Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2015-996-AC1, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global atmospheric budget of simple monocyclic aromatic compounds" *by* David Cabrera-Perez et al.

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Received and published: 18 May 2016

We are grateful to the reviewer for their constructive comments that we believe have helped us to strengthen the manuscript. Below we include the original review (in bold), and the response to each comment.

1. The validation of the simulations has been focused on whether the model can reproduce the observed mixing ratios for aromatics. This is of course useful. A step forward for the validation is to test the model's capability for simulating the observed species: species ratios. For example, we have been using the benzene/toluene, or toluene/xylenes ratios as photochemical clocks to determine the age of air mass, because they are typically co-emitted from similar sources and they all have different photochemical lifetime. This could at least give some indications about how confidence the chemistry is in the model.

We added in the manuscript (in the "evaluation with observations" section) a description of the model representations of the toluene/benzene and xylene/benzene ratios compared to the LITERATURE observations (as this set of observations cover the largest area). The related figures were added in the supplement material since they are not essential for the comprehension of the paper.

2. The speciation of xylenes and trimethylbenzenes, etc. The model lumps isomers and assumes equal proportions of single isomers. It is unclear how the rate coefficients are calculated for the lumped species. And, is it a good assumption to assume equal proportion of single isomers when we know some isomer dominates? Justification is needed here considering these species are typically more reactive than benzene and toluene thus are expected to have larger atmospheric impacts. Other two thoughts about the speciation: 1) how sensitive are observation techniques used in the validation to those isomers? 2) how do the emission inventories used here separate those isomers, and what are their assumptions when they lump species?

The assumption of the equal proportions for xylenes and trimethyl benzene isomers is justified by the lack of information on the emissions relative ratios. Additionally, the RCP VOC speciation does not contain any isomer speciation for xylenes. The rate constant of the three isomers (for both species) present differences below 70%. Thus, we averaged the rate constant (weighted by the branching ratios) and we suppose our error is within an acceptable range. A detailed description of the calculation of the reaction rates for each channel has been added to the supplement material.

Furthermore, there are numerous examples in the literature where full speciation/measurements of the isomers is not provided (e.g Lee et al., 2005; Derwent et al., 2014). Consequently for a consistent comparison between observation and model simulations, we consider our approach reasonable.

3. ‘Anthropogenic emissions represent the largest source of aromatics’ is not something really exciting, because this has been known for a long time. Can the authors provide more sectorial information about these anthropogenic emissions? For example, solvent usage has been considered as the largest source for toluene and (lumped) xylenes but not for benzene in the RETRO inventory. Can this work say something about the importance about the solvent usage? Another example, are emissions from vehicles still an important source for aromatics in urban and rural areas? Insights in such sectorial emissions could really improve the quality of the paper.

We added a paragraph in the anthropogenic emissions subsection describing the contribution of each sector to the total anthropogenic emissions. “[...]. When into the sectors provided by the RCP, we found for benzene 49% of the emissions are originated in the residential sector, followed by the energy sector (29%). In the case of toluene, emissions are evenly split for transportation, energy, solvents and residential. Xylenes emission are similarly distributed as for toluene, however solvents are the leading source with 30% of the emissions and residential only 7%. Trimethylbenzenes are abundantly emitted by the transportation sector (90%), as well as other aromatics (60%).”

4. Can the authors say something quantitatively about the RCP emission inventory for benzene, toluene, and xylenes? Are they good? How good? Are there any regions that need to be improved based on the validation in the paper? What are the weaknesses of this emission inventory for aromatics?

The number of observations used for this paper is limited for an exhaustive evaluation of the emission inventory and furthermore this is out of the scope of this paper. Nevertheless, we can state that the comparison of scenarios shows a better agreement with observation in the RCP case. Previous literature estimates seems to underestimate benzene emissions. In the case of toluene and xylene, the difference is less evident, as the emissions in both scenarios are not dramatically different.

5. Is it really necessary to simulate 666 reactions and 229 species in order to reach the conclusions of the paper? Do the authors have any recommendations for a simplified chemistry for model communities? What are the advantages of comprehensive descriptions about the chemical reactions? The authors need to expand the motivations about this.

On the one hand, the chemical mechanism should be as comprehensive as possible for an accurate description of atmospheric chemistry. On the other hand, it is of course necessary to simplify the mechanism for usage in long-term simulations with global models. To achieve this, we have started to work on skeletal mechanism generation based on the directed relation graph with error propagation (DRGEP) method. However, this is work in progress and not ready for inclusion into the current manuscript. This work serve as a basis for further studies atmospheric impacts of these compounds. The mechanism is planned to be used for future studies related to impacts on ozone, hydroxyl radical and other trace species.

6. Tables 1 and 2 in the supplement are not self-explanatory at all. They will need to be modified.

Explanations for the Tables 1 and 2 have been added in the supplement material.

7. I suggest that the ‘2.3 Sinks ’ should be renamed as ‘2.3 Scavenging and dry deposition’, as ‘2.2 Chemistry’ is considered a part of ‘Sinks’ too.

We changed the naming of the section. Nevertheless, “Chemistry” can either be a sink and/or a source or aromatic VOCs. For example, phenol main source is the oxidation from benzene, producing 4Tg/yr. Therefore, we believe that “Chemistry" should remain in an independent section from “Sinks" in order to avoid possible misleading.

References:

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2015-996, 2016.