

Interactive comment on “Can we use modelling methodologies to assess airborne benzo[a]pyrene from biomonitors? A comprehensive evaluation approach” by N. Ratola and P. Jiménez-Guerrero

Anonymous Referee #3

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General Comments This manuscript describes the use of a chemical transport model (WRF+CHIMERE) to simulate airborne benzo[a]pyrene concentrations over the Iberian Peninsula. The model was driven by emissions generated by EMEP-MSCEast. Ambient concentrations measured at 10 sites through EMEP were used to evaluate the airborne concentrations generated by the model. Modelled deposition was then compared to measured biomonitoring data from pine needles collected at 70 sites. The overall goals of the paper are unclear and the title does not represent its aims. A major error in the modelling framework (lack of O₃ reactivity) makes the model results unreliable. The lack of uncertainty analysis casts doubt on the applicability of the model in generating airborne concentrations and deposition.

Specific Comments A major

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deficiency in the modelling framework is the error made in benzo[a]pyrene reactivity representation. Section 2.4 of the manuscript states, “This CHIMERE version includes gaseous and particulate BaP and its degradation by OH radicals, which represents over 99% of the degradation path for BaP” and this statement is referenced to Bieser et al. (2012) on p.3 of the Supplement. The relevant oxidant for benzo[a]pyrene is in fact ozone rather than hydroxyl. The relevant text in Bieser et al. (2012, p. 1399) is, “For particulate BaP, the reaction rate with ozone is 1 order of magnitude higher than other degradation processes. The main degradation path of gaseous BaP is the reaction with OH radicals. Because 99% of the total BaP is bound to particles, the reaction with ozone can be considered the only effective degradation path of BaP in the atmosphere.” Though the authors have correctly considered the particulate nature of BaP in ambient air, they have mistakenly applied the wrong degradation pathway. As a result, BaP concentrations simulated using their model are likely substantially over-estimated. This error calls into question all further results described in the manuscript.

Technical Corrections General

- The authors should clarify the nature of their reported BaP values at each instance that they are mentioned: measured, modelled or bias-corrected modelled. Also, bias-corrected implies that the result is indeed “correct”. Bias-adjusted is a fairer way to express this.
- Section 2.3 Why include the gaseous equation and discussion when only particulate BaP is being considered? The interested reader can refer to the cited references for full formulations.
- Line 23-24: high molecular weight, not volume
- It would be easier for the reader if all formulae were put into the same form as $C_a = xxx$
- Why was a three-month average temperature used for Koa calculation? How fast is equilibration between air and pine needles? Justify.
- Section 3.1.1 The discussion of the dry deposition flux calculation should be part of the Experimental section rather than the Results
- Describe “corrections have been implemented” as mentioned on line 17 of p. 26490
- There seems to be an implicit assumption that there is 100% uptake of deposited BaP by pine needles. How was modelled deposition flux converted to pine needle concentrations? Is the method used the reverse of that used to convert measured pine needle concentrations

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to those in ambient air? If so, what effect does this symmetry have on the validity of the results? – Explain land use patterns for readers not familiar with this geographic region – Add uncertainty discussion/analysis to all steps in the process (model, bias adjustment to model, measurements in air, measurements in vegetation, deposition conversion to vegetation concentration) in order to constrain conclusions

Table 1 – Specify meaning of values listed under OBS and MOD MEAN (mean plus or minus standard deviation? Standard error?) – Are modelled concentrations bias-adjusted or raw?

Supplement: Modelling Experiment – What equation is used for partitioning? Does it yield the correct benzo[a]pyrene particulate fraction?

Supplement: Model Validation – What is the effect of the time period coverage at each measurement site? Only one covers the entire modelling period. – What is the EMEP sampling method? Frequency of measurement? Duration? Uncertainty in measurements? What is the effect of degradation for measurements that are weekly or monthly averages? – The discussion of the bias correction is unclear and should be reworded so that the reader does not need to consult the original references. What was done exactly? How much were concentrations adjusted? Supplement: Table S3 – Add column “n” to identify number of data pairs at each site – “Bias” appears to be the difference between the observed and modelled means. Check? Suggested Peer Review Aspects 1. Does the paper address relevant scientific questions within the scope of ACP? YES 2. Does the paper present novel concepts, ideas, tools, or data? NO though the way they are put together is novel 3. Are substantial conclusions reached? NO conclusions are not justified due to errors in modelling framework and lack of uncertainty analysis 4. Are the scientific methods and assumptions valid and clearly outlined? NO the flow of the manuscript is generally difficult to follow 5. Are the results sufficient to support the interpretations and conclusions? NO uncertainty analysis is required 6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of re-

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sults)? YES though improvements in wording would improve clarity 7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? YES to credit but NO to indicating their contribution 8. Does the title clearly reflect the contents of the paper? NO 9. Does the abstract provide a concise and complete summary? NO 10. Is the overall presentation well structured and clear? NO 11. Is the language fluent and precise? NO 12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? YES for the most part 13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? YES an overall reworking of the text flow is required 14. Are the number and quality of references appropriate? YES 15. Is the amount and quality of supplementary material appropriate? YES

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 26481, 2015.

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