Anonymous Referee #1

This manuscript presents an evaluation of alternative gas-particle partitioning schemes in a modified version of the WRF-CMAQ model run for Benzo-a-pyrene (BaP). The authors compare modeled BaP concentrations within a European model domain under 5 different gas-particle partitioning schemes and also compare model results to measurement data from EMEP monitoring sites.

The novel aspect of this work seems to be announcing that the different gas-particle partitioning schemes have been incorporated into CMAQ. The more complicated schemes apparently provide the best model performance relative to measurement data, but the authors state that the disagreements between measurements and the model are likely mostly attributable to uncertainties in emission estimates. Therefore it is difficult to draw conclusions about which partitioning scheme is best based just on results reported in this paper.

We appreciate the thoughtful review and the recognition of the novelty of the approach in 3D air quality systems such as CMAQ, despite the limited spatiotemporal coverage of the EMEP network.

I have two scientific concerns about the paper. I think both are significant, but possibly #1 arises from unclear presentation in the paper.

1) I understand Table 2 and the text to say that Scenarios 3, 4 and 5 include both the Junge-Pankow adsorption model *and* the Harner-Bidleman Koa absorption model. Is that correct? If so, I think the model is perhaps adding two redundant descriptions of gas-particle partitioning.

It has been a while since I looked at the JP model, but my recollection is that it includes a fitting parameter that was derived from empirical data under the assumption that all partitioning was by adsorption to aerosol surfaces. But, the assumption that adsorption dominates could not be confirmed. Xiao and Wania (http://www.sciencedirect.com/science/article/pii/S1352231003002139) later showed that vapor pressure and Koa both describe sorption to organic matter equally well. Therefore, I think partitioning schemes that include both vapor pressure and Koa as chemical parameters are likely including redundant information, and could easily be over-fitted.

Reply:

We agree, the parameter c_J (high for strong sorption, low for weak sorption) in the JP model may not be totally free from absorptive contributions. Therefore, adding θ of JP and K_{oa} may eventually lead to overpredictions, but these should be very low (negligible), as the absorptive contribution within JP is very low. It is very low, as the fitting was done for all possible vapors, and many of these were inorganic (almost not soluble in octanol). Unfortunately, no c_J for BAP based on measurements has ever been published. However, we found recently in background aerosol of Central Europe (Kosetice 2012-13, n=162 aerosol samples, c_J derived from experimental S/V data; Shahpoury et al., 2016) that c_J for BaP is very low, namely 0.01-5.46 Pa cm (median = 0.44 Pa cm; and also low for all other PAHs, with one exception, anthracene for which c_J>17.2 Pa cm is found). This finding is not representative for all types of aerosol in the simulation, but for the European background aerosol. It supports the perception that a possible implicit absorptive contribution to PAHs' c_J must be very low i.e., negligible. Therefore, the combined partitioning according to both the JP and a K_{oa} model is justified (note that the Harner-Bidleman version of the K_{oa} model is present only in scenario 3 – Table was modified to avoid confusion).

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New text in the revised version reads (after line 13, page 5): "The empiric JP model, despite assuming adsorption to be the only relevant partitioning process, may eventually not be free from absorptive contributions (to c_J), but these should be very low. The fitting in the empiric parameterisation was done for all possible vapors, many of these not soluble in octanol. No c_J for BAP derived from ambient measurements has ever been published. However, we found recently in background aerosol of Central Europe (Kosetice 2012-13, n=162 aerosol samples, c_J derived from experimental S/V data; Shahpoury et al., 2016) that c_J for BAP is very low, namely 0.01-5.46 Pa cm (median 0.44 Pa cm), supporting the perception that a possible implicit absorptive contribution to c_J must be very low."

Shahpoury P., Lammel G., Albinet A., Sofuoglu A., Domanoğlu Y., Sofuoglu C.S., Wagner Z., Zdimal V.: Evaluation of a conceptual model for gas-particle partitioning of polycyclic aromatic hydrocarbons using poly-parameter linear free energy relationships, Environ. Sci. Technol., Submitted, 2016.

2) The authors state that soot-air partition coefficients were calculated as the ratio of soot-water adsorption constants (Ksw) and the Henry's Law constant. Kai Goss has pointed out that applying a thermodynamic triangle for a solid interface in contact with water and the same solid interface in contact with an adjacent gaseous phase is a conceptual mistake that can lead to very large errors (http://pubs.acs.org/doi/abs/10.1021/es0301370).
I actually do not see a good option for the authors to overcome this problem, since the Lohmann & Lammel (2004) and the Dachs & Eisenreich (2000) papers that are the basis for the gas-particle partitioning scheme applied in this paper are both based on thermodynamic triangles that Goss argues (convincingly!) are invalid.
Reply:
We thank the reviewer for pointing this out. 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 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.