

We thank the referee for her/his valuable comments. Here the comments are repeated with our answers.

In this study, the authors have applied a global chemistry-climate model (in a chemical transport model mode) to study the impact of monocyclic aromatic species on tropospheric gas-phase composition. They assess the changes in the distribution of several air quality and climate relevant chemical species, including OH, O₃, NO_x, HCHO, glyoxal and CO as a result of including aromatics in the chemical mechanism. The authors find that the impact of aromatics is small on a global scale but have a bigger impact regionally. Despite finding a small impact on a global scale, the authors recommend that global models include aromatics.

I do not think that the authors make a convincing case for this. Moreover several global models already include aromatics (e.g., GEOS-chem, MOZART-4). The paper in its current form does not advance our scientific understanding of the role of aromatics in chemistry and climate.

We agree with the referee that many global model do include aromatics. Nevertheless none of the chemistry climate model do represent aromatics at the level of detail of this work, but rather they make strongly use of lumped species also for very simple aromatics. As stated in Porter et al. (2017) until recently in Geos-Chem aromatics only contributed to SOA formation without any real influence on chemistry. Their estimated impact on tropospheric ozone is however combined with the one of monoterpenes (Porter et al., 2017). Moreover, we are not aware of any study with CAM-Chem (or CESM) quantifying the sole impact of aromatics on ozone. Further MOZART-4 (Emmons et al., 2010) does include only one lumped specie for aromatics. Contrary to the current understanding/expectations, our results indicate that aromatics oxidation, as in MCM, does not significantly impact tropospheric ozone on a global scale. If not a scientific advancement, we believe this to be the first real quantification of single aromatics impact on ozone and other species, such as benzene, toluene and xylenes. Therefore the scientific advancement on this work is reflected not only on the impact quantification, but mostly in the relative impact of each simple aromatic tracer.

Specific comments are provided to improve the paper: P6, section 3: Comparison with observations is important to build confidence in the simulation of aromatics by the model. How do we believe the model is simulating aromatics in the right place for the right reasons? Are there any comparisons with observations?

A detailed evaluation with observations for simulated aromatics with EMAC is available in (Cabrera-Perez et al., 2016). As replied to referee #1 we will summarize the main results of that paper in the revised version:

“Aromatic emissions are dominated by anthropogenic sources, followed by biomass burning emissions and finally biogenic emissions play a minor role. The largest sink of aromatics is chemical oxidation, being dry deposition a minor sink and wet deposition a negligible process. The EMAC model is able to represent the spatial distribution and annual cycle of background stations for benzene and toluene. Benzene and toluene have very low bias and root mean square error (below 50%) when compared to the observations, however higher discrepancies are present for toluene when representing the annual cycle. The complete description of the model setup including emissions, the chemical mechanism used, and the evaluation of the AROM scenario—are included in Cabrera-Perez et al. (2016). The complete set of chemical reactions can be found in the supplementary information (supplement in Cabrera-Perez et al., 2016). The aromatic mechanism has been evaluated against the MCM with the CAABA/MECCA box model in Cabrera-Perez (2017). The comparison shows that the aromatic mechanism consistently simulates ozone, OH and VOCs, with relative differences below 10%. The largest differences affect NO₃ and HONO, which are expected, due to the channels added in the aromatic mechanism including the phenoxy radical reactions and nitrophenol photolysis, respectively. Besides the model evaluation for aromatics EMAC has been extensively evaluated for others species, including studies on O₃ Jöckel et al. (2006); Jöckel et al. (2010); Jöckel et al. (2016), CO in Yoon and Pozzer (2014) and, CO and other tracers (VOCs) in Pozzer et al. (2012b, 2010, 2012a).”

P5,L101: Why 2004-2005 time period was chosen? Why not a more recent period as I would imagine that emissions have evolved since 2005.

The main reason is that we found 2005 the year with the largest number of observations available (see Cabrera-Perez et al. (2016)). Besides, since the aim is to evaluate the chemical impacts of aromatics in the system, results do not aim in resolving them for a specific year but rather as a general concept.

P5, L114-115: Which RCP emissions are being used here?

RCP8.5 emissions. This information has been added for clarity in the methods section.

“We used the Representative Concentration Pathways (RCP8.5) inventory for anthropogenic emissions (van Vuuren et al., 2011) [...]”

P6, L146: Despite high levels of surface aromatics simulated over India and central Africa in the AROM run, why is there not a concomitant increase in day time OH over the highly polluted northern Indo-Gangetic plains or central Africa between REF and AROM?

These are areas where OH and O₃ production are limited due to the large amount of VOC concentrations, and where NO_x is acting a limiting species. For example this has been well reported in India by Ojha et al. (2012); Kumar et al. (2012).

60 **P7, L155: Where are these results shown?**

The figure has been added in the supplement information and referenced in the manuscript.

From: "During the night, an increase up to 6% is found between 800–600 hPa in the northern hemisphere."

to: "During the night, an increase up to 6% is found between 800–600 hPa in the northern hemisphere (see Figure 3 in the supplement)."

65 **Figure 3. How significant are the differences, especially the small numbers, shown here. Presumably the black line on the lower right plot shows the tropopause level, please indicate this in the caption.**

We modified the caption from

On the upper left panel, annual average surface concentrations of OH during day time (REF scenario). On the upper right, surface OH relative difference between aromatic and no-aromatic scenarios expressed in %. On the lower left, OH absolute difference between mentioned scenarios. On the lower right, zonal relative differences (in %).

to

On the upper left panel, annual average surface concentrations of OH during day time (REF scenario). On the upper right, surface OH relative difference between aromatic and no-aromatic scenarios expressed in %. On the lower left, OH absolute difference between mentioned scenarios. On the lower right, zonal relative differences (in %). The black line denotes the tropopause level.

The statistical significance for a 95% confidence level is very low (below 2) in terms of annual means for OH and ozone. When studying the seasons, only winter present statistical significant differences (see figure 1) for OH and ozone species. This information has been added to the manuscript.

80 **P8,L171: Please clarify the statement "Although there is ubiquitous decrease in NO_x this does not seem to limit OH formation."**

This sentence is linked to the previous one and it has been clarified: "The increase of OH production via NO+RO₂ through the increase of RO₂ concentrations are due to aromatics oxidation. Although there is a ubiquitous decrease in NO_x, this does not seem to limit OH formation"

85 to

"The increase of OH production (in Europe, China, and US) via NO+RO₂ through the increase of RO₂ concentrations are due the aromatics oxidation. Although a decrease in NO_x mixing ratios in these areas is present once aromatics are introduced (see section 3.3), which could possibly limit OH formation, this is not observed, as the system is in the high NO_x regimes."

90 **P8, L172-174: How significant are the decreases in the remote southern hemisphere oceanic regions?**

As mentioned in previous comments, the relative changes in OH are small as well as the significance for a 95% confidence level. Again, only ozone in winter season shows significant changes for oceanic areas.

Figure 2 shows zero concentrations of aromatics in this region and given their short lifetimes I do not think they are being transported into this region? Possibly CO is being transported. If so, can you please provide a plot of changes in CO distribution with a significance estimate.

95

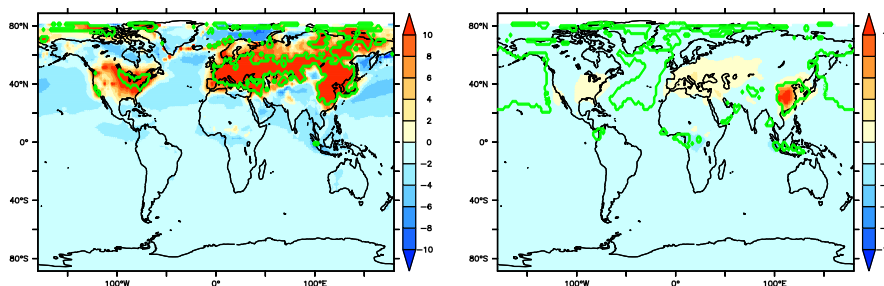


Figure 1. Surface winter relative difference between aromatic and no-aromatic scenarios expressed in %. The area inside the green lines denotes locations where the influence of aromatics is significant.

Benzene concentrations ranges 1 to 7 pptv, it is the only compound whose lifetime allows for the transportations to this region. The plot for CO is provided in the second figure of the supplement. It presents the surface relative change of CO, with an homogeneous mixing ratio increase of 3%.

P8,L174: Figure 5 should be moved to Figure 4 if it is being discussed before the existing Figure 4.

100 The order of the figures has been changed accordingly to the text.

P9, Table 2: Please clarify in the caption that the global methane lifetime refers to lifetime of CH₄ in the global boundary layer. Also suggest explicitly showing how this lifetime is calculated. In particular, how is the boundary layer diagnosed?

The caption has been expanded:

105 “Estimated global averaged OH concentrations in the boundary layer by three different approximations Lawrence et al. (2001) and global boundary layer lifetimes of methane. Calculations for the reference scenario and the aromatics scenario and the differences between them.”

110 The boundary layer diagnosis is fully described in (Pozzer et al., 2009). The PBL is calculated in the model based on the work of (Holtslag et al., 1990). An interactively calculation is performed following the approach of (Troen and Mahrt, 1986), using the Richardson number, the horizontal velocity components, the buoyancy parameters and the virtual temperature. A complete description of this method can be found in (Holtslag and Boville, 1993, sect.3). This information has been added to the manuscript.

P10, L187-188: How do the tropospheric methane lifetime compare against those from other models, for example the ACCMIP models for year 2000?

115 Tropospheric methane lifetime in EMAC is 8.5 years (Lelieveld et al., 2016). This value is within the range 8-10 years, similar to most ACCMIP models. The 17 models that participated in the model intercomparison by Naik et al. (2013) show a range of 7.1–14.0 years, while the multimodel mean of 9.7 years was considered to be 5–10% higher than observation-derived estimates.

P10, L195: High NO_x can also result in titration of ozone.

120 We mentioned this possibility in the text:

from

In areas with high NO_x and VOC mixing ratios, ozone is formed, which under photolysis produces OH (??-??.) to

125 In areas with high NO_x and VOC mixing ratios, ozone is formed, which under photolysis produces OH (R5-R6), although ozone titration may also take place reducing ozone formation (R11).

P10, L203-218: Please point to figures in the supplementary in this discussion.

The text has been modified specifying the figures of the supplement. From:

“For HO₂ a relative decrease of less than 1% in the atmospheric global burden is found. [...]”

130 “Contrary to expectations, we found large increases of HONO mixing ratios in continental areas, generally with the sign of this change opposite to that of OH. These relative changes reached more than 50%, specifically in Africa, South-America, the Arabian Peninsula and South-East Asia. [...]”

to:

“For HO₂ a relative decrease of less than 1% in the atmospheric global burden is predicted (see Figure 2 in the supplement). [...]”

135 “Contrary to expectations, we found large increases of HONO mixing ratios in continental areas, generally with the sign of this change opposite to that of OH. These relative changes reached more than 50%, specifically in Africa, South-America, the Arabian Peninsula and South-East Asia (see Figure 2 in the supplement). [...]”

P 9, Figure 4: The figure shows O₃ changes while the caption mentions OH changes. Please correct the caption.

140 The caption has been corrected:

“On the upper left panel, annual average surface mixing ratios of O₃ during day time (REF scenario). On the upper right, surface O₃ relative difference between aromatic and no-aromatic scenarios expressed in %. On the lower left, O₃ absolute difference between mentioned scenarios. On the lower right, zonal relative differences (in %).”

145 **P13, L259: Is deposition of individual aromatics considered in this study? If so, how are the dry deposition velocities calculated?**

Yes, it is. Calculations are done with the DDEP submodel, based on the big area leaf approach. The complete description of the methodology can be found in Kerkweg et al. (2006)

P18, L348: The van Vuuren et al reference is wrong.

150 This reference has been corrected. From van Vuuren et al. (2008) to van Vuuren et al. (2011)

References

- Cabrera-Perez, C. D.: Simple Monocyclic Aromatic Compounds from a Global Scale Perspective, Ph.D. thesis, Johannes Gutenberg-Universität Mainz, 2017.
- Cabrera-Perez, D., Taraborrelli, D., Sander, R., and Pozzer, A.: Global atmospheric budget of simple monocyclic aromatic compounds, *Atmospheric Chemistry and Physics Discussions*, 2016, 1–25, doi:10.5194/acp-2015-996, <http://www.atmos-chem-phys-discuss.net/acp-2015-996/>, 2016.
- Emmons, L., Walters, S., Hess, P., Lamarque, J.-F., Pfister, G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepfle, T., et al.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geoscientific Model Development*, 3, 43–67, 2010.
- Holtzlag, A. and Boville, B.: Local versus nonlocal boundary-layer diffusion in a global climate model, *Journal of Climate*, 6, 1825–1842, 1993.
- Holtzlag, A., De Bruijn, E., and Pan, H.: A high resolution air mass transformation model for short-range weather forecasting, *Monthly Weather Review*, 118, 1561–1575, 1990.
- Jöckel, P., Tost, H., Pozzer, A., Brühl, C., Buchholz, J., Ganzeveld, L., Hoor, P., Kerkweg, A., Lawrence, M. G., Sander, R., Steil, B., Stiller, G., Tanarhte, M., Taraborrelli, D., van Aardenne, J., and Lelieveld, J.: The atmospheric chemistry general circulation model ECHAM5/MESSy1: consistent simulation of ozone from the surface to the mesosphere, *Atmospheric Chemistry and Physics*, 6, 5067–5104, doi:10.5194/acp-6-5067-2006, <https://www.atmos-chem-phys.net/6/5067/2006/>, 2006.
- Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the modular earth submodel system (MESSy2), *Geoscientific Model Development*, 3, 717–752, 2010.
- Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K.-D., Graf, P., Grewe, V., Kerkweg, A., Kern, B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, *Geoscientific Model Development*, 9, 1153–1200, doi:10.5194/gmd-9-1153-2016, <https://www.geosci-model-dev.net/9/1153/2016/>, 2016.
- Kerkweg, A., Buchholz, J., Ganzeveld, L., Pozzer, A., Tost, H., and Jöckel, P.: Technical Note: an implementation of the dry removal processes DRY DEPosition and SEDImentation in the Modular Earth Submodel System (MESSy), *Atmospheric Chemistry and Physics*, 6, 4617–4632, 2006.
- Kumar, R., Naja, M., Pfister, G. G., Barth, M. C., Wiedinmyer, C., and Brasseur, G. P.: Simulations over South Asia using the Weather Research and Forecasting model with Chemistry (WRF-Chem): chemistry evaluation and initial results, *Geoscientific Model Development*, 5, 619–648, doi:10.5194/gmd-5-619-2012, <https://www.geosci-model-dev.net/5/619/2012/>, 2012.
- Lawrence, M., Jöckel, P., and Kuhlmann, R. v.: What does the global mean OH concentration tell us?, *Atmospheric Chemistry and Physics*, 1, 37–49, 2001.
- Lelieveld, J., Gromov, S., Pozzer, A., and Taraborrelli, D.: Global tropospheric hydroxyl distribution, budget and reactivity, *Atmospheric Chemistry and Physics*, 16, 12477–12493, doi:10.5194/acp-16-12477-2016, <http://www.atmos-chem-phys.net/16/12477/2016/>, 2016.
- Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmospheric Chemistry and Physics*, 13, 5277–5298, doi:10.5194/acp-13-5277-2013, <https://www.atmos-chem-phys.net/13/5277/2013/>, 2013.
- Ojha, N., Naja, M., Singh, K., Sarangi, T., Kumar, R., Lal, S., Lawrence, M., Butler, T., and Chandola, H.: Variabilities in ozone at a semi-urban site in the Indo-Gangetic Plain region: Association with the meteorology and regional processes, *Journal of Geophysical Research: Atmospheres*, 117, 2012.
- Porter, W. C., Safieddine, S. A., and Heald, C. L.: Impact of aromatics and monoterpenes on simulated tropospheric ozone and total OH reactivity, *Atmospheric Environment*, 169, 250 – 257, doi:<https://doi.org/10.1016/j.atmosenv.2017.08.048>, <http://www.sciencedirect.com/science/article/pii/S1352231017305617>, 2017.
- Pozzer, A., Jöckel, P., and Aardenne, J. V.: The influence of the vertical distribution of emissions on tropospheric chemistry, *Atmospheric Chemistry and physics*, 9, 9417–9432, 2009.
- Pozzer, A., Pollmann, J., Taraborrelli, D., Joeckel, P., Helmig, D., Tans, P., Hueber, J., and Lelieveld, J.: Observed and simulated global distribution and budget of atmospheric C₂-C₅ alkanes, *Atmospheric Chemistry and Physics*, 10, 4403–4422, 2010.
- Pozzer, A., Meij, A. d., Pringle, K., Tost, H., Doering, U., Aardenne, J. v., and Lelieveld, J.: Distributions and regional budgets of aerosols and their precursors simulated with the EMAC chemistry-climate model, *Atmospheric Chemistry and Physics*, 12, 961–987, 2012a.
- Pozzer, A., Zimmermann, P., Doering, U. M., van Aardenne, J., Tost, H., Dentener, F., Janssens-Maenhout, G., and Lelieveld, J.: Effects of business-as-usual anthropogenic emissions on air quality, *Atmospheric Chemistry and Physics*, 12, 6915–6937, doi:10.5194/acp-12-6915-2012, <https://www.atmos-chem-phys.net/12/6915/2012/>, 2012b.

- Troen, I. and Mahrt, L.: A simple model of the atmospheric boundary layer; sensitivity to surface evaporation, *Boundary-Layer Meteorology*, 37, 129–148, 1986.
- van Vuuren, D. P., Feddema, J., Lamarque, J.-F., Riahi, K., Rose, S., Smith, S., and Hibbard, K.: Work plan for data exchange between the integrated assessment and climate modeling community in support of phase-0 of scenario analysis for climate change assessment (representative community pathways), assessment (Representative Community Pathways), 3, 4, 2008.
- 215 van Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G. C., Kram, T., Krey, V., Lamarque, J.-F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S. J., and Rose, S. K.: The representative concentration pathways: an overview, *Climatic Change*, 109, 5, doi:10.1007/s10584-011-0148-z, <https://doi.org/10.1007/s10584-011-0148-z>, 2011.
- 220 Yoon, J. and Pozzer, A.: Model-simulated trend of surface carbon monoxide for the 2001–2010 decade, *Atmospheric Chemistry and Physics*, 14, 10 465–10 482, doi:10.5194/acp-14-10465-2014, <https://www.atmos-chem-phys.net/14/10465/2014/>, 2014.