Answer to comment of **Referee#3**

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

Reviewer Comment - OVERVIEW:

The author has shown North America is a major source of north Atlantic free tropospheric aerosol. These aerosol concentrations are enhanced by the propagation of mid-latitude cyclones. Furthermore, there is a seasonality associated with the free tropospheric aerosol composition due to the spatial distribution of aerosol sources in the North Atlantic, and latitudinal shift in the westerly jet from low latitudes in the winter to higher latitudes in the summer. Back trajectories and previous findings on aerosol spatial compositional differences in the United States are linked to explain differences in five years of measured aerosol chemical composition at the Izaña Observatory (which is often exposed to free tropospheric air). Seasonal variations in measured free tropospheric aerosol composition correspond to the variation in the north Atlantic outflow Latitude. The manuscript findings are supported by the results and previous literature. I recommend publication after the following minor comments are addressed.

REPLY:

Thanks to Referee #3 for the useful comments that contribute to improve the original manuscript. Please, find below a point-by-point reply to each question and suggestion.

Minor comments

1. Line 24 Pg 1- "The present study evidences"- reword

   REPLY:
   Thank you very much for your suggestion.

   CHANGES IN THE MANUSCRIPT [R3#C1]:
   Sentence:
   "The present study evidences how long-term evolution of the aerosol composition in the North Atlantic free troposphere will be influenced by air quality policies and the use of soils (potential dust emitter) in North America."
   Reworded as:
   "The present study *suggests that* long-term evolution of the aerosol composition in the North Atlantic free troposphere will be influenced by air quality policies and the use of soils (potential dust emitter) in North America."

2. Line 28 Pg 1 – remove the word "on" both times it is used in this line.

   REPLY:
   Thank you very much for your observation.

   CHANGES IN THE MANUSCRIPT [R3#C2]:
   Sentence:
   "The export of aerosols from their source areas impacts on air quality (Chin et al., 2007) and also on climate related processes (Ramanathan et al., 2001) in downwind receptor regions."
   Reworded as:
   "The export of aerosols from their source areas impacts on air quality (Chin et al., 2007) and climate related processes (Ramanathan et al., 2001) in downwind receptor regions."

3. Line 32 Pg 1- reword the last sentence of this page (which continues onto the next page).

   REPLY:
   Thank you very much for your suggestion.

   CHANGES IN THE MANUSCRIPT [R3#C3]:

Sentence:
"Aerosols are of special interest as they may have an influence on direct radiative transfer and cloud properties by altering the radiative effect and on rain."
Reworded as:
"Aerosols are of special interest as they may have an influence on direct radiative transfer and cloud properties by altering the radiative effect and rain *patterns*."

4. Line 1-5 Pg 2- separate the second sentence into two sentences.

REPLY:
Thank you very much for your suggestion.

CHANGES IN THE MANUSCRIPT [R3#C4]:
Sentence:
"It is estimated that globally, this influence results in mean radiative forcing due to aerosol-radiation and aerosol-cloud interaction of about -0.9 $W \cdot m^{-2}$, with aerosol-radiation contribution of -0.35 $W \cdot m^{-2}$ as a result of net sulphate contributions (-0.4), black carbon (+0.4), nitrate (-0.11), dust (-0.1) and organics (-0.12), according to the Intergovernmental Panel on Climate Change (IPCC, 2013; Myhre et al., 2013)."
Reworded as:
"It is estimated that globally, this influence results in mean radiative forcing due to aerosol-radiation and aerosol-cloud interaction of about -0.9 $W \cdot m^{-2}$. *The* aerosol-radiation contribution (-0.35 $W \cdot m^{-2}$) *is the* result of the net *contribution of* sulphate (-0.4), black carbon (+0.4), nitrate (-0.11), dust (-0.1) and organics (-0.12), according to the Intergovernmental Panel on Climate Change (IPCC, 2013; Myhre et al., 2013)."

5. Line 21 Pg 3 – What is EN-14907?

REPLY:
Thank you very much for your observation, which will clarify this point. The EN-14907 is the European standard gravimetric measurement method for the determination of the mass fraction of suspended particulate matter.

CHANGES IN THE MANUSCRIPT [R3#C5]:
Sentence:
"Concentrations of PM_x were determined by gravimetry following the EN-14907 procedure."
Reworded as:
"Concentrations of PM_x were determined by gravimetry following the *European standard gravimetric measurement method* EN-14907."

6. Line 3 Pg 4 – shown where?

REPLY:
Thank you very much for your observation. We have replaced "shown" by "plotted", and not directly cited the Figures showing the MCAR, because it would imply to not number the Figures when they are cited (which would be out of the ACP standard).

CHANGES IN THE MANUSCRIPT [R3#C6]:
In the sentence:
"In these MCAR, the typical (median) concentration of each aerosol component recorded at Izaña, when the air mass has passed by each pixel of the study region, is shown."
The figure numbers have been included:
"In these MCAR, the typical (median) concentration of each aerosol component recorded at Izaña, when the air mass has passed by each pixel of the study region, is *plotted*."

7. Line 27 Pg 12 – Can you provide references that show evidence of biogenic sources?

REPLY:
Thank you very much for your suggestion, which was also raised by Reviewer#2. This part of the manuscript needed to be complemented as you point. Also note that in p.13 L.5-8 we stated

previous evidences: "Biogenic emissions are among the principal source of OM in the US, followed by three combustion sources that also emit EC (wildfires, fossil-fuel and bio-fuel) (Park et al., 2003); specifically, in South-eastern US, BVOC emissions are mainly isoprene (81%) and monoterpenes (19%) (Goldstein et al., 2009)."

CHANGES IN THE MANUSCRIPT [R3#C7]:
This text was added to the manuscript:
"*This is consistent with previous studies that estimated the contribution of biogenic SOA to OM within the range 50-60% in Southeastern US (Blanchard et al., 2015; Kim et al., 2015; Ying et al., 2015)*".

The following references have been added:
Blanchard, C. L., Hidy, G. M., Shaw, S., Baumann, K., and Edgerton, E. S.: Effects of emission reductions on organic aerosol in the southeastern United States, Atmos. Chem. Phys., 16, 215-238, doi:10.5194/acp-16-215-2016, 2016.

Kim, P. S., Jacob, D. J., Fisher, J. A., Travis, K., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., Jimenez, J. L., Campuzano-Jost, P., Froyd, K. D., Liao, J., Hair, J. W., Fenn, M. A., Butler, C. F., Wagner, N. L., Gordon, T. D., Welti, A., Wennberg, P. O., Crounse, J. D., St. Clair, J. M., Teng, A. P., Millet, D. B., Schwarz, J. P., Markovic, M. Z., and Perring, A. E.: Sources, seasonality, and trends of southeast US aerosol: an integrated analysis of surface, aircraft, and satellite observations with the GEOS-Chem chemical transport model, Atmos. Chem. Phys., 15, 10411-10433, doi:10.5194/acp-15-10411-2015, 2015.

Ying, Q., Li, J. and Kota, S. H.: Significant Contributions of Isoprene to Summertime Secondary Organic Aerosol in Eastern United States, Environ. Sci. Technol., 49(13), 7834–7842, doi:10.1021/acs.est.5b02514, 2015.

8. Figure 10 caption – change "mayor" to "major"

REPLY:
Thank you very much for your observation.

CHANGES IN THE MANUSCRIPT [R3#C8]:
"mayor" changed to "major" in Fig. 10 caption.