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

In this work, the authors compare differences in predicted tropospheric species distributions and relative concentrations between two model simulations: (1) a base case (REF) in which no aromatics are emitted globally to (2) a case in which C6-C11 aromatics are emitted globally (AROM). Both model simulations

5 include detailed aromatic chemistry, but since there are no aromatic emissions in REF, only AROM contains aromatic chemistry impacts.

The base case (REF) is not consistent with recent, stateof-the-art models (GEOS-chem, MOZART-4, etc.), in which benzene, toluene, xylenes, or some parameterized combination of these and their chemistry is included. A comparison between a model simulation in which a subset of aromatics are parametrized

10 as one or two or three species and then compared to the AROM simulation would be more scientifically relevant.

It is likely, however that the differences (globally) of such a model simulation would be insignificant, as the differences in the AROM-REF situation presented here have very little significant globally.

- We thank the referee for pointing this out. It is well know that aromatics play a role on local scale (Tie et al., 2007; Stroud et al., 2008; Jaars et al., 2014), but no studies are really showing their impact on global scale, especially for each specific simple aromatic. In order to enhance this point (see reply to referee #1) additional sensitivity simulations have been performed to quantify the importance of each sector and aromatic tracer. If we understood correctly the referee, a comparison of our chemical mechanism with other mechanisms used in GCM, which are using strongly lumped aromatics, would be more interesting (see also reply to referee #2). This
- 20 is, however, outside the scope of the manuscript and of the journal, being the suggested work a technical study. Additionally, we challenge the point of being such work more scientifically relevant, as the results from simplified chemistry simulations are known to be somehow already flawed due to the lack of details in their chemical degradation scheme. Therefore only their differences, compared to our mechanism, could be estimated, resulting, as mentioned, in a pure numerical exercise which would not fit the journal's scope. Nevertheless, as shown by our
- 25 work, the impact of aromatics on a global scale is somewhat limited and therefore even simplified chemistry could give reasonable results if used at such resolutions, as also correctly mentioned by the referee.

The other issue is that the model resolution is very coarse, which is also probably why there isn't a significant impact from the addition of the larger, more reactive aromatics – they're simply lost into such a coarse grid.

- 30 This is only valid for urban areas, where the resolution is a limiting factor. At global scale T63 resolution is capable to simulate background chemistry as seen in (Cabrera-Perez et al., 2016). Therefore, the results shown are consistent for representing global scale chemistry. We do agree that uncertainties can be located especially for short-lifetime aromatics, but benzene impact should be reasonable simulated by such resolution due to its long lifetime (8 days).
- 35 In general, the comparisons shown simply reinforce that it is worthwhile to include aromatics and aromatic chemistry in global models, but this is not at all novel as aromatic chemistry has been included in global models for years.

As commented for referee #2, the aim of this study is to show the impact of aromatic with a detailed chemical mechanism. (1) The one used in this study is more detailed/complex than those used in other models. (2) besides

40 the work by (Porter et al., 2017) we are not aware of any study with CAM-Chem (or CESM) quantifying the sole impact of aromatics on ozone. Therefore, no previous work has been focusing on the effects of aromatics on tropospheric chemistry at the global scale.

A comparison of a reasonable current chemistry scheme with emissions of a subset of aromatics compared to the full aromatic scheme and expanded aromatics emissions using a finer model grid over a regional scale

45 may be more appropriate to look at the impacts of including larger aromatics (C9+ aromatics) on urban and regional scales, where the reactive aromatics likely have a more significant impact on tropospheric chemistry. This would be a more novel and valuable approach.

The scheme used in this work is a "reasonable chemistry scheme" as now part of the standard set-up simulation in EMAC. As mentioned also to referee #1 we understand that the effect of single aromatic tracers on atmospheric

50 chemistry would be more valuable, and therefore additional sensitivity runs have been performed. The use of regional model and the only focus on urban areas would be out of the scope of this paper.

Throughout the paper, and in particular the introduction, there are awkwardly worded sentences that would significantly benefit from having a careful onceover/proofread/edit by a native or strong English speaker. E.g., Page 1, line 20; Page 3, lines 63-64; Page 4, line 74; Page 6, line 161; Page 10, line 193; Page 11, Table 2 title

55 **11, Table 2 title.** 

Thanks. We agree with that and the sentences have been rewritten.

Page 1, line 20: "[...]and ozone, a major atmospheric pollutant with the capability of modifying the climate forcing and toxic for terrestrial life[...]"

"[...]and ozone, a major atmospheric pollutant with the capability of modifying the climate forcing and a toxic substance for terrestrial life[...]"

Page 3, lines 63-64: "The emissions are primarily anthropogenic, being mainly fossil fuel combustion and leakage, and solvent as the most important sources."

"The emissions are primarily anthropogenic, being the most important sources fossil fuel combustion and leakage, and solvents."

65 Page 4, line 74: "Furthermore, they are toxic compounds (WMO, 2000) and can affect directly and indirectly human health."

"Furthermore, some aromatics are toxic (WMO, 2000) and can affect directly and indirectly human health." Page 10, line 193: "The changes in OH concentrations are dependent on to the  $NO_x$  regimes[...]" "The changes in OH concentrations depend on the  $NO_x$  levels[...]"

70 Page 11, Table 2 title. "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."

"Estimated global averaged OH concentrations in the boundary layer by three different approximations (weighted by mass, volume and methane concentration) Lawrence et al. (2001) and global boundary layer lifetimes of

75 methane. Calculations are performed for the *REF* and *AROM* scenarios and the differences between them." Specific comments Page 1, line 5 – in referring to the inclusion of "aromatic compounds" in the abstract, perhaps state which species and/or class of aromatics are included, and to what degree their chemistry is included. Even stating C6-C11 aromatics would be helpful.

*The abstract has been changed:* "Globally, we found a small annual average net decrease (less than 3%) in global OH, ozone, and NO<sub>x</sub> mixing ratios when aromatic compounds are included in the chemical mechanism."

"Globally, we found a small annual average net decrease (less than 3%) in global OH, ozone, and NO<sub>x</sub> mixing ratios when aromatic compounds are included in the chemical mechanism. The monocyclic aromatics of this study comprises benzene, toluene, xylenes, phenol, styrene, ethylbenzene, trimethylbenzenes, benzaldehydes and lumped higher aromatics (as aromatics with more than 9 carbon atoms)."

80

- Page 1, line 7 Be specific about the changes relative or absolute. Also, here and elsewhere in the paper, the word "found" is used to refer to differences between the base REF case with no aromatic hydrocarbon emissions and the AROM case in which aromatics emissions are included. I would argue that nothing is "found", but rather, a difference in the atmospheric burden of particular species in the simulation results was noted. "Found" implies that measurements were made, and there were no measurements made or
- 90 reported in this paper. I would suggest replacing "found" with "predicted", or something similar (see also page 1, lines 9, 12, etc.)

In line 7, it is now specified that the changes are relative. We completely agree with the referee. Indeed, "found" is not appropriate and scientifically not sound. Because there are several instances of the word "found" we will not list all the modified sentences here.

95 Page 6, line 121 - It would be nice if I didn't have to go read another paper to get all the details about the additional chemistry. A summary or brief description of the detailed aromatic chemistry in Cabrera-Perez et al. 2016 would be nice to include in this paper.

We apologize for the lack of information. As metioned to referee #1 a summary of the main results obtained in Cabrera-Perez et al. (2016) has been added:

- 100 "Aromatic emissions are dominated by anthropogenic sources, followed by biomas 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 discrepan-
- 105 cies 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 mecha-
- 110 nism consistently simulates ozone, OH and VOCs, with relative differences below 10%. The largest differences affect  $NO_3$  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)."

Page 6, line 143 – define "daytime". Also, define "surface".

Daytime is the time frame with an income of solar radiation greater than zero. As the EMAC model uses surface following vertical hybrid pressure coordinates, surface is defined as the lowest model level, i.e. covering a thickness of roughly 120 meters. These definitions have been added to the revised version.

120 **Table 1 – the title stating "This emissions are the same as in ... but for higher aromatics" is both gram**matically incorrect and not clear. This should be a footnote in the table, and more specific, or perhaps, to make it more clear, show the 2016 paper emissions in the table as another column.

This has been modified for clarity:

"This emissions are the same as in (Cabrera-Perez et al., 2016) but for higher aromatics."

125 "These emissions are the same as in (Cabrera-Perez et al., 2016), with the addition of the emissions for higher aromatics."

More importantly, there is an inconsistency between the table and text: the total aromatic emissions in the table are 39.3 TgC/yr of which 3.8 TgC/yr are higher aromatics, while the text (Page 5, line 120) states that it is 35 TgC/yr, of which 3.4 TgC/yr are higher aromatics. This needs to be reconciled.

130 The total emission of aromatic VOCs is 41Tg/yr of which 3.8Tg/yr belongs to higher aromatics. In the text these values are in TgC/yr (hence the apparent inconsistency). For consistency all units have been rewritten in TgC/yr. Also, Trimethyl-benzene should indicate "trimethylbenzenes" (there are three different trimethylbenzenes). Also, what about the ethyltoluenes?

This has been corrected. Ethyltoluenes were not included in the chemistry.

135 Technical corrections Figures - rather than saying "upper left panel", "top panel", etc., simply label the different panels of each figure a, b, c, etc., and then refer to them in the figure caption with (a), (b), (c), etc. When referring to the supplement, refer the reader to a specific Table (S1, S2, etc.) or Figure S1, S2 in the supplement, and not the document as a whole.

The tables and figures have been specified for clarity.

140 **Page 1, line 16 – change "comprises" to "comprise".** *"comprises" has been replaced to "comprise".* 

Page 2, line 25 - RO2 is not "the peroxy radical", but rather "an organic peroxy radical".

"the peroxy radical" has been replace by "an organic peroxy radical".

Page 3, line 57 – change "In contrast, the high NOx..." to "In contrast, O3 in the high NOx regime..." (It is the O3 that is limited by VOC concentration, not the high NOx regime.

"In contrast, the high NOx..." has been replace by "In contrast, O3 in the high NOx regime..."

Figure 2 – there is an extra space after Aromatic VOC in the "title" of the bottom panel. Also, there is inconsistency throughout the paper in the capitalization of figure titles (which is odd – typically figures do not have titles).

150 The extra space has been removed and the figures have been changed accordingly.

Page 7, line 157 and elsewhere – mlc is not an acceptable shortform for molecules. Either spell it out entirely (preferred), or use the somewhat acceptable "molec" as a shortform.

All instances of "mlc" have been replaced by "molec".

Page 9, Figure 4 caption – Internally inconsistent – refers to O3 and OH.

155 *OH has been replace by O3.* 

Page 13, line 246 – add a space between 7 and km.

The space has been changed.

Page 13, line 254 – add a space between e.g. and Butler.

The space has been added.

160 Supplement-Table(S)1. "higher" does not need to be capitalized.

"Higher" has been changed to "higher".

The sentences after the \* in the table title should be in the footnotes. Also, the reference to extra-tropical forests is unclear. Define the references to PTR (naphthalene and C11 Aromatics).

This has been changed. The explanation has been changed. We will omit the information and naming PTR naming as we do not consider relevant for the description. The reference on extra-tropical forest has been clarified:

"We use the average value of Pine-forest understory and coniferous canopy as representative for extra-tropical forest."

"We use the average value of Pine-forest understory and coniferous canopy as the input emission factor representative for extra-tropical forest." 170 Supplement Figure (S)1. Change "(Arom – base)/base, %" to "(AROM-REF)/REF, %" to be consistent with the main text, and this should really be the y-axis title, not the figure title. For the y-axis labels, use decimals instead of commas, or just use integer values.

Titles and labels have been changed.

Figure S2 – units in %? Figures S3 and S4 – the titles are redundant with the figure captions.

175 In Figure S2 % has been specified. Titles have been removed.

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 aro-
- 180 matic 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.
  - Jaars, K., Beukes, J., van Zyl, P., Venter, A., Josipovic, M., Pienaar, J., Vakkari, V., Aaltonen, H., Laakso, H., Kulmala, M., et al.: Ambient aromatic hydrocarbon measurements at Welgegund, South Africa, Atmospheric Chemistry and Physics, 14, 7075–7089, 2014.
- 185 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, Geoscien-
- 195 tific Model Development, 9, 1153–1200, doi:10.5194/gmd-9-1153-2016, https://www.geosci-model-dev.net/9/1153/2016/, 2016.
  - 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.
  - 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., Pollmann, J., Taraborrelli, D., Joeckel, P., Helmig, D., Tans, P., Hueber, J., and Lelieveld, J.: Observed and simulated
  - global distribution and budget of atmospheric  $C_2$ - $C_5$  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.
- 210 Stroud, C., Morneau, G., Makar, P., Moran, M., Gong, W., Pabla, B., Zhang, J., Bouchet, V., Fox, D., Venkatesh, S., Wang, D., and Dann, T.: OH-reactivity of volatile organic compounds at urban and rural sites across Canada: Evaluation of air quality model predictions using speciated VOC measurements, Atmospheric Environment, 42, 7746 – 7756, doi:https://doi.org/10.1016/j.atmosenv.2008.05.054, http://www.sciencedirect.com/science/article/pii/S1352231008005104, 2008.
- 215 Tie, X., Madronich, S., Li, G., Ying, Z., Zhang, R., Garcia, A. R., Lee-Taylor, J., and Liu, Y.: Characterizations of chemical oxidants in Mexico City: A regional chemical dynamical model (WRF-Chem) study, Atmospheric Environment, 41, 1989 – 2008, doi:https://doi.org/10.1016/j.atmosenv.2006.10.053, http://www.sciencedirect.com/science/article/ pii/S1352231006010399, 2007.
  - WMO: Air quality guidelines for Europe, Copenhagen: WHO Regional Office for Europe, 2000.
- 220 Yoon, J. and Pozzer, A.: Model-simulated trend of surface carbon monoxide for the 2001–2010 decade, Atmospheric Chemistry and Physics, 14, 10465–10482, doi:10.5194/acp-14-10465-2014, https://www.atmos-chem-phys.net/14/10465/2014/, 2014.