 against the annual mean B(a)P air concentrations, observed at EMEP monitoring sites. Left diagram (a) is for the sites where deviations between the modelled and observed values are within a factor of 2, and right diagram (b) for the sites where deviations for both models exceed a factor of 2.*

Results of preliminary model simulations, performed by the GLEMOS and CHIMERE models, indicate significant discrepancies between the modelled and observed B(a)P air concentrations for some of the monitoring sites in Belgium, the Netherlands, Germany, Poland, Portugal, and Spain. Similar deviations with measurements were earlier revealed by the analysis of B(a)P modelling results for 2015 (POP Status Report [*Gusev et al.*, 2017]) as well as of modelling results for 2016 (Chapter 2 of this

report). *These deviations may be explained by the effect of several factors. As it was discussed in the Chapter 2, uncertainties in national emission inventories of the EMEP countries as well as uncertainties in the model parameterizations applied for gas-particle partitioning and degradation of B(a)P can lead to over or under-predictions of observed concentrations by the models.* In particular, a number of recent studies for PAHs suggested that phase partitioning of PAHs may not follow the assumption of instantaneous equilibrium between the gaseous and particle phases (that is applied in currently used parameterization in the GLEMOS model). This process can be influenced by secondary organic aerosols, which might protect particle-phase PAHs from degradation and evaporation [*Zelenyuk et al.*, 2012; *Friedman et al.*, 2014]. Furthermore, it was shown that considering the effects of temperature and humidity variations in the atmosphere on multiphase degradation of PAHs is important for better predicting spatial variability of air concentrations and long-range transport [*Mu et al.*, 2018]. The influence of these factors on model predictions requires more detailed analysis that is planned to be carried out using GLEMOS and CHIMERE models in the framework of this B(a)P case study.

3.3. Experimental scenario of B(a)P emissions in the selected EMEP countries

To explore sensitivity of modelling results to possible uncertainties in the emission inventories of the EMEP countries, experimental model simulations with the scenario of B(a)P emissions were carried out. A simple emission scenario was constructed on the basis of scaling of emissions from particular source categories of selected countries. Two emission source categories, namely, the 'Residential combustion' and 'Field burning of agricultural residues', associated with the most significant level of uncertainties, were considered for the scaling. Changes of emissions were performed for the countries where the most significant deviations between the modelled and observed B(a)P air concentrations were found.

The scaling factors, defined for the selected countries, are given in the Table 3.2. The scenario assumes a factor of 0.5 change of B(a)P emissions from the 'Residential combustion' sector for Belgium, the Netherlands, and Germany following the average difference between the modelled and measured B(a)P air concentrations. In case of Spain and Portugal, emissions from agriculture sector were scaled by a factor of 0.2 and 0.4, respectively, following the ratio of officially reported and TNO emissions [*Denier van der Gon et al.*, 2011b]. Emissions from the 'Residential combustion' sector of Poland and France were increased by a factor of 4 and 3, respectively, following the ratio of officially reported and TNO emissions for Poland, and in accordance with the suggestion of national experts for France.

Table 3.2. Definition of experimental B(a)P emission scenario for selected EMEP countries

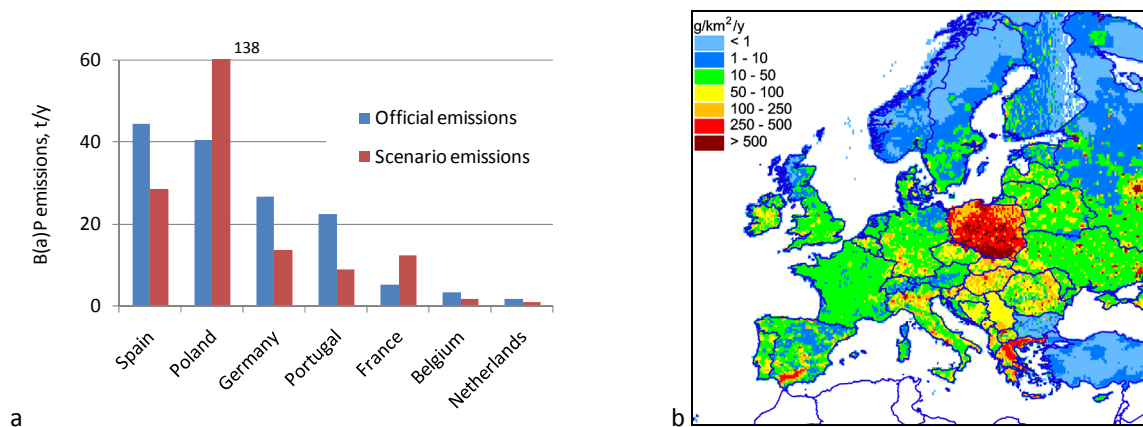| Country	Emission source category	Emission scale factor
Belgium	Residential combustion	0.5
The Netherlands	Residential combustion	0.5
Germany	Residential combustion	0.5
France	Residential combustion	3
Poland	Residential combustion	4
Spain	Field burning of agricultural residues	0.2
Portugal	Field burning of agricultural residues (3F)	0.4

Fig. 3.5. *Changes of national emissions (t/y) of selected countries in the experimental scenario of B(a)P emissions (a) and spatial distribution of annual emission fluxes of B(a)P (g/km²/y) in the experimental scenario for 2015.*

In Figure 3.5a absolute changes of annual total B(a)P emissions in the selected countries in accordance with the prepared scenario are illustrated. Spatial distribution of resulted B(a)P emission fluxes in the EU02 modelling domain is shown in Figure 3.5b. It is seen that the largest absolute change of B(a)P emissions in the scenario took place for Poland followed by Spain, Portugal, and Germany. At the same time, maximum relative changes of emissions were noted for Poland and France.

Annual mean B(a)P air concentrations, simulated by the GLEMOS and CHIMERE models using experimental emission scenario, and measured concentrations of the AIRBASE background rural and remote monitoring stations are presented in Figure 3.6. Results of reference model runs of the two models, carried out on the basis of officially reported B(a)P emissions, are described in the previous section. Evaluation of the effect of emission scaling on modelling results was carried out using comparison of the scenario and reference model simulations with measurements of AIRBASE (Table 3.3). The Table shows noticeable improvement of all statistical metrics, calculated on the basis of annual mean modelled and observed values.
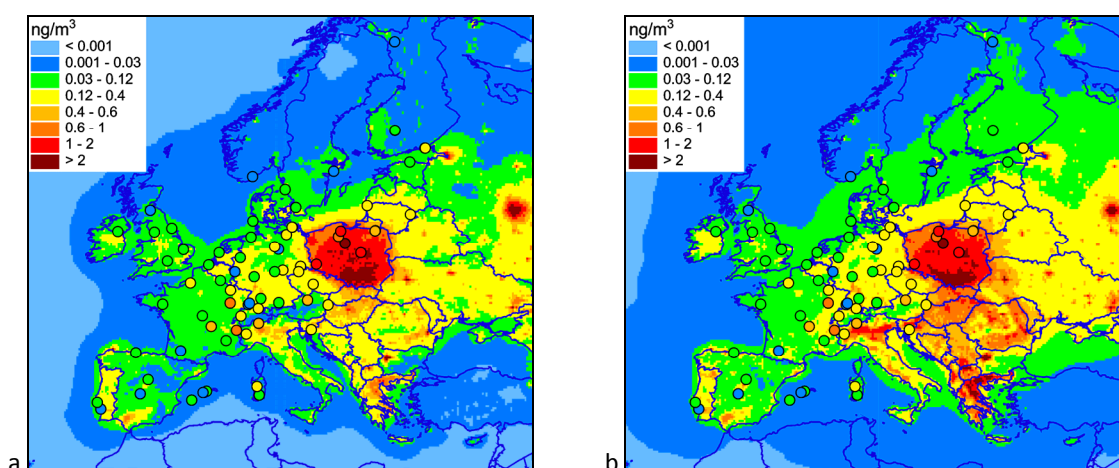

Fig. 3.6. *Annual mean B(a)P air concentrations (ng/m³) in 2015 simulated by the GLEMOS (a) and CHIMERE (b) models for the EU02 domain 0.2°x0.2° using scenario emissions; and B(a)P concentrations observed at AIRBASE background rural and remote monitoring sites, overlaid as coloured circles in the same scale as modelled values.*

The spatial pattern of B(a)P air concentrations, simulated by GLEMOS and CHIMERE using scenario emissions, better captures the spatial distribution of observed B(a)P air concentrations comparing to reference model runs. Spatial correlation between the simulated concentrations and measurements improved from 0.43 to 0.82 for GLEMOS, and from 0.30 to 0.80 for CHIMERE. Similarly, the values of the bias and RMSE metrics considerably decreased for both models results. Reduction of deviations of model estimates from measurements is also reflected in the increased values of F2 and F3 metrics.

Table 3.3. Summary of statistical metrics, calculated on the basis of annual mean B(a)P air concentrations observed at AIRBASE background rural and remote monitoring stations (n=82) and estimated by GLEMOS and CHIMERE in the scenario and reference model runs for 2015.

Model	NMB[a] (%)	R[a]	RMSE[a] (ng/m³)	F2[a] (%)	F3[a] (%)
Reference model run with official emissions					
GLEMOS	-39	0.43	0.41	39	59
CHIMERE	-28	0.30	0.43	45	70
Scenario model run with scaled emissions					
GLEMOS	-12	0.82	0.25	51	72
CHIMERE	-2	0.80	0.27	59	83

[a] *NMB is normalized mean bias; R is the spatial correlation between modelled and observed concentrations; RMSE is the root mean square error; F2 and F3 represent fractions of sites for which deviation between modelled and observed values are within a factor of 2 and 3, respectively.*

Model simulations, performed by GLEMOS and CHIMERE using scenario emissions, showed noticeable improvement of agreement between predicted and observed B(a)P air concentrations in European countries. At the same time, though the level of the disagreement is generally decreased, for some of the monitoring sites, located in Austria, Finland, Germany, and France, the deviations remained significant. It should be noted that this simple scenario was based on the scaling of total sectoral emissions, and did not take into account uncertainties of spatial distribution of emissions that could substantially affect model performance for individual monitoring sites.

These results may indicate that officially reported B(a)P emissions of some of the EMEP countries are subject of significant uncertainties that lead to substantial deviations between the model estimates and observations. To improve the quality of model assessment of B(a)P pollution levels, further refinement of national emission inventories with respect to estimates of total emissions and their spatial distribution is appreciated.

3.4. Analysis of B(a)P emissions from agricultural sources in Spain

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Burning of agricultural residues can be important source of toxic pollutants (e.g. PAHs, PCDD/Fs) releases to the atmosphere [*Viana et al.*, 2008; *Gonçalves et al.*, 2011]. In spite of restrictions, introduced by many European countries for open burning of agricultural wastes, this activity still takes place in southern Europe due to permissions given by local and regional authorities [*AIRUSE*, 2016]. According to the national emission inventory of Spain, emissions from combustion of crop residues accounted for about 45% of national total PAH emitted in 2015 [*IIR*, 2017]. This emission source category considers the burning of herbaceous agricultural cotton residues[5], which are, after the harvest, spread but not fixed to land, and stored in a small area for a local burning. Although burning of cotton residues has significantly decreased during the recent years, emissions from this activity are still important since Spain produces ~21% of the European Union's cotton (being the second largest producer after Greece).

Evaluation of emissions from the combustion of agricultural residues to the atmosphere represents difficulties due to dependency on many factors (e.g. type of residue, its moisture content, conditions of combustion, and meteorological conditions) [*Oanh et al.*, 2011; *Sanchis et al.*, 2014; *Gonçalves et al.*, 2011]. The methodology currently used for this purpose contains some uncertainties that can lead to overestimation of PAH emissions from agricultural activities in southern part of Spain (Andalusia). In this section a summary of information on applied methodology, its uncertainties, and results of test model simulations carried out on the basis of national PAH emission inventory and experimental modelling scenarios, is provided.

Estimation of B(a)P emissions from agricultural sources

To estimate emissions from agricultural sources, the Tier 1 methodology and default emission factors of the EMEP/EEA air pollutant emission inventory guidebook [EMEP/EEA Guidebook, 2016] were applied. The Tier 1 methodology optionally points to the generic approach of the IPCC Guidelines [*IPCC Guidelines*, 2006], which was developed to quantify the amount of burned residues and emissions resulting from biomass burning. According to this approach pollutant emission to the atmosphere from burning of agricultural residues can be evaluated using the following equation:

$$L_{fire} = A \times M_b \times C_f \times G_{ef} \times 10^{-3}$$

where:

L_{fire} the amount of pollutant gaseous emission due to combustion of residues (tons);

A the burnt area (ha);

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[5] According to the SNAP nomenclature this source category relates to SNAP 10 (Agriculture), subcategory 10.3.5, *burning of herbaceous agricultural crop residue*, and in the NFR nomenclature to the source category 3.F, *field burning of agricultural residues*.

This area is calculated based on the cultivated area, multiplied by the burned fraction specific for each crop type. For cotton a value of 0.333 was used (following the [MAPAMA, 2015]).

M_b the specific mass of residues available for combustion (tons ha^{-1});

C_f the combustion factor defined according to default values of IPCC methodology [*IPCC Guidelines*, 2006] (dimensionless);

The combustion factor represents a measure of the proportion of biomass fuel that is actually combusted. This value depends on the size and architecture of the fuel load, the moisture content of the fuel as well as on fire intensity and rate of spread.

The burnt dry combustible mass is calculated as $M_B * C_f$ and values of this product were taken from [*IPCC Guidelines*, 2006]. For the calculation of PAH emissions the value for the aggregated vegetation type '*All Shrublands*', equal to 14.3, was selected.

G_{ef} the emission factor for considered pollutant (kg/kg of burnt dry matter).

The Tier 1 default emission factors for NFR source category 3.F [*EMEP/EEA Guidebook,* 2016] were used for PAHs, which values were based on the study [*Jenkins et al.*, 1996].

Analysis of this methodology, applied for the evaluation of PAH emissions in Spain, indicates that several parameters of this approach can be subject of considerable uncertainties. In particular:

1. The area of burning (A), calculated as a function of the cultivated area, can be overestimated. During the recent years part of the cultivated area in Andalucía has been assigned to an integrated production system, for which the legislation does not allow residues burning. Therefore, calculation of PAH emissions without taking this into account would lead to the overestimation of actual emissions.

2. The value of the burnt dry combustible mass ($M_B * C_f$), selected from the default IPCC estimates [*IPCC Guidelines*, 2006], is evaluated for the aggregated vegetation type and is not specifically determined for the cotton residues. At the same time, there are estimates for several crops in these guidelines, namely, for wheat, maize, rice, and sugarcane residues. Thus, their use might be more appropriate for the evaluation of this parameter for cotton.

3. Values of default emission factors (G_{ef}) for PAHs in the EMEP/EEA guidebook [EMEP/EEA Guidebook, 2016] are not specific for cotton (for example, for benzo(a)pyrene the value of 67.7 mg/kg dry matter is suggested following [*Jenkins et al.*, 1996] which was estimated for other types of crops).

Experimental emission scenarios and results of model simulations

To examine possible effects of the uncertainties mentioned above on model predictions of B(a)P pollution levels, several experimental emission scenarios were prepared. These scenarios are based on the official PAH emission data for 2015, provided by the Spanish Ministry of the Environment. Inventory of PAH emissions is generated for the sum of 4 PAHs without splitting for particular PAH compounds. Therefore, to obtain emissions of B(a)P, its fraction in the emissions of 4 PAHs was taken equal to 14%. This assumption was made in accordance with the information from national experts on emissions, with

reference to the EMEP/EEA Guidebook 2016. The following modifications of B(a)P emissions were considered in the scenarios:

- Scenario 1: Modification of the spatial distribution of emissions applying a spatial mask for areas assigned to the integrated production system. For these areas no burning of residues is expected, so the emissions in the corresponding grid cells are removed, which reduces total emission from agricultural sources by 62%. In this scenario both the spatial distribution and total emission from agriculture sector are changed.
- Scenario 2: Modification of the fraction of burned area for cotton (A). About 85% of fields for cotton are now part of an integrated production system, where no burning is allowed. Thus, the fraction of burned area can be reduced from 0.333 to 0.15, which leads to 55% reduction of emissions from agricultural sources. This scenario assumes changing of total emission from agriculture sector, while spatial distribution is left the same as it is defined in the national emissions inventory.
- Scenario 3: Modification of the value for burnt dry combustible mass ($M_B * C_f$). Though there is no specific value for burning of cotton residues, the IPCC Guidelines provide the estimates for burning of wheat, maize, rice, and sugarcane crop residues. The average value of these estimates, equal to 6.5, can be used for the evaluation of emissions from agricultural sources (instead of the chosen value 14.3), which equates to a 58% emission reduction.

Preliminary model simulations of B(a)P pollution in Spain were performed for the first scenario with the two air quality models, namely, EMEP GLEMOS and CHIMERE (v2013, [*Menut et al.*, 2013]) models. Modelling of B(a)P pollution using the GLEMOS model was carried out for the year 2015. In case of the CHIMERE model the simulations were made with meteorological data for the year 2017 due to availability of necessary resources. For the evaluation of the impact of emission modifications, model predictions of Scenario 1 (SC1) simulations were compared with the base case (BC) simulations. Besides, level of agreement between modelled and observed B(a)P concentrations was evaluated for both sets of modelling results (BC and SC1). Measurements of 12 monitoring sites in Andalusia of different types, including background urban and suburban, industrial, and traffic sites, were selected for the comparison. The GLEMOS and CHIMERE models are not specifically designed for the evaluation of pollution in urban and industrial areas. However, due to the lack of background rural and remote sites in this area, other types of sites (e.g. urban, industrial, and traffic) were also considered in the comparison for the analysis of spatial distribution of pollution.

Spatial distributions of annual mean B(a)P air concentrations for the BC and SC1 simulations, carried out using the CHIMERE model, are shown in the Fig.3.7. Similar results of the GLEMOS model are presented in the Fig.3.8. Model predictions of the CHIMERE model are slightly higher comparing to the results of the GLEMOS model, which can be explained by several reasons. The CHIMERE model, applied in for these simulations, did not consider degradation of B(a)P by ozone. Furthermore, the GLEMOS model includes parameterization of B(a)P gaseous exchange with underlying surface, whereas in the CHIMERE model this process was not considered. Besides, some differences in modelling results can be due to different model setups (e.g. meteorological and geophysical input data). In spite of this, both models showed similar patterns of B(a)P air concentrations.
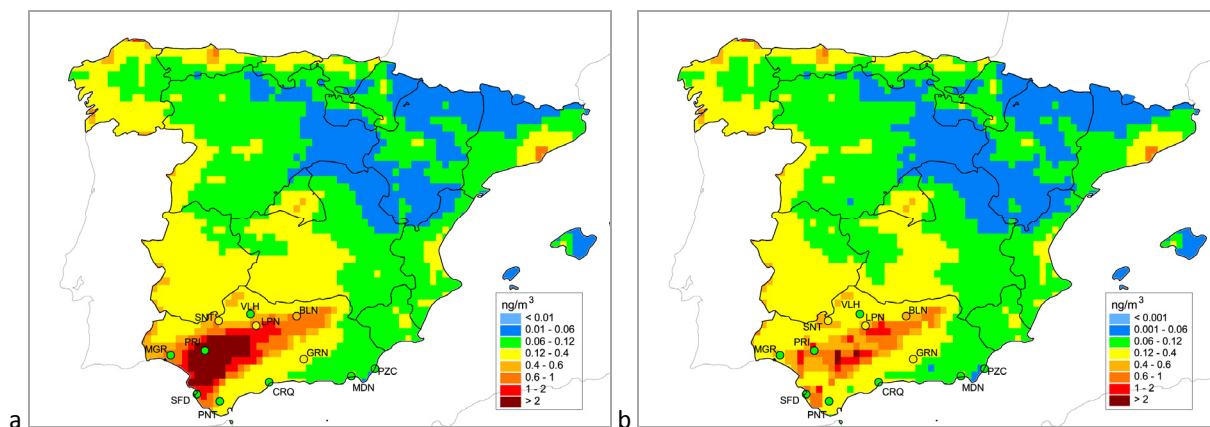
Fig. 3.7. *Annual mean B(a)P air concentrations (ng/m³) in 2017 obtained by the CHIMERE model in the BC and SC1 simulations. B(a)P air concentrations observed at monitoring sites[6] in Andalusia in 2017, are overlaid as coloured circles in the same scale as modelled values.*
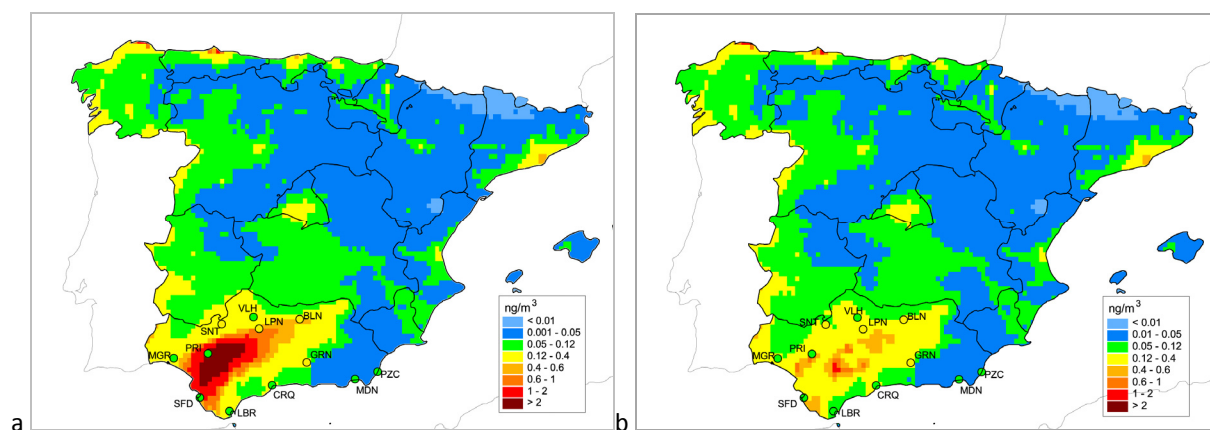

Fig. 3.8. *Annual mean B(a)P air concentrations (ng/m³) in 2015 obtained by the GLEMOS model in the BC and SC1 simulations. B(a)P air concentrations observed at monitoring sites[6] in Andalusia in 2015, are overlaid as coloured circles in the same scale as modelled values.*

In Table 3.4 statistical indicators of the agreement between model predictions of B(a)P air concentrations and measurements are presented. The GLEMOS and CHIMERE models significantly over-predicted observed B(a)P concentrations in the BC simulations. Besides, the spatial correlation between the modelled and measured concentrations was low. The over-prediction can be attributed to possible overestimation of B(a)P emissions from agriculture sector, which is dominating source in this area according to the national emissions inventory. Low correlation might indicate possible inaccuracies in the spatial allocation of emissions.

In case of SC1 simulations both models show substantial decrease of modelled B(a)P air concentrations due to reduction of the burning area and consequently lower emissions from the combustion of cotton residues. The values of average bias (MFB) and error (MFE) decreased comparing to the BC simulations.

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[6]   *Monitoring sites: Moguer (MGR), Principes (PRI) , Sierra Norte (SNT), Villaharta (VLH), Lepanto (LPN), Bailin (BLN), San Fernando (SFD), Puente M (PNT), Los Barrios (LBR), Carranque (CRQ), Mediterraneo (MDN), Pza del Castillo (PZC), Granada Norte (GRN).*

Table 3.4. *Statistics of the comparison of GLEMOS and CHIMERE modelling results with measurements of B(a)P air concentrations made at 12 monitoring sites in Andalucía in 2015 and 2017*

Model (scenario)	Year	Observed, ng/m^3	Modelled, ng/m^3	MFB[a], %	MFE[a], %	Correlation
CHIMERE (BC)	2017	0.119	0.718	123	123	0.02
CHIMERE (SC1)	2017	0.119	0.370	106	108	0.25
GLEMOS (BC)	2015	0.125	0.255	39	109	-0.05
GLEMOS (SC1)	2015	0.125	0.134	11	96	-0.01

[a] MFB is mean fractional bias; and MFE is mean fractional error following [*Boylan and Russell*, 2006].

At the same time, changes in spatial distribution of agricultural emissions did not lead to noticeable improvements of spatial correlation between model predictions and measurements. In particular, in the case of the CHIMERE SC1 simulations the spatial correlation was only slightly increased, while no correlation was obtained in the GLEMOS BC and SC1 simulations.

In Fig.3.9 comparison of monthly mean modelled and observed B(a)P air concentrations is illustrated on the example of GLEMOS modelling results for three monitoring sites Principes, Lepanto, and Moguer. The sites are located quite close to the area of cotton agricultural activities. Besides, model predictions for their locations demonstrated the most pronounced response to the changes of emissions in the scenario SC1. It is seen that despite significant decrease of modelled values, the overestimation of observed concentrations for these sites is still significant.
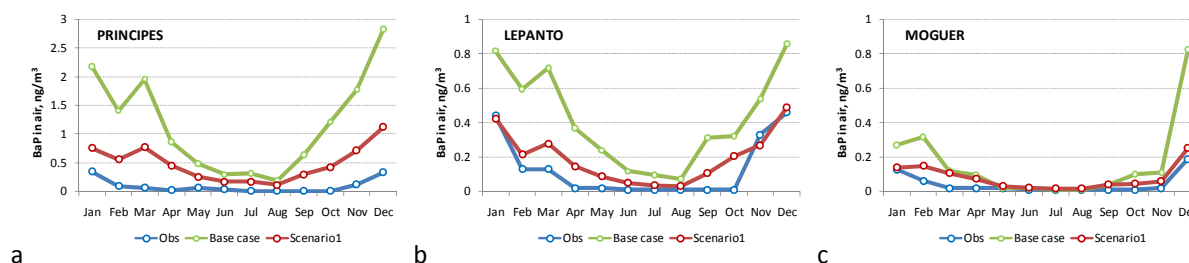

Fig. 3.9. *Modelling results of GLEMOS model with base case and scenario emissions.*

Concluding, it can be noted that high B(a)P concentrations, modelled with both GLEMOS and CHIMERE in southern Spain (Andalucía), might be directly related to the high emissions from burning of herbaceous crop residues in agriculture. Emission scenario assuming reduction of the burning area for cotton residues permitted to improve the agreement between model predictions and measurements for both models. At the same time, the overestimation of observed B(a)P air concentrations for some of the monitoring sites in the scenario model simulations is still significant. Besides, there is a lack of spatial correlation between the simulated and measured concentrations. Thus, *analysis of B(a)P pollution levels in this area needs to be continued with application of additional emission scenarios. Furthermore, results of test model simulations indicate the need of refinement of methodology applied to the evaluation of the emissions from burning of agricultural residues in Spain.*