Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-608-AC1, 2021

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identifying forecast uncertainties for biogenic gases in the Po valley related to model configuration in EURAD-IM during PEGASOS 2012" *by* Annika Vogel and Hendrik Elbern

Annika Vogel and Hendrik Elbern

av@eurad.uni-koeln.de

Received and published: 25 January 2021

We thank the reviewer for the elucidating evaluation and valuable remarks. We did several substantial modifications to the manuscript as requested and we confident to address all remarks in a satisfying way.

This paper presents a very interesting study analyzing a variety of meteorological and physical impacts on biogenic emissions and the resulting ambient con-

Printer-friendly version

Discussion paper

centrations. Gaining a greater understanding of these processes, their impacts and uncertainties is of great importance for air quality modeling. The design of the experiment, changing a variety of model parameterizations in turn, provides a wealth of data for this study. The analysis of the impacts on biogenic emissions (isoprene and lumped aldehydes) is interesting and useful for the community. However, the results on the effect on surface concentrations are presented without any consideration of the atmospheric chemistry that might affect their results. Isoprene reacts with OH very quickly, and OH distributions are likely influenced by the meteorological changes (clouds, humidity). This should at least be mentioned and preferably OH fields also shown, to allow a greater understanding of the changes in isoprene among cases.

Reply1: We are grateful for this clue. In this context, OH provides useful insights in relation to isoprene concentrations and downstream reactive atmospheric chemistry. Therefore, we added an analysis of OH surface concentrations including Fig. 8 (Fig. 1 in this document) and Subsection 5.2 (l. 406-419 new count) in the manuscript (replacing aldehyde concentrations, as described below):

" The hydroxy radical OH is a highly reactive oxidant in the atmosphere acting as most important sink of isoprene (Kaser et al., 2015). Generally, OH may be influenced by the model configuration via reaction with biogenically emitted gases and meteorological conditions. Local meteorology mainly affects OH by changes in radiation related to humidity and clouds. In this specific case, the weather in the Po region was continuously characterized by clear and dry conditions as described in Sect. 2.1. Thus, no significant differences in humidity and cloud coverage are simulated by the model configurations (not shown). This renders the differences in OH concentrations being determined by changed biogenic VOCs.

As expected from atmospheric chemistry, daytime OH concentrations shown in Fig. 8 are reduced in regions of high BVOC concentrations like the central Po valley and the southern Apennines. In contrast, OH concentrations remain comparably high in the mountains and over the ocean were isoprene concentrations are neglectable. This direct dependence of OH to

biogenic gases causes also significant differences in OH concentrations with respect to model configuration. In this case, the effects are most dominant in cases of increased isoprene concentrations with respect to the reference simulation. Significant reduction of OH is induced by excluding drought response ("no SMOIS"), RUC LSM, and less pronounced for MODIS land use in the southern Apennines. While these reductions are persistent in time, increased isoprene concentrations in the central Po valley for GFS global meteorology at 06 UTC result in temporally reduced OH concentrations in this region. "

Related modifications have been made in the conclusions (l.467-470 new count) as well as the abstract (l.12-14 new count):

ABSTRACT: *" As a result, large sensitivities to model configuration are found for surface concentrations of isoprene as well as OH, affecting reactive atmospheric chemistry. "*

CONCLUSIONS: *" Moreover, changes in surface concentrations of biogenic trace gases induce significant differences in OH concentrations affecting reactive atmospheric chemistry. Excluding the emission response to drought stress reduces local OH concentrations by up to a factor of three in this study. "*

It would also be interesting to use the sensitivity studies performed for this work to analyze the intensity of segregation and see how the model parameterizations affect that and thus the chemistry. See Kaser et al., GRL, 2015, doi:10.1002/2015GL066641

Reply2: The investigation of dynamical separation of chemical compounds provides a very interesting aspect in the context of this study. We thank the reviewer for pointing towards this topic which is skillfully exposed in Kaser et al, 2015. As described above in Reply1, we added the evaluation of OH fields in the manuscript which provides useful information in this context. However, - in our study context and related objectives - we believe to perceive two critical points, which hinder an addition of segregation in the

current study:
1. In our current setup, the temporal resolution of emission information is not suitable for calculating fluctuations as required for the intensity of segregation. For computational reasons, MEGAN emissions are not calculated every timestep (currently: every 1800 sec.).
2. According to our opinion, a thorough investigation of segregation as proposed in Kaser 2015 would require a dedicated extra investigation of related processes which would be too voluminous to be added in this study.
Clearly, the Kaser et al. study clearly suggests such an investigation in a follow-up study. At this point we suggest to confine to advise the reader to this item and added a reference to Kaser 2015 as example for the complexity of chemistry-turbulence interactions in the introduction (l. 42-44, new count):

" An example of highly complex chemistry-turbulence interactions is found by Kaser et al. (2015) who investigated effects of local separation of isoprene and OH. "

I have greater concern about the aldehydes results because aldehydes have a large secondary production that is not even mentioned in the paper. So the surface concentrations will be affected not only by biogenic emissions but chemical production (from isoprene and anthropogenic sources). It might have been better to study something like methanol, which has large biogenic emissions, longer lifetime and no secondary production. Thus, in order to keep Section 5 in the paper, much more discussion should be included about the impact of atmospheric chemistry on the surface concentrations.

Reply3: We fully agree with the reviewer that aldehydes are affected by a large number of processes like secondary production. Individual effects of those cannot be separated by the approach used in this study. We therefore decided to withdraw the analysis of aldehyde concentrations from Section 5. Instead, we added the investigation of OH (as described above) and noted this issue in the beginning of Sect. 5 (l.386-389 new

count) as follows:

" The evaluation of biogenic gas concentrations is restricted to isoprene because of its direct dependency on the model processes discussed above. Other biogenic gases are affected by additional processes like secondary production which hamper a detailed evaluation. Instead, resulting OH concentrations are analyzed on their impact on reactive atmospheric chemistry. "

Unfortunately, studying methanol concentrations cannot be pertinently addressed in our modeling system. EURAD-IM uses the RACM-MIM chemistry mechanism which handles methanol in a chemical group (HC3) including other non-biogenic compounds like ethanol and propane. Thus, individual methanol concentrations are not available and a discussion of the whole chemical group would suffer from the same concerns as for aldehydes. Nevertheless, we agree that methanol is an interesting biogenic compound, so we added the biogenic emissions of HC3 (which solely refer to methanol emissions) and other BVOCs in Section 4.1. We thank the reviewer for pointing this out and hope that we could adapt the manuscript in a sufficient way. We updated Fig.2 of the manuscript (Fig. 2 in this document) and included a new figure showing the additional BVOCs (Fig. 3 in this document) which is now the new Fig.3 in the manuscript. The description of Section 4.1 was modified accordingly and reads now (l.272-305, new count):

" The effects of model configurations on biogenic emissions of different gases are given in Fig. 2 and Fig 3. As the changes induced by the different model configurations are similar for all presented biogenic gases, the following description focuses on isoprene and HC3 shown in Fig. 2. Note that biogenic HC3 emissions refer solely to methanol which is the only biogenically emitted compound in this chemical group defined in the model.

Differences between nighttime (03 UTC) and daytime (09 UTC) emissions are more significant for isoprene than for other biogenic gases. This is because isoprene is a direct product of photosynthesis which is mainly limited to daytime conditions. For the reference setup ("ref"), daytime

isoprene emissions are mainly restricted to the Apennine Mountains and two areas within the the central Po valley north of Modena and Bologna. According to USGS land use, these locations are assigned to "Deciduous Broadleaf Forest" and "Crop/Woodland Mosaic", respectively. In contrast to "Dryland Cropland and Pasture" in the rest of the valley, broadleaf trees emit high levels of isoprene. Thus, even small numbers of trees result in significantly increased local isoprene emissions. Biogenic emissions of alpha-pinene, limonene and aldehyde show also increased these regions, but with decreasing characteristic. In contrast, biogenic emissions of methanol and aldehydes almost equally emitted by all vegetation types in this regions. This results in a comparably uniform distribution over most parts of the domain with a significant reduction in the Apennine mountains.

The high dependency on tree coverage is emphasized by comparing reference biogenic emissions to emissions based on MODIS land use ("land use"). In contrast to USGS, MODIS does not indicate any trees within the Po valley, which results in negligible biogenic emissions in this region. Although this effect is most prominent for isoprene, significant emission reduction is found for all considered biogenic gases. At the same time, the whole Apennine Mountains and southern foothills of the Alps are assigned to high coverage of broadleaf trees resulting in high isoprene emissions. The use of GFS global meteorology does not change the general emission patterns ("global"). Caused by different initial- and boundary conditions, all biogenic emissions are slightly reduced in the whole region. The implemented response of biogenic emissions to soil dryness significantly influences biogenic emissions ("no SMOIS"). By neglecting this response, emissions are considerably larger than for the reference case, especially in the southern part of the domain. As soil moisture decreases after sunrise, the largest sensitivities are found at 09 UTC for both gases.

The RUC LSM induces slightly increased biogenic emissions of all considered gases, in most areas ("LSM"). This general increase is overlapped by a drastic reduction to almost zero emissions in the south-eastern parts of the Po valley for all gases - most prominently visible for biogenic methanol and aldehyde emissions. This reduction is caused by low soil moisture predicted by RUC LSM in the morning hours which results in drought induced plant

Printer-friendly version

Discussion paper

stress. Using the ACM2 boundary layer- (PBL) and Pleim-Xiu surface layer (SL) schemes instead of MYJ PBL + Eta SL schemes leads to a reduction of biogenic emissions ("PBL + SL"). This effect affects the biogenic emissions of all considered gases and is largest in the eastern central Po valley. Only minor changes in biogenic emissions due to microphysics- and radiation schemes are visible ("microph.", "rad."). Using TGS microphysics instead of the reference WSM6 does only induce small local effects during nighttime (03 UTC). Although being small, effects of using different radiation schemes after sunrise can be attributed to different formulations of shortwave radiation by the Dudhia and RRTMG schemes. "

Accordingly, a short note on the model variable "HC3" is given in the introduction (l.92, new count):

Methanol is part of the model variable "HC3" which also includes ethanol and propane as not biogenically emitted alkanes.

Additional comments:

- **l.90: aldehydes are also photochemically produced.**

  Reply: Yes indeed, the reviewer is right. We added this information at the referred position manuscript (l. 93-94, new count):
  " The model variable "aldehyde" represents a composite of oxidized BVOCs which are affected by several processes including biogenic- and anthropogenic emissions, photochemical production, atmospheric transport and dry deposition. "

- **l.236+: You might want to refer to Jiang et al., https://doi.org/10.1016/j.atmosenv.2018.01.026 for a discussion of the implementation of drought impact in MEGANv3**

  Reply: Thank you very much for this insightful hint. We are happy to refer to this publication by adding the following sentence in l.258 (new count):
  " Recently, Jiang et al. (2018) formulate drought response in the subsequent version MEGAN 3 as function of photosynthesis and generalized water stress. "

- **Figure 4: It is very difficult to pick out the differences highlighted in the discussion in these tiny panels. It would be nice to find another way to illustrate these differences. Perhaps all except the first column should be differences (percent) from the ref case. Or show just averages over the Po Valley and other regions of particular relevance (not on a map).**

  Reply: We agree that the differences are less obvious for friction velocities compared to the other variables shown. We made attempts with the suggested presentation scheme. However, plotting relative difference (i.e. by factors) shown in Fig. 4 of this document may be misleading as they appear to be dominated by small differences of low absolute values at 3 UTC. Thus, showing absolute values provides a visible measure of the magnitude of differences, also in comparison to the other variables. Therefore, we came to the conclusion that the important aspects are more intuitively visible in the original plot, than for factors.
  This is also the case for the newly added surface concentrations of OH in Fig. 8 of the new manuscript.

- **Figures 6 & 7 are also too small - they could at least be enlarged to the width of the page, but difference plots would help illustrate features.**

  Reply: Figures 7 and 8 (new count) have been enlarged which makes it easier to pick individual features.

- **There are a number of spelling and grammar errors, but the paper is understandable.**
  Here are some corrections: l.112: through → trough
  l.269: "neglectable" → negligible
  l.305: "friction velocities does only" → "friction velocities only"
  l.307: do you mean ?exemplary? (best of its kind) here, or perhaps just "example" or "representative" [similarly elsewhere in the paper]
  l.309: does → do
  l.353: plain → plane
  l.364: hove → have
  l.419 "so large" → "to large"
  l.420 add comma after "common"

Reply: We apologize for our trivial mistakes and are grateful for pointing them out. The manuscript was corrected accordingly.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-608, 2020.

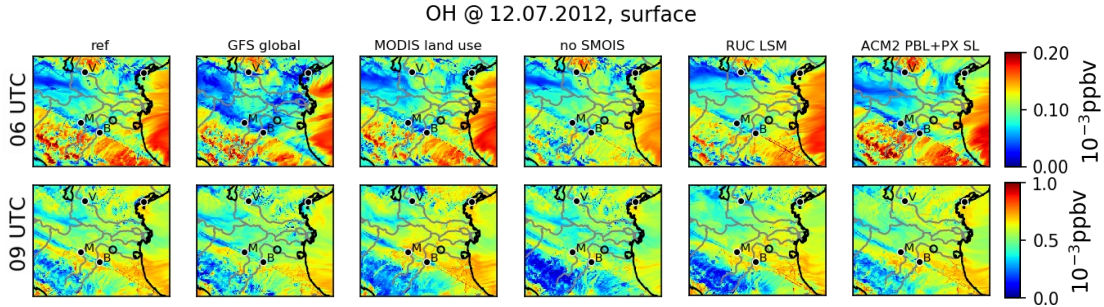

Fig. 1. OH surface concentrations on 12 July 2012 at 06 and 09 UTC (coded by colors) for different model configurations.

Printer-friendly version

Discussion paper

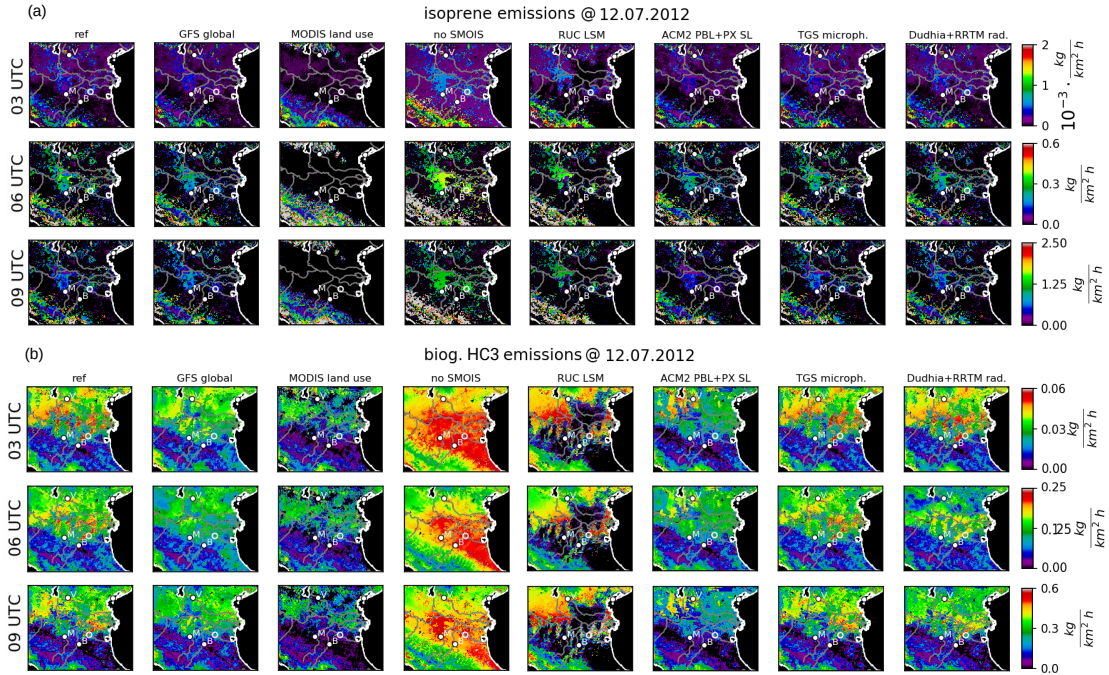

Fig. 2. Isoprene (a) and biogenic HC3 (b) emissions on 12 July 2012 at 03, 06 and 09 UTC (coded by colors) for different model configurations

Printer-friendly version

Discussion paper

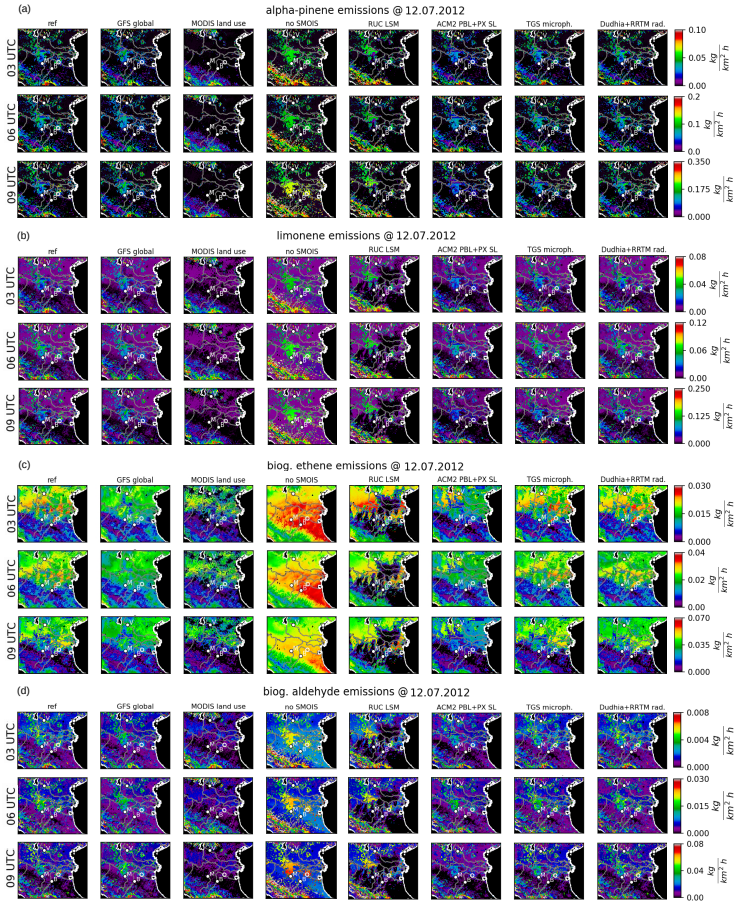

Fig. 3. Alpha-pinene (a), limonene (b), biogenic ethene (c) and biogenic aldehyde (d) emissions on 12 July 2012 at 03, 06 and 09 UTC (coded by colors) for different model configurations.

Printer-friendly version

Discussion paper

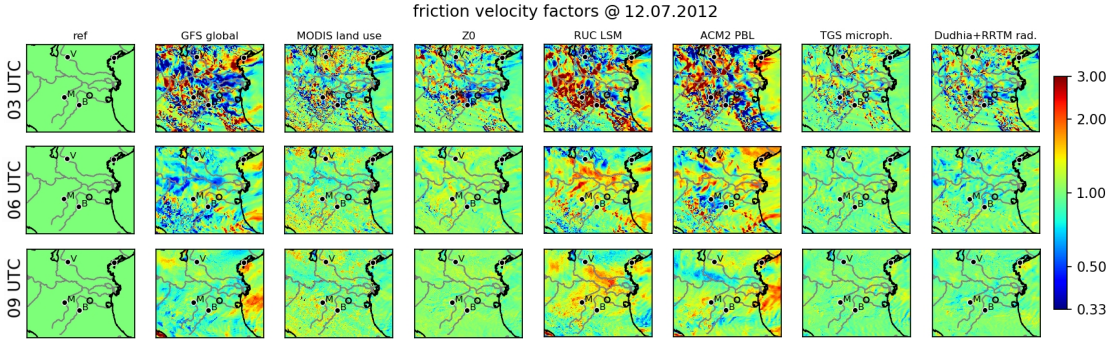

Fig. 4. Friction velocity factors on 12 July 2012 at 03, 06 and 09 UTC (coded by colors) for different model configurations including increased roughness length ("Z0").

Printer-friendly version

Discussion paper