

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

The manuscript by Cabrera-Perez et al. discusses the impact of aromatic species on the tropospheric composition using the global chemical transport model MESSy/EMAC. A model simulations with aromatics included is compared to a simulation without considering aromatics and the impacts on key species such as OH, Ozone, NO_x, NO₃, HNO₃, HCHO and glyoxal are explored. This manuscript is strongly based on Cabrera-Perez(2016), which describes the aromatic chemistry added to EMAC and evaluates the global distribution of modeled aromatic species. While I acknowledge the importance of studying how aromatics impact the chemistry of the troposphere, I see limited scientific advancement in the presented study.

The manuscript rather represents a model description, I see it better suited for a journal like GMD.

Journals like GMD are rather technical and suited for model development/evaluation. Here, impact studies are performed on a global scale and therefore, to our opinion, the topic fits perfectly within the journal's scope.

Major Comments: Aromatics come from a number of different source types including both anthropogenic and natural sources. Simply turning off all aromatic species in the model independent of their origin or their reactivity does provide little practical insight. The study would provide more value if it were to look at individual source types and/or individual aromatic species.

We thank the referee for pointing this issue. We fully agree with her/him, and to improve the manuscript, we performed simulations for studying the relevance of each source (anthropogenic, biogenic and biomass burning) and also including only benzene, toluene and the rest of compounds.

We performed two series of sensitivity simulations. In the first series, three simulations were run, where each of the different sources were switched off, i.e. anthropogenic, biogenic and biomass burning emissions, respectively. In the second series, four simulations were run, where different compounds were removed from the mechanism (benzene, toluene, xylenes, and the rest of aromatics).

In Figures 1 and 2, the sensitivity simulations are compared with the AROM scenario.

In general anthropogenic emissions dominate the impacts over biomass burning and biogenic sources. These emissions also have major relevance on the northern hemisphere for NO₂, NO₃, and Glyoxal. As expected, biomass burning sources have a large impact on specific areas: Siberia, central Africa, and the Amazonian forest. Increases of NO₂ above 10% are estimated in central Africa when biomass burning sources are missing, while Glyoxal decreases more than 10% in central Africa. Biogenic emissions are of minor relevance for the impacts on any species.

Among benzene, toluene, xylenes, and the rest of aromatics the first species has the lowest relevance due to its low reactivity (among the aforementioned aromatics). Toluene is the most important source for glyoxal overall and is also relevant for NO₃ in remote areas. Xylenes seem to have generally a small influence on the studied tracers. Interestingly, the rest of aromatics appear to lead the changes in OH, NO₂, NO₃ and glyoxal (this last only in continental areas) in areas dominated by biomass burning.

The study is conducted at a rather coarse horizontal model resolution (1.875 degrees). At this resolution urban chemistry cannot be represented, yet aromatics play an important role specifically in urban regimes. The authors themselves state in Line 260 that the coarse resolution limits the representation of urban chemistry. So why was the study not done at higher model resolution or with a higher resolution regional model?

It is well known that aromatics have strong impact on local scale (Tie et al., 2007; Stroud et al., 2008; Jaars et al., 2014), and therefore any high resolution urban model simulation would not have added anything to the already present knowledge. On the other side, the aim of this work is to find the chemical impact of aromatics at global scale, as this was never really quantified before for single aromatic species. The T63 resolution was chosen as the highest resolution possible with an affordable computation cost.

Has the aromatic chemistry in EMAC been compared to a fully explicit chemical scheme? If so, how well does it represent the full chemistry? The authors list a large range of uncertainties that impact the presented results and reduce confidence in the value of the conclusions.

The only chemical scheme that can be considered a "fully explicit scheme" for aromatics is the MCM mechanism (Bloss et al., 2005b). A model comparison between the original MCM and the mechanism used in this study has been done using the CAABA/MECCA box model and is fully described in Cabrera-Perez (2017). This information will be added to the methods section of the manuscript, summarizing the main results, i.e.:

"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."

I agree that emissions in general can pose, in parts, large uncertainties. Have the authors looked at other inventories (HTAP, RETRO, NEIs, ...)? What was the reason for selecting IPCC emissions?

HTAP offers no speciation of organics, but rather a lumped NMVOCs emissions. We relied on the speciation provided by IPCC so to assure the correct regional emissions distribution (although this does not guarantee the global total emissions to be correct). Finally, as the IPCC and RETRO use the same speciation algorithm, no real differences are present between the two dataset, in this sense. Finally, RETRO covers only up to 2000, while we were interested in the year 2005 as the biomass burning database (GFAS) covers only from 2003 onwards. Finally IPCC emissions offers the advantages to have a long time coverage (1850-2100) when the historical data and the projections are used.

What is the reason for the high ozone bias in ECAM? Could the chemical scheme be a cause?

This issue in the EMAC model has been addressed in previous work Jöckel et al. (2016); Righi et al. (2015) this is the evaluation of MESSy2. It is not clear what is the cause of this bias, but our work clarifies that the bias is not due by the lack of details in the NMVOCs chemistry.

Line362: Do the authors imply that MCMV3.1 is incorrectly representing ozone chemistry or that the lumped chemical scheme that was developed based on MCMv3.1 and used in the current study has problems? If so, how much can one trust the derived sensitivity of tropospheric chemistry to aromatics? Does including HONO help with this problem? Has this been tested in a box model? Given the large role of SOA in aromatic chemistry, should such a study not have been conducted with a more detailed aerosol scheme that represents SOA formation.

We are confident on the results from MCMv3.1. In our work we summarize what Bloss et al. (2005a) pointed as limitations of MCM when simulating ozone mixing ratios. As mentioned in Cabrera-Perez et al. (2016), there were limitations in order to evaluate xylene concentrations, but the results show that in general EMAC is able to represent observations. The results of our study suggest that aromatics have small influence at the global scale, and therefore the limitations by MCM do not lead to large errors in the global scale. The HONO channels included in the model do not have a large effect on the sensitivity runs.

There is zero evaluation of the model with observations. Aromatic species themselves have been evaluated in Cabrera-Perez (2016), but what about e.g. O3 or CO?

There are several studies covering the evaluation of O3, CO and several organic species. (Jöckel et al 2010, gmd, pozzer et al 2010). As mentioned before, the basic chemical mechanism is the one used in many model evaluation's works. Here detail comparison for O3 can be found in Jöckel et al. (2006); Jöckel et al. (2010); Jöckel et al. (2016), for CO in Yoon and Pozzer (2014) and for CO and other tracers (VOCs) in Pozzer et al. (2012b, 2010, 2012a). We do not believe that adding a detail evaluation of these tracers would give additional informations beside what is already present in the literature.

The following text will be added to the manuscript: "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. (2007, 2010, 2012a)."

Minor Comments: Line 55 (and also Line 195): This is a very simplified description of ozone chemistry. High NOx regimes do not equate net ozone formation as at very high NOx concentrations ozone can actually be lost. The actual urban chemistry cannot be resolved at the coarse model resolution applied in here.

We fully agree that in this description effects of ozone titration are somehow neglected. Although the description given would be essential for the understanding of the manuscript, we understand that it should give a complete description, and therefore will be extended in the revised version. We introduced the following in the Introduction:

"When elevated ozone and NO concentrations are simultaneously present, another possible reaction occurs, the ozone titration (e.g. Kley et al., 1994): $\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$ "

Line125: Given how closely this manuscript is linked to (Cabrera-Perez et al., 2016) I suggest referencing some of the major findings of this study as they apply to this work.

The following text has been added:

"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

115 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).”

120 **Figure 5 and Figure 6: Is this daytime? Daytime and Nighttime? Most Figure Captions require more details on what is shown.**

We thanks the referee for pointing this out. More details have been added to the labels.

Caption in Fig 5	Daytime OH relative difference (expressed in percent) in the boundary layer between the <i>AROM</i> and <i>REF</i> scenarios. In black, values for the northern hemisphere. In red, values for the southern hemisphere.
Caption in Fig 6.	Daytime O ₃ relative difference (expressed in percent) in the boundary layer between the <i>AROM</i> and <i>REF</i> scenarios. In black, values for the northern hemisphere. In red, values for the southern hemisphere.
Caption in Fig 7	Surface daytime NO ₂ (left) and NO ₃ (right) relative difference between aromatic and no-aromatic scenarios expressed in %.
Caption in Fig 8	Surface daytime relative difference (<i>(No phenoxy channel-AROM) / AROM</i>) expressed as a percent for ozone (left) and NO ₃ (right).
Caption in Fig 9	Top. Annual average daytime surface mixing ratios of HCHO in ppt. Bottom, daytime surface relative difference in %.
Caption in Fig 10	Top. Annual average surface daytime mixing ratios of glyoxal in ppt. Bottom, daytime surface relative difference in %.

The authors discuss the role of a number of intermediate species. I suggest adding the chemical reactions for the aromatics as included in MESSy in the Supplement or at least include a reference to where the chemical reactions can be looked up.

125 *A reference to the supplement of (Cabrera-Perez et al., 2016), has been added in the methods sections:.*

“The complete set of chemical reactions can be found in the supplementary information (supplement in Cabrera-Perez et al., 2016).”

References

- 130 Bloss, C., Wagner, V., Bonzanini, A., Jenkin, M. E., Wirtz, K., Martin-Reviejo, M., and Pilling, M. J.: Evaluation of detailed aromatic mechanisms (MCMv3 and MCMv3.1) against environmental chamber data, *Atmospheric Chemistry and Physics*, 5, 623–639, doi:10.5194/acp-5-623-2005, <http://www.atmos-chem-phys.net/5/623/2005/>, 2005a.
- Bloss, C., Wagner, V., Jenkin, M. E., Volkamer, R., Bloss, W. J., Lee, J. D., Heard, D. E., Wirtz, K., Martin-Reviejo, M., Rea, G., Wenger, J. C., and Pilling, M. J.: Development of a detailed chemical mechanism (MCMv3.1) for the atmospheric oxidation of aromatic hydrocarbons, *Atmospheric Chemistry and Physics*, 5, 641–664, doi:10.5194/acp-5-641-2005, <https://www.atmos-chem-phys.net/5/641/2005/>, 2005b.
- 135 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.
- 140 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 Welgegend, South Africa, *Atmospheric Chemistry and Physics*, 14, 7075–7089, 2014.
- 145 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.
- 150 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., Neu-maier, 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.
- 155 Kley, D., Geiss, H., and Mohnen, V. A.: Tropospheric ozone at elevated sites and precursor emissions in the United States and Europe, *Atmospheric Environment*, 28, 149 – 158, doi:[https://doi.org/10.1016/1352-2310\(94\)90030-2](https://doi.org/10.1016/1352-2310(94)90030-2), <http://www.sciencedirect.com/science/article/pii/S1352231094900302>, 1994.
- 160 Pozzer, A., Jöckel, P., Tost, H., Sander, R., Ganzeveld, L., Kerkweg, A., and Lelieveld, J.: Simulating organic species with the global atmospheric chemistry general circulation model ECHAM5/MESSy1: a comparison of model results with observations, *Atmospheric Chemistry and Physics*, 7, 2527–2550, 2007.
- 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.
- 165 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.
- 170 Righi, M., Eyring, V., Gottschaldt, K.-D., Klinger, C., Frank, F., Jöckel, P., and Cionni, I.: Quantitative evaluation of ozone and selected climate parameters in a set of EMAC simulations, *Geoscientific Model Development*, 8, 733–768, doi:10.5194/gmd-8-733-2015, <https://www.geosci-model-dev.net/8/733/2015/>, 2015.
- 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.
- 175 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.
- 180 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.
- 185

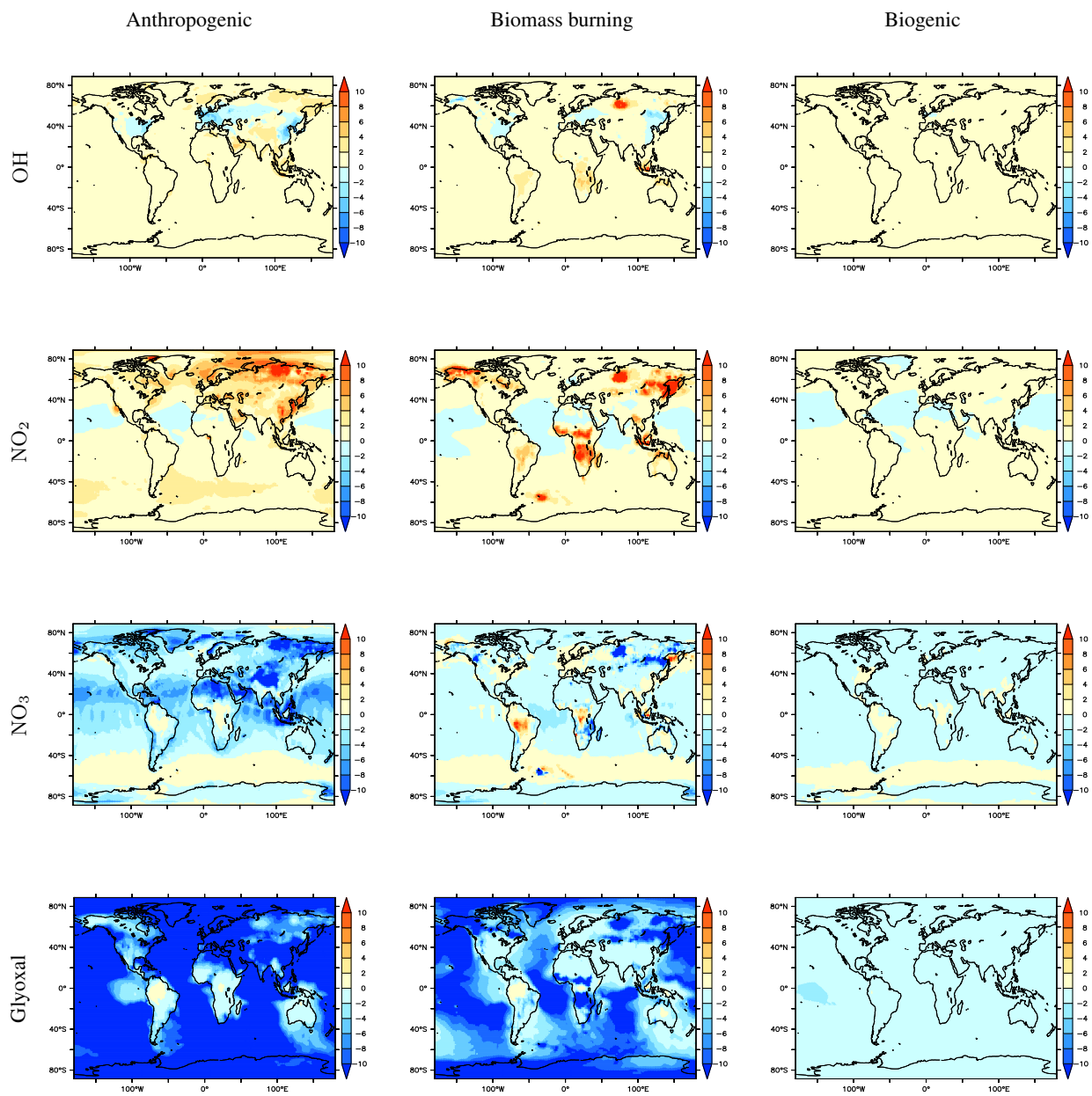


Figure 1. Surface annual mean relative differences between aromatic and the different scenarios expressed in %.

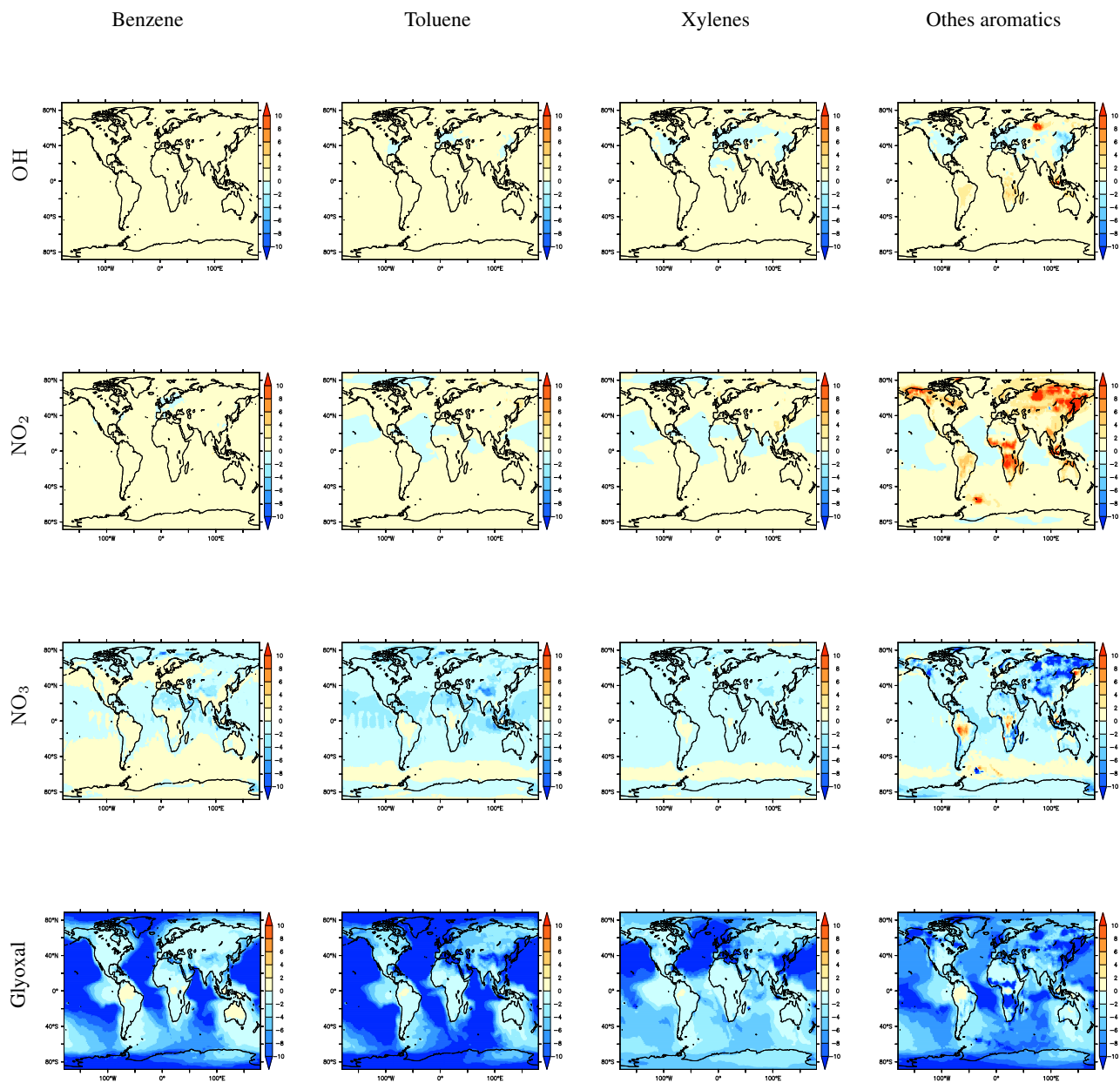


Figure 2. Surface annual mean relative differences between aromatic and the different scenarios expressed in %.