This modeling study presents the aerosol results of the air-quality model Polyphemus applied to western Mediterranean in summer periods during the ChArMEx campaigns. In their previous publication (Chrit et al., 2017) the authors focused on organic aerosols simulated for the same periods. In this manuscript, modelled PM10, PM1, OM1 and inorganic aerosols were evaluated against measurements at the same site in Corsica as in the previous publication. In addition, sensitivity of model results to meteorological model, anthropogenic emission inventory and some model parameterization was analyzed. The air pollution over the Mediterranean is a significant topic and this manuscript might contribute to the research in this area. There are however, some critical issues to be clarified, addressed and revised before the manuscript can be considered for publication.

My main concern is about the anthropogenic emissions. According to the manuscript, emission inventory for 2010 was used for simulations of 2012, 2013 and 2014. There is no description in the manuscript how the 2010 emissions were adjusted to other years. This needs clarification and justification. If they were not somehow scaled, then it should be discussed in which emissions and sectors (traffic, ships, industry, etc.) differences between years are expected and how much this would affect the results.

Another weak point in the manuscript is the airborne evaluation (section 4.4). The method for comparison of model results with measurements is poorly described. Meteorological evaluation is missing for this period which seems to have different wind speed and direction than the other periods. The model performance for meteorology is very important for the interpretation of results. The method chosen to compare the model results with airborne measurements in Figs. 4 and 5 does not seem to be suitable and therefore the interpretation of various sensitivity simulations is difficult.

General comments:

Although they might be published elsewhere, a brief description about the model and input parameters are needed to be given in this manuscript (in Sect. 2) as well (e.g. chemical mechanism, aerosol model, VBS?, particle size (modal or sectional?), boundary conditions, vertical resolution, model top height, which layer is compared with measurements?) Sensitivity simulations for inorganic aerosols should be described earlier in this section (not first in 4.4).

Please do not use SIA (Secondary Inorganic Aerosols) as sum of Cl, NO_3, NH_4. SIA usually refers to ammonium nitrate and sulfate (SO_4, NO_3, NH_4).

Please avoid one-sentence paragraphs (e.g. page 9, lines 17-18).

Specific comments:

Title: Although the title is "*Aerosol sources* …" there is no clear conclusion about the sources. It is more a sensitivity analysis of the model results.

P2, L4: Please consider citing more recent studies.

P2, L9: Di Biagio et al. (2015)

P2, L19-20: *"Difficulties in modeling aerosol concentrations..",* It is not clear what is meant here; all or only inorganic aerosols? Please revise it.

P4: Was ECMWF data in mother domain (0.25^{o}x0.25^{o}) used also for the nested domain with a finer horizontal resolution? If yes, please describe how they were adapted to the finer resolution.

P5, L12: Please add the version of MEGAN used in the simulations. Which meteorological data was used to calculate biogenic emissions? Were biogenic emissions re-calculated using different meteorological data for the sensitivity simulations? Please clarify it and add to the text.

P5, L13-14: Using emissions of 2010 for simulations of 2012, 2013, 2014 needs some discussion. If 2010 data were used without any scaling for other years, it should be justified and it should also be discussed in which sectors (traffic, ships, industry, etc.) differences between years are expected and how much this would affect the results.

P6, L3-4: *"inorganic aerosols (chloride, nitrate, ammonium)"* Please remove *"inorganic aerosols*", just write those species relevant for partitioning.

P8, L2: Please make it clear: results from which model layer were used for comparison with measurements, was any interpolation applied?

P8, L10: '*The inorganic precursors HNO_3, HCl and SO_2 were measured…"* NH_3 is one of the most important precursors for inorganic aerosols. Were there any measurements of ammonia? Some information about ammonia emissions (temporal profiles, uncertainties) would be useful for discussion of SIA.

P 8, L30: *"…Appendix A of comparison…"* please replace "*of*" with "and"

P9, L1: Although the mean values over the whole period look satisfactory, Fig. B1 shows bias as high as about 5K in daily points. Deviation in hourly values might be even higher. Please consider the effect of T bias on especially nitrate discussions.

P9, L6-7: An explanation is needed for why ECMWF performs better in spite of its coarser resolution than WRF.

P9, L17-18: "*Tables 5*": should be "Table 5"

P9, L19-22: Authors might consider using recommended statistical parameters for the discussion in meteorological-model evaluation (EPA 2007). Evaluation of meteorology for summer 2014 is not shown. Since wind direction seems to be different than other summers, it is necessary to know how the model performance is for discussion in Section 4.4.

P11, L22: "*probably because of higher occurrence of transported desert dust in 2012"* Please provide some evidence to support this hypothesis.

P11, L23-24: *".. PM1 concentration is slightly over-estimated during summer 2013.."* On Table 7 however, PM1 in 2013 seems to be under-estimated. Please check and revise the sentence.

P12, L1-13: In this section, it is not clear whether the discussion is about the measured or modeled composition (also in Fig. 3). Please revise it.

P12, L7: BC appears for the first time here. How was BC measured?

P14, L6-9: Justification of underestimated ammonium (even more with EMEP emissions) based only on ship NOx emissions sounds not completely right. It might also be due to differences in NH_3 emissions between the two inventories.

P14, L20-23: Please consider revising the sentence "*For nitrate, the total nitrate (gas + particle phase) is under-estimated between 21 and 26 July 2013 (2.7 μ g m−3 in the measurements and 6.6 μ g m−3 simulated), and most of it is in the gas phase (only 0.4 μ g m−3 in the particle phase in the measurements and 0.2 simulated)"*
If measurements are 2.7 and simulations are 6.6, total nitrate is not underestimated but overestimated. It would also be necessary to show these numbers somewhere.

P15, L2-3: "*By influencing biogenic emissions, meteorology affects the formation of organics and hence the formation of inorganics (because of formation of organic nitrate)."* This sentence needs some justification and references (e.g. for organic nitrate formation Ng et al. 2017, for inorganic nitrate formation Aksoyoglu et al., 2017).
Meteorological parameters affecting biogenic emissions are mainly temperature and radiation. How much can be the difference in biogenic emissions due to using another meteorological model (due to differences in T and radiation) to cause such a significant change in inorganics shown in Table 10? If it is difficult

to provide any data to support the role of meteorology via biogenic emissions, then please add some discussion using other published studies.

P15, L5: *"hence the oxidants and the nitrate formation"* Please make it clear whether it is organic or inorganic nitrate.

P15, L9-18: Evaluation of meteorological parameters for the period of 9-10 July 2014 when airborne measurements were performed is missing. How was the modelled wind speed and direction? Which model layer concentrations were compared to aircraft measurements?

P15, L19-21: Please remove this information from here and give in the following corresponding sections.

P17, L1-2: Can the over estimation of sulfate be due to emissions? How were the emissions from ship stacks treated? If they were vertically distributed, to which model layer were they emitted?

P18, L2: "*ammonium and nitrate and chloride.."* please replace the first "and" with a comma.

P18, L28-29: Please do not use SIA for chloride, nitrate and ammonium.

P18, L30- 31: *"OM1 concentrations are high nearby locations of high biogenic emissions"* Is there any evidence (high biogenic SOA) to support this sentence or any reference for high biogenic emissions in those regions?

P19, L9: This sentence suggests that biogenic emissions were recalculated using different meteorology so that impacts on biogenic emissions were taken into account. If it is the case, this should be described in the methods section.

P20, L5-8: It needs some discussion how sulfate (and other inorganic aerosols) is affected by sea-salt parameterization.

P20, L31: In addition to temperature, other parameters such as radiation (for photolysis) and humidity also affect the reactions and formation of secondary products.

Table 10: Measured and modeled means of nitrate are similar (slight overestimation) while MFB is negative -24. Is this correct?

Figures:
Fig. 1: Is it reasonable to have such a low PBLH at noon over the sea north of Corsica? Can it be validated?

Fig. 3: Are these measured or modeled compositions? Please revise the caption.

Figs. 4-5: This is the first place where “model levels” are mentioned. This has to be described in the methods section.

These figures are difficult to understand. Averaged vertical profiles do not give any information about the location. Looking at the purple lines in Fig 1, it seems to me that the location can be either close to the southern coast of France, over the sea in the middle between French coast and Corsica or close to the Corsican coast. In addition, profiles seem to have a wide range (lines at certain altitudes) which makes the average values very uncertain. It would be more useful to extract model data from the corresponding grid cells and layers along the flight path for comparison with measurements. Because of these uncertainties, discussion in whole section 4.4 about sensitivity simulations does not make much sense.

Fig. B1: WRF Lambert OBSGRID is worst (bias 5K) in spite of nudging, why? Please replace “wind module” with “wind speed”

Fig. B2: What is the reason of less variation in 2013 with respect to 2012?

Fig. C1. S1 cannot be seen in the OM1 figure. Are S1 and S5 overlapped?
Scales in x-axes are different, i.e. PM10 July06-July31, PM1 June06-Aug05, OM1 Jun07-Aug05. It would be better if they were consistent, also with the meteorological figures (B1)

Fig. C2: Measured PM10 SO_4 looks sometimes lower than measured PM1 SO_4 (eg. June 15). Please check and/or justify.

Fig. C3: The modeled PM10 and PM1 NO_3 look exactly the same in all simulations, suggesting that coarse nitrate was not modeled. If this is the case, it should be mentioned in the methods section. Then there is no need to show the figure for PM10 NO_3 (same for NH_4 too).

Fig. D1: Please give the size fraction for the figures in the lower 2 panels.
Map for $Cl+NO_3+NH_4$ (please do not call it SIA) shows high concentrations (up to 24 microg/m^3). Please give some information which species is contributing more to this high level. Considering highly polluted Po Basin and high ammonia emissions in the region, high concentrations are very likely due to ammonium nitrate. It would make more sense to show maps of NO_3 and NH_4 separately and not to sum with Cl.

Fig. D2. It would be useful to see also the absolute differences.

Fig. D3: please separate Cl from NO_3 and NH_4. It is probably dominated by Cl.

Please revise the following references: Chrit et al 2017, Cholakian et al. 2017

References

EPA, U.: Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for ozone, PM2. 5, and regional haze, US Environmental Protection Agency, Office of Air Quality Planning and Standards, 2007.

Aksoyoglu, S., Ciarelli, G., El-Haddad, I., Baltensperger, U., and Prévôt, A. S. H.: Secondary inorganic aerosols in Europe: sources and the significant influence of biogenic VOC emissions, especially on ammonium nitrate, Atmos. Chem. Phys., 17, 7757-7773, https://doi.org/10.5194/acp-17-7757-2017, 2017.

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