Dear Dr. Russell,

I contact you because of the end of the discussion period of the ms <acp-2017-60>. As you can see in the web site, we have already replied to each of the question of the three reviewers. In the reply we have followed the structure (1) comments from Referees, (2) author's response, (3) author's changes in manuscript, recommended by ACP.

We have now prepared a revised version of the manuscript. For preparing this version we have taken into account the comments of the three reviewers. As you can see in the reports, the referees have arisen minor and technical comments and agree publication of this manuscript in ACP. The questions and suggestions of all reviewers have definitively contributed to improve the manuscript.

Please, find attached to this letter (i) the "Authors Response Report", which includes the answers to reviewers 1, 2 and 3, with a reply to each question and the description of the changes in the manuscript as a consequence of each referee comment, and (ii) the "Revised Manuscript and Supplement", where the changes performed (with respect to the ACPD version) are highlighted. We have also followed the structure (1) comments from Referees, (2) author's response, (3) author's changes in manuscript, recommended by ACP, so changes can easily be tracked.

This revised version includes the minor comments and technical changes were introduced to include the comments of the referees.

The latex file of the manuscript and the pdf of the supplement will be uploaded in a further step.

Thanks,
Sergio Rodríguez

Authors Response Report

REPLY TO REVIEWER#1:

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 Rodriguez 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 SO_2 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-SO_4 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) NOx 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 –NH_3 and HNO_3-, 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 (NH_3 linked to emissions in agriculture fields treated with fertilizers and HNO_3 due to oxidation of NOx 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 <Transatlatnic 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".

REPLY TO REVIEWER#2:

Thanks to Referee #2 for his 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. P3. Line 24: The authors wrote, "the uncertainties may be significant below this threshold value." What's the percentage of uncertainties in this method? Does the difference of 15% humidity affect the concentrations of PMx?

REPLY:
For the Izaña sampling conditions (e.g. airflow, duration of the sampling), the uncertainty associated for PM < 10 $\mu g \cdot m^{-3}$ is ± 5 $\mu g \cdot m^{-3}$. The percentage will depend on the sample mass, ranging from 500% (PM = 1 $\mu g \cdot m^{-3}$) to 50 % (PM = 10 $\mu g \cdot m^{-3}$). The standard reference method of Europe set the relative humidity (for filter conditioning and during weighing) to be 45-50%. However, because the sampling at Izaña is usually performed at ambient relative humidity lower than 50%, the long-term aerosol program of Izaña is based on filter conditioning at 30-35% relative humidity. This is already described in section S1 < Uncertainty of the gravimetric method > of the Supplement. The influence of decreasing the conditioning relative humidity on the filter weigh depends on the relative humidity range, such 15% decrease may be relevant when conditioning to rather high relative humidity (50% or higher), but less important when conditioning to low relative humidity (e.g. 20%). The conditioning to 30-35% used in the long-term program of Izaña is suitable for the low relative humidity of the ambient air and is consistent with the measurements of other aerosol properties (optical properties and number size distribution) that are performed after condition the aerosol sample at relative humidity lower than 40% according to GAW standardization.

CHANGES IN THE MANUSCRIPT [R2#C1]:
This short description has been added in section S1 of the Supplement.

"The conditioning to 30-35% used in the long-term program of Izaña is suitable for the low relative humidity of the ambient air and is consistent with the measurements of other aerosol properties (optical properties and number size distribution) that are performed after condition the aerosol sample at relative humidity lower than 40% according to GAW standardization".

2. P6. Line 7: The authors also wrote, "high uncertainty of the manual gravimetric method". How high is the uncertainty?

REPLY:
The uncertainty is described in the Supplement. It was determined by using the procedures described in EN 14907. As example, the following table shows the uncertainty associated with the mean concentrations of PM at Izaña shown in Table 1 of the article (see details in the Supplement):

	Saharan Air Layer		Westerlies	
	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
PM, $\mu g \cdot m^{-3}$	46.42 ± 4.7	21.27± 4.6	2.54±4.6	2.19±4.6

CHANGES IN THE MANUSCRIPT [R2#C2]:
Not needed.

3. P4. Line 7: The authors used "the empirical ratio of Na and sulfate (=0.25) " to determine the sea salt sulfate and non-sea salt sulfate. Please specify the ratio of Na and sulfate. Is it SO_4^{2-}/ Na^+ =0.25? Bonsang et al. [1980] reported that the ratios of SO_4^{2-}-to Na^+ could be greater than 1, and the sulfate contributions could be due to gas-to-particle conversion of the oxidation of marine SO_2. It would be good to state the assumptions of this method.

REPLY:
Thank you very this comment, which is already included in the manuscript (see line 6-9 pag 4).

CHANGES IN THE MANUSCRIPT [R2#C3]:
Not needed.

4. P12. Line 27: "The OM is rich in southeastern US and could be from biogenic emissions". It would be good to add some examples and references of biogenic emissions.

REPLY:
Thanks for your suggestion. This part of the manuscript actually needed to be complemented as you point.

CHANGES IN THE MANUSCRIPT [R2#C4]:
This text was added
"*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.

5. P11. Line1: The author wrote, "because nitrate may experience negative artefacts during sampling we cannot discard underestimations." If there were possible underestimations, would the authors recommend any further methods to improve them?

REPLY:
Some studies have already shown that quartz filters (as those used in the present study) are the most suitable for avoiding nitrate negative artifacts (e.g. Schaap et al., 2004; Tian et al., 2016). Intercomparison campaigns with real-time measurements in other to evaluate the percentage of losses from the filter-based method could be done in a future.

References:
Tian, S., Pan, Y., Wang, J. and Wang, Y.: Concurrent measurements of size-segregated particulate sulfate, nitrate and ammonium using quartzfiberfilters, glassfiberfilters and cellulose membranes, Atmos. Environ., 145, 293-298 , 2016.

CHANGES IN THE MANUSCRIPT [R2#C5]:
Sentence:
"…; because nitrate may experience negative artefacts during sampling (Schaap et al., 2004; Vecchi et al., 2009), we cannot discard underestimations and believe that further intercomparison with artefact-free real-time nitrate measurements should be carried out.."
was re-written as:
"…Because nitrate may experience negative artefacts during sampling (Schaap et al., 2004; Vecchi et al., 2009), we cannot discard underestimations; *further artefact-free real-time nitrate measurements should be included in the long term aerosol measurements program*"

6. P.15. Line 25: The authors wrote, "the aerosol composition…. will be influenced by air quality policies and use of (potential dust emitter) soils in North America." This conclusion points out that the air quality policies and anthropogenic sources are important. I don't find any air quality policies related to dust emission control addressed in this manuscript. The aerosol composition could be influenced by many factors and sources that were addressed in this manuscript. It would be good to emphasize the important contribution.

REPLY:
Thanks for this suggestion. We agree to emphasize on the importance of the use of soil and dust emissions.

CHANGES IN THE MANUSCRIPT [R2#C6]:
This text has been added to the penultimate paragraph of section 3.3.8 <Mass closure of aerosols>:
"*Environmental studies have shown that the conversion of natural lands to agriculture and pasturage fields has had a number of impacts in North America (Nordstrom et al., 2004, Wu et al., 2007). Moreover, research has predicted that this change of land use may increase (MNP, 2006, Lawler et al., 2014), and this suggests an enhancement of dust impacts in downwind regions*"

The last paragraph of the conclusions was reworded as:

"*The overall results indicate that future long-term evolution of the aerosol composition in the North Atlantic free troposphere will be influenced not only by air quality policies applied in urban and industrial areas, but also by the use of potential dust emitter soils in North America, specially those lands linked to agriculture and pasturage activities. These dust emissions should be considered in the regulations on air quality and climate change mitigation*" **[**R2#C6**]**.

7. Abstract line 24: The author wrote, "our results suggest that a significant fraction of organic aerosols may be linked to sources other than combustion." What's the fraction of organic aerosol do authors suggest?

REPLY: The techniques used in this study do not allow obtaining any realistic estimation with a rather low uncertainty. Such estimation should be based on organic aerosol speciation, as the ones used by García et al. (2016; Speciation of organic aerosols in the Saharan Air Layer and in the free troposphere westerlies, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-108) in the Saharan Air Layer and during a biomass burning event that impacted on the Atlantic (unfortunately, winter to spring period was not included in that study). The potentially high contribution of biogenic emissions we suggests is consistent with previous studies, which points to biogenic contributions to OM within the range 50-60%; this has already been included in the revised version of the manuscript [see R2#C4].
CHANGES IN THE MANUSCRIPT [R2#C7]:
Not needed.

Technical correction

8. P13. Line 13: "are Izaña are extremely low" should be "at" Izaña are extremely low.

REPLY:
Thanks for the observation.

CHANGES IN THE MANUSCRIPT [R2#C8]:
It has been already corrected in the text.

REPLY TO REVIEWER#3:

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.

Revised Manuscript

Changes are highlighted in yellow for reviewer#1, in green for reviewer#2, in pink for reviewer#3 and in blue for authors. Brackets indicate the reviewer and comment that prompt the change.

Impact of North America on the aerosol composition in the North Atlantic free troposphere

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Abstract. In the AEROATLAN project we study the composition of aerosols collected over ~5 years at Izaña Observatory (located at ~2400 m a.s.l. in Tenerife, the Canary Islands) under the prevailing westerly airflows typical of the North Atlantic free troposphere at subtropical and mid-latitudes. Mass concentrations of sub10-µm aerosols (PM_{10}) carried by westerly winds to Izaña, after transatlantic transport, are typically within the range 1.2 and 4.2 µg·m^{-3} (20th and 80th percentiles). The main contributors to background levels of aerosols (PM_{10} within the 1st - 50th percentiles = 0.15 – 2.54 µg·m^{-3}) are North American dust (53%), non-sea-salt-$SO_4^{=}$ (14%) and organic matter (18%). High PM_{10} events (75th - 95th percentiles ≈ 4.0 – 9.0 µg·m^{-3}) ~~and~~ [Authors] are prompted by dust (56%), organic matter (24%) and nss-$SO_4^{=}$ (9%). These aerosol components experience a seasonal evolution explained by (i) their spatial distribution in North America and (ii) the seasonal shift of the North American outflow, which migrates from low latitudes in winter (~32ºN, January–March) to high latitudes in summer (~52ºN, August–September). The westerlies carry maximum loads of nss-sulphate, ammonium and organic matter in spring (March–May), of North American dust from mid-winter to mid-spring (February–May) and of elemental carbon in summer (August–September). Our results suggest that a significant fraction of organic aerosols may be linked to sources other than combustion (e.g. biogenic); further studies are necessary for this topic. The present study suggests that [R3#C1] 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.

1 Introduction

The export of aerosols from their source areas impacts on air quality (Chin et al., 2007) and ~~also on~~[R3#C2] climate related processes (Ramanathan et al., 2001) in downwind receptor regions. Exposure to aerosols – or particulate matter (PM) – and reactive gases in ambient air pollution is associated with ~3.7 million deaths a year~~-1~~ [Authors], mostly due to ischaemic heart disease (~40%), stroke (~40%), chronic obstructive pulmonary disease (~11%), lung cancer (~6%) and acute lower respiratory infections in children (~3%) according to the World Health Organization (WHO, 2014). 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 [R3#C3]. 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) [R3#C4].

North America is a major source of aerosols and trace gases (Li et al., 2004; Park et al., 2003, 2004). The export of trace gases to the North Atlantic in the so-called North American outflow (Li et al., 2005) is enhanced by mid-latitude cyclones (Dickerson et al., 1995; Merrill and Moody, 1996; Moody et al., 1996). These cyclones frequently form on the lee side of the Rocky Mountains and propagate eastward, with associated cold fronts southeastward across the eastern United States (US; Whittaker and Horn, 1984; Zishka and Smith, 1980). The cyclones occur every 5 days – on average – in summer (Li et al., 2005), although in spring that frequency may be even higher. Four airstreams are associated with mid-latitude cyclones: the warm conveyor belt ahead of the cold front, the cold conveyor belt, the dry airstream subsiding behind the cold front, and the post cold front boundary layer airstream (Cooper et al., 2002a, 2002b). The northeastward ascending airstream represented by the warm conveyor belt prompts the upward transport of pollutants from North America to the free troposphere over the North Atlantic (Eckhardt et al., 2004), where it may connect with the westerly circulation at the north of the Azores High (Li et al., 2005) prompting the transatlantic transport of pollutants; this has been documented for relatively long lifetime (LT) trace gases, such as CO (LT~ 60 days) and O_3 (LT~25 days) (Honrath et al., 2004; Owen et al., 2006). Convection is also an important mechanism for ventilation of the boundary layer; the convective outflow prompts the upward transport of pollutants (Dickerson et al., 1987; Talbot et al., 1998), which may remain over North America for several days prompting ozone production and its subsequent export to the North Atlantic (Li et al., 2005). This mechanism is important in the south-eastern United States in summer, as the warm conveyor belt of the mid-latitude cyclones is shifted northward (Li et al., 2005). There are a number of observation-based evidences on the large-scale impact of the CO and O_3 pollution events in the North Atlantic linked to North American fires and pollution export (Parrish et al., 1998; Moody et al., 1996; Honrath et al., 2004; Owen et al., 2006).

Although aerosols have been less studied, some research has found evidence of their export to the Atlantic in the cyclone modulated North American outflow (Li et al., 2005), even if they have a relatively short lifetime (LT~15 days). By ground based and airborne lidar measurements, Ancellet et al. (2016) detected the transatlantic transport of North American biomass burning aerosols and dust to the Mediterranean. At Pico Observatory in the Azores, free troposphere transport of North American black carbon aerosols linked to boreal fires (Val Martin et al., 2006) and sulphate, nitrate, elemental carbon and organic aerosols (such as biomass burning) has been detected (Dzepina et al., 2015). Modelling studies have also shown evidence of [R1#C1] intercontinental transport of aerosols (Park et al., 2004; Chin et al., 2007).

Previous studies on transatlantic transport of North American aerosols have reported on events detected in intensive campaigns, typically lasting from weeks to a few months. In this study we have used a complementary approach, based on long-term records. We analysed the long-term aerosol chemistry register of the Izaña Observatory – located at ~2400 m a.s.l. on the island of Tenerife – with the aim of identifying the composition, potential sources and origin of the aerosols transported by westerly winds across the North Atlantic. To our knowledge, this is the first study addressing the issue.

2 Methods

2.1 Sampling site

The Izaña Global Atmospheric Watch (GAW) observatory is located on a mountain ridge (~2400 m a.s.l.) lying almost permanently above the temperature inversion and marine stratocumulus layer typical of the marine boundary layer (MBL) top in the subtropics. Buoyant upslope winds develop during daylight, with minimum impact on the aerosol mass concentrations (Rodríguez et al., 2009). At night, upslope winds cease and Izaña is basically exposed to the prevailing westerly free troposphere subsiding airflow.

2.2 Sampling and chemical composition

This study is based on a long-term record of chemical composition of PM smaller than 10 µm (PM_{10}) and 2.5 µm ($PM_{2.5}$) aerodynamic diameters at Izaña Observatory. A total of 401 PM_{10} and 315 $PM_{2.5}$ samples were collected and chemically analysed from February 2008 to August 2013.

The samples of PM_{10} were collected on quartz micro fibber filters (150 mm diameter) pre-heated at 205ºC for 5 h~~ours~~ [Authors]; this procedure removes potentially adsorbed volatile carbon. Aerosol sampling was performed at 30 $m^3 \cdot h^{-1}$ flow rate overnight (22:00 to 06:00 GMT), under the influence of free troposphere airflows. One PM_x sample was collected every 3 days, except in August, when sampling was daily. Concentrations of PM_x were determined by gravimetry following the European standard gravimetric measurement method EN-14907 [R3#C5] (except that filter conditioning was performed at 30–35% relative humidity instead of 50%). The manual gravimetric method is considered optimal for PM_x concentrations > 10 $\mu g \cdot m^{-3}$ (EN-14907), so uncertainties are higher below this threshold value (details in section S1 of the supplement). Blank weighing [R1#C2] room and blank field filters were collected and weighed [R1#C2] as part of the quality assurance / quality control (QA/QC) protocol.

The methods used in the long-term (~ [Authors] 30-years) aerosol chemical composition record of Izaña Observatory are described in detail in previous articles (Rodríguez et al., 2015). Briefly, in the study period (2008–2013) soluble species were determined by ion chromatography ($SO_4^{=}$, NO_3^-, Cl^-; detection limits 0.113, 0.113 and 0.505 $\mu g \cdot m^{-3}$, respectively) and selective electrode (NH_4^+; detection limit 0.056 $\mu g \cdot m^{-3}$). Elemental composition was determined by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES, IRIS Advantage TJA Solutions, THERMO™) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS, X Series II, THERMO™) after acid digestion of the samples. Organic and elemental carbon (detection limits 0.8 and 0.032 $\mu g \cdot m^{-3}$, respectively) were analysed by thermal-optical transmittance (TOT, Sunset Laboratory Inc.™) following the EUSAAR2 protocol (Cavalli et al., 2009).

Because quartz microfiber filters may adsorb volatile carbon very easily due to the high active surface (Chai et al., 2012), the more unstable part of the organic carbon was discarded based on the results of the field blank filters analysis. Sulphate was split into sea salt sulphate (ss-$SO_4^{=}$) and non-sea salt sulphate (nss-$SO_4^{=}$) using the empirical ratio of Na and sulphate ($SO_4^{=}/Na^{+}$ = 0.25) in seawater (Gravenhorst et al., 1978), which assumes there is no sulphate enrichment due to gas-to-particle conversion of the oxidation of marine SO_2 (Bonsang et al., 1980). Organic matter (OM) was determined by using the ratio OM/OC = 1.8 observed in the North American aerosols collected at Pico Observatory in the Azores (Dzepina et al., 2015). Blank field filters were subject to gravimetry and chemical analysis, and mean values were subtracted from the PM_x samples.

The chemical composition data were used for a mass closure of PM_x (table 1). The undetermined fraction of PM, i.e. the difference between the gravimetrically determined PM_x and the sum of the chemical compounds, increased under low PM_x conditions. This has already been observed in previous studies (Ripoll et al., 2015) and is attributed to inaccuracies of the manual gravimetric method under low PM concentrations (< 10 $\mu g \cdot m^{-3}$) and to the relatively higher contribution of water not fully removed during filter conditioning.

2.3 Meteorology, back-trajectories and MCAR plots

We analyzed meteorological re-analysis data from the National Centre for Environmental Prediction / National Centre for Atmospheric Research (NCEP/NCAR) (Kalnay et al., 1996) to study the processes involved in the export and transatlantic transport of aerosols from North America. The analysis includes geopotential heights, winds and omega (vertical wind) at several standard levels (925, 850 and 700 hPa) and precipitation rates.

Three-dimensional 10-day back-trajectories were computed at 00:00 GMT for Izaña using the meteorological input data from the European Centre for Medium-Range Weather Forecasts (ECMWF) and Lagrangian model FLEXTRA (Stohl et al., 1995; Sthol and Seibert., 1998). These back-trajectories were used as input in a self-developed Matlab script (The Mathworks, Natick, USA) which segregates air masses coming from North America and the North Atlantic, from those from Africa, attending to the latitude and longitude values along the transport path towards Izaña (details in section S2 of the supplement). The frequency of the westerlies and of the Saharan Air Layer at Izaña is shown in Fig. 1C. Samples of PM_{10} and $PM_{2.5}$ were associated with westerlies and SAL according to the back-trajectories (Fig. S2).

We determined the Median Concentrations At Receptor (MCAR) plots for the main PM_{10} chemical component using the method described by Rodríguez et al. (2011). 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 [R3#C6]. The MCAR plots were calculated with back-trajectories representative of transatlantic transport from North America. Events linked to (i) back-trajectories from North Africa or (ii) associated with Saharan dust re-circulated over the North Atlantic (exported from North Africa westward and then re-circulated eastward, e.g. as described by Ancellet et al., 2016) were

removed; to identify the latter type of events we also used the output forecasts of the BSC-DREAM8b model (Pérez et al., 2006; Basart et al., 2012) storages at the Barcelona Supercomputing Centre website (http://www.bsc.es/earth-sciences/mineral-dust-forecast-system/bsc-dream8b-forecast/north-africa-europe-and-middle-ea-1). Similar plots were used to analyse seasonality of the frequency of westerlies reaching Izaña, which is linked to the export of North American pollutants. Monthly Transport Route Frequency (TRF) plots for the period 2008–2013 were calculated in a similar way to the MCAR plots, but instead of the median concentration, the total number of back-trajectories passing by each cell grid is represented.

2.4 Complementary data

We used the Global Fire Emissions Database Version 4 including small fires data (GFEDv4.1s; Randerson et al., 2015) to estimate the average burned fraction of each 0.25º x 0.25º grid cell in North America during the study period (2008–2013). The data set was downloaded from the Oak Ridge National Laboratory Distributed Active Archive Center (ORNL DAAC) for biogeochemical dynamics (https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1293). We also made use of Level 3 UV Aerosol Index (AI) data, from the Ozone Monitor Instrument spectrometer onboard satellite Aura (OMI 2008–2013), to study the spatial and temporal variability of dust in North America during the study period (2008–2013). The data set was downloaded from the Giovanni online data system of the NASA Goddard Earth Sciences Data and Information Services Center (GES DISC; http://disc.sci.gsfc.nasa.gov/).

3 Results and discussion

3.1 Chemical characterisation

This study focuses on aerosols transported by the westerlies, i.e. the westerly airstream that flows from North America across the North Atlantic at subtropical and mid-latitudes. Previous studies have shown that the Saharan Air Layer (SAL), i.e. the dusty airstream that expands from North Africa to the Americas, is the most important carrier of aerosols in the tropical and subtropical North Atlantic (Prospero and Carlson, 1972); so first we did a brief comparison of the characteristics of the aerosol composition in these two air streams, with the aim of illustrating the huge differences between them. The frequency of the westerlies and of the Saharan Air Layer at Izaña is shown in Fig. 1C; the westerlies occur with high frequency throughout the year, with a maximum in April–May and a minimum in July–August when Izaña is mostly within the SAL.

Table 1 shows the median chemical composition and mass closure of the PM_{10} and $PM_{2.5}$ aerosols in samples collected at Izaña under the SAL and the westerlies (Fig. 1A–B). The transport of particulate pollutants in the SAL had already been studied by Rodríguez et al. (2011). The SAL impacts on Izaña in July and August, and is linked to the northern shift of the Harmattan – trade winds (Fig. 1A). The summer SAL occurs 1–5 km a.s.l. off North Africa; it is associated with air from the Mediterranean flowing south-westward to the Sahara resulting in the emissions and export of dust to the Atlantic above the marine

boundary layer. The westerlies occur throughout the year and are associated with airstreams from North America that, in some cases, may have circulated around the Azores High (Fig. 1A).

Concentrations of bulk PM_{10} and $PM_{2.5}$ are ~ 20 and 10 times higher in the SAL than in the westerlies, respectively (table 1). Mass closure of PM_x accounts for a rather low fraction of the gravimetrically determined PM_x concentrations under westerly conditions (~ 50–70% of PM_x, table 1), compared to the SAL (70–90% of PM_x, table 1 [Authors]). This is attributed to the relatively high inaccuracy of the manual gravimetric method under low PM_x concentrations described above (see details in section S1 of the supplement). Thus, the sum of the main chemical components (∑ in table 1) is probably a better proxy of the actual bulk PM_x concentrations in the westerlies than the gravimetric PM concentrations.

In the SAL, the PM_{10} aerosol population (median of ∑ – sum of the main chemical components – ~ 41 $\mu g \cdot m^{-3}$; table 1) is basically constituted by dust (78%: 36 $\mu g \cdot m^{-3}$) mixed with organic matter (4.5%: 2.1 $\mu g \cdot m^{-3}$), sulphate (3.8%: 1.8 $\mu g \cdot m^{-3}$), nitrate (1.8%: 0.8 $\mu g \cdot m^{-3}$) and ammonium (0.4%: 0.2 $\mu g \cdot m^{-3}$). In contrast, PM_{10} aerosol in the westerlies (median of ∑ – sum of the main chemical components – ~ 1.8 $\mu g \cdot m^{-3}$) is predominantly constituted by dust (44.5%: 1.1 $\mu g \cdot m^{-3}$), organic matter (12.4%: 0.32 $\mu g \cdot m^{-3}$), nss-$SO_4^{=}$ (9.8%: 0.25 $\mu g \cdot m^{-3}$) and ammonium (2.2%: 0.06 $\mu g \cdot m^{-3}$). Nitrate in the SAL mostly occurs in the coarse range as non-ammonium salt coating dust particles (see details in Rodríguez et al. 2011). In the westerlies nitrate concentrations tend to be extremely low; in the few observed nitrate events, it tended to occur in the sub-2.5 µm range, attributed to an ammonium salt. In the SAL, about ¼ of non-sea-salt-sulphate (nss-$SO_4^{=}$) is present as ammonium sulphate (a-$SO_4^{=}$) linked to anthropogenic sulphur emissions, with the remaining ¾ being non-ammonium sulphate (na-$SO_4^{=}$) most probably linked to soil emissions of gypsum / anhydrite soil minerals in beds of Saharan dry lakes (Rodríguez et al., 2011; Pérez García-Pando et al., 2016) [R1#C3].

Aerosols in the SAL and in the westerlies also exhibit differences in terms of size distribution. PM mass mostly occurs in the sub-2.5µm in the westerlies and in the coarse 2.5–10 µm range in the SAL (table 1).

3.2 North American large-scale meteorology and airstreams

The meteorological scenarios that prompt pollutant export events from North America are described in previous studies (Merril and Moody, 1996; Moody et al., 1996; Stohl et al., 2002; Li et al., 2005; Owen et al., 2006); here a complementary view is provided. We analysed how large-scale circulations over North America evolve over the year, more specifically how they may influence ~~on~~ [R1#C4] the export of aerosols to the Atlantic. The monthly values of key meteorological fields (geopotential heights, winds and omega at several standard levels, 925, 850 and 700 hPa and precipitation rates, e.g. Fig. 2A) were determined with the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis data (Kalnay et al., 1996). To facilitate interpretation on how the variability in meteorology may influence the export of aerosols we also plotted (i) the latitudinal range of the westerlies over the eastern coast of North America (observed in the monthly NCEP/NCAR wind fields; Fig. 3A), (ii) the spatial distribution of SO_2 according to Fioletov et al. (2016) (Fig. 3B) and of major aerosol components according to Park et al. (2003, 2004) (Fig. 4) and (iii) the monthly mean values

of the omega vertical component of wind in selected domains (Fig. 3C and 3D). Finally, in order to link the export of North American pollutants with transatlantic transport, the Transport Route Frequency (TRF) field was determined for each month based on back-trajectories (Fig. 2B). Figure 2 shows examples for illustrative months (January, April, August and November); additional material is presented in the Supplement (Fig. S3).

During January and February, the North Atlantic anticyclone shifts southward, expanding over the Caribbean, resulting in an intense geopotential / pressure gradient (Fig.S2A) and westerly winds over most of [R1#C7] North America (Fig.2A1). The main stream of the westerlies (which we will refer to as 'westerly jet') flows from Western Canada (~55ºN) to Eastern US entering the North Atlantic at relatively low latitudes (36–38ºN in the 850hPa standard level; Fig. 2A1). We refer to the westerly jet over the eastern coast of North America as 'North American outflow', whose latitudinal position over the year is plotted in Fig. 3A. The TRF analysis shows that during this period air from Central and Southern US reaches Izaña (Fig. 2B1).

From March to June, the North Atlantic anticyclone progressively intensifies (Fig.S2B) and the western side of its clockwise atmospheric circulation expands from the inner Gulf of Mexico northward to Central and then to Eastern US, resulting in an airstream that we have called 'the Gulf inflow', which is observed in the wind fields at the 925, 850 and 700hPa levels (Fig. 2A2). The Gulf inflow is observed from March, when the trade winds building up results in an northward inflow across the coast of Texas, which subsequently turns northeastward over Central US (Arkansas to Tennessee–Indiana) and then eastward resulting in a westerly outflow to the Atlantic by the Eastern coast of North America following the clockwise circulation of the North Atlantic high (Fig. 2B2). The analysis of the (vertical) omega component, at the 925, 850 and 700 hPa levels, shows that upward movement of air occurs in the regions affected by the Gulf inflow of warm and humid air from Texas to Indiana, in such a way that air masses from the continental boundary layer of Central to North-eastern US may be exported in the westerlies to the North Atlantic free troposphere (Fig. S3B). Observe in Fig. 3C how omega decreases to negative values (net upward movements) from March to May in Central US (domain 1 in Fig. 3D) under the influence of the Gulf inflow; the decrease in omega in this season is also observed to the East (domain 2 in Fig. 3D). This is consistent with the fact that the storm season occurs in this period (March to June) between Central US (Northern Texas–Kansas) to the west of the Appalachians (Tennessee–Kentucky–Indiana), as reported by NOAA (http://www.ncdc.noaa.gov/climate-information/extreme-events/us-tornado-climatology [R1#C8]; section S4 of the supplement). Along the path of the Gulf inflow there are a number of sources of aerosols and their precursors – including coal-fired power plants (Fioletov et al., 2016) – whose emissions may be lifted to the mid-troposphere during convective processes (Dickerson et al., 1987; Talbot et al., 1998), and then exported to the North Atlantic free troposphere by the westerly circulation, which in this period tends to occur 35–45ºN (Fig. 3A). The export of pollutants from Eastern US to the Atlantic is enhanced by eastward moving cyclones, whose tracks typically occur south of 40ºN in this season (Cooper et al., 2002a); Fig. 3B shows an illustration of this scenario (Cooper et al., 2002a, 2002b) and the mean SO_2 spatial distribution observed by satellite (Fioletov et al., 2016); a dotted blue line shows the typical eastward track of the cyclones ~~typical~~ [R1#C9] in March–April, whereas the blue

arrows indicate a simplified scheme of the associated circulation; the satellite detection of SO_2 over the ocean off the coast of Virginia to New Jersey (35–40ºN in Fig. 3B) evidences the importance of the export of this aerosol sulphate precursor to the Atlantic. The back-trajectory based TRF analysis shows that air masses from Central US (e.g. domain 1 in Fig. 3D) and Eastern US (e.g. domain 2 in Fig. 3D) are regularly transported to Izaña (Fig. 2B2). Spring (March-April) is the season of maximum frequency [R1#C10] of the westerlies at this observatory (23–27 days·month^{-1}; Fig. 1C1) and that has implications for the export of major aerosol components, whose concentrations are high in North-eastern US (Fig. 4).

In July and August, the North Atlantic anticyclone shifts northward (Fig.S2C) resulting in intense trade winds over the Caribbean; the Gulf inflow continues blowing northward across the great plains up to Canada where it connects with the main westerlies jet, whereas southern winds prevail along the Eastern coast of the US (Fig. 2A3). In September, the trade winds, the resulting Gulf inflow and southern winds over the Eastern coast weaken. In this season, the westerlies and the resulting North American outflow shift northward (45–55ºN; Fig. 2A3–2B3, Fig.3A); this is consistent with previous studies showing that cyclone tracks and the resulting warm conveyor belts linked to the export of pollutants tend to occur over Canada (Merryill [Authors] and Moody, 1996; Cooper et al., 2002a). This scenario is illustrated in Fig. 3B, where the cyclone track is highlighted by a dotted red line and the circulation by the red arrow. This is consistent with the seasonal evolution of the omega vertical wind component, which shows the lowest (and negative) values in August and September in Eastern Canada (domain 4 in Fig. 3C and Fig. 3D), indicating upward movements of the North American outflow. At Izaña, the westerlies occur with a minimum frequency in July (12 days) and August (9 days; Fig.1C1), the period in which the observatory is frequently within the easterly Saharan Air Layer (Rodríguez et al., 2011, 2015). Observe how the air masses from inner North America have a lower impact at Izaña compared to at other periods (Fig.2B3).

From October to December, the North Atlantic high shifts southward, expanding over Southeastern US (Fig.S1D); the Gulf inflow weakens and the westerly wind band shifts progressively southward prompting the transport of air from Central US to the North Atlantic (Fig. 2A4–2B4).

An overall analysis indicates marked seasonality in the atmospheric circulations with potential implications for the export and transatlantic transport of major aerosol components. The westerlies (Fig. 2A–2B), including the North American outflow (Fig. 3), occur at lower (subtropical) latitudes in winter than in summer (mid-latitudes). This seasonal shift is also associated with the upward transport of air, which is important in Central US to Eastern US in March–May (domains 1 and 2 in Fig. 3C and 3D) and shifts northward along Eastern North America through spring and summer (domains 2 to 4 in Fig. 3C and 3D), reaching maximum intensity when the North American outflow occurs over Eastern Canada in August and September (domains 4 in Fig. 3D). This affects which source regions of North America impact downwind North Atlantic free troposphere; air masses from Southern US are transported across the Atlantic in winter (Fig. 2B1), from Central US in spring (Fig. 2B2) and from Canada in summertime (Fig. 2B3). This is consistent with the seasonal shift of cyclone tracks, westerlies and warm conveyor belt described in previous studies (Stohl, 2001; Cooper et al., 2002a). Of special relevance is the spring, when

the westerly jet blows over SO_2 source regions coupled with upward movements able to transport aerosols emitted near ground to altitude above the boundary layer (Fig. 3B).

3.3 Transatlantic transport of North American aerosols

We studied the seasonal variability of the sub-10 µm aerosol components under westerly airflow conditions at Izaña and its connection to the transatlantic transport from North America. The chemical composition of 126 samples of PM_{10} collected at Izaña (2008–2013) under westerly airflow conditions, whose back-trajectories are plotted in Fig.S1A, were used.

Under westerly airflow conditions, the time series of the aerosol components typically show a low background level and sporadic peak episodes; for example, nss-$SO_4^=$ shows a background of 0.05–0.15 µg·m^{-3} and peak events 0.5–1.5 µg·m^{-3}, organic matter increase from 0.01–0.2 µg·m^{-3} background level to 1–2.5 µg·m^{-3} peak events, whereas elemental carbon has a background < 0.01 µg·m^{-3} and peak events within the range 0.03–0.1 µg·m^{-3}. For each aerosol component, we determined the seasonal evolution of the monthly 30th, 50th and 80th percentiles as representative of the background levels, central position of concentration distribution and high concentration episodes, respectively. The 30th and 80th percentiles plots are presented in the supplement (S5). The Median Concentrations at Receptor (MCAR) plots were determined for the study of the connection of peak events of aerosol components at Izaña with episodes of North American aerosol export.

3.3.1 Sulphate

Figure 5A1 shows the MCAR plot for nss-$SO_4^=$, whereas Fig. 5B1 shows the value of the 50th percentile (50thP) concentration for each month at Izaña. The monthly 50thP of nss-$SO_4^=$ shows high levels from March to July (0.28–0.41 µg·m^{-3}), with a maximum in March–May (0.33–0.41 µg·m^{-3}), and low levels from September to February (0.06–0.24 µg·m^{3}). The March to July period can be considered the high-sulphate concentration season, given that both the monthly background (30thP) and median (50thP) levels are high in this period (Fig. S5A).

The MCAR plot represents the nss-$SO_4^=$ concentration recorded at Izaña (median value, i.e. 50thP) when the airflows (tracked by back-trajectories) have passed by each pixel of the study domain (Fig.5A1). Regions with relatively high nss-$SO_4^=$ concentrations (0.3–0.5 µg·m^{-3} in yellow to red scale) compared to the background (< 0.1 µg·m^{-3} in blue) are connected to potential transport routes. The MCAR plot suggests that there are two preferential transport paths. The first route points to the transport of nss-$SO_4^=$ from the North-East US at ~40ºN; this is consistent with the high SO_2 emissions (Fig. 3B) and high nss-$SO_4^=$ concentrations (Fig. 4A) typical of this region (Fig. 3B) associated with coal burning power plants (Mann et al., 2010; Fioletov et al., 2011, 2016) and the North American outflow (Fig. 3). Because the North American outflow occurs over this region (NE–US) during a great part of the year (Fig. 3), this is probably the most important nss-$SO_4^=$ export region to the Atlantic. The correlated seasonal evolution of omega in this region (domain 2 in Fig. 3C and 3D) and nss-$SO_4^=$ at Izaña (Fig. 5B1) indicates enhanced upward movements of air from March to July enrich the North Atlantic free troposphere in sulphate

aerosols. Maximum nss-$SO_4^=$ occurs from March to May (Fig. 5B1), when upward air movements associated with the Gulf inflow (Fig. 3), cyclones (Cooper et al., 2002b) and the occurrence of the North American outflow over this region enhances the export of regional pollutants (Fig. 3A and Fig. 4A). A second transport pathway is associated with transatlantic transport at higher latitudes (50ºN) and anticyclonic circulation around the Azores High (Fig. 5A1); this route is associated with the occurrence of the North American outflow over Northern US and Canada, from mid-summer (August) to mid-autumn (November; Fig. 3A1). Observe how the drop in the median (50^{th}P: from 0.30 to 0.20 µg·m^{-3} Fig. S5A2) and of the background (30^{th}P: from 0.26 to 0.16 µg·m^{-3}; Fig. S5A3) nss-$SO_4^=$ concentrations from July to August is associated with the northward shift of the main westerly stream and the North American outflow (Fig. 3A1); from August on the westerly jet occurs at higher latitudes, over Canada, in regions with less [R1#C11] nss-$SO_4^=$ compared to NE–US (Fig. 4A), as a result less nss-$SO_4^=$ is exported and transported across the Atlantic.

3.3.2 Nitrate

The MCAR plot and the monthly 50^{th}P of NO_3^- at Izaña are plotted in Fig. 5A2 and 5B2, respectively. Nitrate was present in extremely low concentrations most of the time. Concentrations were < 0.05 µg·m^{-3} in 97 samples, and > 0.1 µg·m^{-3} in only 21 samples; for this reason the 50^{th}P was ~zero for most months (Fig. 5B2). High NO_3^- concentration events (80^{th}P > 0.1 µg·m^{-3}) were mostly recorded in winter and early spring (January–April, Fig. 5B2), when high NO_3^- was within the range 0.2–0.8 µg·m^{-3}. This is typical behaviour for ammonium nitrate, which mostly forms under low temperature conditions, whereas gaseous nitric acid prevails in warmer environments (Squizzato et al., 2013). The MCAR plot shows transport of NO_3^- at low latitudes 30–35ºN (Fig. 5A2), which is consistent with the circulation of the westerlies (e.g. Fig. 2A1 and Fig 2B1) and the North American outflow in winter months (e.g. Fig. 3A1), when most of the above-described high NO_3^- events occur (Fig. 5B2). 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 (NH_3 linked to emissions in agriculture fields treated with fertilizers and HNO_3 due to oxidation of NO_x linked to fossil fuel combustion) and (ii) suitable thermodynamic conditions (rather low temperature and enough high relative humidity) [R1#C12]. Our results suggest that nitrate export events from North America may be associated with NW winds (e.g. as the main stream of the westerlies in winter, Fig. 2A1) over this high nitrate region (Fig.4B) followed by export at 35–30 ºN (Fig. 5B1) under geopotential / pressure systems that should be studied in future research. The MCAR plot also suggests a second transport route similar to that observed for nss-$SO_4^=$, i.e. transatlantic transport at high latitudes (50ºN) and circulation around the Azores High (Fig. 5B1) which is probably associated with the autumn events (Fig. 5B2), when the North American outflow occurs over Canada (Fig. 3A1). The nitrate concentrations we observe at Izaña are similarly low to those registered by Dzepina et al. (2015) at the Pico free troposphere site in the Azores linked to long range transport from North America. Because nitrate may experience negative artefacts during sampling (Schaap et al., 2004; Vecchi et al., 2009), we cannot discard underestimations; further artefact-free real-time nitrate measurements should be included in the long term aerosol measurements program [R2#C5].

3.3.3 Ammonium

The MCAR plot and the monthly 50thP of NH_4^+ at Izaña are shown in Fig. 5A3 and 5B3, respectively. Median (50thP) and 80thP concentrations present a maximum in April–May (Fig. 5B3 and Fig. S5C1), as nss-$SO_4^=$. The MCAR plot for NH_4^+ shows two main transport pathways which resemble those of nss-$SO_4^=$; one transport pathways from North-eastern US at ~40 ºN and a second transport route pointing to the occurrence of the North American outflow by Canada and subsequent transatlantic transport at high latitudes and circulation around the Azores High (Fig. 5A3). Because of the prevalent extremely low levels of nitrate, ammonium is attributed to ammonium-sulphate in most events. The transport (north-eastward export) routes we observe for nss-$SO_4^=$ and NH_4^+ in Eastern US (Alabama–Tennessee–Virginia; Fig. 5A1 and 5A3) are similar to those associated with the passage of spring cyclones and front in the region, prompting the export of pollutants to the Atlantic (Cooper et al., 2002b).

3.3.4 Elemental Carbon

Figures 6A1 and 6B1 show the MCAR plot and the monthly 50thP and 80thP of EC, respectively. The analysis of the monthly 50thP and 80thP values discloses two relevant periods, associated to high (May–September) and low (January–April) EC events. We associated this seasonal variability of the EC transported by the westerlies to Izaña (i) with the spatial distribution of the EC source regions in North America (Fig. 4D) and (ii) with the seasonal shift of the westerlies (Fig.2A1–2A4) and the North American outflow (Fig. 3). The highest surface concentrations of EC are estimated to occur in what we have called "EC-rich NE–US regions", which include large urban areas placed 40–45 ºN south of the great lakes to the Atlantic coast (e.g. Chicago, Detroit, New Jersey, Philadelphia and New York; Fig. 4D), linked to fossil fuel combustion (mostly diesel exhaust emissions and coal burning) according to Park et al. (2003).

The season of high EC concentrations at Izaña occurs from May to September, when the westerlies shift northward from 40ºN to 55ºN (Fig. 3A) affecting the "EC-rich NE–US regions" (40–45ºN, Chicago to New York, Fig. 4D). This seasonal shift is associated with a rise of the upward air movements in Eastern US (domains 2 and 3 in Fig. 3C and 3D), including the "EC-rich NE–US regions" (domain 3 in Fig. 3C and 3D), that enhances the export of EC to the North Atlantic free troposphere in the North American outflow. The highest median EC concentrations at Izaña are observed in August and September (~0.03 $\mu g \cdot m^{-3}$; Fig. 6B1 and Fig. S5D1), when the eastward propagating cyclones over Canada prompt the export of pollutants from these "EC-rich NE–US regions" to the Gulf of Maine and the Atlantic (Merrill and Moody, 1996), in a scenario illustrated in Fig 3B; the lowest values of omega in Eastern Canada (domain 4 in Fig. 3C and 3D) occur in August and September, which indicates a great potential to lift boundary layer air to the North Atlantic free troposphere. This interpretation is consistent with the MCAR plot for EC, which shows a clear transport pathway (at high latitudes, ~50ºN) from Canada and these "EC-rich NE–US regions" to the Gulf of Maine and then to the Atlantic with subsequent circulation around the Azores High (Fig. 6A1); this EC transport route is similar to the prevalent transport pathway of August and September (Fig. 2B3).

Low EC concentrations at Izaña occur between January and April, when the westerlies and the North American outflow occur at low latitudes (<40ºN; Fig. 3A), to the south of the EC source regions (40–45ºN; Fig. 4D).

Figure 7 shows the mean burnt fraction (%) of each 0.25º x 0.25º grid cell associated with fires, which occur mainly in Southeastern US in January–February (Fig. 7A); it then spreads northward from March on over Central US to Southern Canada (Fig. 7B) and over Canada and NW US in June–September (Fig. 7C) and then shifts southward to US (Fig. 7D). Boreal fires prompt high EC concentrations in Canada (Park et al., 2005) in NW and North-Central US (Washington, Oregon and Nevada; Park et al., 2003) that can be exported to the Atlantic in the uplifting North American outflow, potentially contributing to the EC records at Izaña in August and September (Fig. 6B1).

3.3.5 Organic Matter

Figures 6A2 and 6B2 show the MCAR plot and the monthly 50thP of OM. This aerosol component shows very marked seasonal evolution, with high levels from January to July, and a maximum from March to May (Fig. 6B2, Fig. S5E1–E3). This is consistent with the MCAR plot (Fig. 6A2), which shows a transport route from Southeastern US to Izaña at low latitudes (30–40ºN), a common circulation of winter and spring (Fig. 2A1–2A2). From August to December, OM concentrations transported by the westerlies to Izaña are low, associated with the occurrence of the westerlies over North America at high latitudes (Fig.2A3–2A4).

The seasonal evolution of OM is very different (~opposite) to that of EC (Fig. 6A2 and 6B2). Air masses transported from Southeastern US to Izaña (January to April) are rich in OM and relatively poor in EC (Fig. 6A2 and 6B2), whereas the air transported from NE US to Izaña (typically from July to September) is poor in OM and rich in EC (Fig. 6A2 and 6B2). This is consistent with the spatial distribution of these aerosol species in the US (Fig. 4D and 4E) and suggests that in Southeastern US there is a significant contribution to OM of sources that are not related to combustion, but probably to biogenic emissions [R1#C13]. 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) [R2#C4 and R3#C7]. Globally, biogenic volatile organic carbon emissions (BVOCs), some of which are [Authors] precursors of secondary OA, are comprised of isoprene (~50%), methanol, ethanol, acetaldehyde, acetone, α-pinene, β-pinene, t-β-ocimene, limonene, ethene, and propene (~30%), and other compounds (mostly terpenoids; ~17%) (Guenther et al., 2012). Biogenic emissions are among [Authors] the principal sources [Authors] 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)~~, with biogenic secondary OA predicted to contribute around 10-20% of $PM_{2.5}$ mass (Liao et al., 2007)~~ [R2#C4 and R3#C7]. A scenario of biogenic emissions higher in SE–US than in NE–US is consistent with global distribution of the secondary organic aerosols, whose concentrations are usually higher near to the tropics than at mid-latitudes (Guenther et al., 2012; Sindelarova et al., 2014). The importance of the spatial variability of the OM and EC sources and the latitudinal shift of the westerlies over Eastern North America is illustrated

from July to August, when a drop in OM concentrations (0.85 to 0.20 μg·m^{-3}; Fig. 6B2) and an increase in EC concentrations (0.005 to 0.03 μg·m^{-3}; Fig. 6B1) is associated with the northern shift of the North American outflow (43º to 50ºN; Fig. 3A).

3.3.6 Mineral dust

Figures 8A and 8B show the MCAR plot and the monthly 50thP of calcium, aluminium and the associated bulk dust concentrations. These aerosol components exhibit high concentrations from February to May (Fig. 8B1–B3; Fig. S5F–S5H). The MCAR plot shows a pattern of North American dust export at low latitudes (~35ºN, through North Carolina) towards the north-east which, once over the Atlantic, follow the anticyclonic circulation around the Azores High to Izaña (Fig. 8A1–A3). We attribute these events to dust emissions in a region that expands from SW Texas northward throughout the High Plains, and subsequent dust export to the Atlantic. Figure 8C shows the Major Dust Activity Frequency (MDAF) detected by satellite; dust activity is observed in SW Texas (Chiguagua–Big Bend Desert) in February, through March to May the activity expands northward across the High Plains (western Texas to Nebraska); other dust sources with lower potential to impact on the North Atlantic are also observed in western US (Great Basin, Mojave Desert and Colorado Plateau). The High Plains are among the major dust sources in North America, and these sources are considered anthropogenic (linked to agriculture), with maximum activity between February and May (Ginoux et al., 2012). Dust emissions and eastward mobilisation is associated with the intense westerly winds linked to eastward moving cyclones, which also prompt the upward transport of dust to several ~~kilimetres~~ km [Authors] above ground, according to Novlan et al. (2007); this scenario is illustrated in Fig. 8A3, showing how the associated air mass track is consistent with the dust export and transatlantic transport route to Izaña observed in our analysis. During these spring events high dust concentrations (100s of μg·m^{-3}) are lifted to altitudes 6–12 km a.s.l. over Southern US (Talbot et al., 1998). Upward transport of dust is also associated with convective activity in Central US, Colorado and Oklahoma, in May-June (Corr et al., 2016). The correlation we found between the seasonal evolution of omega in domain 1 (Fig.3C and 3D) and dust at Izaña (Fig.8B3) supports the idea that from February to May westerly winds and the uplifting of air in the High Plains enrich the North American outflow and the westerly jet in the in dust aerosols.

3.3.7 Sea salt

Sea salt concentrations at [R2#C8] Izaña are extremely low, with monthly 50thP values 0.07 to 0.22 μg·m^{-3} between December and May (Fig. 9). These low concentrations are typical of free troposphere sites; in fact, sea salt at Izaña (average = 0.25 μg·m^{-3}, median 0.16 μg·m^{-3}) is about two orders of magnitude lower than in the marine boundary layer of the Canary Islands (average ~ 11 μg·m^{-3}, Querol et al., 2004). The extremely low concentrations of this marine aerosol at Izaña supports our interpretations, i.e. the aerosols transported by the westerlies to Izaña are mostly linked to emissions and upward transport in continental regions of North America, and not over the ocean.

3.3.8 Mass closure of aerosols

Figure [Authors] 10 shows the contribution of each species to bulk PM_{10} aerosol mass in samples collected in the westerlies at Izaña; data are classified from the highest to the lowest levels. We considered two approaches: including and excluding mineral dust. When mineral dust is not included (Fig. 10A), the most important contributors to bulk PM_{10} are by far nss-$SO_4^=$ and OM. In the 1st–50th percentile range for the sum of aerosol components (a proxy of background levels) - 0.05 to 1.0 $\mu g \cdot m^{-3}$ - the most important contributors are nss-$SO_4^=$ (0.19 $\mu g \cdot m^{-3}$ on average, accounting for 38% of the sum of chemical species) and OM (0.14 $\mu g \cdot m^{-3}$, 30%). In the 75st–95th percentile range (a proxy of high load of aerosol events) - ~2.0 to 3.6 $\mu g \cdot m^{-3}$ - the most important contributors are OM (1.43 $\mu g \cdot m^{-3}$, 57%) and then nss-$SO_4^=$ (0.48 $\mu g \cdot m^{-3}$, 19%). These results are consistent with previous studies that did not include dust, such as Park et al. (2004, 2005), who focused on the composition of aerosols in the background boundary layer of US, and Dzepina et al. (2015), who studied the aerosols transported from North America to the North Atlantic free troposphere at Pico observatory in the Azores.

When mineral dust is considered, it becomes the most important contributor to sub-10µm aerosol mass (Fig. 10B). In the 1st–50th percentile range (a proxy of background levels), bulk PM_{10} = 0.15 – 2.54 $\mu g \cdot m^{-3}$, the most important contributors to bulk aerosol mass are dust (0.78 $\mu g \cdot m^{-3}$ on average, accounting for 53% of bulk mass), nss-$SO_4^=$ (0.21 $\mu g \cdot m^{-3}$, 14%), OM (0.27 $\mu g \cdot m^{-3}$, 18%) and NH_4^+ (0.07 $\mu g \cdot m^{-3}$, 5%). In the 75th–95th percentile range (a proxy of aerosol events), bulk PM_{10} = 3.9–8.9 $\mu g \cdot m^{-3}$, the most important contributors to bulk aerosol mass are dust (2.8 $\mu g \cdot m^{-3}$, 56%) and OM (1.23 $\mu g \cdot m^{-3}$, 24%) followed by nss-$SO_4^=$ (0.47 $\mu g \cdot m^{-3}$, 9%) and NH_4^+ (0.1 $\mu g \cdot m^{-3}$, 2%). The lack of previous studies on transatlantic transport of North American dust make the comparison with previous data difficult; it should be highlighted that Ancellet et al. (2016) detected this transatlantic transport in June (out of the seasonal maximum). Environmental studies have shown that the conversion of natural lands to agriculture and pasturage fields has had a number of impacts in North America (Nordstrom et al., 2004, Wu et al., 2007). Moreover, research has predicted that this change of land use may increase (MNP, 2006, Lawler et al., 2014), and this suggests an enhancement of dust impacts in downwind regions [R2#C6].

Our overall results evidence that dust and organic matter are the most abundant aerosols transported from North America to the North Atlantic free troposphere.

4 Conclusions

A ~5-year record of aerosol chemistry at Izaña Observatory (located at ~2400 m.a.s.l. in Tenerife, the Canary Islands) was used to study the transatlantic transport of aerosols. This study shows that North America is a major source of aerosols, which are transported by the westerly winds across the North Atlantic free troposphere at subtropical and mid-latitudes. The composition of aerosols carried by the westerlies experiences a marked seasonal evolution which is influenced by (i) the spatial distribution of the aerosol sources in North America and (ii) the seasonal variability of the large-scale meteorology in North America. Of special meteorological relevance is the seasonal shift in the westerly jet and the North American outflow, which migrate from low latitudes in winter (~32ºN, January–March) to high latitudes in summer (~52ºN, August–September). The export of boundary layer air laden in aerosols to the North

Atlantic free troposphere is enhanced by the occurrence of cyclones that move eastward with the westerly atmospheric circulation.

We found that the westerlies carry high loads of:

- mineral dust from February to May, associated with dust emissions in a region that expands from SW Texas (Chiguagua–Big Bend Desert) northward through the High Plains (western Texas to Nebraska), and subsequent dust export to the Atlantic associated with eastward moving cyclones, westerly winds and the North American outflow, which in this period migrate from 35ºN in February to 40ºN in May,

- non sea-salt-sulphate and ammonium from March to May, when cyclones and the associated outflow occur over North-eastern US, where the highest SO_2 emissions occur in North America,

- organic matter from February to May, when cyclones and the associated outflow occur over regions of Eastern US rich in organic aerosols according to previous studies,

- elemental carbon in August and September, when cyclones, the westerly jet and the North American outflow occur at high latitudes (50 to 55ºN) favouring the export of boundary layer air from the regions where the highest concentrations of elemental carbon occur in North America according to previous studies (Chicago to New York, 40–45ºN),

The concentrations of sub10-µm aerosol mass (PM_{10}) that reach Izaña Observatory after transatlantic transport typically range between 1.2 and 4.23 $\mu g \cdot m^{-3}$ (20^{th} and 80^{th} percentiles). The most important contributors to background aerosols (when PM_{10} is within the 1^{st}–50^{th} percentiles = 0.15–2.54 $\mu g \cdot m^{-3}$) are North American dust (53%), organic matter (18%) and non sea-salt-$SO_4^{=}$ (14%). High PM_{10} events (75^{th} – 95^{th} percentiles = 3.9–8.9 $\mu g \cdot m^{-3}$) are prompted by dust (56%), organic matter (24%) and nss-$SO_4^{=}$ (9%). Our results suggest that a significant fraction of organic aerosols may be linked to sources other than combustion (e.g. biogenic) and that North American dust may be linked to anthropogenic dust sources linked to the use of soil.

The overall results indicate that future long-term evolution of the aerosol composition in the North Atlantic free troposphere will be influenced not only by air quality policies applied in urban and industrial areas, but also by the use of potential dust emitter soils in North America, specially those lands linked to agriculture and pasturage activities. These dust emissions should be considered in the regulations on air quality and climate change mitigation [R2#C6].

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Table 1. Mass closure and median concentrations of PM_{10} and $PM_{2.5}$ components in samples collected when Izaña was (i) within the Saharan Air Layer (SAL) and (ii) within the westerlies (WES). Only days when PM_{10} and $PM_{2.5}$ were sampled simultaneously were taken into account. The percentage of the corresponding parameter with respect to the PM is shown.

	Saharan Air Layer				westerlies			
	PM_{10}	*%*	$PM_{2.5}$	*%*	PM_{10}	*%*	$PM_{2.5}$	*%*
NS	146		146		96		96	
mass closure								
PM, $\mu g \cdot m^{-3}$	46.42		21.27		2.54		2.19	
$\sum$, $\mu g \cdot m^{-3}$	40.87	***88***	15.59	***73***	1.78	***70***	1.05	***48***
undetermined, $\mu g \cdot m^{-3}$	5.55	***12***	5.68	***27***	0.75	***30***	1.14	***52***
median concentrations								
dust, $\mu g \cdot m^{-3}$	36.07	***77.7***	13.18	***62.0***	1.13	***44.5***	0.50	***22.8***
sea salt, $\mu g \cdot m^{-3}$	< 0.01	***~0***	< 0.01	***~0***	0.01	***0.5***	0.01	***0.5***
EC, $\mu g \cdot m^{-3}$	< 0.01	***~0***	< 0.01	***~0***	0.02	***0.6***	0.01	***0.3***
OM, $\mu g \cdot m^{-3}$	2.07	***4.5***	1.00	***4.7***	0.32	***12.4***	0.26	***11.7***
NH_4^+, $\mu g \cdot m^{-3}$	0.18	***0.4***	0.17	***0.8***	0.06	***2.2***	0.06	***2.7***
NO_3^-, $\mu g \cdot m^{-3}$	0.82	***1.8***	0.17	***0.8***	< 0.01	***~0***	< 0.01	***~0***
$SO_4^=$, $\mu g \cdot m^{-3}$	1.73	***3.8***	1.06	***5.0***	0.25	***10.0***	0.22	***10.0***
sulphate speciation								
ss-$SO_4^=$, $\mu g \cdot m^{-3}$	< 0.01	***~0***	< 0.01	***~0***	< 0.01	***~0***	< 0.01	***0.1***
nss-$SO_4^=$, $\mu g \cdot m^{-3}$	1.72	***3.7***	1.05	***4.9***	0.25	***9.8***	0.22	***10.0***
a-$SO_4^=$, $\mu g \cdot m^{-3}$	0.48	***1.0***	0.45	***2.2***	nd	***nd***	nd	***nd***
na-$SO_4^=$, $\mu g \cdot m^{-3}$	1.22	***2.6***	0.55	***2.6***	nd	***nd***	nd	***nd***

NS: number of samples. **PM**: Particulate matter obtained with the gravimetric method. $\sum$: summation of the major chemical species (dust + sea salt + EC + OM + NH_4^+ + NO_3^- + $SO_4^=$). **nd:** not determined.

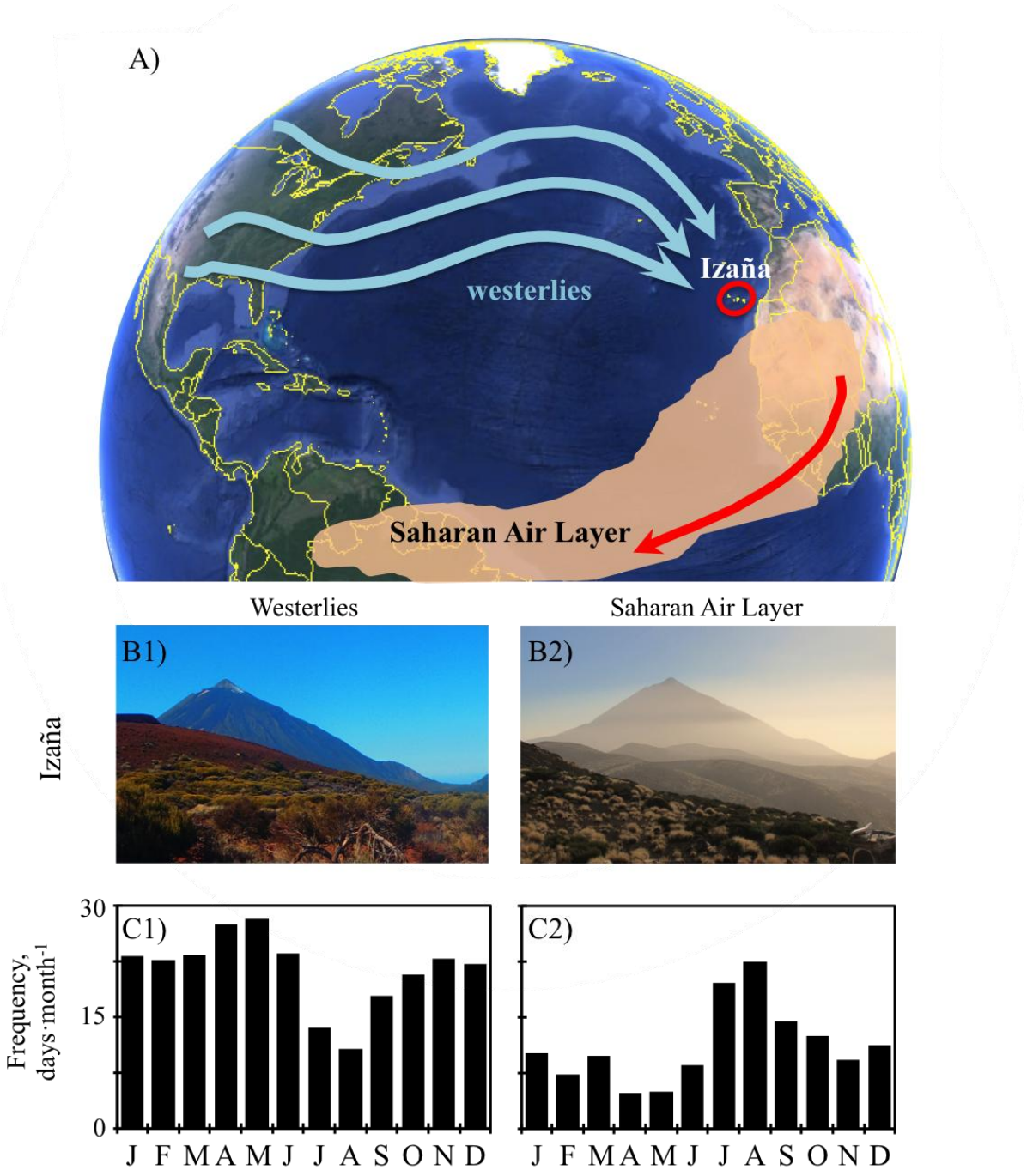

Figure 1. (**A**) Location of the Izaña Observatory with an illustration of the Saharan Air Layer and the westerlies. (**B**) View from the Izaña Observatory to the west under the westerly and Saharan Air Layer conditions. (**C**) Monthly frequency (number of days per month) of westerly and Saharan Air Layer at Izaña based on backtrajectories.

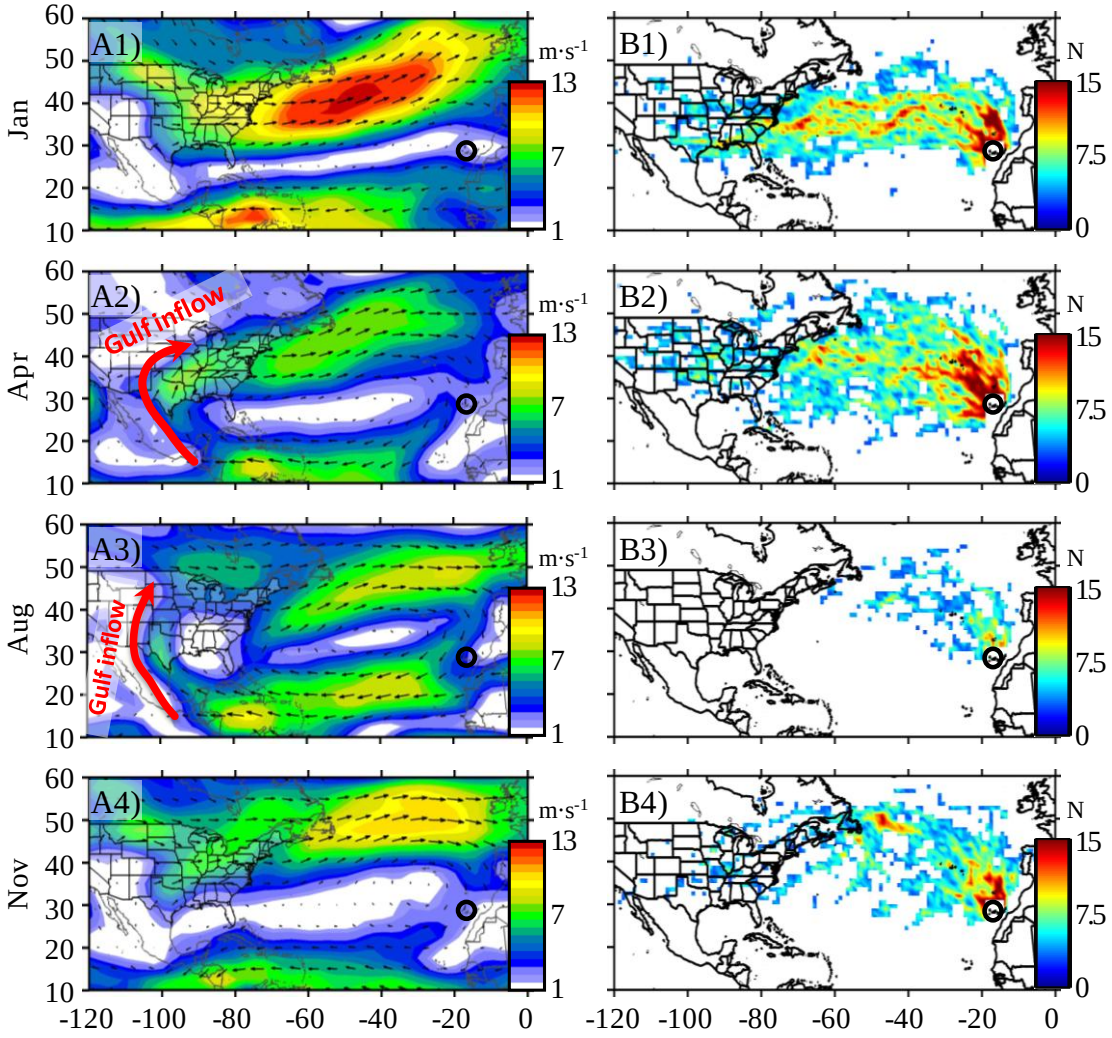

Figure 2. (A) Wind vector at 850 mb and (B) Transport Route Frequency (TRF) for January (Jan), April (Apr), August (Aug) and November (Nov) of the period 2008–2013. The Gulf inflow is highlighted as well as the location of Izaña (black circle) [R1#C5].

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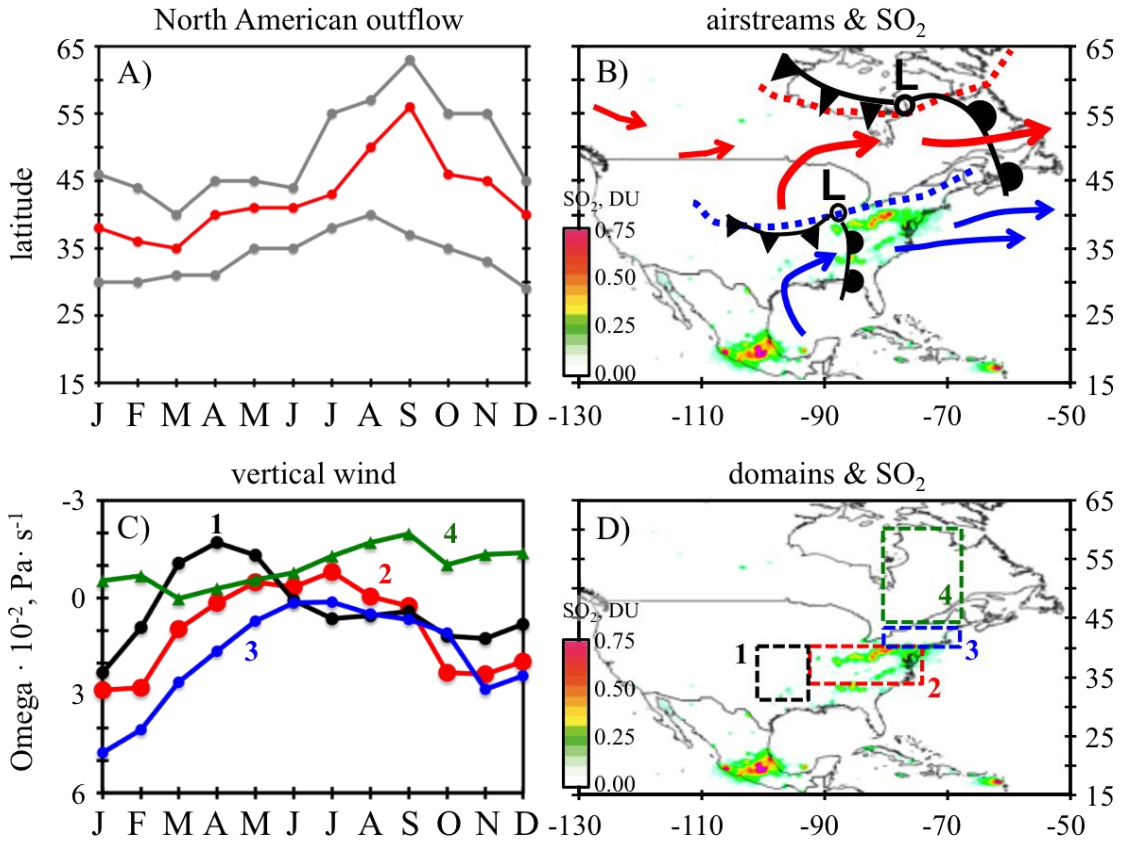

Figure 3. (A) Latitudinal ranges at which the westerlies occurs over the Eastern coast of North America. Grey circles: maximum and minimum latitude of the outflow. Red circles: centre of the outflow (**B**) Meteorological scenarios associated with export of pollutants (according to Cooper et al., 2002a, 2002b, Merryill [Authors] and Moody, 1996) and circulations (blue: Jan-May, red: Jul–Aug) illustrated over mean SO_2 values observed by satellite by Fioletov et al., (2016) - Copyright by Author(s) 2016. CC Attribution 3.0 License -. **(C)** Monthly average values of the omega vertical wind component at the 850hPa level (negative values indicate upward movements) calculated for the domains [R1#C6] illustrated in plot (D). **(D)** Domains 1 (32–40º N, 90–100º W), 2 (35–40º N, 75–90º W), 3 (40–43º N, 70–80º W) and 4 (46–60º N, 70–80º W).

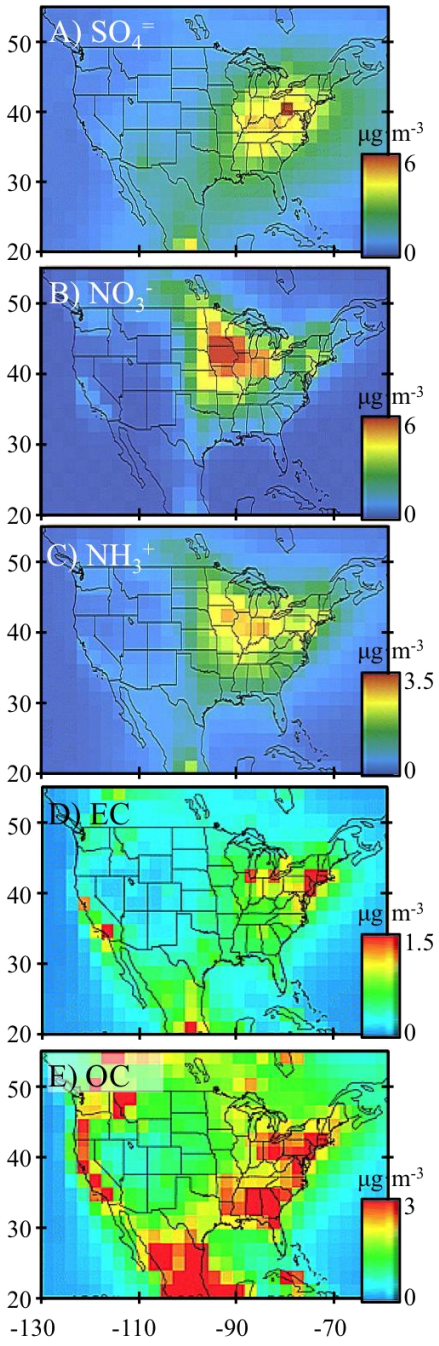

Figure 4. Mean surface concentrations of the **(A)** $SO_4^=$, **(B)** NO_3^-, **(C)** NH_3^+, **(D)** EC and **(E)** OC in North America obtained in previous studies by GEOS-CHEM modelling validated with the observations in the network IMPROVE (Park et al. 2003, 2004, Copyright by the American Geophysical Union).

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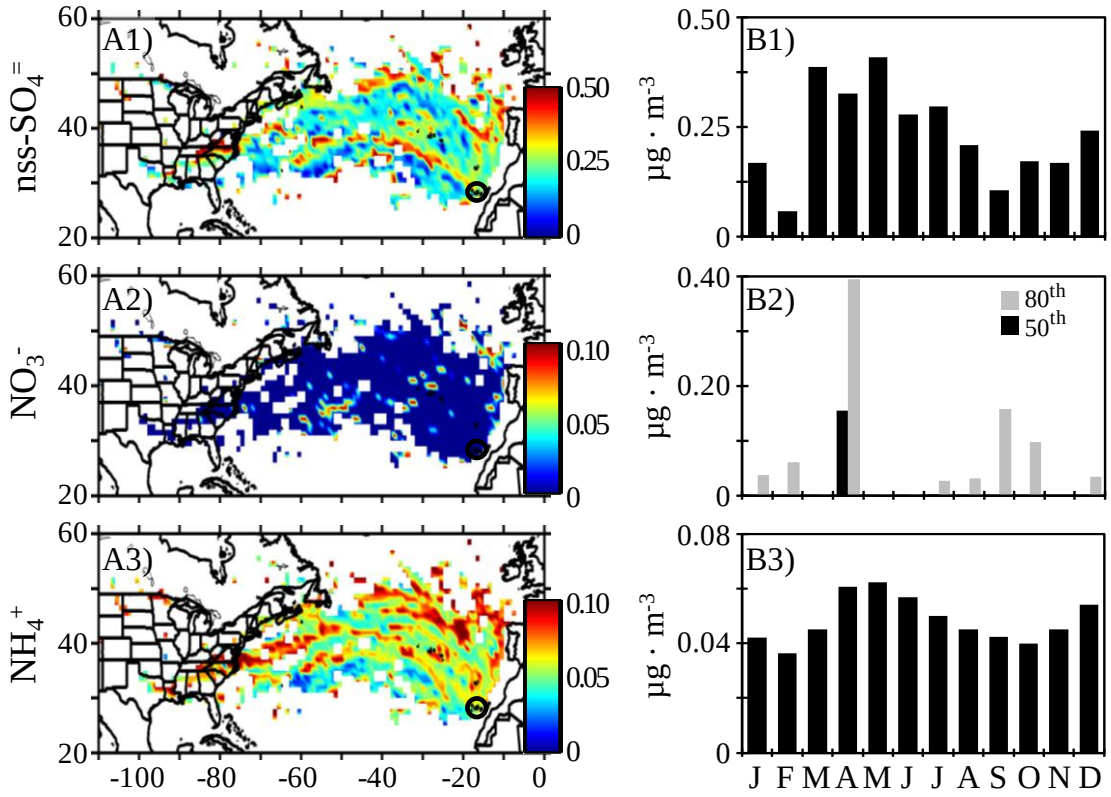

Figure 5. (A) Median Concentration At Receptor (MCAR) plots and (**B**) Monthly median distribution for nss-$SO_4^{=}$, NO_3^-, and NH_4^+ for the study period. Percentiles 50 and 80 are shown for NO_3^-. The MCAR plots maximum concentration tick label includes concentration higher than this upper limit. The location of Izaña is highlighted (black circle) [R1#C5].

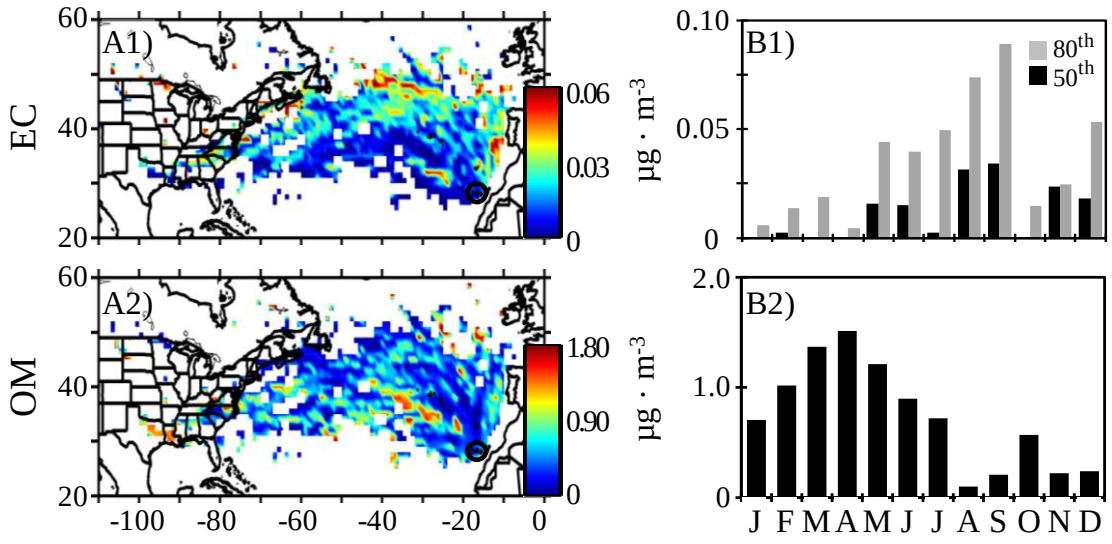

Figure 6. (A) Median Concentration At Receptor (MCAR) plots and (**B**) Monthly median distribution for elemental carbon (EC) and organic matter (OM) for the study period. Percentiles 50 and 80 are shown for EC. The MCAR plots maximum concentration tick label includes concentration higher than this upper limit. The location of Izaña is highlighted (black circle) [R1#C5].

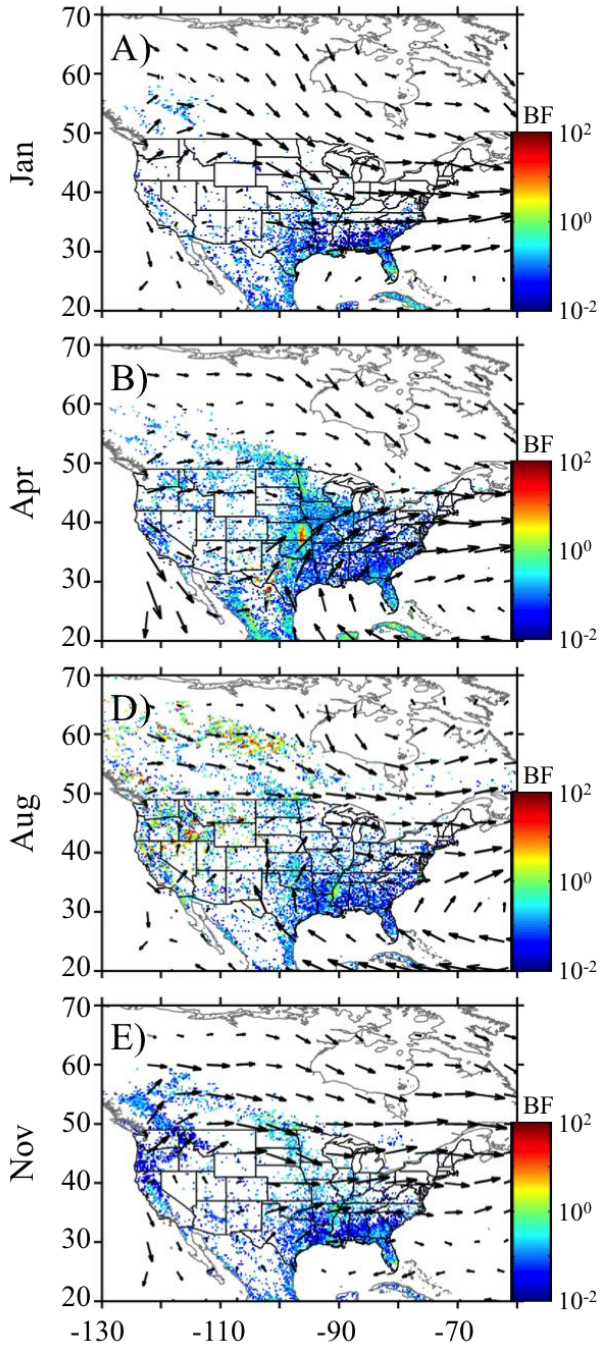

Figure 7. Burned fraction (BF) – of each 0.25º x 0.25º grid cell – and vector wind at 850 mb averaged from 2008 to 2013 for (**A**) January, (**B**) April, (**C**) August and (**D**) November. The Global Fire Emissions Database Version 4 including small fires data (GFEDv4.1s; Randerson et al., 2015) was downloaded from the Oak Ridge National Laboratory Distributed Active Archive Centre (ORNL DAAC) for biogeochemical dynamics (https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1293).

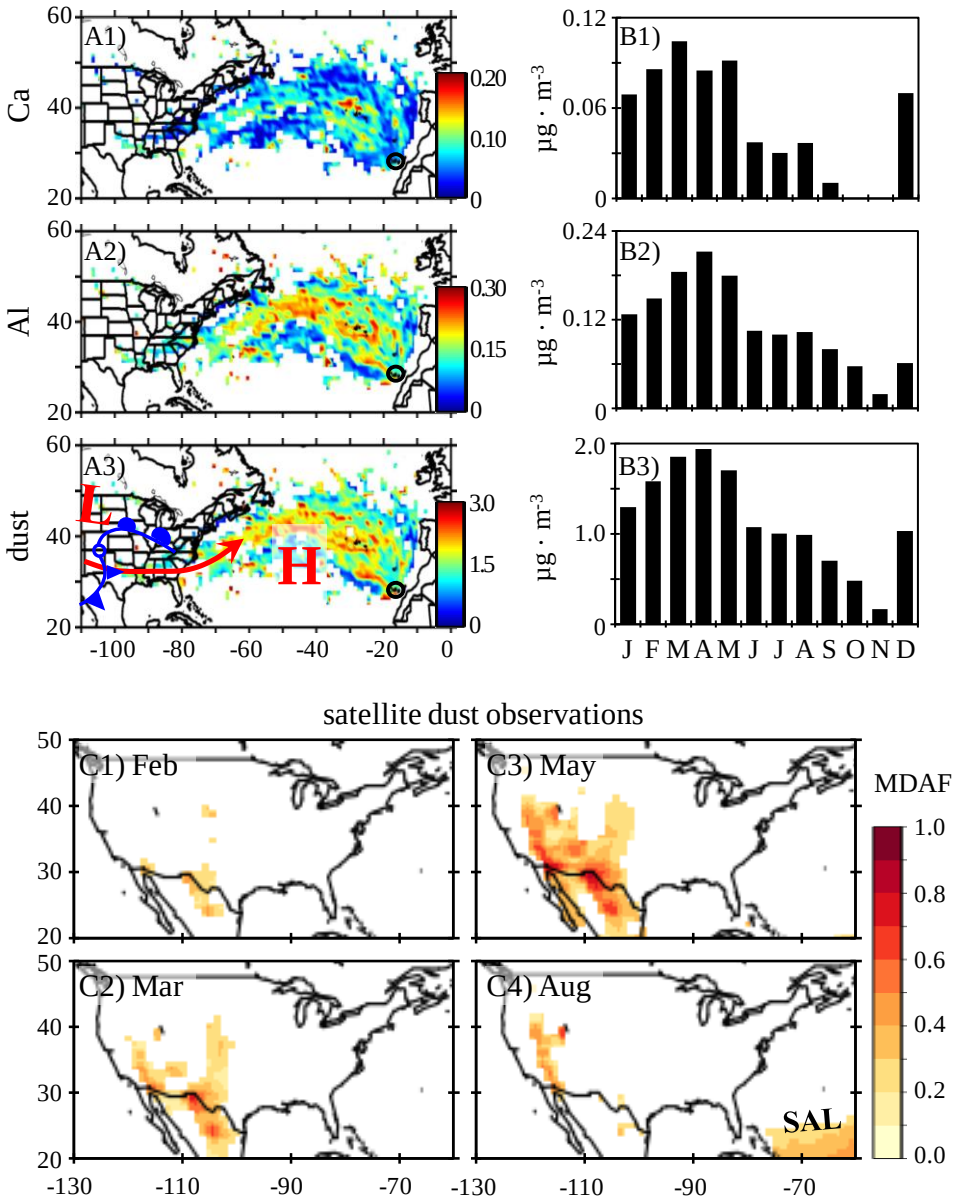

Figure 8. (A) Median Concentration At Receptor (MCAR) plots and (**B**) monthly median distribution for calcium (Ca), aluminium (Al) and dust, and (**C**) major dust activity frequency (**MDAF**) for the study period: the number of days with AI values > 1 divided by the total number of days with available AI data in % [R1#C14] (data source: http://disc.sci.gsfc.nasa.gov/). The MCAR plots maximum concentration tick label includes concentration higher than this upper limit. The location of Izaña is highlighted (black circle) [R1#C5].

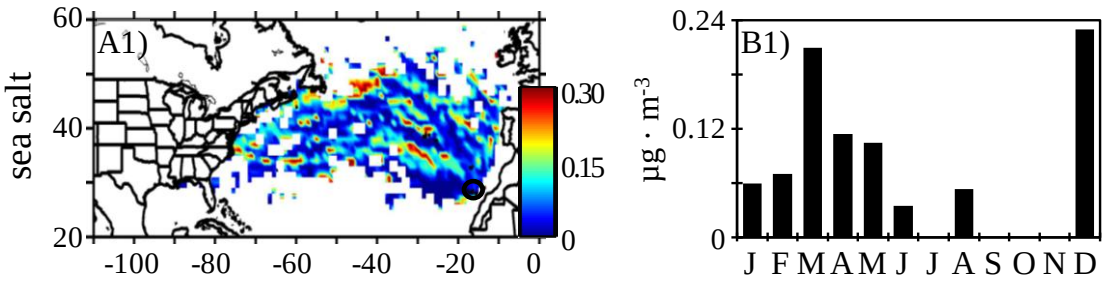

Figure 9. (A) Median Concentration At Receptor (MCAR) plots and (**B**) Monthly median distribution for sea salt for the study period. The MCAR plots maximum concentration tick label includes concentration higher than this upper limit. The location of Izaña is highlighted (black circle) [R1#C5].

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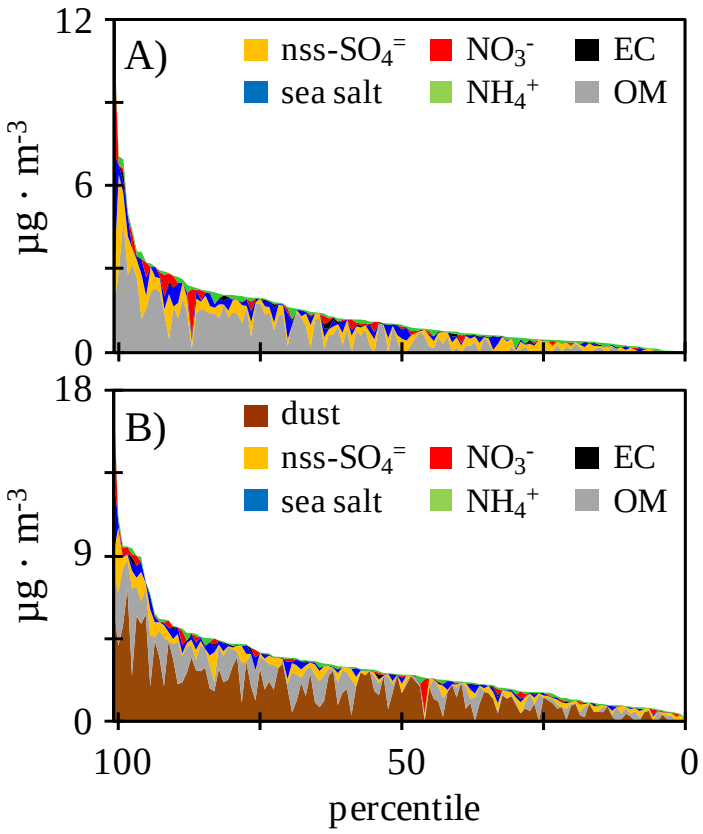

Figure 10. Contribution of each aerosol specie to bulk PM_{10} in samples collected at Izaña under westerly airflow conditions; data classified from the highest to the lowest levels**. (A)** Considering major [R1#C16 and R3#C8] components except dust. **(B)** Considering all major [R1#C16 and R3#C8] components including mineral dust.

Revised Supplement

Changes are highlighted in yellow for reviewer#1, in green for reviewer#2, in pink for reviewer#3 and in blue for authors. Brackets indicate the reviewer and comment that prompt the change.

S1 Uncertainty of the gravimetric method

Concentrations of PM_x were determined by gravimetry following the EN-14907 procedure (except that filter conditioning was performed at 30–35% relative humidity instead of 50%). The combined standard uncertainty (uc), associated to a specific PM_x concentration, is expressed as the combination of the individual sources of uncertainty identified in EN-14907; by multiplying uc by the coverage factor k=2, the expanded uncertainty (U) is obtained. U implies that there is a 95% probability that the true value lies within ± U of the measured value and it was calculated for individual samples, with the Izaña sampling conditions (sampling time: 8h; sampler flow: 30 $m^3 \cdot h^{-1}$), as represented in Fig. S1. The expanded uncertainty associated for PM_x<10 $\mu g \cdot m^{-3}$ is ± 5 $\mu g \cdot m^{-3}$. U (%) will depend on the sample mass, with U>50% for PM_x<10 $\mu g \cdot m^{-3}$.

The European standard procedure sets the relative humidity (for filter conditioning and during weighing) to 50% to avoid the effect of water absorption by the filter material, and therefore in the filter mass. The conditioning to 30-35% used in the long-term program of Izaña is suitable for the low relative humidity of the ambient air and is consistent with the measurements of other aerosol properties (optical properties and number size distribution) that are performed after condition the aerosol sample at relative humidity lower than 40% according to GAW standardization [R2#C1].

S2 Westerlies and Saharan air Layer

Westerlies (WES) and Saharan Air Layer (SAL) air masses were separated attending to the back-trajectories computed with FLEXTRA. A self-developed software, programmed in MATLAB 7.4 (The Mathworks, Natick, USA), scanned the latitude and longitude points for each day classifying samples as WES or SAL. This sorting depended on if the air masses passed over Africa or came from North America and the Atlantic Ocean. After the classification, we used the output of the Barcelona Supercomputing Centre-DREAM8b – atmospheric dust forecast system – in order to verify that the westerlies associated back-trajectories were not Saharan dust re-circulated events. From the total of the 401 PM_{10} samples, 126 were collected under the westerlies (Fig. S2A), 177 under the SAL (Fig. S2B) and 98 were Saharan dust re-circulated events (Fig. S2C). Figure S1A shows the back-trajectories used as input for the Median Concentrations At Receptor (MCAR) plots of the PM_{10} samples collected under the westerlies influence.

S3 Meteorological scenario

Monthly values of key meteorological fields – geopotential heights, winds and omega (at 925, 850 and 700 hPa standard levels) and precipitations rates – were determined with the National Centre for Environmental Prediction/National Centre for Atmospheric Research (NCEP/NCAR) reanalysis data. Figure S3 shows the averaged (2008–2013) geopotential height at 850 hPa for the most representative months (January, May, August and November) in order to illustrate the latitudinal shift in the North Atlantic anticyclone.

S4 North America storm season

The records of tornado climatology provided by the National Weather Service (National Oceanic and Atmospheric Administration, NOAA) show that North America is affected by a "tornado season", which usually occurs from spring to early summer (March to July). The regional frequency of the tornados is related to the progression of the warm season, moving north from spring to summer (http://www.ncdc.noaa.gov/climate-information/extreme-events/us-tornado-climatology) (Fig. S4A). In early spring, tornados affects to Gulf States – such as Mississippi and Louisiana – and then moves northward affecting Kansas, Nebraska, and the Tennessee Valley region during late spring. Into summer, most of "Tornado Alley" – south central United States – is active, and then shifts back southward into the late autumn. Tornados, the most violent of atmospheric convective storms, significantly influence the vertical distribution of aerosols by transporting them from the boundary layer to the troposphere (Barth et al., 2015).

Ginoux et al. (2012) identified the Great Plains – located between the east of the Rocky Mountains and the Mississippi River – as the largest dust source in United States with a main anthropogenic origin. The maximum monthly average of tornadoes occurrence, from Southern (Texas, Oklahoma, New Mexico) to Central (Kansas, Colorado) plains, is from April to June (Fig. S4B) – during the study period (2008–2013) – according with the maximum recorded high dust concentrations at Izaña.

S5 Percentiles of the inorganic compounds concentration

To explain the seasonal evolution of the background (30^{th}P), median (50^{th}P) and high (80^{th}P) concentration events, monthly distribution for the aerosol components concentrations are shown in Fig. S5. The aerosol background (30^{th}P) composition changes significantly thought the year, although nss-$SO_4^{=}$, NH_4^{+} and Al are always part of the total bulk. The remaining chemical species exhibit certain seasonality. Two seasons – associated to the latitudinal ranges of the North American outflow and the spatial distribution of the source regions – can be well differentiated: (i) Jan-May (~35–40ºN) and Jun-Dec (~40–55ºN). The first season is characterized by an increase of the concentrations of most chemical species (nss-$SO_4^{=}$, NH_4^{+}, OM, Ca, Al and dust), whereas the second season is mainly associated to an increase in the concentration of EC.

Supplement references

Barth, M. C., Cantrell, C. A., Brune, W. H., Rutledge, S. A., Crawford, J. H., Huntrieser, H., Carey, L. D., acGorman, D., Weisman, M., Pickering, K. E., Bruning, E., Anderson, B., Apel, E., Biggerstaff, M., Campos, T., CampuzanoJost, P., Cohen, R., Crounse, J., Day, D. A., Diskin, G., Flocke, F., Fried, A., Garland, C., Heikes, B., Honomichi, S., Hornbrook, R., Huey, L. G., Jimenez, J., Lang, T., Lichtenstern, M., Mikoviny, T., Nault, B., O'Sullivan, D., Pan, L., Peischl, J., Pollack, I., Richter, D., Riemer, D., Ryerson, T., Schlager, H., St. Clair, J., Walega, J., Weibring, P., Weinheimer, A., Wennberg, P., Wisthaler, A., Wooldridge, P., and Zeigler, C.: The Deep Convective clouds and Chemistry (DC3) Field Campaign, B. Am. Meteorol. Soc., 96, 1281–1309, doi:10.1175/BAMS-D-13-00290.1, 2015.

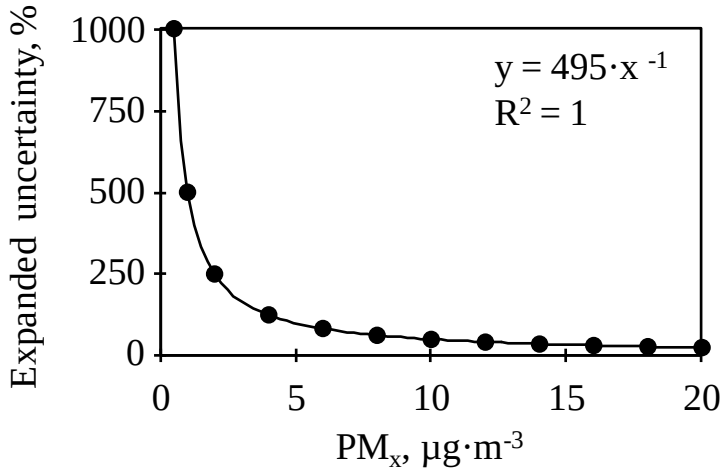

Figure S1. Expanded uncertainty (U) of the gravimetric method for individual PM_x samples as described in EN-14907.

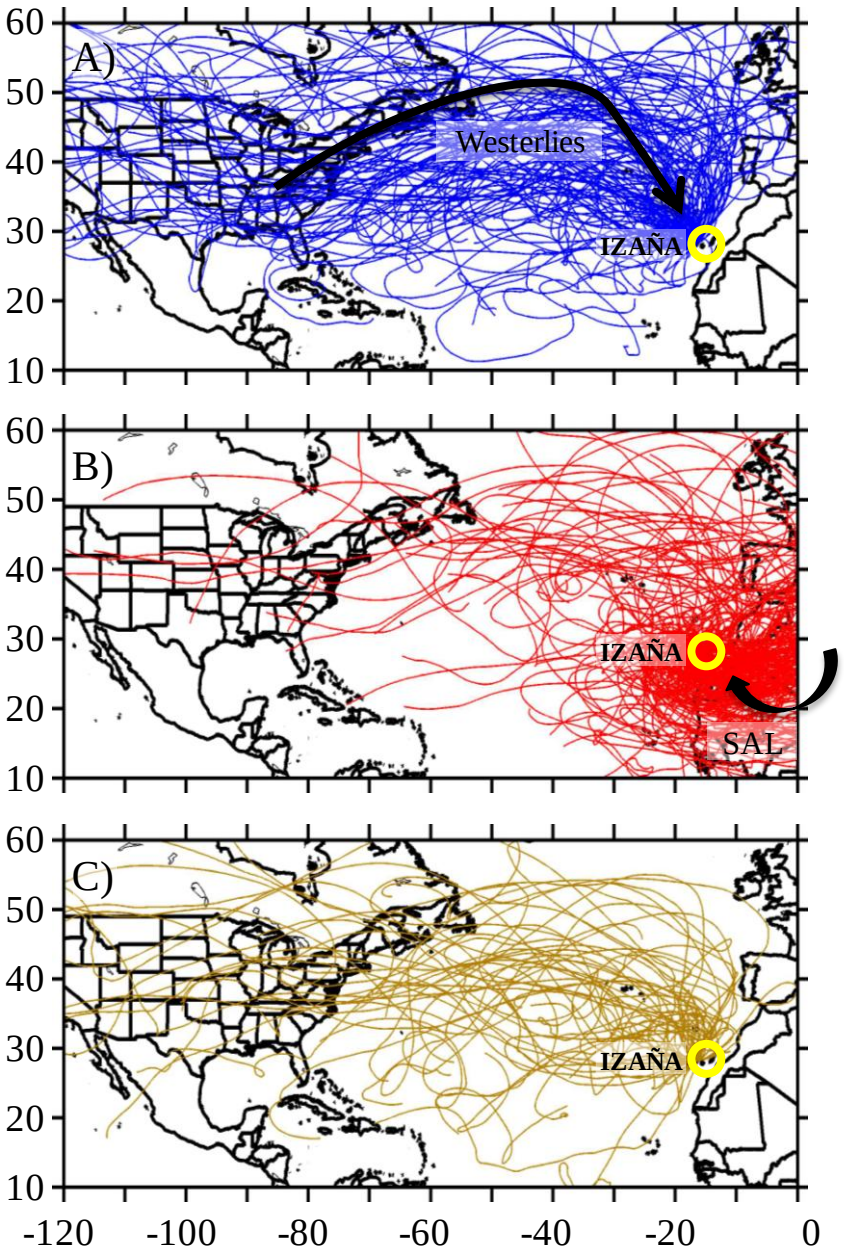

Figure S2. 10 days back-trajectories from 2008 to 2013 of the chemically analyzed samples collected under (**A**) the westerlies, (**B**) the Saharan Air Layer and (**C**) Saharan dust re-circulated over the North Atlantic conditions.

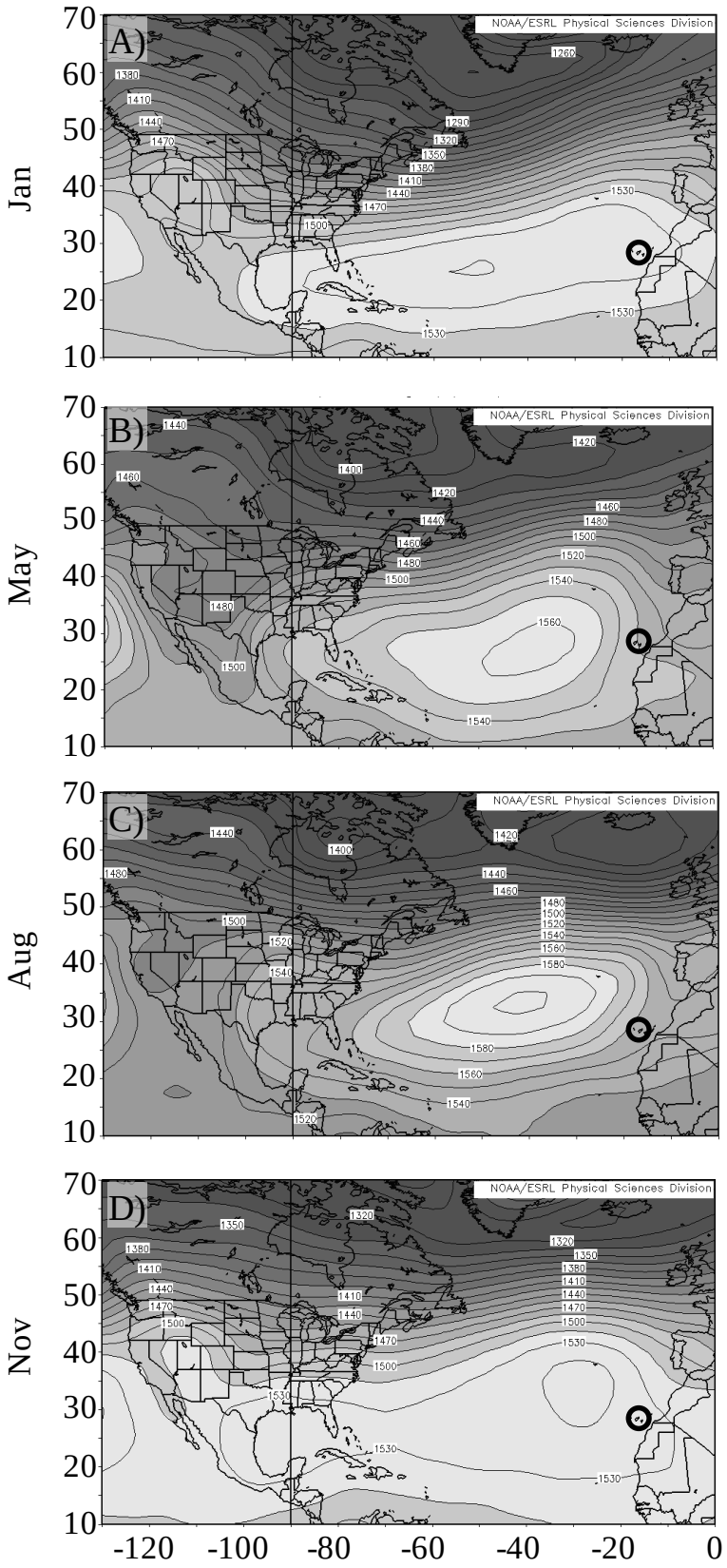

Figure S3. Geopotential Height at 850 mb averaged from 2008 to 2013 for (**A**) January, (**B**) May, (**C**) August and (**D**) November. The location of Izaña is highlighted (black circle) [R1#C5].

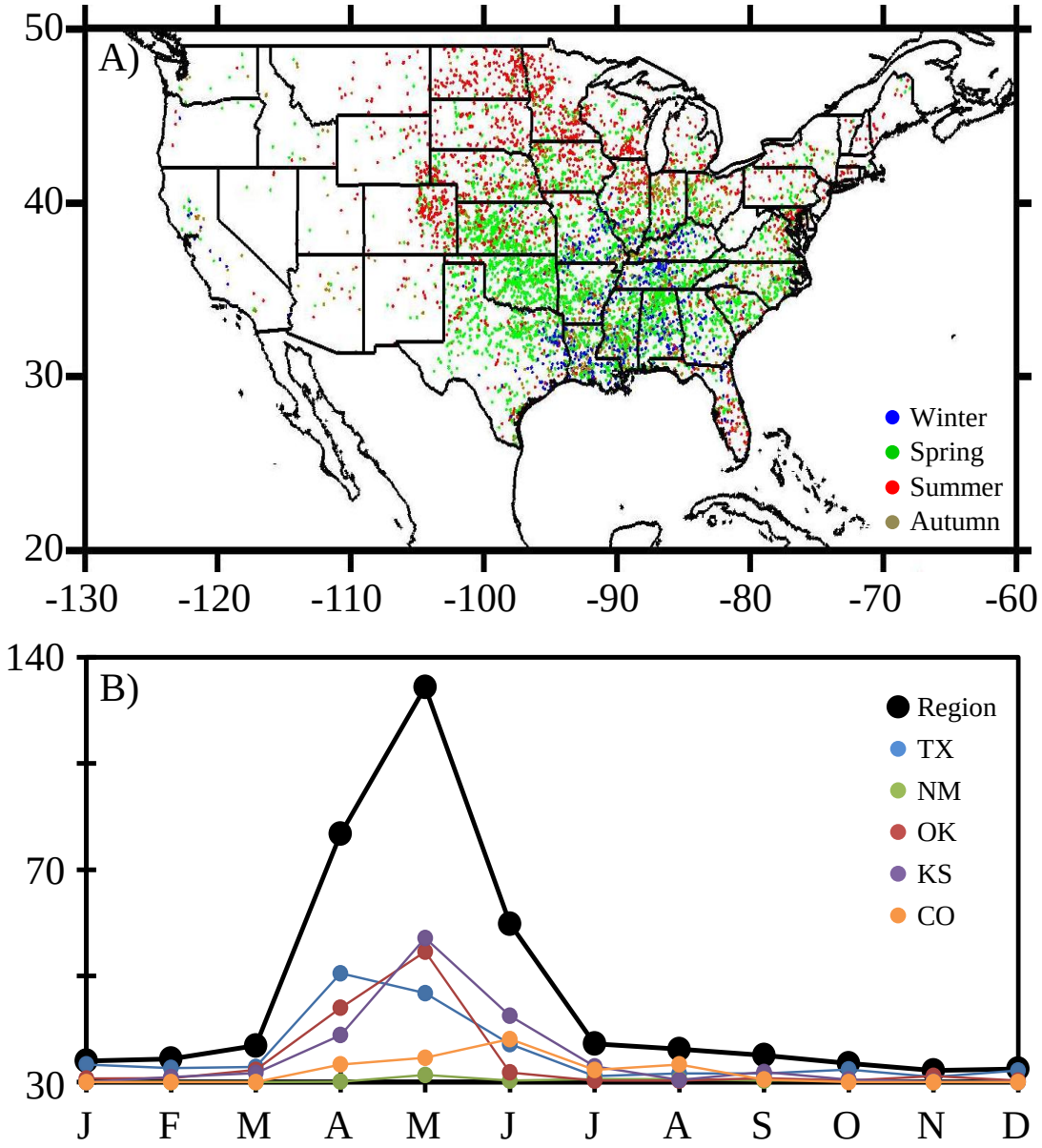

Figure S4. (A) Tornadoes which took place in U.S.A during the study period (2008–2013). The starting point is represented by colours depending on the season of occurrence. **(B)** Monthly average number of tornadoes during the study period (2008–2013) in the selected States and the whole Region (TX, NM, OK, KS and CO). **TX**: Texas; **OK**: Oklahoma; **NM**: New Mexico; **KS**: Kansas; **CO**: Colorado. Data source: http://www.spc.noaa.gov/wcm/index.html#data.

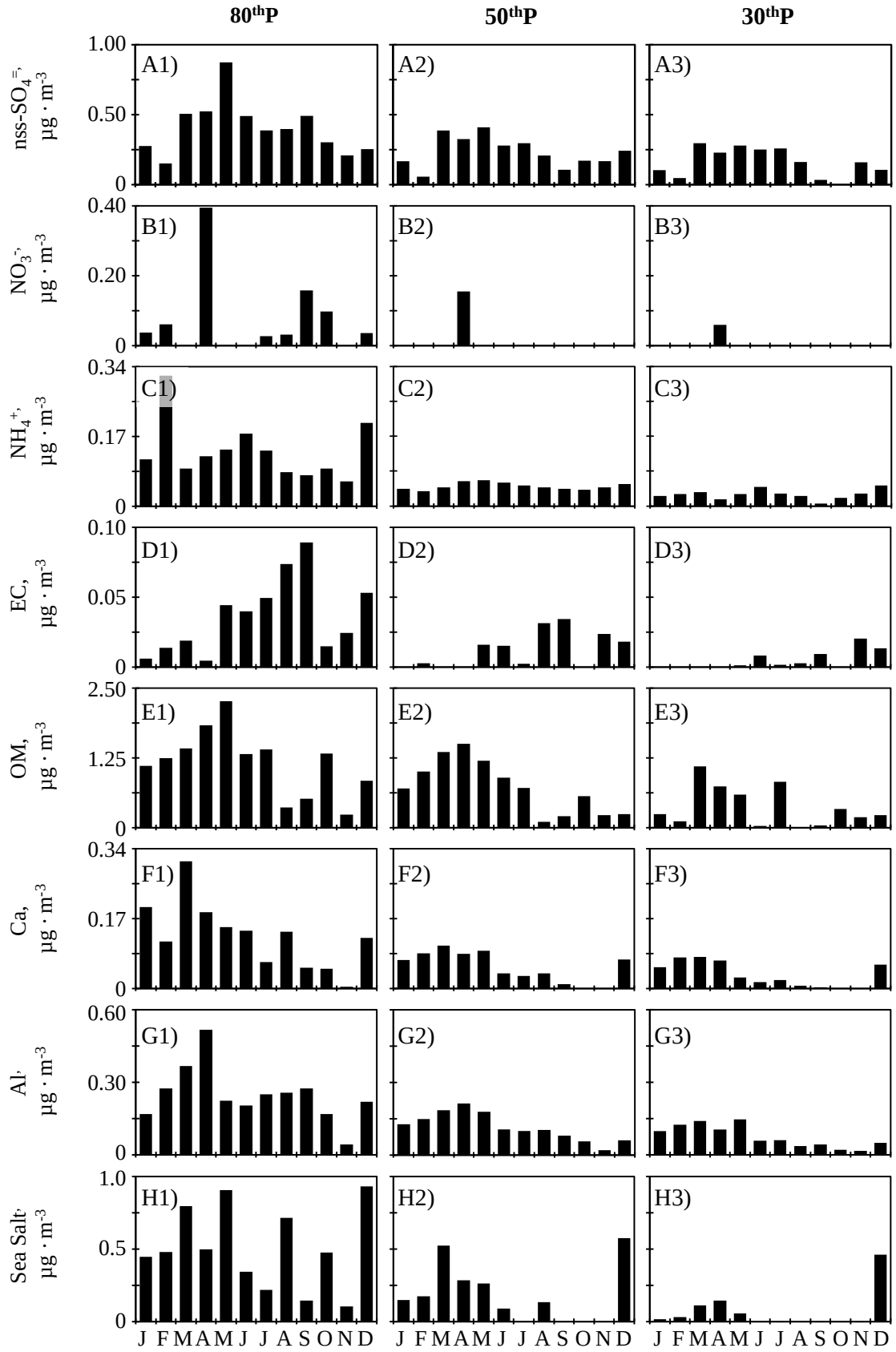

Figure S5. Monthly distribution of the percentiles 80, 50 and 30 for the concentrations of (**A**) nss-$SO_4^{=}$, (**B**) NO_3^-, (**C**) NH_4^+, (**D**) EC, (**E**) OM, (**F**) Ca, (**G**) Al and (**H**) sea salt, from Jan 2008 to Aug 2013.