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

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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: 8 h; 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 $\mu g \cdot m^{-3}$.

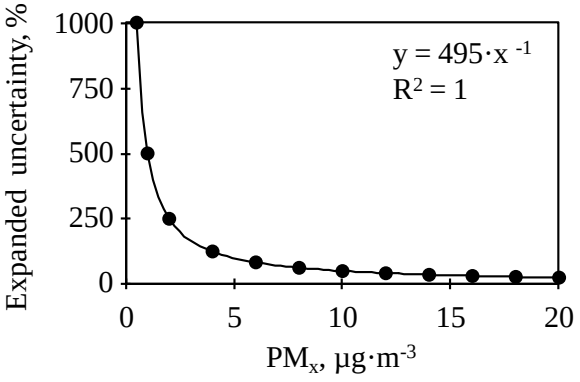

Fig. S1: Expanded uncertainty (U) of the gravimetric method for individual PM_x samples as described in EN-14907.

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.

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.

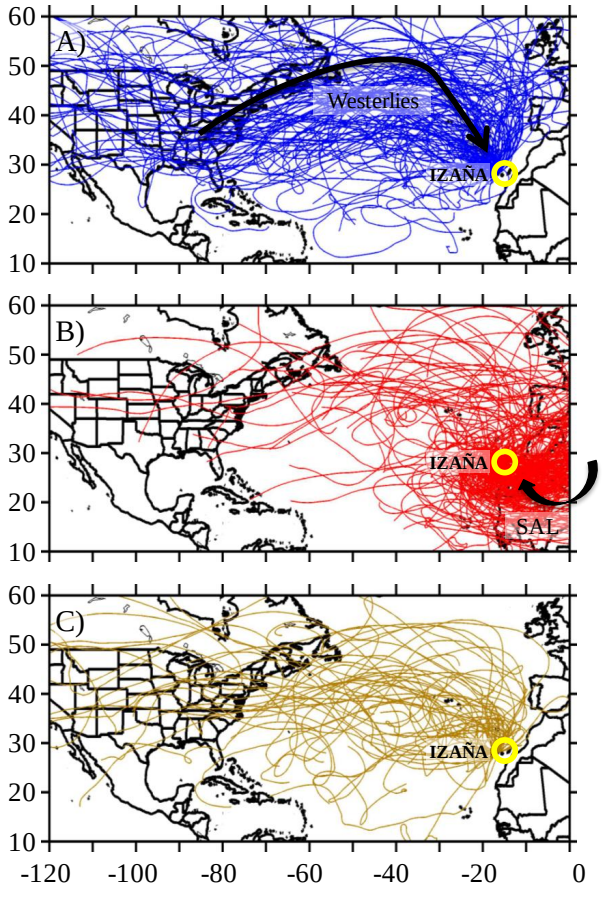

Fig. S2: 10 days back-trajectories from 2008 to 2013 of the chemically analyzed samples collected under (**A**) the westerlies, (**B**) the Saharan Air Layer and (**C**) Saharan dust re-circulated over the North Atlantic conditions.

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.

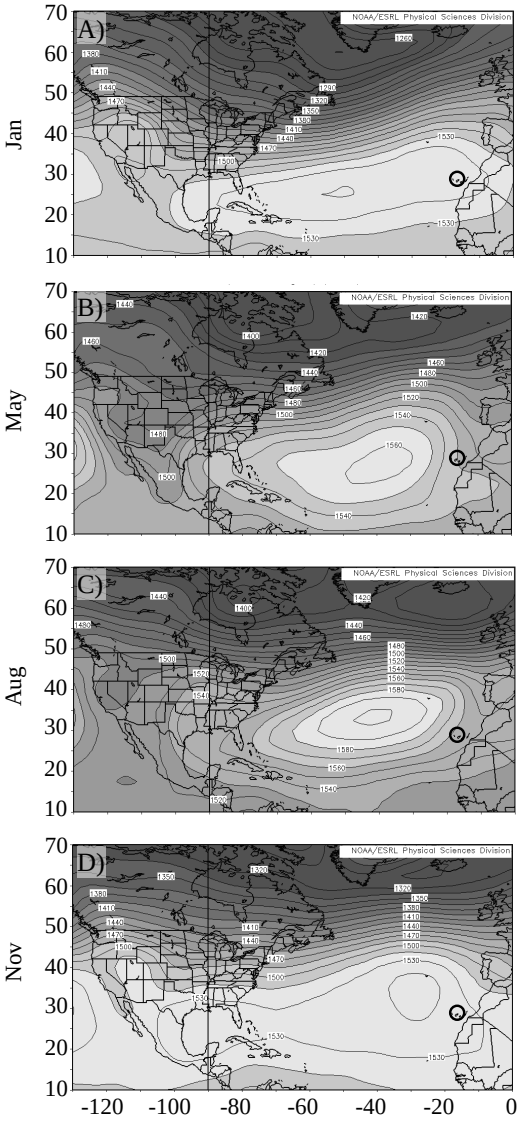

Fig. S3: Geopotential Height at 850 mb averaged from 2008 to 2013 for (**A**) January, (**B**) May, (**C**) August and (**D**) November. The location of Izaña is highlighted (black circle).

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.

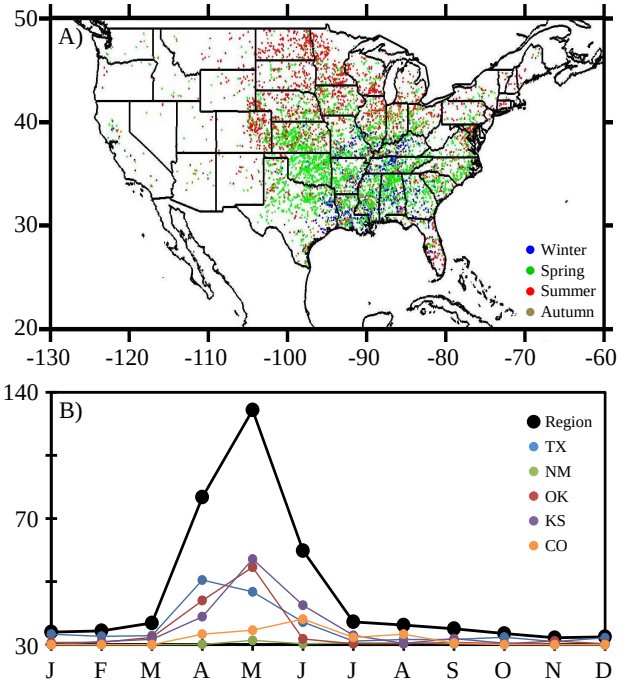

Fig. S4: (**A**) Tornadoes which took place in U.S.A during the study period (2008–2013). The starting point is represented by colours depending on the season of occurrence. (**B**) Monthly average number of tornadoes during the study period (2008–2013) in the selected States and the whole Region (TX, 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.htmldata.

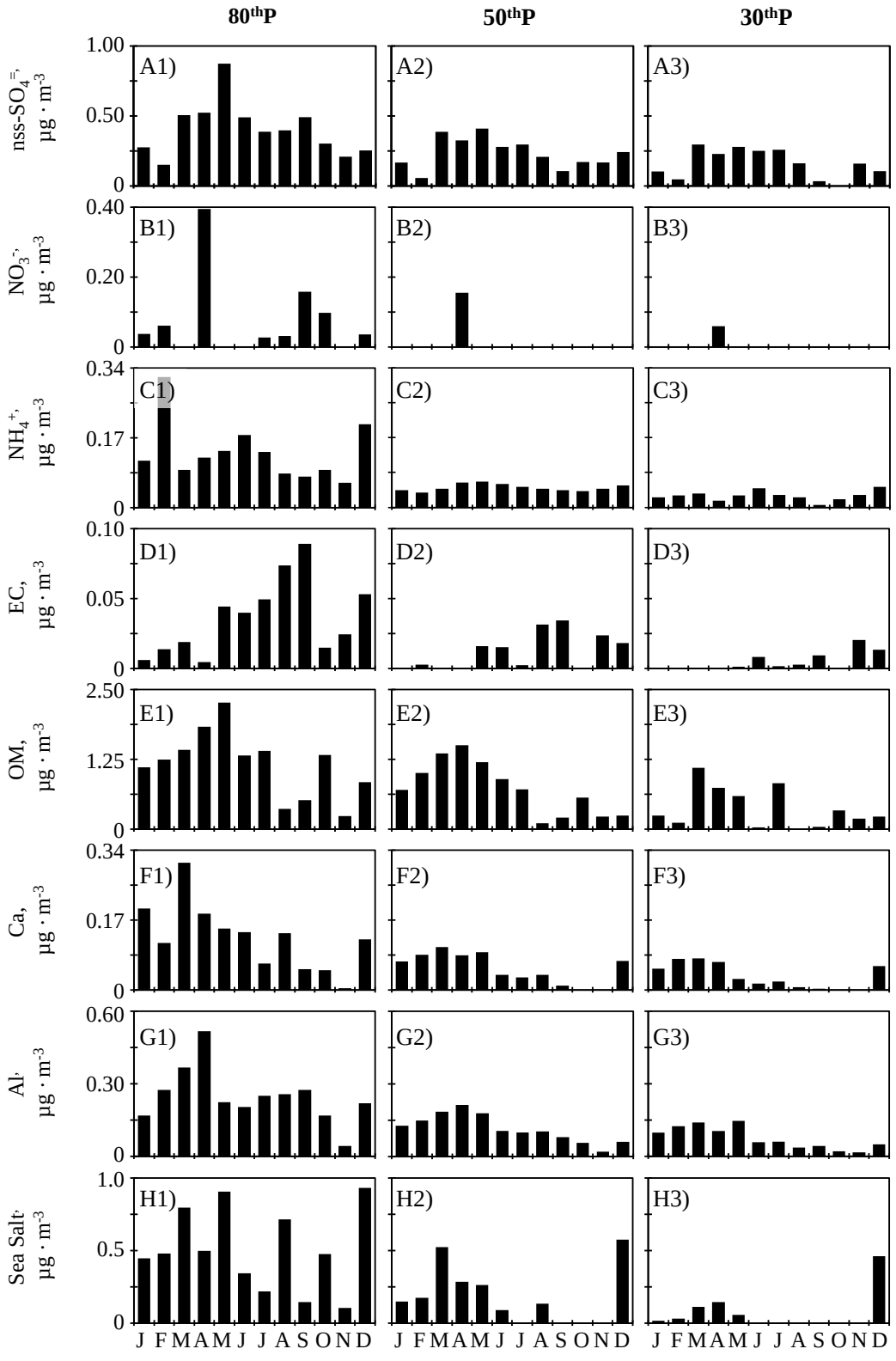

Fig. S5: Monthly distribution of the percentiles 80, 50 and 30 for the concentrations of (**A**) nss-$SO_4^=$, (**B**) NO_3^-, (**C**) NH_4^+, (**D**) EC, (**E**) OM, (**F**) Ca, (**G**) Al and (**H**) sea salt, from Jan 2008 to Aug 2013.

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 ($\sim$35–40°N) and Jun-Dec ($\sim$ 40–55°N). 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.

Supporting References

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