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Atmospheric Chemistry and Physics Discussions

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Interactive comment

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

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1. Reviewer #3: The paper gives important contribution to address the source apportionment study regarding ozone episode occurred in Spain. The paper is well structured and presents a complete analysis of the modelling results. However, there are some major points that should be addressed before recommended for publication. Besides that, English should be revised along the manuscript, there is some inconsistencies and grammatical errors. See below major and minor comments.

Authors: We thank the reviewer #3 for the comments and suggestions for improvement. We have corrected errors and omissions, and introduced as much as possible the reviewer's suggestions.

Please, find below the item-by-item response. For more details on the review process, we have uploaded the manuscript with track-changes.

2. Reviewer #3: Abstract; Line 15 (Page 4/Line 2): there is recently studies that showed that source-apportionment methods are not adequate to investigate plans and mitigation measures, in particular for non-linear pollutants like ozone, and that for that purpose "scenario analysis" based on "brute-force" are recommended. Authors should revise the text along the manuscript where it is mentioned the purpose of "designing plans", which should not be the final objective of this source-apportionment study. (see Clappier, A., Belis, C., Pernigotti, D., Thunis, P. Source apportionment and sensitivity analysis: two methodologies with two different purposes. Geosci. Model Dev. Discuss. 10, 4245-4256 (2017))

Authors: We totally agree with the reviewer's point of view and with the content of Clappier et al., (2017). The main goal of this study is not the design abatement policies, but it is to provide a first quantitative estimation of the contribution of the main anthropogenic activity sectors to peak O3 events in Spain relative to the contribution of imported O3.

Actually, source apportionment techniques alone cannot be used to the design abatement policies. Subsequent source sensitivity analyses tailoring the identified main contribution sources could predict how O3 will respond to reductions in precursor emissions. Both, source apportionment and source sensitivity are complementary and essential studies to define the most efficient O3 abatement strategies in the Western Mediterranean Basin. The manuscript highlights now this idea in different sections that we recap as follows:

Page 4 – Line 26-30: "Quantifying the contribution of emission sources during acute O3 episodes is a prerequisite for the design of future mitigation strategies in the region. In this framework, the main goal of this study is to provide a first quantitative estimation of the contribution of the main anthropogenic activity sectors compared to the imported concentration (regional and hemispheric) to peak O3 events in Spain.”

Page 22–Line 23-27: “We note that our results cannot predict whether emission abatement will have either a positive or negative effect in O3 changes due to the non-linearity of the O3 generation process. Subsequent source sensitivity analyses tailoring the identified main contribution sources could predict how O3 will respond to reductions in precursor emissions, which are essential to define the most efficient O3 abatement strategies in the Western Mediterranean Basin.”

Page 23-Line 4-7: “This work has quantified the local and imported contributions to O3 during an episode in a particular area in sourthwestern Europe. In addition, we have provided a perspective about the potential use of source apportionment method for regulatory studies in non-attainment regions. Further O3 source apportionment studies targeting other nonattainment regions in Europe are necessary prior to design local mitigation measures that complement national and European-wide abatement efforts.”

In order to be clear in the source apportionment applications and limitations of certain methods, we have added in the revised version of the manuscript a comment taking into account the recent findings in Clappier et al. (2017) as follows:

Page 4-Line 7-11: “Brute force is simple to implement, as it does not require additional coding in the CTM. However, as it quantifies the contribution of each source based on its absence, it does not reproduce actual atmospheric conditions, and therefore it is susceptible to inaccuracies in the prediction of O3 peaks under non-linear regimes (Cohan and Napelenok, 2011). Actually, brute force is not suitable to retrieve source contribution when the relationship between emissions and concentration is non-linear, but it is useful for analysing the concentration responses to emission abatement scenarios (Clappier et al., 2017).”

3. Reviewer #3: Page 4/Line 7-9: please review this sentence according to what has been commented before.

Authors: The statement in Page 4/Line 7-9 has been expanded following the reviewer's suggestion as shown in the previous authors' answer (see Page 4-Line 9-13).

4. Reviewer #3: Page 5/Line 26: the authors should comment about the representativeness of the 2009 emissions to the 2012 SA study presented. From 2009 to 2012 several changes happened in society and economy which was reflected in emission data.

Authors: Our methodological choice has been to use a detailed bottom-up emission inventory instead of a typical top-down regional emission inventory. BottomâĂŘup emissions, estimated using sourceâĂŘspecific emission factors and activity statistics, accurately characterise pollutant sources and allow obtaining more realistic results than the ones reported by top-down or regional emission inventories. However, they require very large efforts to be compiled and consequently the updating processes cannot be implemented year-to-year.

In HERMESv2 emissions are based on 2009 data, which was the closest year with updated information on local emission activities at the time this work started.

To understand the impact of the use of 2009 data to study year 2012 we revised the EMEP Centre on Emission Inventories and Projections (EMEP-CEIP), which collects and reviews the national emission inventories from Parties to the Convention on Long-range Transboundary Air Pollution (CLRTAP). Between 2009 and 2012 total NOx and NMVOC emissions in Spain decreased by -10.6% and -10.7%, respectively (EMEP CEIP, 2019). For NOx, around 80% of this reduction is linked to a reduction of road transport emissions, whereas in the case of NMVOC $\sim$50% of the reduction is due to a decrease of industrial emissions. NOx emissions from shipping in Europe have also decreased in the period 2009-2012 by 15%.

For our modelling study, we consider these differences as small and acceptable, and not creating any major inconsistency. The difference of 10-15 % in emissions for certain precursors between 2009 and 2012 is within the typically larger ranges of uncertainty in emission inventories. We also note that all our results are thoroughly evaluated and critically assessed using observations.

In any case, we have followed the reviewer's suggestion, and we have discussed in the manuscript the potential impact of these differences when the contribution of each emission sector is analysed:

Page 17–Line 32-33: "[. . .] This factor, added to the 15% decrease of NOx shipping emissions observed in Europe between 2009 (HERMESv2.0 base year) and 2012 (EMEP CEIP, 2019) could explain the discrepancies observed."

Page 18–Line 12-14: "[. . .] it has been estimated that between 2009 and 2012 energy production in coal-fired power plants increased from 13.1% to 19.4% (UNESA, 2012), which implied an increase of NOx emissions from the power industry sector of around 19.5% (EMEP CEIP, 2019)."

The Section 4 of the revised version of the manuscript includes now a comment on the methodological implication of using 2009 emissions for O3 source apportionment studies in an episode in 2012 as follows:

Page 5-Line 31-32: "HERMESv2.0 is currently based on 2009 data, which is the closest year with updated information on local emission activities at the time this work started."

Page 21-Line 31: "Our methodological choice has been to use a detailed bottom-up emission inventory instead of a typical top-down regional emission inventory. BottomâĂŘup emissions, estimated using sourceâĂŘspecific emission factors and activity statistics, accurately characterise pollutant sources and allow obtaining more realistic results than the ones reported by top-down or regional emission inventories. To understand the impact of the use of 2009 data to study year 2012, we revised the EMEP Centre on Emission Inventories and Projections (EMEP-CEIP), which collects and reviews the national emission inventories from Parties to the Convention on Long-range Transboundary Air Pollution. Between 2009 and 2012, total NOx and NMVOC emissions in Spain decreased by -10.6% and -10.7%, respectively (EMEP CEIP, 2019). For NOx, around 80% of this reduction is linked to a reduction of road transport emissions, whereas in the case of NMVOC ∼50% of the reduction is due to a decrease of industrial emissions. For our modelling study, we consider these differences as small and acceptable, and not creating any major inconsistency. The difference of 10-15 % in emissions for certain precursors between 2009 and 2012 is within the typically larger ranges of uncertainty in emission inventories."

Reference:

EMEP CEIP, 2019. Officially reported emission data. Available at: http://www.ceip.at/ms/ceip_home1/ceip_home/data_viewers/official_tableau/ (last access February 2019)

5. Reviewer #3: Page 7, Line 9: SNAP2 activity can be a particular important source for ozone precursors. Authors should comment about that when they mentioned that SNAP2 is aggregated with other activity sectors.

Authors: One limitation of the current version of CMAQ-ISAMv5.0.2 is that the number of tagged sources increases the computational time. In the current version of CMAQv5.0.2 the increase of the computational resources does not decrease the computational time. A more computationally efficient version of the ISAM will be released with the final version of CMAQv5.3 in Spring 2019. Based on that limitation, we configured our first study tagging the energy, industrial, road transport and non-road transport sectors (Fig. 1b), which account for 92% of the total NOx emissions in Spain. The remaining emission sectors are lumped in the OTHER tag. This selection criterion is explained in the manuscript as follows:

Page 7- Line 20-23: "The selected (tagged) SNAP categories in this study are the energy, industrial, road transport and non-road transport sectors (Fig. 1b), which account for the 92% of the total NOx emissions in Spain. An additional tracer (OTHR) gathers the remaining emission categories that were not explicitly tracked (i.e., SNAP2, 5, 6, 9, 10 and 11).”

We have seen in other studies performed over different domains (e.g., Portugal) that SNAP2 can be an important contributor to O3 precursors. We have mentioned this fact in the manuscript as follows:

From Page 13- Line 33 to Page 14-Line 2: “The high OTHR concentration around the biggest cities in Portugal may be related to precursors emitted by the residential sector (SNAP2 and 9) and biogenic emissions, as found in other source apportionment studies over Portugal (Borrego et al., 2016; Karamchandani et al., 2017).”

6. Reviewer #3: Page 7, Lines 25-30: in the scope of FAIRMODE – Forum for Air Quality Modelling in Europe – tools were developed, namely the DELTA-Tool, to evaluate air quality models and conclude about their suitability to be used for legislation purposes. The authors should consider the application of this tool to evaluate model performance instead of calculating the traditional statistical indicators. In any case, authors should justify why they decided not using this tool.

Authors: The evaluation with the Delta tool has been taken into account in previous evaluation studies of the CALIOPE system, although it has not been shown in the manuscript. We have added this comment to complement this information:

Page 11-Line 2-6: “CALIOPE has been evaluated in detail elsewhere (Pay et al., 2014 and references therein). Furthermore, the system has been evaluated using the tool developed by the Forum for Air Quality Modelling in Europe, named DELTA-Tool (Thunis and Cuvelier, 2014) to support and harmonize the model evaluation in the frame of the Air Quality Directive. Valverde et al. (2016a; 2016b) used the DELTA-Tool v4.0 and showed that the CALIOPE system accomplishes the quality objectives as defined in the Air Quality Directive for 78% to 91% of the NO2 and O3 monitoring stations in summer conditions in 2012.”

References: Thunis, P., Cuvelier, C., 2014. DELTA Version 4.0. Joint Research Center, Ispra (http://aqm.jrc.ec.europa.eu/DELTA/assessment/data/DELTA_UserGuide_V4_0.pdf).

Valverde, V., Pay, M.T., and Baldasano, J.M.: Ozone attributed to Madrid and Barcelona on-road transport emissions: characterization of plume dynamics over the Iberian Peninsula, Sci. Total Environ. 543, 670–682, doi: 10.1016/j.scitotenv.2015.11.070, 2016a. Valverde, V., Pay, M.T., and Baldasano, J.M.: A model-based analysis of SO and NO dynamics from coal-fired power plants under representative synoptic circulation types over the Iberian Peninsula, Sci. Total Environ., 541, 701-713, doi: 10.1016/j.scitotenv.2015.09.111, 2016b.

7. Review #3: Page 13, Lines 16-17: this information (model performance in terms of o3 peaks) should be presented and discussed in the model validation section.

Authors: The information provided in these lines is not a dedicated evaluation analysis on O3 peaks. It is devoted to provide a general perspective on source apportionment results shown in Figure 7. In order to make it clear, we have rewritten this sentence as follows:

Page 14-Line 15-16: "Figure 7 indicates that during exceedances of the MDA8 target value there is a good agreement (r = 0.79) between the sum of apportioned O3 and the observed concentrations over the receptor regions."

8. Reviewer #3: Page 13, Line 22: this sentence should be completed with information about the area where this impact (up to 8%) is verified.

Authors: The reviewer is right. We have rewritten the sentence as follows:

Page 14 – Line 20-21: "Shipping emissions in the MED region contributed up to 8% of the total O3".

9. Reviewer #3: Page 15, Lines 4-5: The authors should quantify how "model reproduces reasonably well"

Authors: We agree with the reviewer and we have quantified the model performance at this station as follows:

Page 15-Line 33: “At the urban station, the model reproduces the O3 traffic cycle (r = 0.66 and MB=22.5 $\mu$gm$-$3) featuring the typical low O3 concentrations (< 40 $\mu$gm-3) in the early morning and in the afternoon due to O3 titration (Fig. 8a).”

10. Reviewer #3: Page 15, Lines 9-10: Please clarify the sentence “The NO2 overestimation correlates with the highest road transport contribution”. Page 15, Lines 11-12: Please explain why: “The results point towards a poor representation of the vertical mixing during the stagnant conditions”

Authors: In order to clarify this sentence, we have rewritten this paragraph as follows:

Page 16-Line 2-5:”O3 was overestimated (MB type D) during daytime peaks due to the overestimation of the NO2 morning peaks during stagnant conditions, coincident with the highest road transport contribution for both pollutants. The results point towards a poor representation of the meteorological condition in the city during the stagnant conditions as shown in the meteorological evaluation (Sect. 4 in the supplement)”

11. Reviewer #3: Page 16, Lines 4-10: this should be placed in the model validation section.

Authors: We agree with the reviewer that evaluation results should be addressed in the dedicated section. However, the interpretation of the source apportionment results benefits from model evaluation, and at the same time the source apportionment results support enhanced model evaluation. We have added a comment on this in the Section 2.4 Evaluation method to clarify this issue as follow:

Page 8-Line 24-27: “Evaluation results are discussed together with the source apportionment results. On the one side, the interpretation of the source apportionment results benefits from model evaluation. On the other side, the source apportionment results support enhanced model evaluation as it allows identifying potential errors in



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emission estimates for specific sectors and/or in the chemical boundary conditions."

12. Reviewer #3: Page 16, Lines 13-14: how can the authors conclude that the model is able to reproduce all these different processes? Can the authors support better this statement?

Authors: As shown in Fig. 8d, modelled O3 peaks (> 120 $\mu$gm$-$3) are in a good agreement with observations (Fig. 8d), which suggests that overall the model reproduces the main transport paths, photochemical processes, and relative contributions from different sources. We have rewritten this statement as follows:

Page 16-Line 32: "At the rural station, modelled O3 peaks (> 120 $\mu$gm$-$3) are in a good agreement with observations (Fig. 8d), which suggests that overall the model reproduces the main transport paths, photochemical processes, and relative contributions from different sources."

13. Reviewer #3: Page 18, Line 4: since only one rural station is analysed, the authors should not generalize as "In rural background areas"

Authors: The reviewer is right; we have rewritten the sentence accordingly:

Page 18-Line 8-9: "Despite the O3 biases during stagnant conditions, the modelled O3 concentration is in general agreement with observations at the rural background station (Fig. 9d)"

14. Reviewer #3: Page 18, Lines 30-33 to Page 19: the authors analyse the vertical profile in a single point, but this will be not representativeness of the all study domain. Authors should change the text according to this limitation and comment it, or increase the number of points analysed.

Authors: The Figure 10 does not show a vertical profile, but it shows the O3 distribution over a vertical cross section crossing the IP from the west to the east at the centre of the domain (approximately at a latitude of 40.38° N). Although Figure 10 is not representative of the whole domain of study, it helps to understand the vertical variability of both pollutants with the PBL dynamics as schematized by Millán et al. (1996). We have clarified this point in the manuscript as follows:

Page 19-Line 1-3: "Figure 10 shows the vertical cross-sections at 6, 12, and 18 UTC for O3 and NO2 at a constant latitude (40.38° N) on July 25th, 28th and 30th. It helps to understand the vertical variability of both pollutants according to the dynamics of the PBL as schematized by Millán et al. (1996)."

15. Reviewer #3: Figure 2: please review the figure caption "Number of days exceeding the O3 target value (120 ug.m-3) by each day of the episode"

Authors: We have rewritten the caption of Figure 2 following the reviewer's suggestions as follows: Figure 2-caption: "Number of stations exceeding the O3 Target Value (120 $\mu$g/m3) per episode day"

Note that in the reviewed version this figure corresponds to the Figure S3 in the Supplement.

16. Review #3: Page 1/Line 20: write 4x4 km2 instead of 4x4 km (please correct this along the manuscript)

Authors: We have amended that.

17. Reviewer #3: Authors should refer the modelling system (CALIOPE) in the abstract.

Authors: The suggestion of the reviewer has been included in the new version of the manuscript as follows:

Abstract (Page 1-Line 21-24): "Our study applies and thoroughly evaluates a countrywide O3 source apportionment method implemented in the CALIOPE air quality forecast system for Spain at high resolution (4 x 4 km2) to understand and quantify the origin of peak O3 events over a 10-day period covering the most frequent synoptic summer conditions in the Iberian Peninsula."

18. Reviewer #3: Page 3, Line 4: The following reference should be added, since it is

the biggest ozone episode occurred in IP region: “Monteiro A., Gama C., Candido M., Ribeiro I., Lopes M. (2016) Investigating ozone high levels and the role of sea breeze on its transport. Atmospheric Pollution Research 7, 339-347.

Authors: We have added Monteiro et al. (2016) to the list of studies as an example of a large ozone episode occurred in IP region.

19. Reviewer #3: Page 7, Line 23: please indicate how many stations measure both O3 and NO2 pollutants.

Authors: We have mentioned this information latter on when we introduce the NO2/O3 source apportionment time series:

Page 8-Line 20-22: “This work will only discuss in detail the source apportionment plots at key O3 receptor regions, given the high number of stations (260) that simultaneously measure O3 and NO2.”

20. Reviewer #3: Page 10, Lines 17, 24, 29: please add “average” when mentioning “hourly O3” (the values presented are an average of different locations and not an “hourly O3 data”

Authors: The suggestion of adding “average” when mentioning “hourly concentration” has been implemented in the whole manuscript.

21. Reviewer #3:- Page 12, Lines 2-7: the following reference should be added to support this part: Borrego C., Monteiro A., Martins H., Ferreira J., Fernandes A.P., Rafael S., Miranda A.I., Guevara M., Baldasano J.M. (2016). Air quality plan for ozone: a case-study for North Portugal. Air Quality, Atmosphere & Health 9 (5), 447–460.

Authors: The reviewed version of the manuscript includes the reference Borrego et al., (2016) over northern Portugal to support the fact that, under stagnant conditions, imported O3 is depleted and O3 photochemical production is enhanced around the largest industrial/urban areas. Page 13-Line 4-6: “In a source attribution study over northern Portugal, Borrego et al. (2016) also found a reduction of imported O3 and the





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subsequent O3 formation by local sources under similar meteorological conditions."

22. Reviewer #3: Page 14, Line 30: please review the English.

Authors: The reviewed version of the manuscript has rewritten this paragraph as follows:

Page 15-Line 28-29: "The following sections analyse the source apportionment results at key regions (see Fig. 5) with a high on-road traffic contribution (i.e., CIP and NEIP) and a high contribution from industry and energy production (i.e., NWIP and Guadalquivir Valley)."

23. Reviewer #3: Page 17, Lines 19-24: authors should consult and use the following reference that compares the different shipping emission inventories mentioned: Russo M.A., Leitão J., Gama C., Ferreira J., Borrego C., Monteiro A. (2018) Shipping emissions over Europe: a state-of-the-art and comparative analysis. Atmospheric Environment 177, 187–194.

Authors: The reviewed version of the manuscript include the reference Russo et al. (2018) to discuss the uncertainties on shipping emissions over Europe:

Page 17-Line 28-32: "A recent review on the state-of-the-art of marine traffic emissions (Russo et al., 2018) indicates that STEAM appears as the most reliable and detailed emissions inventory since it is based on Automatic Identification System data and specific vessel information, with a resolution of 2.5 x 2.5 km2 (Jalkanen et al., 2016). A comparative analysis indicates that EMEP gridded inventories are overestimated, in particular over hotspots in the Mediterranean shipping routes, and underestimated in secondary routes."

References:

Jalkanen, J.-P., Johansson, L., and Kukkonen, J.: A comprehensive inventory of ship traffic exhaust emissions in the European sea areas in 2011, Atmos. Chem. Phys., 16, 71-84, https://doi.org/10.5194/acp-16-71-2016, 2016.

Russo, M. A., Leitão, J., Gama, C., Ferreira, J., & Monteiro, A.: Shipping emissions over Europe: A state-of-the-art and comparative analysis. Atmospheric Environment, 177, 187-194, 2018.

24. Reviewer #3: Page 18, Line 22: please replace "These O3 ..." by "The results presented before ... "

Authors: We have amended that the suggestion.

25. Reviewer #3: Figure 3: Please review the units used along the manuscript, like "m/s"

Authors: We have harmonize the use of units as ms-1.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-727/acp-2018-727-AC2-supplement.zip

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