Long-term aerosol measurements in Gran Canaria, Canary Islands: Particle concentration, sources and elemental composition

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[1] There are very few sets of long-term measurements of aerosol concentrations over the North Atlantic Ocean, yet such data is invaluable in quantifying atmospheric dust inputs to this ocean region. We present an 8-year record of total suspended particles (TSP) collected at three stations on Gran Canaria Island, Spain (Taliarte at sea level, Tafira 269 m above sea level (a.s.l.) and Pico de la Gorra 1930 m a.s.l.). Using wet and dry deposition measurements, the mean dust flux was calculated at 42.3 mg m^{-2} d^{-1}. Air mass back trajectories (HYSPLIT, NOAA) suggested that the Sahara desert is the major source of African dust (dominant during 32–50% of days), while the Sahel desert was the major source only 2–10% of the time (maximum in summer). Elemental composition ratios of African samples indicate that, despite the homogeneity of the dust in collected samples, some signatures of the bedrocks can still be detected. Differences were found for the Sahel, Central Sahara and North of Sahara regions in Ti/Al, Mg/Al and Ca/Al ratios, respectively. Elements often associated with pollution (Pb, Cd, Ni, Zn) appeared to share a common origin, while Cu may have a predominantly local source, as suggested by a decrease in the enrichment factor (EF) of Cu during dust events. The inter-annual variability of dust concentrations is investigated in this work. During winter, African dust concentration measurements at the Pico de la Gorra station were found to correlate with the North Atlantic Oscillation (NAO) index.

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1. Introduction

[2] Mineral aerosols are the focus of increasing scientific attention due to their direct effect on the radiative budget and their important role in biogeochemical cycles [*Arimoto*, 2001; *Jickells et al.*, 2005; *Satheesh and Krishna Moorthy*, 2005]. Arid regions in the Sahara and Sahel provide large quantities of soil dust, resulting in a westward flow of material over the North Atlantic Ocean accounting for some 30–50% of global dust output [*Schütz et al.*, 1981; *d'Almeida*, 1986]. Atmospheric dust deposition is an important source of limiting nutrients and metals to the ocean [*Measures et al.*, 1986; *Duce et al.*, 1991; *Baker et al.*, 2003; *Mills et al.*, 2004; *Jickells et al.*, 2005] and recent reports suggest that dust inputs may promote nitrogen fixation [*Mills et al.*, 2004; *Duarte et al.*, 2006; *Moore et al.*, 2009]. Atmospheric iron inputs may play an important role in stimulating the growth of microorganisms. For example, an unusual bloom of the marine diazotrophic cyanobacterium, Trichodesmium erythraeum, was reported in the Northwest African upwelling during a period of exceptionally warm weather and following a massive African dust outbreak [*Ramos et al.*, 2005]. In addition, intense African dust intrusion episodes have important environmental impacts in the Canary Islands [*Criado and Dorta*, 2003; *Griffin and Kellogg*, 2004] with repercussions for the local economy, particularly affecting agriculture, where atmospheric dust causes temperature increases and coats the leaves of crops [e.g., *Criado and Dorta*, 2003], and human health, since the inhalation of dust can trigger or aggravate respiratory conditions [e.g., *Julià-Serdà et al.*, 2005].

[3] Situated close to the African continent and within the "dust belt" [*Prospero and Lamb*, 2003] the Canary Islands are affected by two large sources of Aeolian soil dust: the desert regions of the Sahara and the Sahel. They are therefore an ideal location to study the impact of African dust episodes in the subtropical North Atlantic Ocean [*Gelado-Caballero et al.*, 1996; *Torres-Padrón et al.*, 2002; *Neuer et al.*, 2004]. The transport mechanisms of African dust are well-understood and there are large seasonal and spatial variations in mineral dust concentration over the Atlantic Ocean [*Prospero*, 1999; *Goudie and Middleton*, 2001]. Dust plumes arriving to the islands are generally up to 4 km in height, but the top of the so-called Saharan Air Layer (SAL)

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can reach as high as 6 km. Dust transport over the islands is greatly affected by the seasonal latitudinal shift of the Intertropical Convergence Zone (ITCZ). During the boreal summer, the ITCZ shifts northward over the northwestern African continent (from approximately 5°N in winter to 19°N in summer), creating a thermal low over the surface of the African continent. This causes the North African high-pressure system to be located at higher altitude and consequently dust transport occurs high in the atmosphere. Dust emerging from the African coast follows the path of easterly waves, arriving to the archipelago from the S-SW [*Sancho et al.*, 1992]. In winter, the ITCZ is located in its most southerly position and dust transport is controlled by high pressure systems located over the northern African continent. Dust arrives to the archipelago from the east at relatively low altitudes and is injected mainly within the marine boundary layer (MBL) [*Díaz et al.*, 2006].

[4] The Marine Boundary Layer (MBL) is limited by the base of the thermal inversion layer at altitudes that range between 500 m above sea level (a.s.l.) near the Azores High and 2000 m a.s.l. in the trade wind region close to the tropics. In subtropical regions, the upper limit of the marine boundary layer is normally bounded by the trade wind inversion layer (TWI). In the Canary Islands, the location of the TWI follows a seasonal pattern, occurring in summer between 770 and 1380 m a.s.l. with an average thickness of about 560 m, while in winter the layer is found at higher altitude (1360 to 1850 m a.s.l.) and with a reduced average thickness of about 360 m [*Torres et al.*, 2001]. During summer, when African dust is generally transported at higher altitude, the TWI is thought to play an important role in limiting the amount of dust reaching the marine boundary layer [*Alonso-Pérez et al.*, 2007].

[5] It has been suggested that the year-to-year variations in Saharan dust transport are related to large-scale meteorological phenomena. Over the Northern Hemisphere, the North Atlantic Oscillation (NAO) exerts a major influence on weather and climate variability [*Hurrell and Deser*, 2009]. It is likely that the NAO directly controls the winter dust export through changes in strength and location of the Azores anticyclone, which is known to be an important agent for the transport of dust from Africa in the trade winds layer [*Chiapello et al.*, 1995]. High positive NAO Index corresponds to drier weather in North Africa and therefore an increase in dust mobilization, while a negative Index value corresponds to increased rainfall and a corresponding reduction in dust mobilization. Some studies have shown the influence of the NAO on the year-to-year variability of dust export from Africa, as observed using TOMS/Nimbus-7 and Meteosat/VIS satellite measurements [*Moulin et al.*, 1997; *Chiapello and Moulin*, 2002]. *Moulin et al.* [1997] suggested that the inter-annual variations in dust transport over the Atlantic Ocean and Mediterranean Sea are well correlated with the climatic variability defined by the NAO. They found that the annual mean dust concentrations at Barbados generally follow variations in the NAO index. However, the 36-year record of dust concentrations at Barbados indicates that there is no correlation between surface dust concentrations in winter and the NAO index [*Ginoux et al.*, 2004].

[6] Most work on the characterization of mineral aerosols tries to relate the chemical composition and mineralogy of the particles to potential source regions in the Sahara and Sahel [*Chiapello et al.*, 1997; *Moreno et al.*, 2006]. A correlation has been observed between aerosol calcium content and the calcite content of source region soils [*Desboeufs and Cautenet*, 2005; *Kandler et al.*, 2007]. Mineralogical and chemical analyses of aerosol samples collected in the Canary Region indicate that particles >1 μm diameter are dominated by mineral dust (aluminosilicate particles), while sulphates and carbonaceous material become more important at particle sizes below 1 μm diameter [*Hoornaert et al.*, 2003; *Kandler et al.*, 2007]. *Viana et al.* [2002] studied samples of aerosols collected on different islands in the Canary Islands and found that during winter African dust outbreaks there is a very significant increase in the concentrations of Si, Al, K, Ti, Ca, Fe, Mn, Mg and Ba and concluded that such species can be used as excellent tracers of African origin.

[7] In the Canary Islands a number of studies have focused on the inter-annual and seasonal variability of atmospheric dust concentrations [*Torres-Padrón et al.*, 2002; *Viana et al.*, 2002; *Alonso-Pérez et al.*, 2007]. African dust events are especially intense and more frequent in winter and early spring, with maximum PM10 and TSP concentrations of up 5238 μg m^{-3} (4th of March 2004) and 5000 μg m^{-3} (6th of January 2002) being recorded, respectively [*Gelado-Caballero et al.*, 2005; *Hernández et al.*, 2005]. However, with the exception of a few short-term studies [e.g., *Viana et al.*, 2002; *Alastuey et al.*, 2005; *Kandler et al.*, 2007] no studies in the region to date have included measurements of elemental composition.

[8] This current study represents the first reported long-term measurements of bulk aerosol and atmospheric trace metal concentrations in the Canary Islands. A large data set of particulate atmospheric trace metal measurements (Al, Fe, Mn, Ti, Co, Ca, Mg, Na, Pb, Cd, Cu and Zn) has been produced from samples collected in Gran Canaria over an 8-year period in order to determine the relative contribution of different African source regions to the mineral aerosols collected. Furthermore, the use of different sampling sites on the island enables differences in dust concentration composition and concentration with altitude to be investigated. A multivariate statistical method, principal component analysis (PCA), has been applied to gain insight into the common factors influencing the elemental concentrations, while differences in aerosol characteristics in relation to sampled air masses have been interpreted using a combination of elemental data and air mass back-trajectories. Finally, the effect of the NAO on atmospheric dust concentrations has been investigated.

2. Methodology

2.1. Sampling Sites

[9] Three sampling sites were used in this study in order to investigate the variation of aerosol concentration and chemical composition with altitude (Figure 1). Pico de la Gorra (PG) (1930 m a.s.l.; 27° 56′ N, 15° 33′ W) is a rural station free of direct influence from local sources of pollution, Tafira (TF) (269 m a.s.l.; 28° 06′ N, 15° 24′ W) and Taliarte (TL) (close to sea level; 27°59.5′ N 15°22′ W) are urban background sites subject to anthropogenic influence with the latter situated on the coast with a strong and direct influence from marine aerosols. Both TL and TF stations are situated within the marine boundary layer (<1800 m a.s.l).

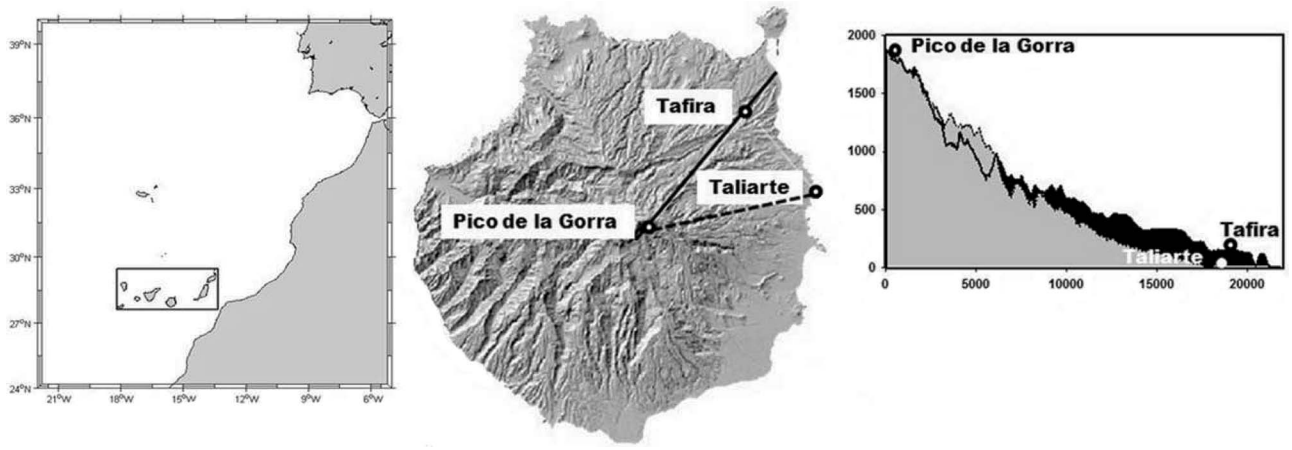

Figure 1. Location of the three sampling sites in the Canary Islands: Pico de la Gorra (PG) (1930 m a.s.l., 27°56′N, 15°33′W), Tafira (TF) (269 m a.s.l, 28°06′N, 15°24′W) and Taliarte (TL) (close to sea level; 27°59.5′N 15°22′W).

[10] At PG, the aerosol samplers are positioned on top of a tank 2 m above ground level; at TF, on the roof (15 m above ground) of the Chemistry Department of the University of Las Palmas; and at TL, on the roof (10 m above ground) of the Instituto Canario de Ciencias Marinas.

2.2. Aerosol Sampling

[11] High volume (68 m^3 h^{-1}) aerosol collectors (MCV, model CAV-A/M) were used to collect aerosol samples for metal analysis on acid-washed cellulose filters (Whatman 41) and for TSP measurement on glass fiber filters (Whatman GF/A). PM10 measurements were made using, a PM1025-CAV (MCV) head (30 m^3 h^{-1} flow rate) and glass fiber filters (Whatman GF/A) were used. TSP and PM10 concentrations were measured by weighing the filters before and after sampling. The Whatman 41 filters were washed with 10% HCl and 1% HNO_3 in order to eliminate metal traces. One eighth of each filter was used for the trace metal analysis. The collection efficiency of Whatman 41 is effectively 100% for dust [*Savoie*, 1984].

[12] To minimize the risk of sample contamination, exposed filters were transferred to individual zip-lock plastic bags immediately after collection and subsequently handled under a laminar flow clean bench. Samples for trace metal (TM) analysis were stored frozen (−20°C) until analysis.

[13] A TSP concentration time series has been recorded from 1 December 1996 to 31 December 1998 [see *Torres-Padrón et al.*, 2002] and 1 December 2001 to 31 December 2009 at the PG sampling station and from 1 December 2003 to 31 December 2009 at TF. Samples for trace metal analysis were collected at PG (one sample per week) between June 2006 and November 2009 and at TL (daily during African dust episodes) between April 2004 and December 2009 (there was no sampling between October 2005 and May 2006 due to construction work at the site). Simultaneous PM10 and TSP samples were collected at TF station from 5 August 2004 to 31 December 2009.

[14] Due to the remote location of the station at PG, filters were changed once per week. Seven samplers were installed at PG, of which 6 were loaded with glass-fiber filters and used to obtain a continuous data set of daily measurements of TSP over each 7-day period (with 5 samplers programmed to collect on individual days, and the sixth collecting over a 2-day period). The remaining sampler was loaded with cellulose filters and used to collect a sample for trace metal analysis on one day per week. Since the elemental composition of particles of African mineral dust was of particular interest, the sampling day was selected based on the output of dust prediction models (such as DREAM, see section 2.5) in an attempt to sample air masses of African origin where possible.

[15] At TF, samples were collected daily for TSP with no sampling for trace metal analysis. At the third station, TL, no routine samples were collected, but cellulose filters for trace metal determinations were deployed daily during dust events. Procedural blanks were also collected at the three sites during non-dust days.

[16] To avoid pollution from local sources (mainly traffic emissions), sampling was conducted during the night (from 6:00 P.M. to 6:00 A.M. UTC). More than 5000 samples were collected over an 8-year period, along with 446 filters for metal determinations. Wind direction and velocity, temperature, humidity and pressure were continually monitored (Vantage Pro2 Plus) during sample collection in order to aid the identification of African air masses.

2.3. Particle Deposition Measurement

[17] For dry and wet deposition flux measurement, an automatic wet and dry sampler (ARS 1000, MTX Italy) with cubic containers having a surface area of 660 cm^2 and equipped with a rain sensor was used. The dry atmospheric deposition was collected throughout the period from December 2003 to December 2008 at Tafira, in 61 samples representing time periods ranging from 5 to 50 days. Wet depositions (96 samples) were collected after each rainfall over the same period.

[18] Water surfaces have been proposed to improve the measurements of dry deposition fluxes of atmospheric gases and particles [*Yi et al.*, 1997]. Therefore, a water deposition sampler was tested simultaneously over a two-year period in order to compare with the dry deposition measurements of the surrogate surfaces (data not shown). This equipment was

Table 1. Procedural Blanks Values and Detection Limits for Aerosol Sampling Substrates[a]

	Blanks	Instrumental Detection Limits
Al	208	83
Fe	20	16
Mn	<2.8	2.8
Co	<0.056	0.056
Ti	0.86	0.12
Mg	10	9.5
Na	64	11
Ca	327	86
Cu	<0.25	0.25
Ni	<0.37	0.37
Cr	<5.8	5.8
Cd	0.02	0.0021
Pb	<0.38	0.38
Zn	1.5	0.031

[a]Values in ng m^{-3} and calculated using a typical air volume of 719.8 m^3.

made by modifying an ARS 1000 instrument according to *Morselli et al.* [2008]. We found that the water deposition sampler underestimated dry deposition values for long collection periods (7–21 days), due to difficulties with filtration arising from possible algal growth and an accumulation of aerosol particles in the sampler.

[19] The aerosols collected were quantified by gravimetry after filtering. For dry flux measurements, ultrapure water was used to rinse the bucket and filtered through a 0.45 μm pore size Nucleopore filter (Whatman). Rain samples were filtered through a 0.2 μm pore size Nucleopore filter (Whatman). The filtered rainwater was placed in acid clean bottles and frozen (−20°C) until analysis.

[20] Membrane filters were dried and weighed to an accuracy of ±0.01 mg (Sartorius CP225D). They were washed with 0.1 M hydrochloric acid (Suprapure, Merck) prior to use.

2.4. Chemical Analysis

[21] Chemical characterization of metal composition was carried out using Whatman 41 filters collected at Taliarte and Pico de la Gorra. Analyses of Al, Fe, Mn, Ti, Co, Ca, Mg, Na, Pb, Cd, Cu and Zn in TSP were carried out after total acid digestion with Suprapur-grade (or equivalent) acids: HF (37%, Fluka), HNO_3 (65%,PANREAC) and $HClO_4$ (70%, PANREAC). Acid was added to the sample in the volume ratio 7:5:4 ($HF:HNO_3:HClO_4$), the vessel was sealed and the sample was heated at 120°C overnight before the lid was removed and the sample evaporated to near dryness. Acid was re-added and the process repeated a further four times. For full details of the acid digestion protocol, see *Guieu* [1991]. The digested samples were diluted to volume of 50 mL using 0.5 M HNO_3 Suprapur.

[22] After digestion, solutions were analyzed for trace metals by Graphite Furnace Atomic Absorption Spectroscopy (GFAAS, SpectraAA 220Z with Zeeman background correction) and Flame Atomic Absorption Spectroscopy (FAA, SpectraAA 220FS). To prevent contamination, all containers had been previously cleaned in 3M HNO_3 (1 week) and 0.1M HCl (1 week), rinsing thoroughly with ultra high purity water after each step. Calibration lines were obtained using standard additions. The limits of detection and procedural blanks for each metal are presented in Table 1.

[23] The metal concentrations in the blank filters were usually less than 10% of the sample concentrations. The analytical precision was estimated to be approximately 6% of the values reported, except for Zn and Co for which the precision was approximately 9%. The accuracy of the methods was evaluated on the basis of analyses of International Standard Reference Material, MESS-3 Sediment (NRCC). Results for all elements were within 5% of the certified values.

[24] Soluble metals were extracted using an ammonium acetate leach at pH 4.7 for 2 h [*Sarthou et al.*, 2003]. This pH was chosen primarily to mimic the release of trace metals from aerosols in rainwater, but is also similar to a procedure employed by *Bruland et al.* [2001] to examine the bioavailable Fe content of upwelled particulate matter off the coast of California. Sample extracts were analyzed for Al, Fe, Ti, Mn, Co and Cu using Graphite Furnace Atomic Absorption Spectroscopy (GFAAS, SpectraAA 220Z with Zeeman background correction). The concentrations of blank filters were usually less than 10% of sample concentrations. Soluble P and N were extracted using 100 mL Milli-Q water, following the approach of *Buck et al.* [2006] and measured on a standard segmented-flow nutrient autoanalyzer for nitrate + nitrite measurements [*Kirkwood*, 1996] and a segmented flow autoanalyzer equipped with a liquid waveguide capillary cell for the measurement of nanomolar concentrations of phosphate [*Patey et al.*, 2008].

2.5. Air Mass Identification

[25] The origin of air masses for each day sampled was interpreted using 5-day isentropic back trajectories (finishing at altitudes of 750, 1500, 2000, 2500, 3000 and 3500 m in 6 h steps) calculated twice a day (00:00 and 12:00 UTC) using the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model [*Draxler and Hess*, 1997; *Draxler*, 1999] and FNL (Global forecast system final analysis) and GDAS (Global Data Assimilation System) meteorological data set. In addition, specific dust outbreaks were interpreted with the aid of satellite imagery and forecast models, including a daily evaluation of TOMS (NASA) [*Herman et al.*, 1997], SeaWiFS-NASA satellite images (provided by the SeaWiFS Project, NASA/Goddard Space Flight Center and GeoEye), MODIS images (courtesy of MODIS Rapid Response Team, NASA GSFC), and the Dust Regional Atmospheric Model (DREAM) [*Nickovic et al.*, 2001].

3. Results

3.1. TSP Levels and Seasonal Variation

[26] The results in Figure 2 show the large variation of the daily concentrations of TSP at the TF station (269 m a.s.l.) and PG station (1930 m a.s.l). Peaks in particle concentration were observed all year-round; however the most intense events tended to occur during the winter and early spring (December–March). In this data set, the largest African dust intrusion in the last decade occurred on 5 January 2002 (5586 μg m^{-3} measured at the coastal TL station). Other significant peaks in concentration were recorded in winter and early spring: January 2005 (2314 μg m^{-3}), March 2004 (2295 μg m^{-3}) and February 2005 (2020 μg m^{-3}).

[27] During summer, the mean TSP concentration is higher due to dust transport occurring at high altitudes. This produces a marked seasonality (from July to September) in the dust record at the PG station (see Figure 3). It has been suggested that certain features of geographical relief in the

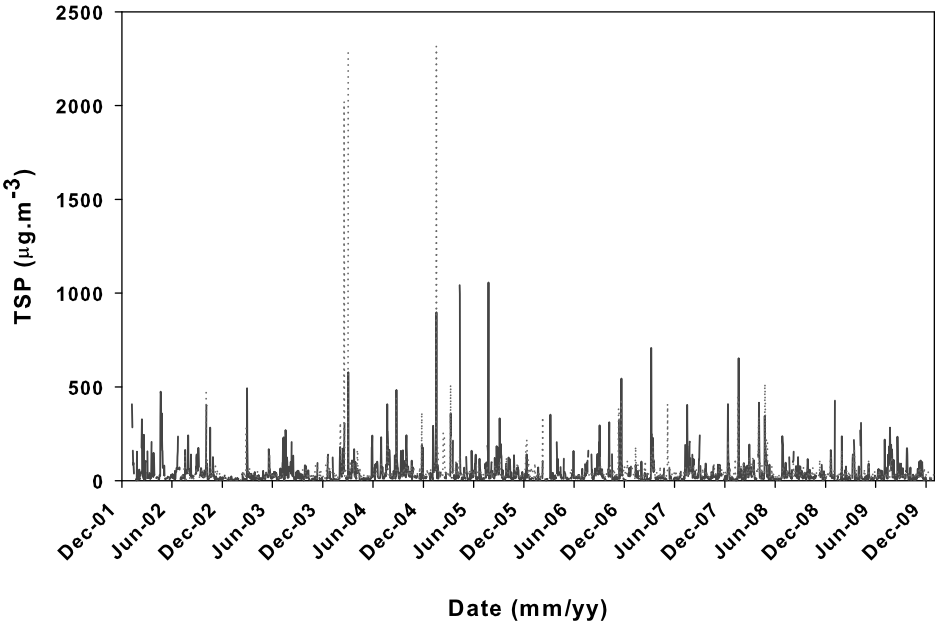

Figure 2. Concentration of TSP (in μg m^{-3}) at Pico de la Gorra (solid line, from December 2001–2009) and Tafira stations (dashed line, from January 2004 to December 2009).

Canary Islands may cause local alterations in the altitude of dust transport [*Falke et al.*, 2001]. Moreover, isolated outliers were registered at PG from January to April but without seasonal pattern. The TF station did not show a clear seasonal pattern, although higher standard deviations were found during the winter months (from December to March) corresponding to months during which strong dust events occurred (see auxiliary material).[1]

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011JD016646.

[28] Table 2 shows the statistical values for TSP concentrations at both stations. The 75th percentile is similar to the average while the 90th percentile shows that, despite the highest daily TSP values being recorded in TF, PG experiences a greater number of days where the concentration is more than 100 μg m^{-3} due to the summer events.

[29] TSP background air concentrations have been obtained by averaging daily TSP for non-African days at TF and PG stations. The calculated mean TSP background concentrations were 11.4 $\pm$ 8.6 μg m^{-3} (n = 688 days) at PG and 28.4 $\pm$ 12.1 μg m^{-3} (n = 728 days) at TF, which appear

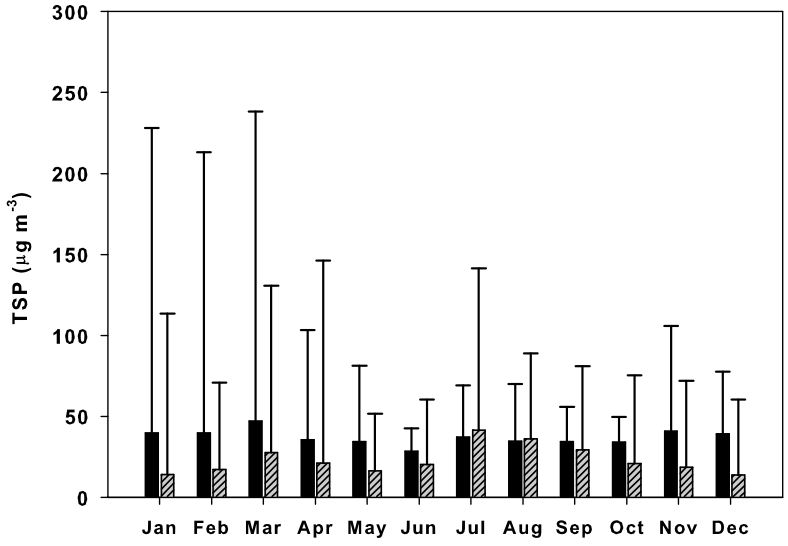

Figure 3. Geometric mean monthly TSP (μg/m3) measured between 2002 and 2009 at Pico de la Gorra (gray bars) and Tafira (black bars). Error bars represent one standard deviation calculated from all samples falling within a given month for all years.

Table 2. Statistical Summary of Atmospheric Concentrations (μg m^{-3}) at Pico de la Gorra and Tafira Stations[a]

	Pico de la Gorra	Tafira
	TSP	
Average (SD)	45 (82)	49 (92)
P-10	6	20
P-25	10	25
P-75	47	49
P-90	105	71
Median	19	34
Geometric mean	22	35
	TSP Background	
Average (SD)	11 (9)	28 (12)

[a]TSP, total suspended particles; SD, standard deviation; TSP Background, TSP for non-African dust outbreak days.

to correspond approximately to the 10th and 25th percentiles of TSP concentrations (Table 2). The difference between the background values at the two stations is consistent with more anthropogenic influence at the TF station. An increase in background PM10 has previously been reported for other stations with urban influences, such as for Arinaga and Playa del Inglés, in Gran Canaria [*Viana et al.*, 2002]. Background TSP concentrations at PG are similar to an average of 14 μg m^{-3} TSP reported for a rural station (Rio) in Tenerife [*Alonso-Pérez et al.*, 2007].

[30] During the period of simultaneous PM10 and TSP measurement at TF from 5 August 2004 to 31 December 2009, the mean PM10/TSP ratio was 0.63. This ratio is similar to previously reported data in urban background stations [*Viana*, 2003].

3.2. Aerosol Transport and Air Mass Back-Trajectories

[31] Chemical aerosol properties can be related to the origin and trajectory of the aerosol-laden air masses. The origin of the air masses that affect the troposphere in the northeast Atlantic around the Canary Islands have previously been investigated using isentropic back–trajectories, with classification of the air mass origin using a k-means clustering strategy [*Díaz et al.*, 2006], and by using the African Index that indicates the fraction of time that an air mass remained over a particular African region at one of the possible height intervals of the lower troposphere [*Alonso-Pérez et al.*, 2007].

[32] For this work, a classification of the origin of the air masses was made for samples collected between 2002 and 2009 with the objective of relating the chemical composition of transported mineral aerosols to their sources. The origin of air masses was established with regard to the geographic sectors crossed before reaching the Canary Islands by analyzing the back-trajectories. The sectors are defined with respect to a system centered on the station of Pico de la Gorra. This classification principally takes into account the influence that the ocean and continents exert on the composition of the air masses.

[33] We considered 5 geographic sectors: NS (Northern Sahara: 38°–30°N, 18°W–15°E, North Morocco, North Algeria and Tunisia), WCS (West and Central Sahara: 20–30°N, 18°W–20°E), SH sector (Sahel: 0°–20°N, 18°W–20°E), MAR (maritime aerosol, trajectories over the Atlantic Ocean) and EUR (European and maritime aerosol, trajectories that cross European continent and Atlantic Ocean) (see Figure 4). Regardless of the residence time of air mass over each sector, we have considered that all sectors crossed may contribute to the characteristics and the composition of the aerosol. For this reason, air masses that cross more than one sector are assigned a mixed origin category (i.e., NS-WCS and WCS-SH) to allow for the possibility that the chemical composition of dust aerosol may be different from individual sectors and show contributions from all areas traveled during transport.

[34] Figure 4 and Table 3 shows the seasonal origin of air masses for the period studied (2002–2009). Back trajectories were calculated and classified according to the aforementioned sources. Considering only African days, where samples contain some contribution from one or more of the African sectors (NS, WCS and SH), dust is most frequently transported from the NS and WCS sectors (32–50% of total African days). An increased frequency of the air masses originating in SH sector was observed during summer and autumn (10% of African days) but the Sahara continues to be the major source (37% of African days).

[35] African dust outbreaks during winter are characterized by intrusions in the lowest atmospheric levels mainly originating from the Sahara area. NS and WCS are the most significant sources of dust during the winter (49%) with the SH sector only contributing to around 2% of dust that reaches the Canary Region during this season.

3.3. Chemical Characterization of Dust Episodes

[36] An analysis was undertaken of total metal concentrations in dust samples collected at the PG and TF stations to investigate the chemical composition of African dust and to interpret its variability in relation to source region. Samples containing contributions from African source regions (NS, WCS or SH) were selected for the analysis based on calculated air mass back-trajectories. More than the 80% of selected samples corresponded with a TSP concentration in the range 10–200 μg m^{-3}, with many of the selected filters showing a characteristic orange-brown color.

3.3.1. Chemical Composition

[37] Table 4 lists the arithmetic mean, standard deviation, minimum and maximum concentrations and Enrichment Factor (EF) of all analyzed elements during African dust outbreak days at the two stations. High standard deviations from the mean concentrations of each element are observed during the studied outbreaks, due to the strong variability of mineral aerosol concentration in the samples analyzed. PG station results show lower total concentrations for elements with significant anthropogenic and marine sources whereas concentrations of crustal-derived elements are very similar at the two stations.

3.3.2. Enrichment Factors

[38] Enrichment factors (EFs) are often used to compare the composition of particulate aerosol or rainwater with the composition of the upper continental crust and they constitute a first approximation of the possible origin (crustal or anthropogenic) of metals in the atmosphere. Aluminum was chosen as the reference crustal indicator and the average chemical composition of the upper continental crust used was that reported by *Taylor and McLennan* [1995].

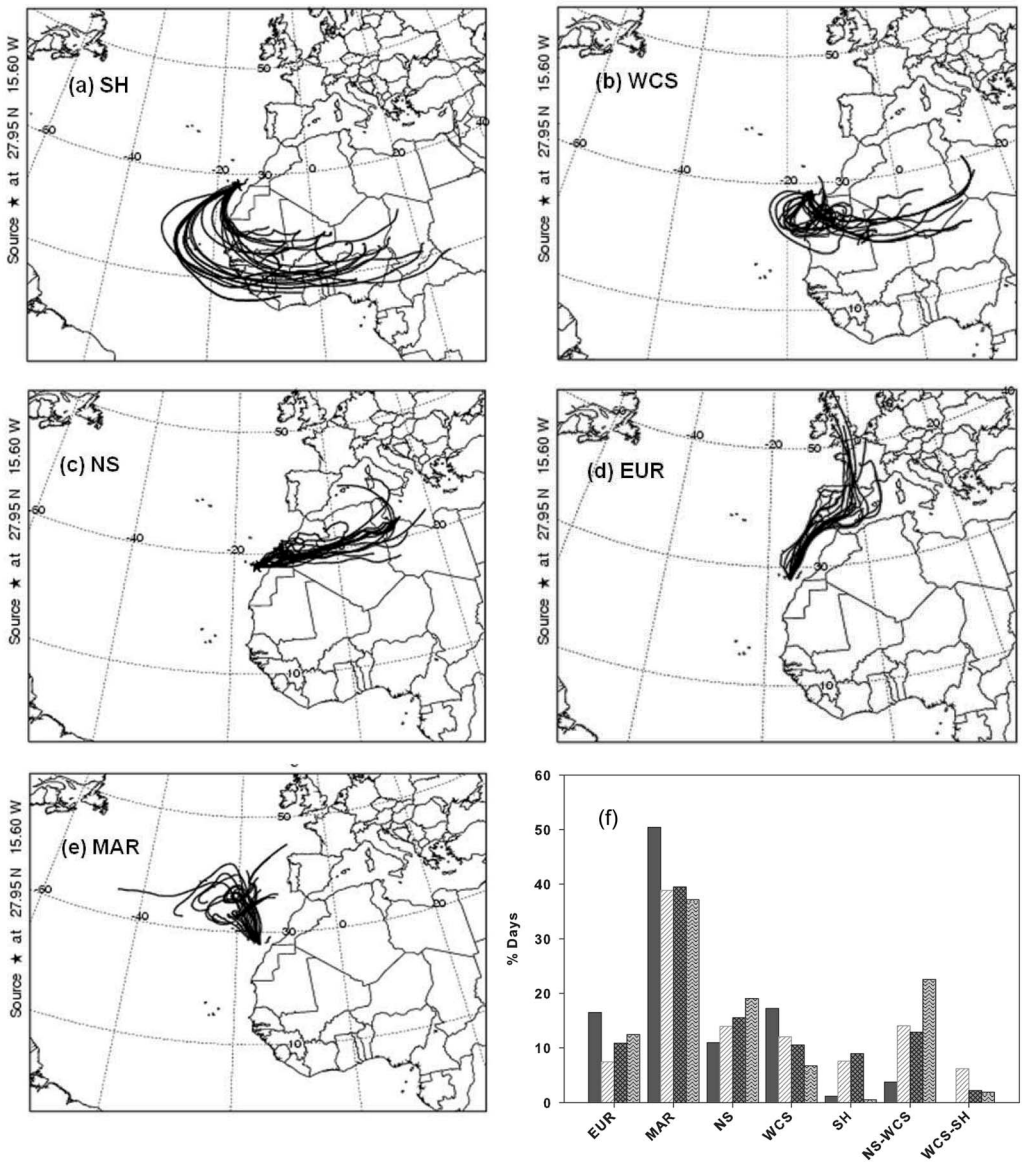

Figure 4. Classification of different back trajectories of the air masses with end point at Gran Canaria. Geographic sectors: (a) SH (Sahel: 0°–20°N, 18°W–20°E), (b) WCS (West and Central Sahara: 20°N–30°N, 18°W–20°E), (c) NS (North of Sahara: 38°N–30°N, 18°W–15°E), (d) EUR (air masses over Europe continent and Atlantic Ocean) and (e) MAR (trajectories over the Atlantic ocean). (f) Seasonal frequency of back trajectories of air masses arriving at the Canary region (2002–2009): Spring (gray area), Summer (white striped area), Autumn (dark gray checkered area) and Winter (light gray striped area).

[39] The EF value for an element (X) in aerosol sample is then calculated according to the equation:

$$(EF)_x = ([X]/[Al])_{aerosol}/([X]/[Al])_{crust}$$

where $([X]/[Al])_{aerosol}$ and $([X]/[Al])_{crust}$ are the concentration ratios of the trace metal (X) and the indicator (Al) in aerosol and the average crustal material. EFs were determined for the samples collected at TL and PG stations (Table 4). An EF values greater than 10 is taken to indicate significant non-crustal enrichment attributable to anthropogenic inputs. Metals such as Mn, Fe, Co and Ti are of predominantly crustal origin at both stations (EF < 10). Na, Ca and Mg showed an EF > 10 in TL station due to the influence of sea-salt particles. Ni, Zn, Pb, Cd and Cu are mainly associated with particles of non-crustal origin and showed the highest EF values. The EF values are slightly higher for most elements for the non-dust samples with a larger standard deviation (data not shown) than those observed for the dust samples. At PG station, EF (Cu) values decrease from anomalous enrichment values (>100) to values <10 of crustal elements [e.g., *Chester et al.*, 1999] during the dust events.

3.3.3. Principal Component Analysis

[40] Common factors influencing the elemental concentrations were examined by principal component analysis (PCA) with Varimax Rotation using the SPSS statistical software package software. Table 5 shows the variable

Table 3. Classification of Air Mass Origin Between 2002 and 2009, Showing Percentage Contribution From Each Sector for Each Season[a]

Source	Spring	Summer	Autumn	Winter
EUR	16.6	7.5	10.9	12.4
MAR	50.4	38.8	39.4	37.2
NS	10.9	13.9	15.4	19.0
WCS	17.2	12.0	10.5	6.6
SH	1.1	7.5	8.8	0.4
NS-WCS	3.8	14.1	12.8	22.5
WCS-SH		6.2	2.2	1.9

[a]Sources: NS (Northern Sahara: 38°N–30°N, 18°W–15°E, North Morocco, North Algeria and Tunisia), WCS (West and Central Sahara: 20°N–30°N, 18°W–20°E), SH sector (Sahel: 0°–20°N, 18°W–20°E), MAR (maritime aerosol, trajectories over the Atlantic Ocean) and EUR (European and maritime aerosol, trajectories that cross European continent and Atlantic Ocean). Seasons: Spring (March–May), Summer (June–August), Autumn (September–November) and Winter (December–February).

loadings on the first four principal components, which explain 82.7% of the total variance. The first component, which explains 42.4% of the total variance, has a high correlation coefficient (>0.75) with typical crustal tracers (Al, Mn, Fe, Ti, Co and Ca). Cu, Pb, Cd, Zn and Ni show no correlation with this component. The second component shows a strong correlation with components of sea aerosol (Mg and Na). We also found a correlation between Zn and these elements probably due to reactivity of Zn with sea salt in the MBL [*Roth and Okada*, 1998]. Components 3 and 4 are highly correlated with the anthropogenic metals. Correlation between Pb, Cd and Ni suggests a common origin (emission) and/or transportation patterns for these metals. We believe that the high copper component can be explained by local factors associated with the different influences at the two stations TL (anthropogenic influence) and PG (rural). This is supported by the reduction in the EF (Cu) during African dust days at PG station.

3.3.4. Aerosol Source Patterns

[41] Elemental characterization of the collected mineral aerosol and back trajectories of the air masses are used to distinguish regional African sources of dust. Metal/Al ratios are presented in Table 6 and compared with reported values for African aerosols and soils measured in the western North Atlantic area. Dust aerosol samples from NS, WCS and SH sectors show different Ca/Al, Ti/Al and Mg/Al ratios.

[42] Ca/Al and Mg/Al ratio were estimated using only data from PG due to the high proportion of marine salts in TL samples. Table 6 shows that the events originating from the NS region contain high Ca/Al ratios compared with those observed for dust originating from the WCS and SH sectors. Similar results have been found for samples collected at Cape Verde [*Chiapello et al.*, 1997] and at Tenerife [*Kandler et al.*, 2007]. The decrease in calcium content from north to south is consistent with the soil maps given by *Desboeufs and Cautenet* [2005] and the reported composition of African soils [*Schütz and Sebert*, 1987].

[43] The three African regions show different mineralogical composition both in soils and aerosols [*Goudie and Middleton*, 2001; *Moreno et al.*, 2006]. Differences in the chemical characteristics of aerosols collected in Cape Verde

Table 4. Statistical Summary of Atmospheric Metal Concentrations (ng m^{-3}) and Enrichment Factors (EF) During African Dust Outbreak Days at Pico de la Gorra and Taliarte Stations[a]

	Mean	SD	Geo. Mean	Minimum	Maximum	P-10	P-90	EF (R = Al)	SD (EF)
					Pico de la Gorra				
Al	4580	5687	2207	230	33280	446.	11136		
Fe	2919	3229	1308	50.0	17180	221	6574	1.9	0.6
Ca	2496	3389	1189	86	22110	201	6936	2.3	1.8
Mg	1206	1635	494	10.0	9380	82.5	2527	2.1	1.3
Na	1141	1092	716	30.0	5389	203	2744	2.0	2.1
Ti	197	240	93.7	10.0	1460	22.3	408	2.5	1.6
Zn	146	381	37.3	9.7	2990	10.8	276	140	471
Mn	54.5	56.9	21.8	2.8	293	4.3	94.5	2.1	1.2
Cu	12.6	22.2	7.5	1.5	119	2.4	48.3	7.5	3.4
Ni	4.4	4.9	2.0	0.4	26.6	0.5	7.9	9.7	17.2
Pb	3.5	4.9	1.9	0.4	30.7	0.5	6.0	6.8	5.4
Co	1.4	1.6	0.5	0.06	9.4	0.09	2.5	2.8	1.9
Cd	0.2	0.2	0.1	0.04	1.7	0.06	0.6	128	177
					Taliarte				
Al	3278	4334	1895	110	26200	553	9279		
Fe	2256	5386	1051	90.0	60810	293	4860	1.5	0.4
Ca	9651	65101	3319	230	786100	991	11203	6.1	5.0
Mg	12897	46136	5295	30.0	429390	553	16267	67.1	272
Na	61820	86782	34714	270	802480	7846	119743	167	284
Ti	234	455	111	10.0	3720	29.1	416	2.6	1.7
Zn	417	1061	185	16.3	11920	34.7	897	290	393
Mn	38.4	52.5	21.6	2.9	328	5.9	87.1	2.1	1.8
Cu	21.9	126	8.5	0.7	1642	2.6	30.0	62.1	100
Ni	6.6	7.3	3.6	0.2	35.1	0.8	17.3	13.5	17.3
Pb	11.8	19.2	7.3	0.4	185	2.7	20.1	27.2	32.2
Co	1.1	1.2	0.7	0.03	8.0	0.2	2.4	4.5	3.3
Cd	0.4	0.5	0.2	0.01	2.8	0.09	0.8	151	213

[a]SD, standard deviation; Geo. Mean, geometric mean; P-10 and P-90, 10th and 90th percentile, respectively.

Table 5. Varimax Rotated Component Matrix

	1-Crustal	2-Marine	3-Anthropogenic	4-Cu
Mn	0.897	−0.022	0.281	0.226
Fe	0.889	−0.155	0.341	0.124
Co	0.809	−0.002	0.268	0.453
Al	0.909	−0.166	0.233	−0.177
Ca	0.815	0.263	−0.371	−0.046
Ti	0.753	−0.101	0.550	0.124
Ni	0.342	0.283	0.668	0.042
Mg	0.001	0.892	0.222	−0.069
Na	−0.171	0.865	0.178	0.077
Pb	0.355	0.048	0.741	−0.018
Cu	0.138	−0.020	0.006	0.981
Cd	−0.023	0.496	0.660	0.044
Zn	0.029	0.758	−0.028	−0.034

[*Chiapello et al.*, 1997], Canary Islands [*Arimoto*, 2001; *Kandler et al.*, 2007] and Puerto Rico [*Formenti et al.*, 2003] could reflect either differences between the potential sources or be the result of mineral fractionation during the transport of aeolian soil dust. *Kandler et al.* [2007] have found differences in the elemental mass ratios (i.e., Ca/Al and Fe/Al) between the aerosols collected at Izaña and Puerto Rico.

[44] The differences between the values of Fe/Al ratios in the three African sectors were not statistically significant. The Fe/Al ratio was 0.57 ± 0.15 (n = 266) in the total particulate samples analyzed from the African days in this study. This is similar to other values reported: a crustal average of 0.44 [*Taylor and McLennan*, 1995] and the mass ratios observed in the atmosphere over the North Atlantic 0.58 in Tenerife [*Kandler et al.*, 2007] and 0.53 in Cape Verde [*Formenti et al.*, 2003]. The average Fe/Al ratio was 0.66 ± 0.27 (n = 21) in the dust samples collected during days with back trajectories originating in the Sahel (SH) region.

3.4. Deposition Measurements

3.4.1. Particle Flux Calculations

[45] Dry and wet deposition fluxes measured at the TF station are given in Figure 5. The total flux calculated from the measurements of dry and wet deposition was 42.3 mg m^{-2} d^{-1}. Higher deposition fluxes were observed in winter, in agreement with the higher mean atmospheric concentration of TSP during that season. The wet deposition flux was approximately 5% of the total flux in the studied period from December 2003 to December 2008. The experimentally determined total deposition flux is consistent with that

Table 6. Elemental Ratios in Dust Collected at Gran Canaria (Average (st dsv)) Compared With Other Reported Values Measured Around Northern Africa

Region	Author	Ca/Al	Mg/Al	Fe/Al	Ti/Al	Type of Sample	Sampling Site	Period of Sampling
NS	*Coudé-Gaussen et al.* [1987]	3.3		1.25		aerosols	Fuerteventura	18 Apr1984
	Chiapello et al. [1997]	0.60		0.54		aerosols	Cape Verde	Dec1991–Dec1994
	Lafon et al. [2006]	0.16		0.53		aerosols	Cape Verde	Jan1993
	Guieu et al. [2002]			0.66		soils (<20 μm)	Morocco	
	Linke et al. [2006]	1.85	0.31	0.57	0.16	soils (<20 μm)	Morocco	
	Moreno et al. [2006]	2.86	0.4	0.66	0.12	soils	Morocco	
	This work	1.15(0.41)	0.36(0.1)	0.64(0.12)	0.11(0.004)	aerosols	Gran Canaria	2002–2009
WCS	*Chiapello et al.* [1997]	0.36		0.51		aerosols	Cape Verde	Dec1991–Dec1994
	Stuut et al. [2005]			0.51	0.075	aerosols (samples 5–9)	Atlantic Ocean	Feb–Mar1998
	Lafon et al. [2006]	0.09		0.45		aerosols	Cape Verde	Feb1994
	Guieu et al. [2002]			0.7		soils (<20 μm)	South of Algeria	
	Moreno et al. [2006]	0.21	0.14	0.52	0.11	soils	South of Algeria	
	This work	0.37(0.13)	0.18(0.02)	0.60(0.07)	0.070(0.005)	aerosols	Gran Canaria	2002–2009
SH	*Adepetu et al.* [1988]		0.14	0.71	0.08	aerosols	Niger-Harmattan	Dec–Feb (1981–1984)
	Chiapello et al. [1997]	0.20		0.54		aerosols	Cape Verde	Dec1991–Dec1994
	Viana et al. [2002]	1.15	0.48	0.88	0.06	aerosols	Izaña, Tenerife	Jun2001
	Stuut et al. [2005]			0.54	0.06	aerosols (samples 10–18)	Atlantic Ocean Oceanographic cruise	Feb–March1998
	Lafon et al. [2006]	0.12		0.44		aerosols	Cape Verde	Jan1992
	Formenti et al. [2008][a]	0.40–0.60	0.14–0.30	0.59–0.70	0.08–0.10	aerosols (ground-based and airbone)	Niger and Senegal	2006–2007
	Guieu et al. [2002]			0.79		soils (<20 μm)	Niger	
	Moreno et al. [2006][a]	0.04–0.18	0.05–0.1	0.5–0.64	0.08–0.13	soils	Bodélé depression and Niger	
	Linke et al. [2006]	0.08	0.04	0.23	0.04	soils (<20 μm)	Niger	
	This work	0.43(0.19)	0.25(0.07)	0.62(0.10)	0.080(0.002)	aerosols	Gran Canaria	2002–2009
African Aerosols[b]	*Kandler et al.* [2007]	0.42	0.21	0.59	0.084	aerosols	Izaña, Tenerife	Jul–Aug2005
	Alastuey et al. [2005]	0.4	0.17	0.52	0.052	aerosols	Izaña, Tenerife	Jul2002
	Alastuey et al. [2005]	1.01	0.33	0.66	0.087	aerosols	Sta. Cruz, Tenerife	Jul2002

[a]Range of ratios for samples collected at different places and time.
[b]Mean values for samples collected in the Canary Islands and originating from a variety of source regions.

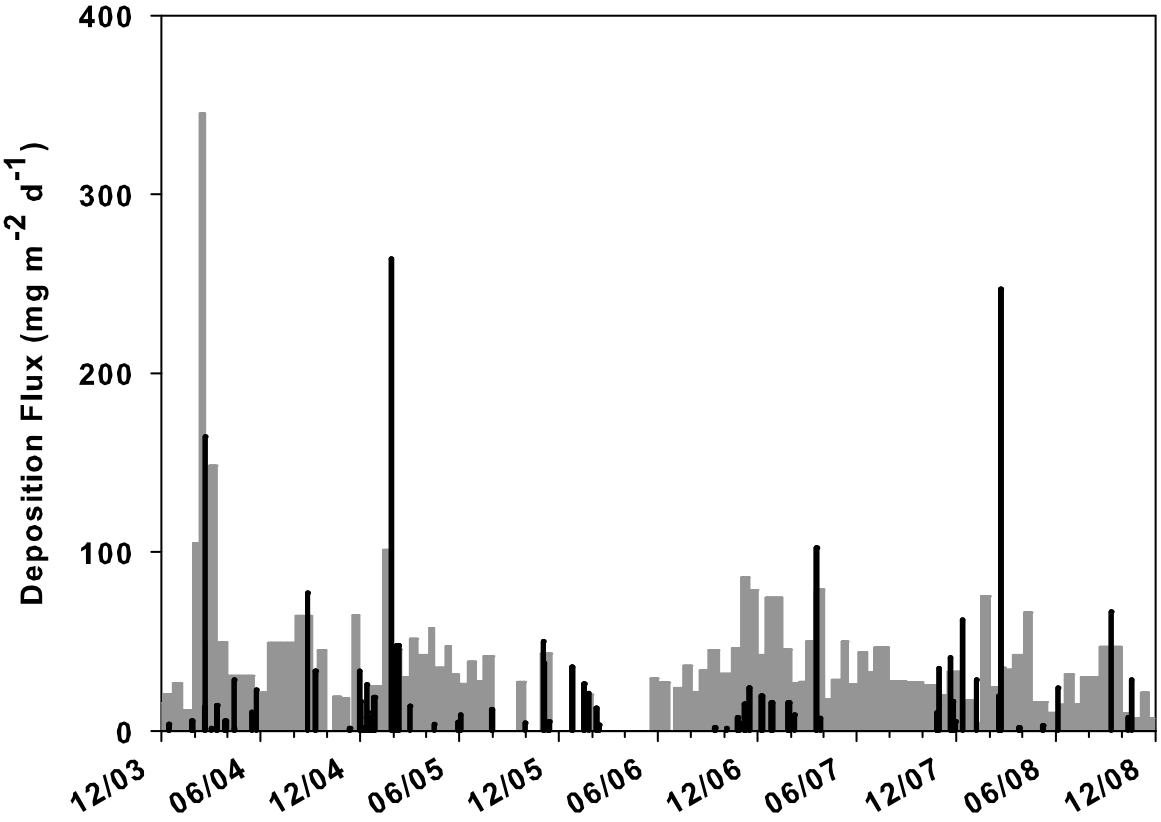

Figure 5. Dry (gray area) and wet (black area) deposition fluxes at Tafira station (from January 2003 to December 2008).

calculated from theoretical models [see, e.g., *Duce et al.*, 1991]. On the basis of these models, we have estimated that the main contribution (>75%) to the annual particle fluxes at PG and TF stations is from dust inputs occurring during periods of African dust transport.

[46] The experimentally determined dry deposition velocity was about 0.9 ± 0.4 cm s^{-1}, reaching values up to 2.4 cm s^{-1} during intense dust events. This velocity is consistent with the observed size distribution of particles with approximately 80% of particles in the 1.3–10 μm size range [*Torres-Padrón et al.*, 2002] and similar to that used by *Tegen and Fung* [1994] to model the deposition of "small" silt (corresponding to 6.1 μm) and by *Duce et al.* [1991] for estimating deposition of aerosols larger than 2 μm (V_d = 2 cm s^{-1}).

3.4.2. Soluble and Total Fluxes of Trace Metals

[47] As an alternative to actual measurements, dry fluxes (F_d) often are estimated from measured trace element concentrations in air (C_i) and model-derived or estimated dry deposition velocities (V_d) [*Jickells et al.*, 1987; *Duce et al.*, 1991].

[48] The dry deposition fluxes (F_d) of trace element were estimated using the following equation:

$$F_d = C_i \times V_d$$

where C_i is the geometric mean particulate trace metal concentration in the atmosphere of the element concerned (i) and V_d is the elemental dry settling velocity, which varies with particle size. A significant limitation with the use of constant dry deposition velocities is that the effects of meteorological conditions are neglected [*Slinn*, 1983]. Nevertheless, these average depositional velocities appear to be appropriate for predicting dry deposition velocities [*Spokes et al.*, 2001; *Koçak et al.*, 2005], and have been widely applied to the geochemical study of the dry deposition of mineral dust and its constituent trace elements [e.g., *Arimoto et al.*, 2003].

[49] Geometric means were calculated using all available trace metal data from the PG station. To check whether the selection of sampling days for trace metals at this station (see Section 2.2) may have introduced a bias in the calculated mean concentrations, the TSP concentrations measured on days when samples for trace metals were taken (166 samples) was compared with all TSP concentrations measured between 2002 and 2009 (2611 samples). Application of the Mann-Whitney U test showed no significant differences between the two distributions of measurements (U = 208625, significance = 0.115), indicating that the trace metal measurements were representative of long-term TSP measurements at the site. However, the geometric mean TSP concentration for the whole TSP data set was 80% of the corresponding value for days with trace metal sampling, with a geometric mean of 21.85 μg m^{-3} for all samples, compared with 27.30 μg m^{-3} for the days sampled for trace metals. To compensate for this bias, the geometric mean concentration of each element used for flux calculations was scaled to 80% of the measured geometric mean.

[50] Estimated metal dry deposition fluxes are presented in Table 7. The dry fluxes of Al, Fe, Ti, Mn and Co have been estimated assuming that the elemental V_d for coarse particles

Table 7. Estimated Trace Metal Deposition Fluxes in Gran Canaria[a]

	Al	Fe	Ti	Mn	Cu	Co	Zn	Pb	Cd	Reference
F_T	770	372	29	7.1	0.22	0.18	0.9	0.04	0.004	This work
	130–1655	228–1164		3.0–10	1.4–1.9	0.03–0.27	0.5–65	0.87–2.47	0.021–0.4	*Guieu et al.* [1997, 2010]
F_S	14	8.1	0.23	3.1	0.13	0.076				This work
	10–102	32–140		1.8–12	0.8–1.3	0.02–0.13				*Guieu et al.* [1997, 2010]

[a]Total dry flux of metals (F_T) and total soluble flux of metals (F_s) in mg m^{-2} y^{-1}. Comparison with estimated dry deposition fluxes in the Mediterranean, adapted from *Guieu et al.* [1997, 2010].

depositing to the ocean less than 1000 km from land is 2 cm s^{-1} [*Duce et al.*, 1991]. For Cd, Pb, Zn and Cu which are found primarily associated with sub-micrometer particles a mean value of 0.1 cm s^{-1} was applied as described by *Duce et al.* [1991]. Estimated atmospheric deposition of soluble Al, Fe, Ti, Mn, Co and Cu in the region have also been calculated assuming that the soluble fractions of these metals are 1.8(±0.8), 2.2(±1.2), 0.8 (±0.6), 44.2(±19.5), 41(±22.6) and 60.7(±18.3) %, respectively (using mean values calculated from 54 dust samples in this study and with quoted uncertainties of ±1 standard deviation). Values are broadly comparable with data reported for the Mediterranean by *Guieu et al.* [1997, 2010] (listed in Table 7).

4. Discussion

[51] Comparing the average concentrations of TSP at the PG and TF stations with those from a 6-year study at the Rio station on the island of Tenerife [*Alonso-Pérez et al.*, 2007], higher values have been measured in this work. Although the sampling periods are different, the difference appears to indicate the spatial variability of the events between the islands. Factors such as differences in relief between the islands [*Falke et al.*, 2001] and island-wide weather patterns seem to be the cause of this variation between sampling stations.

[52] The data sets collected at these stations show consistency in the observed seasonal pattern, with peaks in dust concentration in winter and summer. However, the figures for the winter events are slightly lower at PG. Events lasting more than one week are more common at PG during the summer. However, the reverse pattern is recorded at TF with larger events during the winter. This can be explained by the difference in the height of the air masses that carry dust relative to the altitude of these stations, as during the winter dust transport is predominantly at low altitudes, as can be seen from the study of air mass back-trajectories performed. The dust arrives to the Archipelago from the East at relatively low altitudes and is concentrated mainly within the marine boundary layer (MBL).

[53] The outbreaks last between 1 and 3 days in more than 50% of cases. The most intense events have occurred in TF and PG in winter and early spring. This is consistent with the observation of an increased dust deposition flux in winter (30%). Wet deposition (which contributes 3% to the total deposition flux) occurs almost exclusively during the months of December to January.

[54] It has previously been proposed, based on two years of sampling at the PG station, that the highest mean annual dust concentrations could be related to years with a strongly positive NAO index [*Torres-Padrón et al.*, 2002]. To examine the inter-annual variability of dust distribution at Pico de la Gorra in relation to the NAO index, values were taken from http://www.cgd.ucar.edu/cas/jhurrell/indices.data.html. *Hurrell* [1995] calculated the NAO index by taken the difference between normalized sea level atmospheric pressures between Lisbon, Portugal and Stykkisholmur, Iceland. High NAO (NAO+) leads to drier weather and increased dust mobilization, while during periods of low NAO (NAO−), precipitation is likely to be greater over northern Africa [*Moulin et al.*, 1997]. Values of the winter NAO index were used because the NAO is stronger during this season.

[55] Figure 6 shows the year-to-year variability of the NAO in winter (December–March) and the winter mean of TSP between 1996 and 2010. The correlation coefficient (R) between them is 0.59 (p = 0.05), which is similar to reported values for TOMS and Meteosat dust records (R = 0.67 and 0.74 respectively [*Chiapello and Moulin*, 2002]), simulated and observed concentrations at Barbados (R = 0.67 and 0.5, respectively [*Ginoux et al.*, 2004]) and summer dust loadings at Barbados (R = 0.49 [*Moulin et al.*, 1997]). During the last 5 years of the data set, where the NAO has higher absolute values, there is much stronger correlation (R > 0.9). Nevertheless, the relatively weak relationship observed between TSP and the NAO index suggests that during some periods the NAO does not exert a strong influence on dust production. Other inter-annual and monthly relationships were not found between the NAO index and dust loading. For the measurements made at the TF station no correlation between dust concentrations and the NAO was observed, maybe due to the location and altitude of the station (within the marine boundary layer) making it susceptible to other important influences.

[56] An examination of the variability of the major crustal metals Al, Fe, Ti, Mg and Ca in samples from different geographic areas was made in order to establish possible relationships between the elemental composition of aerosols reaching the Canary Islands and their places of origin. The t-test was applied to the different ratios between the metals Fe, Al, Ti, Ca and Mg and the results indicate no statistically significant difference between the regions for ratios Fe/Al, Fe/Ti and Ca/Ti. However, the ratios of Ti/Al, Ca/Al and Mg/Al in samples can be used to distinguish samples of SH, NS and WCS provenance. The Ti/Al ratio is significantly different in the three sectors (Student's t-test NS_WCS $p < 0.001$; NS_SH $p = 0.003$; WCS_SH $p = 0.04$), while the Ca/Al (Student's t-test NS_WCS $p < 0.001$; NS_SH $p = 0.004$) and Mg/Al (Student's t-test NS_WCS $p < 0.001$; WCS_SH $p = 0.003$) only show significant differences between the NS and WCS sectors, respectively.

[57] The values found for Fe/Ti in this study are similar to those reported for soil samples of Bodélé depression

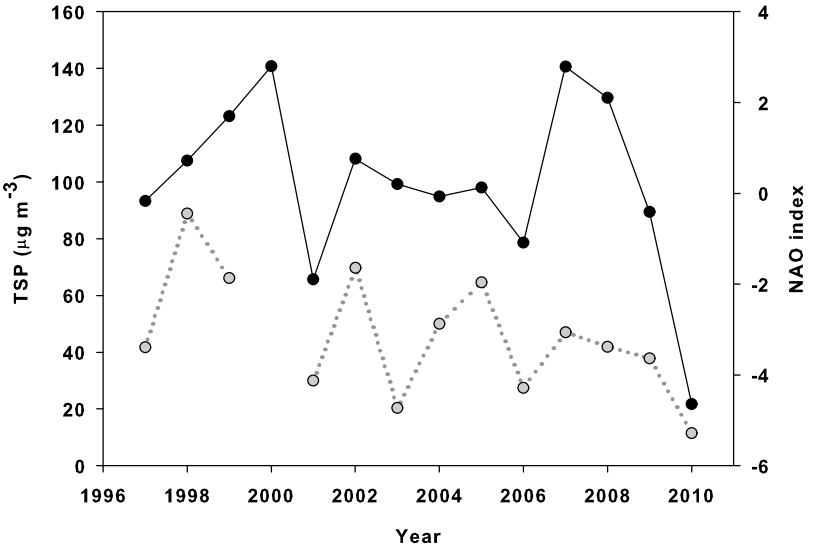

Figure 6. Comparison between the winter North Atlantic Oscillation (NAO) index (black spots and continuous line) and the mean of winter TSP observed at Pico de la Gorra station (gray spots and dashed line), from 1997 to 2010.

[*Moreno et al.*, 2006] and aerosol samples collected in Tenerife [*Alastuey et al.*, 2005; *Kandler et al.*, 2007]. There were no signs of relative enrichment of iron between any of the three African regions. The value of Fe/Al in samples originating in SH is only slightly higher than in northern areas. Since only a very small percentage of dust events reaching the Canary Islands originate in the SH sector and often also contain a significant contribution from the WCS sector it is difficult to have a significant number of samples of pure SH origin. The long transport paths in samples from these sectors (typically more than five days) may also be a factor in altering the chemical composition of the aerosols. Longer transport of dust would favor both homogenization of the aerosols by mixing and size fractionation of the aerosols, resulting in a preferential selection of finer-grained minerals, particularly clays, as coarser minerals such as quartz are lost through deposition [*Glaccum and Prospero*, 1980; *Schütz*, 1989]. The relative observed constancy of the ratio Fe/Al and Fe/Ti in the samples of all Africans sectors also points toward possible dust homogenization. This is also consistent with model studies which suggest that dust is well mixed in iron [*Hand et al.*, 2004].

[58] The analysis of metal concentrations during the African dust events reflect a relative constancy in the distribution of elements such as Fe, Al and Ti, which throughout the study period does not vary significantly with season or aerosol source. This agrees with the observation that the SH sector is the least common among African sectors and virtually all of the dust that is transported to the islands arrive via air masses crossing the NS sectors and WCS, often with air masses passing through more than one sector.

[59] Measured soluble nutrient concentrations in the dust samples were between 0.4 and 2.9 nmol m^{-3} phosphate and 21–64 nmol m^{-3} nitrate + nitrite, resulting in mean N/P, Fe/P and Fe/N ratios of 59, 1.4 and 0.03, respectively. In all samples the N/P, Fe/P and Fe/N ratios were significantly higher than the Redfield values (data not shown). The relatively low abundance of P in atmospheric inputs appears to be related to the observed P limitation in the NE Subtropical Atlantic Ocean [*Baker et al.*, 2003; *Duarte et al.*, 2006].

5. Conclusions

[60] The Canary Islands are subject to significant dust inputs throughout the year, with more frequent episodes in the months of June, August and September for transport at higher altitudes while the highest concentrations were registered in winter and early spring in the lower layers. This study has demonstrated that the selection of different sampling stations in the islands can be helpful for the maintenance of long series of mineral aerosol measurements but these sites should be placed with caution taking into account local features such as the influence of altitude of the trade wind inversion layer (TWI) which determine the height of the transport of the different air masses. During the summer, dust events could only be recorded at the high altitude PG station. However, especially during the winter and early spring dust events, the collected samples in TF and TL stations presented higher concentrations than samples obtained at the PG station. Therefore, simultaneous sampling at different heights allows a more detailed determination of the seasonal pattern of the transport of mineral aerosols in the region.

[61] A comparison of elemental enrichment factors in trace metal samples collected at PG and TL reveals clear contributions from sea-salt (Na and Mg) and local anthropogenic influence (Cu) at the coastal TL site, with enrichment factors at this site significantly higher than measured at the more remote PG station. In this respect the PG station is a better location to conduct a long time aerosol series because due to the low pollution background and practically non-existent sea salt aerosol.

[62] The interannual variability of the observed events is related to the NAO index although some discrepancies are observed due to the location and altitude of the sampling stations and the effect of the MBL on the Islands. The data presented in this work show a good correlation between dust concentrations in Pico de la Gorra and the winter NAO index. However, it is important to consider other factors that influence dust concentrations, since there is also a large amount of within-season variability in the patterns of atmospheric circulation of the North Atlantic [*Hurrell and Deser*, 2009].

[63] Although many authors have found differences in mineralogical and chemical composition of African dust depending on the lithology in the source regions [*Bergametti et al.*, 1989a, 1989b; *Ganor*, 1991; *Chiapello et al.*, 1997; *Caquineau et al.*, 1998, 2002], our results suggest that a homogenization of African aerosols appears to occur with respect to the total iron content of the aerosol particles. Some recognizable signatures were found in SH, WCS and NS provenances due to characteristic variations in the content of Ti, Mg and Ca, respectively. There is no sign of enrichment of Fe for the samples originating in areas with relatively Fe-rich soils, such as the Chad Basin [*Lafon et al.*, 2004]. The lack of any such signal suggests the operation of mineral selection processes during atmospheric transport.

[64] Deposition fluxes obtained in this study constitute a first geochemical estimate of the magnitude of dust inputs in the region based on sampling with high temporal resolution. The total flux is dominated by the dry deposition component. The dry deposition velocities indicate that the flux is mainly dominated by mineral dust and super-micrometer particles not associated with the sea salt (according to the sizes of the major aerosol types) [*Duce et al.*, 1991]. Aerosol deposition is an important supply of macronutrients and trace metals (Fe, Co, Mn, Cu and Al) to the NE Subtropical Atlantic Ocean which may stimulate the autotrophic components (nitrogen fixation and diatoms). However, the aerosols are relatively depleted in soluble P relative to other nutrients (soluble Fe and N) with respect to phytoplankton requirements for these elements.

[65] This work represents a valuable contribution toward understanding of the transport and chemistry of mineral aerosols over the North Atlantic Ocean, where there is a great shortage of in both the temporal and spatial resolution of measurements, particularly in the Eastern Boundary. Given the sporadic and highly variable nature of dust inputs, such long-term data sets are essential in order to further understanding of the composition and properties of African mineral dust as well as to constrain models of dust transport and deposition in the region.

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