Anonymous Referee #2

General comments

The manuscript describes the application of community air quality model CMAQ to study atmospheric transport of B(a)P and effects of gas-particle partitioning and degra- dation. Several gas-particle partitioning models were implemented in the CMAQ model extending its capabilities with respect to modeling of PAHs. Though the subject of the study is of importance with regard to existing air quality problems in Europe, description of this study suffers from obscurity and thus requires substantial revisions.

First of all, I would mention not clear enough description of parameters applied in the equations 10 and 11, related to gas-particle partitioning (e.g. KOA, KSA). Secondly, evaluation of the effects of several GPP mechanisms through their incremental testing (incremental testing is defined in section 3.2 and Table 2) is not quite clearly described with regard to sensitivity of CMAQ model output to particular partitioning mechanisms in model simulation scenarios. In some cases abbreviations, defined for GPP models and scenarios and used throughout the analysis, are mixed like e.g. DE model and DE scenario. This leads to problems with understanding what is presented, for example, in the second column of tables 4 and S8 called 'Model'. There are both abbreviations for scenarios and models (for example, JP-W and DE, HB, etc.). Finally, the underestimation of observed B(a)P concentrations in air is explained by low emissions. At the same time, there is no discussion of potential problems of emission data used, no comparison of total annual emissions with other studies, for example, with similar studies e.g. Aulinger et al. (2007) and others.

Reply:
We appreciate the very thorough and detailed review along with the recognition of the potential relevance of the presented study. The first two points of concern have been addressed in a revised manuscript. Regarding the last area of concern, we feel that the manuscript adequately discusses the limitations of emission data used, which have been the major topic of work published by the authors (Bieser et al., 2012).

Specific comments

Page 3, line 12-13: "CMAQ contains modules representing advection, eddy diffusion, in-cloud, and precipitation processes". It would be better to use 'in cloud and below- cloud scavenging with precipitation'.

Reply: Corrected

Page 3, line 26: 'gas phase reactions' instead of 'gas reactions'.

Reply: Corrected

Page 4, line 23: Though the equation 2 follows the publication of Aulinger et al. (2007), it is not clear how the particulate fractions in each mode i, fi, are obtained, because in the cited work (Cooter and Hutzell, 2002) similar equation is written for the sum of partition coefficients, but not for fractions of a compound in particulate phase.

Reply: According to equations 1-4 in Cooter and Hutzell (2002), particulate fractions are defined for each mode i of CMAQ aerosol.

Page 5, equation 7: fOM is used without index i. Does it mean that the fraction of organic matter in aerosol particles is the same in each of three modes?

Reply: Corrected throughout the text

Page 6, line 3: assumptions used here for ratio of activity coefficients and ratio of mean molar weights of organic matter of the particles and octanol (that they are equal unity) need to be discussed with regard to their uncertainties as it was shown in e.g. (doi: 10.5194/acpd-14-21341-2014).

Reply: More explanations will be added. The new text reads (after line 7, page 6): "Assuming that octanol imitates organic matter in PM, Harner and Bidleman (1998) suggested that the ratio of γ_{oct}/γ_{OM} and M_{oct}/M_{OM} can be assumed to be 1. However, it was later suggested that M_{OM} could be much higher, particularly in secondary organic aerosols containing polymeric structures (Kalberer et al., 2004); a mean value of 500 g mol^{-1} was later suggested by Götz et al. (2007), which results in M_{oct}/M_{OM} of 0.26."

Götz, C. W., Scheringer, M., MacLeod, M., Roth, C. M. and Hungerbühler, K.: Alternative approaches for modeling gas−particle partitioning of semivolatile organic chemicals: model development and comparison, Environ. Sci. Technol., 41(4), 1272–1278, doi:10.1021/es060583y, 2007.
Kalberer, M., Paulsen, D., Sax, M., Steinbacher, M., Dommen, J., Prevot, A. S. H., Fisseha, R., Weingartner, E., Frankevich, V., Zenobi, R. and Baltensperger, U.: Identification of polymers as major components of atmospheric organic aerosols, Science, 303(5664), 1659–1662, doi:10.1126/science.1092185, 2004.

Page 6, line 14-15: Soot-air partition coefficients were calculated as the ratio of soot water adsorption constants KSW and the inverse Henry's Law constant (HC), with KSA values adopted from Bärring et al. (2002). Concerning the way of deriving the KSA it would be important to provide estimates of uncertainties that would be introduced by this assumption. Moreover, it is unclear how this adopting was performed for B(a)P since the publication of Bärring et al. (2002) was focused on experimental determination of the soot–water distribution coefficients for PCDDs, PCDFs, and PBDEs.

Reply: K_{sa} was calculated based on K_{sw} (typo fixed) following the approach described in the footnotes of Table 1 in Lohmann and Lammel (2004). Despite the publication title not mentioning PAHs, this value is also referenced in Table 4 of Bärring et al. (2002). This information will be included in Table 1 for clarity. We agree that the method applied is imperfect, but it is the best available and, hence, commonly accepted in atmospheric modelling (as cited by the referee, Galarneau et al., 2014, and others). The only other option available (suggested by Dachs et al., 2004, and van Noort, 2003) parameterises $K_{soot-air}$ as a function of the BC specific surface area, which is basically unknown and certainly transient while aging (typically changing by at least one order of magnitude within one day following emission from combustion sources). Dachs concludes that prediction of K_{sa} by either method provide reasonably similar results.

New text in the revised version (following a suggestion of the other reviewer) reads (after line 24, page 6):
"This method is subject to uncertainties (Goss, 2004), but is accepted and suitable (Dachs et al., 2004; besides others)."

van Noort, P.: A thermodynamics-based estimation model for adsorption of organic compounds by carbonaceous materials in environmental sorbents, Environ. Toxicol. Chem., 22(6), 1179–1188, doi:10.1002/etc.5620220601, 2003.
Dachs, J., Ribes, S., van Drooge, B., Grimalt, J., Eisenreich, S. J. and Gustafsson, Ö.: Response to the comment on "Influence of soot carbon on the soil−air partitioning of polycyclic aromatic hydrocarbons," Environ. Sci. Technol., 38(5), 1624–1625, doi:10.1021/es0307118, 2004.
Goss, K.-U.: Comment on "Influence of soot carbon on the soil−air partitioning of polycyclic aromatic hydrocarbon," Environ. Sci. Technol., 38(5), 1622–1623, doi:10.1021/es0301370, 2004.

Page 6, line 16: There would be a need to describe more clearly these two parameterizations for K_{oa} (e.g. to give equations, to show difference).

Reply: The two K_{oa} parameterisations are described in the cited publications. Previous modelling studies of B(a)P followed either Beyer et al. (Aulinger, 2007) or Odabasi et al. (Galarneau, 2014). The relevant text was amended to explicitly state that Odabasi et al. determine K_{oa} as a function of temperature.

Page 10, line 6: It would be better to provide the difference (relative or absolute) between the maps in Figures 2a and 2b instead of direct comparison.

Reply: The goal of Figure 2 is to provide an illustation of B(a)P distributions and gradients across Europe. For better clarity we selected logarithmic scale. The difference is in fact provided in Figure 3 that provides the absolute difference comparison plot, discussed in the following paragraph.

Page 21, Table 1: it is not shown from where the values for $K_{soot-water}$, $K_{soot-air}$, and OH reaction rate were taken.

Reply: Corrected

Page 22, table 2: Table 2 does not correspond to its description in section 3.2 for 4th and 5th scenarios.

Reply: Corrected

Page 27, figure 3: Please, correct the abbreviations JB-W and DL, as they were not defined earlier. The same for figure S5. Concerning the maps, it would better to show the difference between them in relative units.

Reply: Corrected the abbreviations uniformly throughout the text. However, no visual benefit when moving from absolute to relative units was noticeable in the figures, therefore omitted.

Table 4 and Table S8 do not present mean modeled and observed B(a)P concentrations. In the column ‘Model’ both abbreviations for scenarios and for GPP models are used (for example, JP-W and DE, HB, etc.). Please, provide equations or references for IOA etc.

Reply: Corrected the abbreviations throughout the text and according to previous comment. The metrics of performance have been calculated based on the openair R package (Carslaw and Ropkins, 2012) as noted in section 3.2 (equations/references can be found therein).