Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-727-RC1, 2018

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Ozone source apportionment during peak summer events over southwestern Europe" *by* Maria Teresa Pay et al.

Anonymous Referee #2

Received and published: 16 October 2018

General comments: The manuscript "Ozone source apportionment during peak summer events over southwestern Europe" presents a source apportionment study based on the application of an Eulerian chemical-transport model for the Iberian Peninsula for a specific summer episode in 2012. The research is interesting and it may constitute a valuable contribution towards a better understanding of O3 dynamics and potential options to improve ground-level concentration values in that region. Nonetheless, the outcomes of their research may potentially be of relevant for other areas and thus, of interest for a wider community. The paper is clearly structured and well written but before it can be considered for publication in Atmospheric Chemistry and Physics there are some obscure points and potential flaws that should be carefully addressed to guarantee that the results and their interpretation are correct. Some specific suggestions to

do so are provided below.

Specific comments Introduction In the introduction the authors provide a good background of the ozone related issues in the Iberian Peninsula including an overview of meteorological conditions typically associated to high ground-level O3 concentrations along with a number of relevant references. They also briefly discuss the trends and justify the need for their research. The topic is timely and interesting not only from the scientific point of view but also considering the legal implications and the need to identify potential interventions that may help alleviate O3 pollution in the Mediterranean Basin. Given the limitations of brute force methods for source apportionment studies of secondary pollutants, the authors apply the Integrated Source Apportionment Method (ISAM) implemented within the Community Multiscale Air Quality (CMAQ) with 4x4 km2 resolution for the whole Iberian Peninsula during a 10-day specific episode in summer 2012. The rationale and approach is clear but it would be interesting to explicitly state the main purpose of the study since this is relevant to understand whether the experiment design is appropriate and what are the limitations that can be expected from potential conclusions.

P3.Line 18: when discussing the possible reasons for the observed increase of O3 in some urban areas in the Iberian Peninsula, the authors assume a VOC-limited situation. Reductions on NOX emissions have necessarily reduced NO titration but I suggest them to remove that assumption regarding the VOC-NOX regime because it may be an oversimplification and not necessarily true in all cases/seasons.

P4.Line 29: the authors claim that the period analysed (between July 21st and 31st) is representative of typical summer synoptic conditions in that particular region. That is quite a strong statement and it would require substantial discussion and evidence to demonstrate to what extent that is true. Regardless of that, although the period may be characteristic in terms of synoptic conditions, O3 dynamics as previously stated, is strongly conditioned by both long-range transport and local conditions, including emissions of O3 precursors, initial chemical conditions, etc. Although the paper constitutes a valuable contribution to the understanding of O3 pollution in the Iberian Peninsula, I think that it cannot be assumed that the outcomes of the study may provide a source apportionment comprehensive description. Consequently, the insight to support the design abatement policies is limited and caution should be used to avoid extracting incorrect conclusions.

Methodology: An illustrative description of the CALIOPE and HERMES systems is provided in section 2.1. along with a number of relevant references. The authors state, however that emissions are based on 2009 since that is the most recent year with updates information on local emission activities. That statement is hard to understand and it may deserve further clarification. In addition, this section would benefit from a more consistent discussion on how this methodological choice may impact the results and to what extent potential inconsistencies with meteorology and boundary conditions for that specific modelling period are compatible with the research specific aim (which is not clearly identified). This issue may be acceptable to gain a general understanding of O3 contribution over a long period of time but for a short, high-O3 episode this may be a potential flaw that should be carefully addressed. VOC emissions are particularly relevant input for this analysis. However, our current understanding of VOC emission is limited, especially in urban areas (Lewis, 2018) which makes it difficult to accurately apportion contributions to tropospheric O3. It is also widely accepted that biogenic VOCs play a major role on atmospheric photochemistry. For this study, the authors rely on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.04 (P5.Line 30). This version was revised and extended through version 2.1 (Guenther et al., 2012) that also includes some code fixes. I'd strongly suggest the authors to perform a sensitivity run to understand whether using an outdated version of MEGAN may introduce relevant biases into their simulation. Species tagging and emission categories selection described in section 2.3 seem sensible although VOC emission shares should be reviewed taking into account the previous comment. I'm also concerned about a potential double counting and/or erroneous spatio-tempral allocation of VOCs emissions from agriculture since plant functional types considered in

MEGAN include crops. From previous literature, a share of 70% of total VOCs from SNAP 11 (nature) seems too high and may support that shortcoming. Please, double check this potential issue since it may bring about a considerable bias the outcomes of the study. The study makes advantage of a remarkably dense network of monitoring sites in the area of study to assess model performance through the computation of a series of common statistics (appendix B). The results however are difficult to interpret in their current form. Please, see corresponding suggestion in the results section. Refs:
-Lewis, A. C. The changing face of urban air pollution. Science (New York, N.Y.), 16 February 2018, Vol.359(6377), pp.744-745. DOI: 10.1126/science.aar4925 -Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471-1492. DOI:10.5194/gmd-5-1471-2012, 2012

Results: The authors discuss that the episode at hand concentrated an important percentage of exceedances in Spain (if I understood correctly; the first paragraph may be reviewed for the sake of clarity). However, the inspection of panel a) in Fig 2. does not seem to indicate that this episode was particularly severe since the distribution of MDA8 is not dissimilar to those of previous years, even when they reflect the concentrations over a 6 month period. In addition, the outliers apparently have moderated values. It would be interesting to make the point for such comparison. I'm not completely sure that it is sound to identify a high pollution episode at national level since O3 largely depends on regional features. If O3 levels were actually high all over the modelling domain, the influence of exported ozone (influenced by synoptic conditions) may be too high and thus, the representativeness of the results and the potential implications policy-wise, rather limited. Please, reflect on that. They also claim that the episode affected the central and north of the IP, but Fig. 2c shows high concentration spots all over the domain (or maybe the colour scale is not clear enough). It is also hard to see what the influence of Madrid and Barcelona plumes is (something consistent, to my understanding, with dominant stagnant conditions). Please, try to clarify. Fig 3 illustrate the meteorological conditions through some WRF -ARW outputs. I understand that a thorough model evaluation is not the main purpose of the paper but a minimal check (also through a statistic evaluation) of the credibility of the meteorological simulation (mainly wind fields) may substantially help the authors to gain a better insight of their results. For instance, that would help them to contrast hypotheses such us the one made in P10.Line 25. and following, where they attribute O3 nighttime overestimation to the underestimation of vertical mixing during nighttime stable conditions. In that case, a comparison of observed (where available) and modelled PBLH may be useful. As for the discussion of the general meteorological conditions, I'm not sure that the attempt to discriminate between ITL and NWadv situations is nor illustrative or needed. The discussion is hard to follow and the application of deterministic CTM may make that effort redundant.

The results of the statistical evaluation for CMAQ outputs are summarized in Table 2. Some suggestions for this table: - Please include in the caption whether the exceedance column refers to observed or modelled values (the missing one may be also included either way) - Two or three decimal points for r may be used - MNB (%) may be more illustrative than MB (that can be derived directly from MM – MO) - It may be misleading to pool together the statistics for different types of monitoring stations. As the authors discuss, it is arguable that outputs from a 4x4 km2 model exercise should be compared against observations at traffic locations - It is unclear why some monitoring sites wouldn't fit into any of the categories considered. Please elaborate and state the rationale to include them in the analysis It may be interesting to put these results into perspective by comparing them with those from other modelling exercises based on similar model suites in the IP or elsewhere (besides referring to previous applications of CALIOPE itself).

P11.L10-14. The description of the performance-based categories is hard to follow and it is already condensed in Fig. 5. Please, simplify or simply remove that passage. I'd suggest the authors to re-compute statistics and assessment with the alternative

BVOC emissions model run mentioned earlier. If mention to specific cities or areas is made (e.g. Ebro Valley, Lleida Plain), please identify them in any of the maps in the manuscript.

I'd generally advice the authors to condense the discussion and try to highlight their main findings not to unnecessarily extend this section. The information summarized in Fig. 7 is interesting although the concept of "receptor regions" is unclear. It seems reasonable in terms of geographical location but differences regarding contributions from different sectors are not evident, especially if the results are put into the perspective of the typical uncertainties of modelling exercises that can be inferred from the model evaluation previously presented. It is interesting noting a relatively large contribution from the SNAP 8 sector. The share of mobile sources is particularly important in the SIP area, which would be consistent with the discussion regarding the influence of shipping. However, other areas such as GV or even CIP present a non-negligible contribution. Could the authors elaborate on that? Maybe the discussion in section 3.4 is too profuse and should be substantially shortened. I encourage the authors to summary here their findings and provide the region-by-region discussion as supplementary material, including Fig. 8 and Fig. 9. Oppositely, the rationale for the station sub-set selection may deserve further explanation. In general, the section is abundant in hypotheses and subjective interpretation that are not clearly supported by evidence. Personally, I don't think this contribution really benefits from such approach. The paper may be restricted to a more solid and consistent analysis and discussion of the findings from the application of CMAQ-ISAM. That is novel and interesting enough and further attempts to relate the results with detailed regional dynamics and atmospheric patterns may very well be addressed in future specific studies (using more specific methods and data, e.g. better resolved emission inventories).

Discussion and conclusions: The authors claim that the modelling exercise presented allowed an in-depth evaluation of the modelling system applied. This relates to my last comment regarding the results section. The paper may lack a well-defined objective and presents a huge amount of information without a clear purpose. For example, if the main interest was to assess the modelling system capabilities and identify options for improvement, the results and analysis should gravitate towards a more detailed statistical analysis within a better defined methodological framework. I acknowledge a valuable study but I think the authors should revise their manuscript under a clearly defined scientific question avoiding an excessive spread in their discussion that may lead to inconsequential or cursory analyses and reflect that also in this section. As for the discussion on model uncertainty I find particularly important to take into account the observations regarding biogenic emissions, although the mismatch between emissions and meteorological conditions may also hinder the discriminating power of the results. In any case, caution should apply since the timespan of the period analysed makes it difficult to extract general conclusions. This is particularly important for the regulatory implications that may be derived from this study. As the authors conclude, I find reasonable to base recommendations for abatement strategies in more specific, regional scale, detailed analyses. Consequently I'd keep such conclusions to a minimum in this contribution.

Technical issues and typos: Please revise equations 1 to 4 for a better readability P7.Line 4: SNAP3 and 4 P8.Lines 12-13: the brackets are not needed P11.Line 11: "for 93% of the stations" instead of "for the 93% of stations" P12.Line 2: "extremely low winds"? P20.Line 8: "O3at" is missing a space P34.Line 5 (Fig. 2 caption): I guess the authors mean "Number of stations" instead of "Number of days"

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-727, 2018.