

We thank the reviewer for his/her comments. Here they are reported (in bold) with our replies.

The authors presented a comprehensive evaluation of an updated and expanded EMAC model. The implementation of explicit mechanisms and the gas-particle condensation module to the chemistry GCM can be very useful for studying organic vapors and their chemistry in the atmosphere. The model evaluation was made on OH, CO, some key VOCs, AOD and the chemical components of PM2.5. Significant biases present in OVOC simulations, whilst the model performance for aerosol composition is similar to that of previous model configurations. The reasons that lead to the model biases were explored but I feel there are still somewhat insufficient which may be improved with more discussions. Overall I think this is a relevant, important paper and recommend it to be accepted after addressing the following comments.

We thank the referee for the positive feedback.

(1) Line 81: Are those emissions reasonable compared with other global model inputs or estimates? It is better to add a few sentences to discuss it. It looks like the work did not apply regional emission inventories (e.g., NEI for US, EMEP for Europe, MIX for East Asia or MEIC for China). Would this affect the simulations in anthropogenic-influenced regions? In particular, for aerosol composition, I think the regional inventories may matter significantly.

The emissions database used in this work has been extensively compared with other global emission databases by Crippa et al. (2018), and therefore it is not repeated here. It can be mentioned that differences can be up to “43% for BC” (Crippa et al., 2018). As we needed to have a global emissions dataset, we adopted the one we considered most realistic at the time of the simulation, although different global emissions dataset are possible within the EMAC model (see for example Jöckel et al., 2016; Reifenberg et al., 2021). As mentioned by the referee, we did not apply any regional emission inventories: as we were not focusing on specific locations, we preferred to adopt a homogeneous global emission dataset rather than over-imposing regional emissions datasets. It must, however, be stressed that the uncertainties in these global emissions are often similar to the regional ones. For example Saikawa et al. (2017) compared 2 global and 3 regional emissions dataset for China, showing that “large disagreements exist among the five inventories [...]”. As a comparison between different emissions datasets is outside the scope of this study, we will simply refer to the work of Crippa et al. (2018) for the details.

(2) Section 3.2.5: I think the authors only used the EANET data for 2010. Please specify.

Yes, we used the data for the year 2010. Thanks for pointing this out. We will correct this in the revised version.

(3) Line 238-239: What kind of biogenic CO precursors?

An example would be methanol CH_3OH , as presented by Rosanka et al. (2021b) and in Sect.4.2.3 in our manuscript. Also CH_3COCH_3 (see Sect.4.2.4) is overestimated in these regions. We will refer to these sections in the revised version of the manuscript.

(4) Line 280-281: How high? I am surprised that temperature difference can be significant between the model parameters and the observations. If the overestimation

is due to excess of biogenic VOCs, it is better to compare the surface concentrations of isoprene etc. Some measurements are available in those tropical areas and the data are published (e.g., from AMAZE-08).

The referee is correct. The bias in temperature is indeed very limited, as shown by Hagemann and Stacke (2015). Nevertheless, it must be stressed that even a small overestimation can have a strong impact on the emissions. As shown by Guenther et al. (2006, Fig.4), the MEGAN model shows a strong response of the emissions on changed temperature. Once plants are exposed for several days to temperatures above 300 K, MEGAN predicts strong dependencies of isoprene emissions on actual temperature, so that even a difference of a few degrees implies large emission differences. Following the referee's suggestion, we compared the simulated isoprene mixing ratios in the Amazon rainforest with observations from the AMAZE-08 campaign (Martin et al., 2016) and from the ATTO tower (Yáñez-Serrano et al., 2015). In comparison to the AMAZE-08 campaign, the simulated isoprene measurements are overestimated by more than a factor of 3 (6.1 ± 1.2 ppb simulated and 1.9 ± 1.4 ppb observed), while the overestimation factor for the ATTO tower is on average approximately 1.6, being higher in February/March and lower in October/November.

(5) Consistent model underestimation present in mid- and high latitudes of Northern Hemisphere for CH_3OH , CH_3COCH_3 , HCOOH , CH_3COOH , and PAN. The authors made some explanations. For example, Biogenic or biomass burning VOCs emissions might be too low during summertime in those regions. In-cloud chemistry may happen. In what regions, such in-cloud chemistry can be a significant source (e.g., HCOOH)? This model bias is too consistent, which made me wonder how this is related to underrepresented emission inventories of VOCs in EDGAR (e.g., contributed by outflow of polluted areas). If biomass burning is not well represented, what does the model-satellite comparison look like for different seasons? Or if it is biogenic, would the simulation become better in winter? I think the current discussion about the potential bias is a bit too general.

Indeed the model bias is very large for those species at high latitude. We agree with the referee that the anthropogenic emissions could be underestimated, but this would unlikely explain such a homogeneous underestimation. As example, Jacob et al. (2002) showed that anthropogenic emissions would roughly contribute only to 25% of the total CH_3COCH_3 budget. The underestimation in our model at high latitude is roughly of 50%, i.e. larger than the total anthropogenic contribution. Furthermore, if anthropogenic emissions would be the main source of error, we would find a constant underestimation also near source regions, which is, again, not the case for CH_3COCH_3 . On contrary, for CH_3OH , the anthropogenic emissions can easily be distinguished in the model results, both, in North America and in South Asia (see Fig.6), and they are clearly higher than the satellite observations suggest.

In Fig. 1, the total column from EMAC and IASI data are compared for latitudes between 30° and 65°N . As the patterns of the comparison between the different VOCs and the model results are different, we can suggest also different reasons for the underestimation. While for the acids HCOOH and CH_3COOH a clear major source is missing (see for example Franco et al., 2021), for PAN and CH_3COCH_3 the observed seasonality is not reproduced, suggesting a more fundamental problem in the numerical simulation of such tracers. It must be stressed that at high latitudes in winter, the poor observational conditions reduce significantly the spectral signal recorded by IASI and hampers meaningful retrievals of the VOCs (see Franco et al., 2018). Hence, the temporal and spatial coverage of the satellite measurements is limited at these latitudes, and the results must

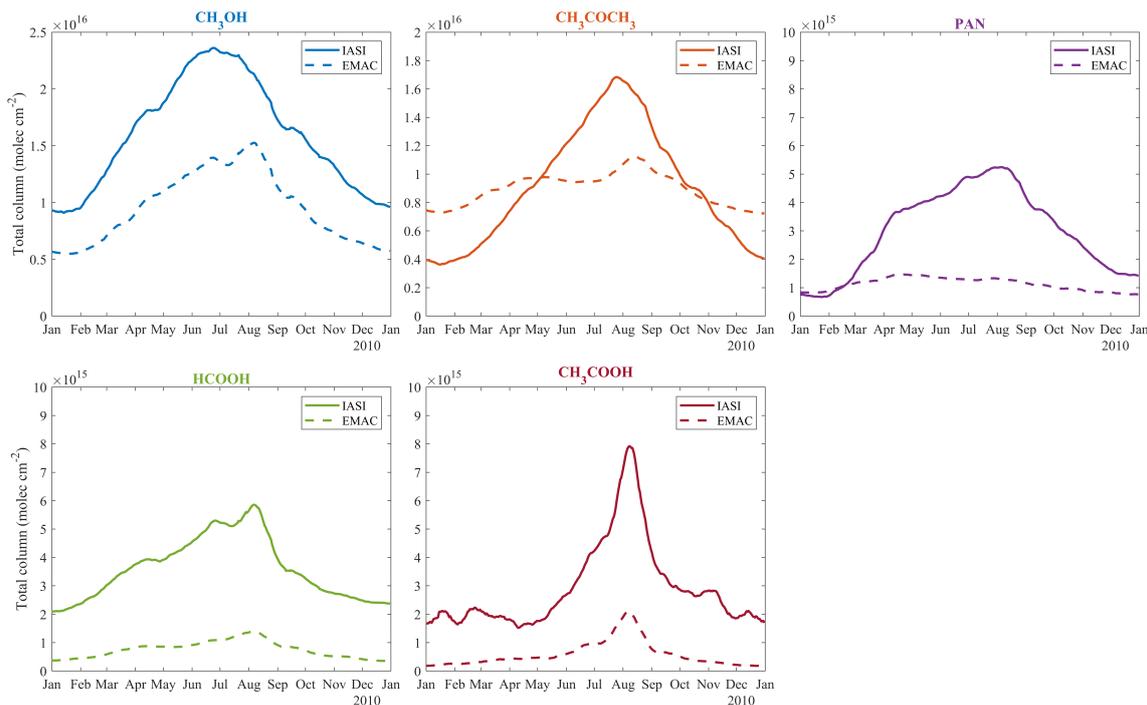


Figure 1: Daily and spatially averaged VOC columns from IASI and EMAC between 30° and 65°N throughout the year 2010. Data were considered only when both IASI and model results were simultaneously available.

be taken with caution. Finally, as the biomass burning period is also in phase with large biogenic emissions (due to high temperature and solar radiation), it is difficult to draw a general conclusion here.

In summary, due to their different budget, a more detailed analysis is necessary to understand the reason of the discrepancies between model results and observations for the mentioned trace gases, which is well beyond the scope of this study. This work should, however, present the weakness in representing such gases, so that future work could use this study as a starting point.

(6) Line 389-390: What are those concentrations? The magnitudes seem not matching with annual mean surface concentrations of OH.

We are indebted to the Reviewer for spotting this typo (units factor exponent should read "5" instead of "15") which occurred during the typesetting of the manuscript. Indeed, typical average tropospheric concentrations of are in the order of 10^5 – 10^6 molecules cm^{-3} .

(7) Section 4.3: What about the model performance of surface OH in different regions? This would affect VOC chemistry and SOA formation significantly. Surface OH concentrations can be quite high in polluted areas (e.g., in China). A recent paper pointed the model underestimation of OH because of the lack of HONO source [Miao et al., 2021]. Would that be a problem in the updated model presented here?

Here we intend to show that the model is reproducing OH similar to that in previous simulations with EMAC. A detailed study on OH and its reactivity on a global scale from this model results is planned in the near future. It has been also shown that the model HO_x budget agrees well when compared to observation in the free troposphere (Tadic et al., 2021). Nevertheless, we

agree with the referee that a comparison in polluted regions at the surface could be problematic, also because of the apparent missing HONO source (see for example Elshorbany et al. (2014) for the approach mimicking HONO sources in EMAC, not adopted in this study). We, however, refer to the answer to referee#2: in an evaluation, of a global model of such complexity as the one presented here, the background conditions should be evaluated at first, showing that the model is able to reproduce such conditions. More specific studies on polluted areas should be conducted with dedicated simulation(s) and with more focused analyses.

(8) Section 4.5: Is the model performance lower in winter than in summer? Residential coal burning in China seems to be a big source of PM_{2.5} associated with large uncertainty in the inventory. Would that affect the comparison in Asia?

Sadly this cannot be investigated with the dataset of Hammer et al. (2020), as these data are provided as annual average (albeit at high spatial resolution). On the other side, we believe the referee to be completely right: as shown by Pozzer et al. (2012, 2017), and more recently by Miao et al. (2020), BC and OC are very important for the PM_{2.5} budget in East and South Asia. We think that the large uncertainties associated with such emissions do strongly affect our results in these regions.

(9) Line 470-473: What do the model values in higher vertical layers compare with the observations?

We have compared SO₄²⁻ concentrations with observations from aircraft campaigns, as compiled by Heald et al. (2011), similarly to Fig. 26 of the manuscript. The results are presented in Fig.2. As mentioned in the manuscript, the stations present at high altitude were not representative because of the difficulties in reproducing the correct orography by a model of such resolution. Nevertheless, the simulated sulfate concentrations agree well with the aircraft observations in the free and lower troposphere, with an underestimation in a few cases (see Fig. 2, ITCT-2K4, ADI-ENT or IMPEX campaigns), but always within the measurement standard deviations.

(10) Line 487-488: Nitrate overestimation seems a general problem in chemical transport models. A main reason could be the inappropriate deposition parameters [Miao et al., 2020; Zhai et al., 2021]. But more pronounced bias in winter than in summer is inconsistent with the other studies.

Indeed, the referee is correct that a more pronounced bias in winter is inconsistent with other studies, and this was acknowledged in the manuscript (see lines 482-484). Due to the intricate chemistry involved in the aerosols budget, we prefer not to draw any conclusions or hypothesis for the behaviour of the simulated nitrate.

(11) Section 4.6.5: It would be good to make some connections for model biases on species. For example, how would the significant biases for the oxygenated organics discussed in the paper affect the OA simulations? So does OH as I mentioned in 7.

Following the referee's previous comments, it is clear that the possible underestimation of OH over polluted regions will result in an underestimation of Organic Aerosols (OA) in the same region. This is visible in Fig. 25 of the manuscript, where the few locations in highly polluted regions over east Asia are strongly underestimated. Nevertheless, it is hardly possible to comment on what effect could have the underestimation of the oxygenated organics mentioned in the paper to the OA. First of all, the discussed trace gases are too light to contribute significantly to OA directly,

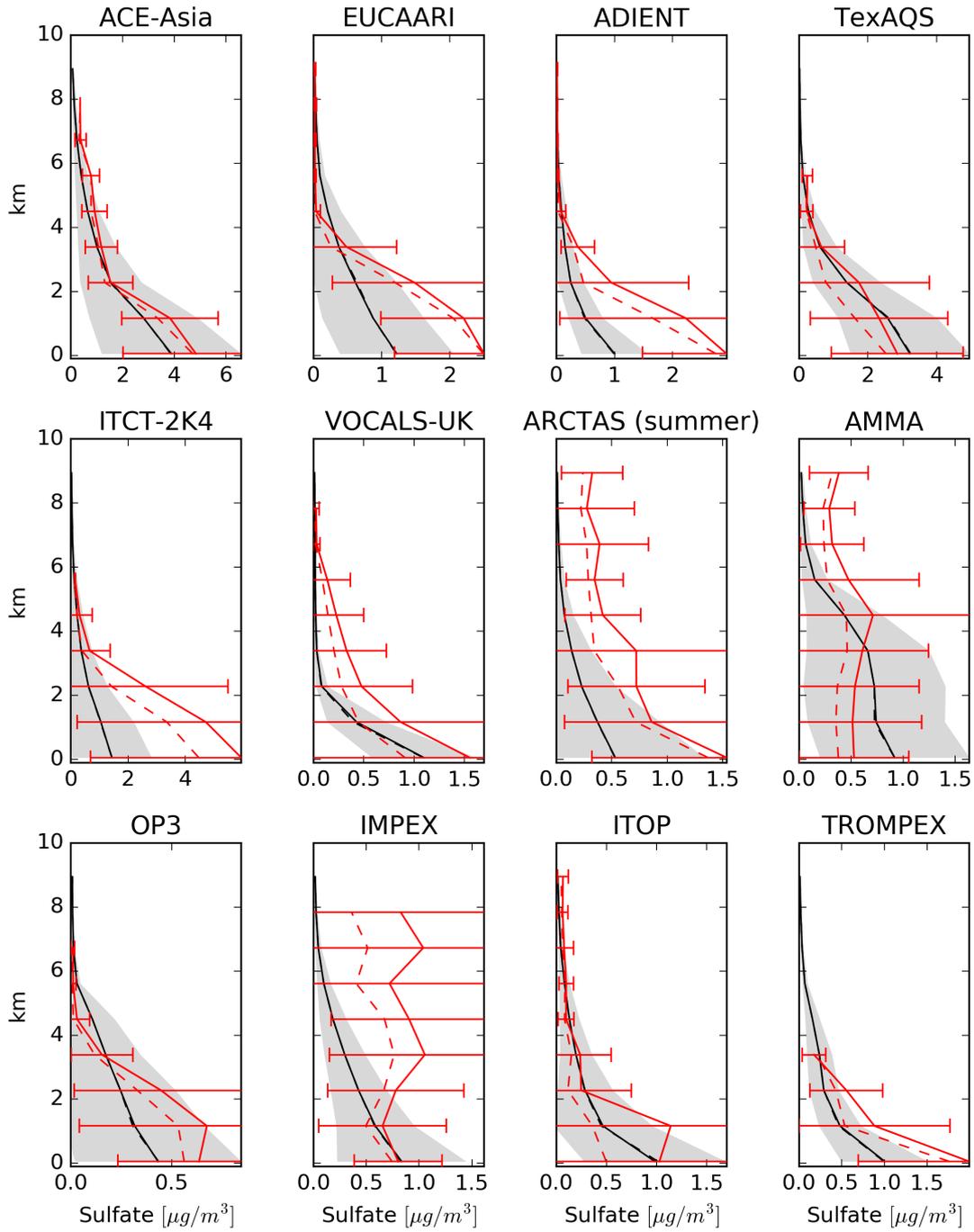


Figure 2: Model results of mean vertical profile of sulfate for selected field campaigns in black, and spatio-temporal standard deviation as grey area. The observed mean values are depicted in solid red, with the bars representing the standard deviation of the observations. The observed median is presented as red dashed line.

even for products of their oxidation chain. Although some of these might contribute to OA mass through aqueous phase chemistry, this process is not (yet) included in the model. Secondly, no OA observations are present for the regions of underestimation, so it cannot be confirmed that the simulated OA deviates from the observations in these regions. Any discussion on this point would

be extremely speculative and we prefer to avoid it.

(12) Line 553- 562: The observations in Asia used here for aerosol composition are quite limited. Studies have suggested the emission inventories for semivolatile and intermediate volatility organic compounds are insufficient and lead to the majority of the model biases in simulating OA [Miao et al., 2021; Zhao et al., 2016] in regions that are influenced significantly by anthropogenic emissions. In Europe, residential wood burning also contributes a lot to the primary OA in winter. Would that be a possible bias? For the rural areas, fragmentation of OA is not included in the model, which may contribute to the overestimation over North America. OA includes both primary OA and SOA. It might be more clear if the two cases are separated discussed.

We thank the reviewer for pointing out these potential sources of discrepancy between our model and observations. In contrast to other observational networks (e.g., EMEP, IMPROVE), EANET does not perform measurements of OA. Therefore, the observations presented here are based on individual field campaigns, which usually last for approximately one month, and not from a network of observational sites that routinely collect samples throughout the year. Therefore, given that the model simulation lasts for only one year, the number of observations used are limited compared to other components (e.g., sulfates from EANET) or other regions (e.g., OA from EMEP in Europe or from IMPROVE in USA). This is emphasized now in the revised text.

ORACLE simulates the contribution of SVOC and IVOC emissions from fuel combustion and biomass burning to OA formation, including additional IVOC emissions that were not accounted for by the traditional emission inventories. However, we do agree with the reviewer that the SVOC/IVOC emission factors used are highly uncertain and can add to the model bias.

Residential wood burning has been identified as the main source of discrepancy in model bias over Europe during winter (Tsimpidi et al., 2016). This is already mentioned in the text (i.e., as biomass burning). In the revised text, this is clarified by replacing the "biomass burning emissions" in line 557 with "biomass combustion emissions from residential heating".

The effect of fragmentation is included in the aging scheme of ORACLE. The oxidation of organic compounds can result in products with different volatilities due to functionalization (reducing the volatility) or fragmentation (increasing the volatility). To minimize the computational cost, ORACLE uses a simple photochemical aging scheme that efficiently simulates the net effects of fragmentation and functionalization of the organic compounds. However, even the conservative aging scheme of ORACLE assumes that the net volatility transformations point to only one direction (functionalization) which can lead to increasingly higher OA concentrations in lower-volatility bins. This may result in an overestimation of OA at long aging timescales downwind of the source regions. This overestimation is partially offset by the use of MOM for simulating explicitly the degradation of the traditional VOCs.

Our manuscript focuses primarily on the evaluation of the model performance for the tropospheric gas phase composition and secondarily on the general properties of the aerosol composition. We believe that an in-depth analysis of the simulated organic aerosol composition along with AMS factor analysis data can provide valuable insights on the benefits of the proposed modeling approach and we have left this analysis for a planned future study.

Technical remarks [...]

All the technical remarks will be taken into account in the revised version of the manuscript

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