Título: ORIGIN OF ATMOSPHERIC AEROSOLS TRANSPORTED ACROSS THE NORTH ATLANTIC FREE TROPOSPHERE

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Localización: ORIGIN OF ATMOSPHERIC AEROSOLS TRANSPORTED ACROSS THE NORTH ATLANTIC FREE TROPOSPHERE

Resumen: This study deals on the origin of atmospheric aerosols transported across the North Atlantic, paying special attention on identifying the impact of human activities. It is based on records of aerosol and other atmospheric components collected at Izaña Global Atmospheric Watch Observatory in Tenerife Island (2367 m a.s.l.).

The aerosol composition has been studied in the two main airflows of the North Atlantic free troposphere: the westerlies ¿ prevailing airstreams that flows from North America eastward across the North Atlantic at subtropical and mid-latitudes ¿ and the Saharan Air Layer ¿ the warm, dry and dusty airstream that expands

from North Africa to the Americas at tropical and subtropical latitudes ¿ . The results of this study provide new insights on the processes that contribute to the variability of the aerosols composition in the North Atlantic free troposphere.

The seasonal evolution observed in the composition of the aerosols carried by the westerlies is influenced by (i) the spatial distribution of their sources in North America, and (ii) the seasonal shift of the westerly jet and the associated eastward moving cyclones. The westerlies carry maximum loads of (i) mineral dust from February to May, associated with the occurrence of the westerly jet at rather low latitudes (35º¿40º N), dust emissions in a region that extends from Southwest Texas to the High Plains, and subsequent dust export to the Atlantic, (ii) none-sea-salt-sulphate and ammonium from March to May, linked to the presence of the westerly jet and to the export of polluted air from Northeastern United States, where the highest sulphur dioxide emissions in North America occur, (iii) organic matter from February to May, associated with the westerly jet and export from regions of Eastern United States rich in organic aerosols, (iv) elemental carbon in August and September, associated with the occurrence of the westerly jet at rather high latitudes (50º ¿55º N) and the consequent export from the regions (Chicago to New York) where the highest concentrations of elemental carbon occur in North America.

Results evidence how dust is a major component of the aerosols transported from North America across the North Atlantic by the westerly winds. The concentrations of sub-10 µm aerosols that reach the Izaña Observatory, after transatlantic transport from North America, typically ranges between 1.2 and 4.2 µg·m¿3 (20th and 80th percentiles). The main contributors to background levels (1st¿50th percentiles = 0.15¿2.54 µg·m¿3) are North American dust (53 %), non-sea-salt-sulphate (14 %) and organic matter (18 %), whereas aerosol composi- tion during high PM10 events (75th¿95th percentiles = 3.9¿8.9 µg·m¿3) is dominated by North American dust (56 %), organic matter (24 %) and nss-sulphate (9 %).

About the 64 % of the organic aerosol transported in the summer westerlies has been chemically identify. The most abundant organic compounds are (i) secondary organic aerosols from isoprene oxidation and (ii) dicarboxylic acids (mainly succinic and phthalic, indicating aged aerosols after the long-range atmospheric transport). Compounds linked to soil emissions (i.e. saccharides) are found but in minor amounts. High concentrations of organic matter and of some organic species (e.g. levoglucosan, succinic and malic acids) are linked to North American fires; air mass impacted by fires has the highest contribution of aged organic aerosols (di-acids formed during the long-range atmospheric transport) and the lowest contributions of secondary organic aerosols from oxidation of isoprene and ¿-pinene (further oxidized under the biomass burning air). This long-range atmospheric transport of biomass burning plumes from North America supports the idea that, under certain conditions, levoglucosan is stable in the atmosphere to experience long range transport. In the westerlies, the organic matter sources are associated with soils, biomass burning and combustion.

This research also provides novel results on the composition and sources of organic aerosols exported over the Atlantic Ocean in the summer Saharan Air Layer. The speciation of the organic matter shows how most of this aerosol has a natural origin. Organic species are associated with (i) primary compounds of surface soils (i.e. saccharides) and terrestrial higher plants (i.e. nC27, nC29 and nC31 n-alkanes), (ii) secondary organic aerosols linked to the oxidation of biogenic isoprene and ¿-pinene, (iii) primary vehicles emissions (i.e. hopanes), and (iv) bio-accumulative and toxic organic compounds (i.e. polycyclic aromatic hydrocarbons). Saccharides from soils

and secondary organic compounds derived from the oxidation of isoprene and ¿-pinene accounts for more than a 70 % of the organic matter. In the Saharan Air Layer, the sources of organic aerosols have been associated mostly with soils and combustion. Of special relevance is the enhancement in the formation of biogenic secondary organic aerosols due to interaction with anthropogenic NOx emissions ¿ as suggested by the correlation between nitrate and secondary organic aerosols from isoprene oxidation ¿, which may exert a large-scale impact as a result of the synoptic scale of the Saharan Air Layer.

This study also includes a research on new particle formation, which has been found to be a frequent source of aerosols above Tenerife. The climatology of the new particle formation events at Izaña shows that these episodes occur a 30 % of the days, with a clearly marked new particle formation season (May¿August). Monthly mean values of the formation and growth rates in this season exhibited values within the ranges of 0.49¿0.92 cm3·s¿1 and 0.48¿0.58 nm·h¿1, respectively. New particle formation is observed during periods of upslope winds that bring gaseous precursors from the boundary layer to the interface with the low free troposphere. Sulphuric acid played a key role as gas precursor, contributing with a 70 % to the observed growth rate. Organic species, such as oxidation products of biogenic volatile organic compounds, probably also contribute. It was found that year-to-year variability of the frequency of new particle formation events is correlated with mean sulphur dioxide concentrations. This study also has found an interaction between dust and the freshly formed new particles in the Saharan Air Layer; dust may play a significant role acting as coagulation sink of freshly formed nucleation particles.

The results of this study provide a comprehensive view of the sources and atmospheric processes that influence on the composition of the aerosol transported across the North Atlantic free troposphere. The conceptual models presented will be useful in further studies on transboundary air pollution, long-term evolution of aerosols, their effects physical properties and their influence on processes related to climate.