

Answer to comment of **Referee#1**

on “*Impact of North America on the aerosol composition in the North Atlantic free troposphere*” by M.I. García et al.

Reviewer Comment - OVERVIEW:

In this paper, a 5-year record of chemical composition of $PM_{2.5}$ and PM_{10} aerosol at Izaña Observatory is presented. The composition time series is analyzed in the context of meteorology, calculated back trajectories, and source-receptor plots. The paper shows very clearly that North America is the major source of aerosols sampled at this free tropospheric site. Measured aerosol composition varies seasonally due to the spatial distribution of aerosol sources in North America and seasonally varying large scale meteorology. The paper should be published in ACP once the comments below have been addressed.

REPLY:

Thanks for the review and the useful comments (listed below) that definitively contribute to improve the original manuscript. Please, find below a point-by-point reply to each question and suggestion.

1. Page 2, line 34: Do you mean “evidence of” instead of “interest in”?

REPLY:

Yes, thanks.

CHANGES IN THE MANUSCRIPT [R1#C1]:

“interest in” changed by “evidence of”.

2. Page 3, lines 24 – 25: Change “weighting” to “weighing”. Also “weighted” to “weighed”.

REPLY:

Thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C2]:

“weighting” changed by “weighing” and “weighted” by “weighed”.

3. Page 6, lines 27 – 29: Can the hypothesis that 3/4 of the non-ammonium sulfate is linked to soil emissions of gypsum be verified by ratios of calcium or other dust-containing elements to this nss-sulfate?

REPLY:

The scatter plot of Ca versus none-ammonium-sulfate shows a high correlation ($r^2=0.8$) with a slope of about 1.4 (g/g), which is higher than the theoretical one for Ca/SO_4 in gypsum (0.4 g/g) due to the presence of Ca with other minerals such as calcite, which is an abundant mineral in Saharan dust (Claquin et al., 1999, Modeling the mineralogy of atmospheric dust sources, Journal of Geophys. Res., 104, 22243-22256); this was discussed in Rodríguez et al. (2011) and more recently in Pérez García-Pando et al. (2016, Predicting the mineral composition of dust aerosols: Insights from elemental composition measured at the Izaña Observatory Authors, Geophys. Res. Lett., 43, no. 19, 10520-10529, doi:10.1002/2016GL069873).

CHANGES IN THE MANUSCRIPT [R1#C3]:

We have added the reference “Pérez García-Pando et al. (2016)” to the main text: “...in beds of Saharan dry lakes (Rodríguez et al., 2011; Pérez García-Pando et al., 2016)...”

4. Page 7, line 4: change to “may influence the export”.

REPLY:

Thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C4]:

“may influence on the export” changed by “*may influence the export*”.

5. Figure 2: A symbol indicating the location of the sampling site should be added to each plot – especially Figure 2A1-A4 – and all similar plots in the paper.

REPLY:

Thank you very much for your suggestion, which facilitate the interpretation of the figures.

CHANGES IN THE MANUSCRIPT [R1#C5]:

The sampling site (Izaña) has been highlighted in Fig.2, Fig.5, Fig.6, Fig.8, Fig.9, and Fig.S3 of the supplement.

6. Figure 3 caption: It is not clear how the monthly average values of the omega vertical wind component at the 850hPa level is “illustrated in plot (D)”. Figure 3D appears to be satellite derived SO₂ values (based on color bar) and domains.

REPLY:

Thank you very much for your observation, the caption of that figure in the original version of the manuscript was not enough clear, so we have reworded as follow:

CHANGES IN THE MANUSCRIPT [R1#C6]:

The text (caption):

“Monthly average values of the omega vertical wind component at the 850hPa level (negative values indicate upward movements) illustrated in plot (D)”

was replaced by:

“Monthly average values of the omega vertical wind component at the 850hPa level (negative values indicate upward movements) *calculated for the domains illustrated* in plot (D)”

7. Page 7, line 17: change to “over most of North America”.

REPLY: Thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C7]:

“over most North America” changed by “over most *of* North America”.

8. Page 8, line 2: A reference should be cited in the main text for the statement “as reported by NOAA”.

REPLY:

Thank you very much for your suggestion.

CHANGES IN THE MANUSCRIPT [R1#C8]:

We have included the reference of the data source “reported by NOAA (<http://www.ncdc.noaa.gov/climate-information/extreme-events/us-tornado-climatology>; *section S4 of the supplement*)”

9. Page 8, line 9: change to “shows the typical eastward track of the cyclones in March-April”

REPLY:

Thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C9]:

The text:

“the typical eastward track of the cyclones typical in March-April”

was reworded as:

“the typical eastward track of the cyclones in March-April”.

10. Page 8, line 13: change to “Spring (March-April) is the season of maximum frequency ...”

REPLY: Thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C10]:

“This is the season of maximum frequency of”

changed by

“*Spring (March-April) is the season of maximum frequency*”.

11. Page 10, line 20: I’m not sure that “depleted” is the correct word. Change to “. . .in regions with less nss-SO₄ compared to NE-US”

REPLY:

We agree; thanks for your observation.

CHANGES IN THE MANUSCRIPT [R1#C11]:

“depleted in” changed by “*with less*”.

12. Page 10, line 31: State the source of the high nitrate concentrations in the Central north region of North America.

REPLY:

Thanks for highlighting this issue that will help to clarify this part of the manuscript. There are three factors that contribute to the high concentrations of ammonium nitrate in the Central north region of North America (United States Environmental Protection Agency, 2000, Park et al., 2004): 1) high concentrations of ammonia linked to the livestock and fertilizers, 2) NO_x emissions linked to combustion, and 3) thermodynamic conditions favoring the reaction and condensation of ammonia and nitric acid as ammonium nitrate (i.e. enough high gas phase precursor –NH₃ and HNO₃-, low temperature and enough high relative humidity). This has been cited in the revised version of the manuscript according to this suggestion.

CHANGES IN THE MANUSCRIPT [R1#C12]:

Text:

“...The highest concentrations of nitrate in North America occur in the Central North region (Fig.4B); our results...”

was replaced by:

“...*High concentrations of nitrate in North America occur in the Central North region (Fig.4B), where conditions favorable for the formation of ammonium nitrate concur (US EPA, 2000; Park et al., 2004): (i) enough high concentrations of gas phase precursors (NH₃ linked to emissions in agriculture fields treated with fertilizers and HNO₃ due to oxidation of NO_x linked to fossil fuel combustion) and (ii) suitable thermodynamic conditions (rather low temperature and enough high relative humidity)...*”

The following reference has been added:

United States Environmental Protection Agency, National air pollutant emissions trends, 1900–1998, EPA-454/R-00-002, Office of Air Qual. Planning and Stand., Research Triangle Park, N. C, 2000.

13. Page 13, lines 1-3: Do you mean to say that there are sources of OM that are not related to combustion but, rather, biogenic in origin?

REPLY:

Yes, it is what we wanted to say. We have rewritten it in a more simple way.

CHANGES IN THE MANUSCRIPT [R1#C13]:

The text:

“there are sources of OM that are not, on the other hand, important in EC, and are most probably biogenic emissions”

was reworded as:

“there is a significant contribution to OM of sources that are not related to combustion, but probably to biogenic emissions”.

- 14.** Figure 8 caption: Supply the full name of “MDAF” shown in the color bar for C3. Also what is “Aerosol Index averaged”?

REPLY:

Thank you very much for pointing this issue. Fig. 8 caption has been corrected as you point.

CHANGES IN THE MANUSCRIPT [R1#C14]:

“Mean 2008-2013 Aerosol index averaged” changed by *“Major dust activity frequency (MDAF) for the study period: the number of days with AI values > 1 divided by the total number of days with available AI data in %”.*

- 15.** Page 14, lines 5-10 and Figure 9: The size range of sea salt discussed and shown should be provided.

REPLY:

All results shown in section 3.3 <Transatlantic transport of North American aerosols> is based on PM10 chemistry, including section “3.3.7 Sea salt” and “Figure 9”. It is described in section “2.2 Meteorology, back-trajectories and MCAR plots” and first paragraph of section 3.3.

CHANGES IN THE MANUSCRIPT [R1#C15]:

Not needed.

- 16.** Figure 10 caption: Change “mayor” to “major”. Also – it is difficult to see differences in the mass fractions for the non-OM and non-dust components. A logarithmic scale on the y-axis would help.

REPLY:

Thanks for your suggestions in which we have spend a time working in. However, this change is not possible in an areal format plot since the log a sum is not the sum of logs. Moreover, that change would smooth the variability of dust and OM, which actually are the most relevant contributors. The fact that the contribution of non-OM and non-dust is rather difficult to see is due to the fact that these species are present in low concentrations (as discussed in the text) and that is the relevant result.

CHANGES IN THE MANUSCRIPT [R1#C16]:

“mayor” changed by “major”.