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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraining the ship contribution to the aerosol of the Central Mediterranean" *by* S. Becagli et al.

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Anonymous Reviewer #1

Dear Editor, this manuscript presents an assessment of shipping contributions to PM10 aerosols in two locations in the Mediterranean Sea. It presents a very interesting integrated approach combining different tools such as analysis of the chemical composition of PM10, tracer analysis, back-trajectories and ship inventories and databases. Whereas certain of these tools are not novel and suffer from limitations (e.g., tracer methods), the combination of all of them provides very interesting results. Over all, the paper is of interest to the scientific community and merits publication after revision.

Specific comments are provided below: - lines 110-113 would fit better in the Methods section

Figure 1 and the corresponding text were moved to the methods section as suggested.

- line 276: very good, it is very important that the authors highlight this kind of limitation.

- line 292: "smaller" should be "lower". In general, the English is correct but a review by a native speaker would be helpful.

The sentence was corrected. The paper was revised by a native English speaker.

- Figure 2: the nitrate contribution at LMP is surprisingly high, considering the thermal instability of this species. Can this be due to an artefact or high uncertainty of the measurements? The nitrate concentrations measured in this campaign are in agreement with the long term measurements performed at Lampedusa (e.g., Calzolai et al., 2015) and with data from other remote sites in the western (Mallorca; e.g. Simo et al., 1991) and eastern Mediterranean (Finokalia; e.g. Mihalopoulos et al., 1997). The presence of artefacts in the nitrate sampling are documented in literature. These artefacts cause lower concentrations in the sample than in the atmosphere, in polluted regions due to NH4NO3 volatilization deposited on the filter. We believe than in relatively clean conditions like those incurring in Lampedusa and Capo Granitola the impact of artefacts is small, and the derived values are reliable. These values also suggest that ship emissions may play a large role. A comment was added to the text.

- line 340: also, EC concentrations are much higher in CGR, supporting this interpretation (the dominance of anthropogenic sources of carbonaceous aerosols in CGR)

We agree with the reviewer (see also comment 11 of reviewer 2). The sentence was changed and the differences were discussed..

- Figure 4 is interesting but not so useful, it could be removed if the authors encounter space limitations

We agree with the reviewer; the discussion of the sea breeze is interesting, but not directly related with the paper main topic. We moved the figure to the supplementary material.

- line 402: about the representativeness of this kind of ratios: it should be stated that this ratio may have a large variability due to varying fuel composition and engine operating conditions

We agree with the reviewer. The sentence was modified.

- line 424: LCR=1-5, how specific are these values for the specific refinery in the Moreno et al study? Can they really be extrapolated to other refineries? What can the uncertainty be for other refineries? Please discuss

Section 3.1.2 was largely modified, also taking into account the different comments of the reviewers. The discussion on the LCR variability was improved and new references to support the discussion were added. In particular, we have reported range of values for ship, refinery, and crustal components of the aerosols, also with reference to literature results.

- line 430: 24% of samples with LCR>1 and 8% >2, this is a very high contribution (32%), can the contribution from refineries be so high at LMP? From where? Or is this an example of the limitation mentioned above, the uncertainty of these LCR values when applied to other study areas? Please discuss

We have double checked the calculated LCR values. As correctly suggested by reviewer 2 (see his/her point 13), we did not take into account the uncertainty and the detection limit of La and Ce measurements. The instrumental detection limit on these determination was added in section 2.1. The very high values of LCR correspond with very low, in some cases below the detection limit, La and/or Ce concentrations. In these cases the uncertainty on LCR exceeds 100%. The elimination of these data points filters out most of the very high LCR values. We have thus redrawn Figure 5 (that becomes fig 4) accordingly, also indicating the range of values which are expected for the crustal component. This figure is reported below.

- Figure 6: again, the authors show only one point per type of dust, what is the uncer-

tainty? The authors' results seem very conclusive with regard to their own samples, but it would still be useful to see the potential variability in the other (refinery, UCC) types of dust. If no other data is available for this kind of sources, please mention as a limitation

The Lanthanoid content in Saharan dust measured in several aerosol size classes and from different areas of Sahara, as well for other compounds are reported in figure 6 (figure 5 in the revised version). The discussion on the variability of the composition was expanded (see also answer to the two previous comments). The figure is reported below.

- line 575: it is also possible that this is due to the larger aerosol dilution during transport towards LMP?

We agree with the reviewer. Dilution may be a cause of lower V concentration at LMP. A sentence on the possible role of dilution was added

- line 666: "two times higher than...", isn't this strongly dependent on fuel and engine operating conditions?

We agree. The sentence was changed accordingly.

- line 702: "component" should be "components"

The typo was corrected.

- line 703: the term "rough estimate" is more adequate than the "unambiguous identification" used in other parts of the text

In the text we use "rough estimation" referring to the quantitative estimation of the minimum contribution of ship aerosol (i.e., using the method of the minimum values reported in section 3.4). However, we believe that the combination of the different methods discussed in the paper leads to an unambiguous attribution of the measured aerosol to the ship source.

- line 722: "50%", again, this is very variable depending on engine conditions, meteorology (oxidation rate of SO2...), etc. Therefore this 20% difference can be expected

We agree. This is one of the reasons why we may estimate only the lower limit for sulfate contribution. The sentence was corrected.

- line 741: the higher % contributions obtained should not be considered a negative result, due to the potentially large variability of these contributions. It is correct to compare the order of magnitude with the previous literature, but the precision of the authors' method (and of other currently available methods) is not sufficient for them to carry out a detailed comparison of the results obtained. The results presented are quite positive, in my opinion.

We fully agree. The sentence is explaining possible reasons for the observed differences. - line 781: should "as expected", be added at the start of the sentence? We agree with the reviewer's comment, and added "as expected" at the beginning of the sentence.

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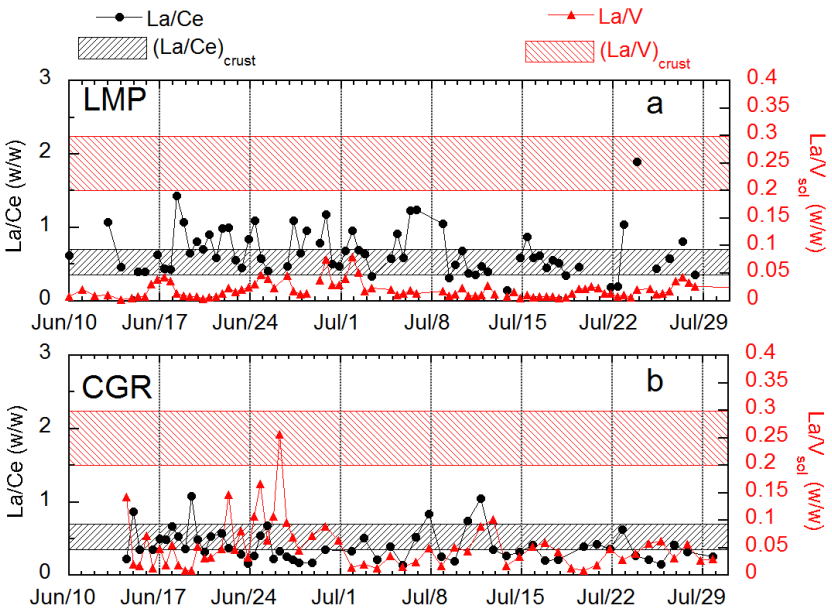

Fig. 1. Figure 4

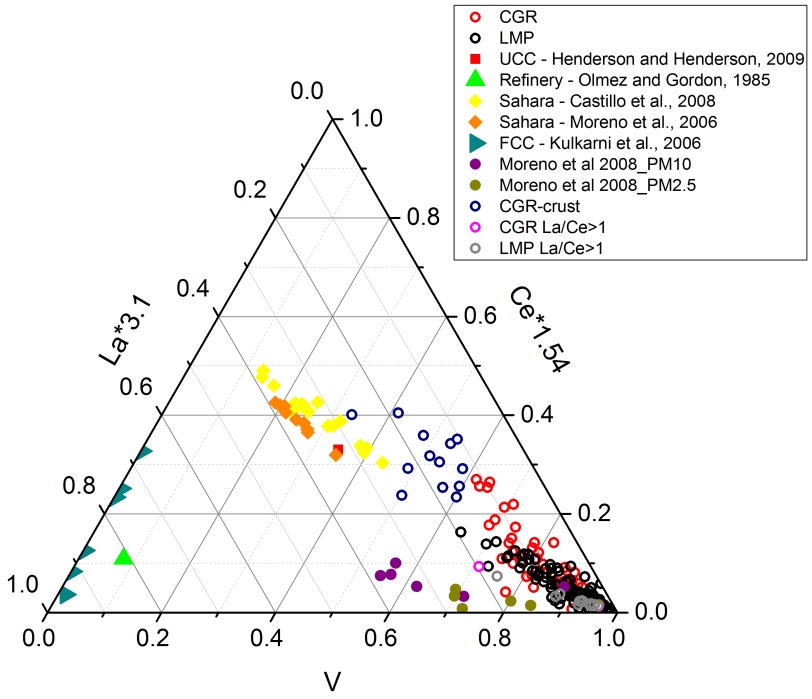

Fig. 2. figure 5