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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air quality over Europe: modeling gaseous and particulate pollutants and the effect of precursor emissions" *by* E. Tagaris et al.

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My major concerns with the current version of this manuscript are listed below. Comment: 1. I think that the focus of this manuscript (scaling and impact of precursor emissions) should be emphasized after certain changes are made to the modeling system. In accordance with Dr. Odman's comments, I think that updating the treatment of organic aerosol in the model is necessary before even looking at particulate pollutants and PM2.5. The inclusion of the volatility basis set (VBS) scheme, which treats both primary and secondary OA as semi-volatile and chemically reactive is nowadays a common practice to all new papers using regional models (that are at least published

in journals such as ACP). This cannot be disregarded here. The use of the VBS will change concentration levels of PM2.5 but also of gaseous pollutants as the availability of oxidants will change (Zhang et al., acpd, 2012; Athanasopoulou et al., 2013; Bergström et al. 2012).

Response: First of all, we would like to point out that that there must be a misunderstanding here; the focus of the manuscript was not the scaling of precursor emissions which was clearly stated in the manuscript. However, agknowledging the uncertainties that the use of such an approach may introduce, in the revised version of the manuscript the section related to scaling the emissions has been removed. We are a little bit confused by the reviewer suggestions for updating the organic aerosol formation in the model which he/she says that is in accordance with Dr. Odman suggestion. Dr. Odman proposed to include a discussion after scaling only NOX and SO2 in order to further explore uncertainties related to NOX and SO2 emissions and not updating model mechanism for aerosol formation. CMAQ model is a state of the art air quality model and the version used here is a quite up-to-date version where several new pathways for secondary organic aerosol (SOA) formation have been implemented (Edney et al., 2007; Carlton et al, 2008), as mentioned in the manuscript. Very recently some groups have started using the volatility basis set (VBS) scheme proposed by the reviewer. We suppose that the reviewer does not suggest working in the direction for changing aerosol modules in the CMAQ accounting VBS schemes. However, we think that this is not an issue because: 1. We clearly state in the manuscript the modeling system used and CMAQ is a state of the art model widely used for air quality simulations. 2. Using VBS schemes is still under evaluation. For example in the reference suggested by the reviewer (i.e., Bergström et al. 2012) where a modeling work of organic aerosols over Europe (2002–2007) using a VBS framework is applied the authors say that they cannot reproduce the winter level of organic aerosols in Europe, suggesting an issue probably related to the emission inventories Furthermore, they state that: "However, given the limitations of any OA-modelling scheme, including uncertainties surrounding emissions, formation, and other modeling issues, there is of

course a danger that results are improved for the wrong reasons. As one example, an increase in BVOC emission rates might give comparable effects to an increase in aerosol yields or aging rates. Another example would be that the high ASOA yields predicted by VBS might be masking problems in POA emissions. Untangling these effects is a real challenge." 3. Tsimpidi et al. (2010) has published in ACP modeling results using a VBS approach with PMCAMX for Mexico City. Later, Tsimpidi et al., (2012) has published in AWMA modeling results for an air pollution episode in northwestern United States using CMAQv4.7 (i.e., the version of the CMAQ model that we have used) without mentioning a limitation in their results because of the absence of the VBS approach.

Comment: 2. In relation to 1, there is no further model development in the specific modeling system used here, compared to previous efforts (application of CMAQ in the US domain) by the same author a few years ago. The WRF model is now preferred for the meteorological description as it is considered an improved version of MM5. Why the authors are still using MM5 ? I think that updating to WRF will be an important part of further model development which would strengthen the paper.

Response: We are very glad that the reviewer is familiar with our previous work. Indeed, we use again Models 3 as in our previous modeling work for the US domain, however the modeling setup is completely different since: a different version of CMAQ is used (v.4.7 instead 4.4) with a different chemical mechanism (CB05 instead of SAPRAC99) in a different domain (Europe instead of USA) with different emissions inventory and meteorological fields. What we would like to point out here, is that in the current work we are not doing model development but an application of a modeling system for air quality studies. We agree with the reviewer's comment that WRF is now a commonly used meteorological model and would definitely be the model of our choise if we were going to work on air quality forecasting or to simulate a new episode. However, in the framework of the AQMEII exercise, readily available meteorological fields have been delivered to us by Laboratoire des Sciences du Climat et de l'Environment, IPSL,

CEA/CNRS/UVSQ (see acknowledgment) and the related discussion on meteorology used has been presented by Vautard et al., (2012), as mentioned in the manuscript. Given that MM5 is still considered a highly credible meteorological model, we believe that its use in the current manuscript does not deplete the value of our outcomes.

Comment: 3. A significant limitation of the manuscript is the lack of related literature review. There are other regional models that have been applied to the European domain in a similar way. The authors should cite these as well as try to compare their results with those of others. Furthermore, there is even the same model (CMAQ v4.7) that has been recently applied in Europe (although in a smaller domain), however it is not referenced at all here. Some well known regional air quality models that have been recently applied in the European domain and are not referenced here are the CHIMERE model (Zhang et al., acpd, 2012), the EMEP (Bergström et al., 2012), the PMCAMx (Fountoukis et al. 2011), etc. Moreover, Im et al., Atmos. Environ, 2012, and Im and Kanakidou, acp, 2012 have used CMAQ in Europe. The authors should reference these papers as well as comment on differences and similarities between their version of CMAQ and the above CMAQ application. Most of the above model applications have used the MEGAN model for the calculation of biogenic emissions. The authors are using a different one. A paragraph is needed explaining in what basis this choice was made and what are the differences (in numbers) in the calculated biogenic emissions in Europe. I am assuming that there are large differences given the associated uncertainties.

Response: There are a huge number of papers related to air quality studies for the European regions. Since our manuscript is not a review paper to cite as much papers of the literature as possible we tried to cite papers closely related to our work focusing on modeling applications for the whole Europe. Moreover we compare our results with two other modeling studies over whole Europe using the CMAQ model (i.e., Pay et al., (2010) and Appel et al., (2012)). The reason for not including the proposed by the reviewer papers in our manuscript is because they cover a very small part of our

domain (i.e, the two papers by Im et al., focused on Greece and Turkey while Zhange et al., focused on Greater Paris Region) or we do not have common pollutants (i.e., the paper by Fountoukis et al. focused on PM1) or different approaches (i.e., Bergström et al. examines the performance of a new organic aerosol module and four different volatility basis set (VBS) schemes). However, in order to comply with the reviewer suggestions we have cited four more air quality studies over Europe. With all the respect to the reviewer's comment we cannot understand his/her concept asking to comment on why we have used a different model from other groups in estimating biogenic emissions. BEIS is a well known model widely used for biogenic emissions. Moreover, SMOKE modeling system includes the BEIS model for computation of hour-specific, meteorology-based biogenic emissions from vegetation and soils. Indeed, there are differences between the models but a discussion about biogenic emissions is out of the scope of that manuscript since a manuscript focused on biogenic emissions is ongoing. However, for an accurate comparison between different models a work focusing on that is necessary where the same data will be used since biogenic emissions are temperature and light dependent. In this way, a comparison against aircraft-based measurements in the eastern United States and Texas suggest that the emissions from MEGAN were higher than the emissions modeled from the isoprene measurements, whereas emissions from BEIS were lower. Moreover, MEGAN over-estimated isoprene by up to a factor of 2 compared to BEIS (Warneke et al., 2010 at JGR).

Other comments.

Comment: P6683: L9: The abstract should be self-explanatory. This sentence doesn't make much sense.

Response: This sentence (i.e., P6683: L9) is not part of the "Abstract" but belongs to the "Introduction" part of the manuscript.

Comment: L15: Which model?

Response: The model has been added in the manuscript.

Comment: L23: Not clear. Re-phrase.

Response: Done.

Comment: L28: Not clear. Re-phrase.

Response: Done.

Comment: Comment: P6694: L15: "..the effect of precursor emissions." On what?

Response: It has been removed from the revised version of the manuscript (the reviewer is referred on page 6684).

Comment: L16-19: But you don't make a big effort towards comparing these results with others.

Response: In the revised version of the manuscript we have extended the comparison of our results with the results from similar studies.

Comment: P6686: L8-10: What are the default values?

Response: We have added the reference.

Comment: L11: what is the prevailing direction?

Response: In order to avoid confusing the readers, the statement has been removed and re-phrased.

Comment: L12: 10 days? Why such a large spin up time is needed? Isn't 2-3 days enough?

Response: We believe that a real issue would be the opposite case (i.e., if we have used 2-3 days of spin up time instead of 10 days). Since the domain is big and we have available data we have used a 10 day spin up time.

Comment: L13-14: What are the new pathways? Please describe.

Response: The related references are stated.

Comment: L24: By how much was the overestimation.

Response: This is referred to the work published by Vautard et al., (2012) and a detailed analysis is presented in the reference. We have stated, here, the general trend discussed in their paper and we do not want to present in detail their findings since it is not the scope of our work.

Comment: P6687, section 2.2: What kind of data are the station data? Daily ?

Response: For NO2 and SO2 hourly, for Max8hrO3 and PM2.5 daily. Although it was clearly stated in the caption of the related figures and tables we have now included it in section2.2.

Comment: What are the future goals of the authors in terms of using this model in Europe?

Response: With all the respect to the reviewer's comment we don't want to discuss our future plans in the manuscript.

Comment: P6688, section 2.3: L3: True, but what kind of feedback does this paper give back to emission people?

Response: Section 2.3 has been removed from the revised version of the manuscript.

Comment: Is the PM2.5 observed, dry or wet? What about the predicted?

Response: It is known that aerosol measurements supposedly remove all water content from samples to consider only dry aerosols (although an issue has been raised with residual water that may exist). As such the simulated aerosol water was not taken into account in the simulating results.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 6681, 2013.