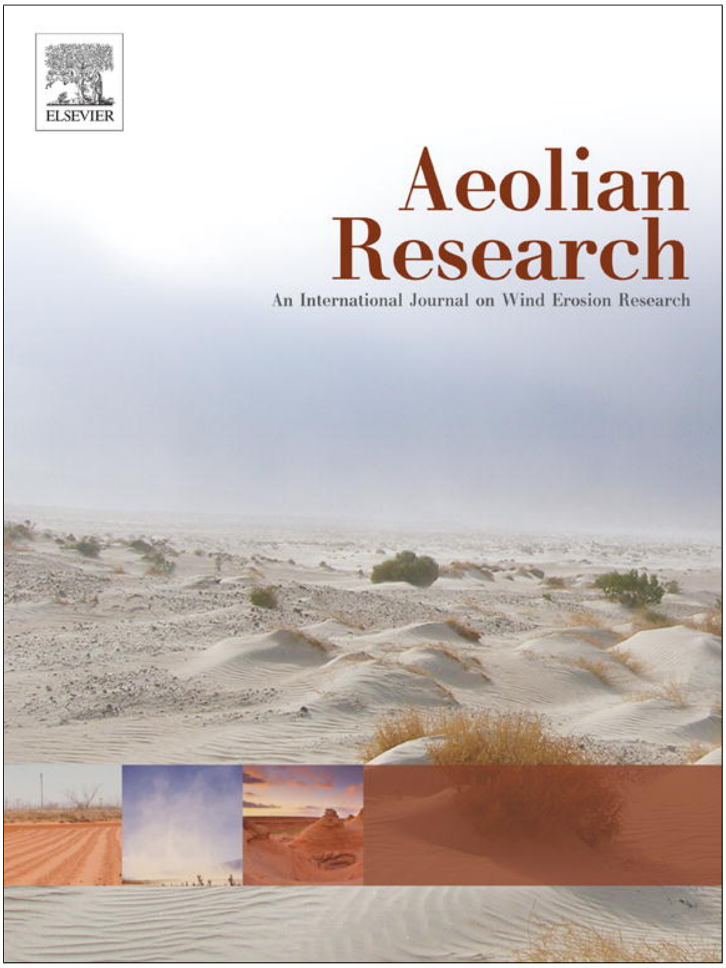
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Review Article

A review of methods for long term in situ characterization of aerosol dust

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ABSTRACT

Desert dust emitted and transported to distant regions influence on the climate, biogeochemistry and air quality of our planet. Long term (multi-decadal) measurements of aerosol dust with in situ characterization techniques are crucial for understanding the interactions of dust with the Earth system. We reviewed the programs which have been performing such long term dust monitoring. First long term observations started in the North Atlantic during the 1960s. Although dust programs rapidly expanded during the 1980s and 1990s, mostly across the Pacific and Atlantic oceans, only a few sites that have been active in the last 20–30 years continue operative nowadays. We reviewed the techniques that are available for in situ aerosol dust characterization. Techniques for long term measurement of mass concentration, size distribution, bulk composition, optical properties are nowadays available. Other techniques that are not regularly used in (or designed for) long term programs are also described; these may be incorporated into long term programs in specific periods (e.g. dust season on a year-to-year basis). Their implementation in long term programs allow us to study potential changes in the sources of some key minerals linked to changes in the strength and location of dust sources, and/or the mixing state of dust with pollutants related to the evolution of anthropogenic emissions. Given that some of these methods are not specifically designed for dust, some considerations should be addressed when the techniques are used for aerosol dust measurements. Accuracy, sources of errors, corrections and features of dust measurements are reviewed.

Contents

Abbreviations: AA, atomic absorption; AC, automated colorimetry; ACE, aerosol characterization experiment; AMMA, African monsoon multidisciplinary analyses; AMS, aerosol mass spectrometer; APSTOFS, aerodynamic particle size – time of flight spectrometer; ATOFMS, aerosol time of flight mass spectrometer; DMA, differential mobility analyser; DRUM, davis rotating-drum universal-size-cut monitoring; EC, elemental carbon; EDX, energy dispersive X-ray; EMA, electrical mobility analyser; EUSAAR, European supersites for atmospheric aerosol research; FAA, flame atomic absorption; GPC, gas proportional counting; HR, relative humidity; HR-ICP-MS, high resolution mass spectrometry; IC, ion chromatography; ICP-AES, inductively coupled plasma atomic emission spectrometry; ICP-MS, inductively coupled plasma mass spectrometry; INAA, instrumental neutron activation analysis; ISE, ion selective electrode; LSS, liquid scintillation spectrometry; MC-ICP-MS, multi collector-inductively coupled plasma mass spectrometry; MINATROC, mineral dust and tropospheric chemistry; NIOSH, national institute for occupational safety and health; OC, organic carbon; OPC, optical particle counter; PIXE, proton induced X-ray emission; PLS, polystyrene latex spheres; PM_x, mass concentrations of particulate matter with an aerodynamic diameter smaller than x microns (50% efficiency at x microns). Terms PM_{10}, $PM_{2.5}$ and PM_1 are used here; PRIDE, Puerto Rico dust experiment; SAMUM, Saharan mineral dust experiment; SEM, scanning electron microscopy; TEOM, tapered element oscillating microbalance; TIMS, thermal ionization mass spectrometry; TOR, thermo-optical reflectance; TSP, total suspended particles; XRF, X-ray fluorescence.

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1. Introduction

Large amounts of dust particles are emitted from arid soils of the Earth (500–1500 Mt/y; Ginoux et al., 2004) mostly located in the so-called ‘dust belt’, which extends throughout North Africa, Middle East, Central and South Asia to China (Prospero et al., 2002). Secondary sources are placed in South and North America, Namibia and Australia. These emissions exert a considerable influence on the climate, biogeochemistry and air quality of our planet (IPCC, 2007; Monks et al., 2009). The following four topics are of major interest today (Fig. 1):

- Because of its ability to scatter and absorb radiation (Redmond et al., 2010), dust interacts directly with incoming solar radiation. Scattering is influenced by dust differentiated features (with respect to other atmospheric aerosols; Reid et al., 2003b; Mishra et al., 2008): shape (sharp-edged instead of spheroids), coarse nature, desert-to-desert variable mineralogy, and change in mineralogy with particle size (e.g. small clays vs. large quartz). Absorption depends on the amount of iron oxides present in the dust (Alfaro et al., 2004). Moreover, dust particles are very frequently constituted by agglomerates, which hampers the determination of dust basic physical and optical properties (Reid et al., 2003a,b).
- Dust is involved in the formation and evolution of clouds. It is considered an effective cloud nucleation and ice nuclei (DeMott et al., 2003; Sassen et al., 2003; Herich et al., 2009). It may even affect the formation of tropical cyclones in the North Atlantic (Zhang et al., 2007). Dust deposition substantially increases the albedo of snow and ice surfaces (Aoki et al., 2006).
- About 25% of global dust emissions are deposited in oceans (Shao et al., 2011) and it is considered that the dissolved fraction of iron and phosphorus plays a key role in the primary production of the ocean and consequently in the atmospheric CO_2 absorption (Martin and Fitzwater, 1988; Falkowski et al., 1998; Mills et al., 2004).
- Poor urban air quality due to the arrival of dust from distant deserts frequently occur in Southern Europe (Rodríguez et al., 2001; Escudero et al., 2007; Mitsakou et al., 2008; Querol et al., 2009a,b), United States (Perry et al., 1997; Prospero, 1999; Prospero et al., 2001), Asia (Chan et al., 2008; Lee et al., 2010), Australia (McGowan and Clark, 2008) and New Zealand (Marx et al., 2005; Cowie et al., 2010). During dust events an increase in mortality (Pérez et al., 2008; Jiménez et al., 2010; Sajani et al., 2010) and in respiratory (Cowie et al., 2010) and cardiovascular diseases (Chan et al., 2008; Middleton et al., 2008) has been observed. Transport of pathogens on dust surface may have an adverse effect on public health (Griffin et al., 2001).

The physicochemical properties of the ‘aerosol (airborne) dust’ may differ from those of the original soil dust (before emission) or freshly emitted dust (Fig. 1). At least three processes are involved:

- Mixing with other chemical species. In the air, dust mixes with pollutants. In Asia, inner continental dust is transported eastward, where it encounters industrial pollutants before being advected to the Pacific Ocean (Perry et al., 1999; Arimoto et al., 2004). In Africa, pollutants emitted in Northern industrial areas are transported to the inner Sahara, where it mixes with dust before being exported to the Atlantic Ocean (Rodríguez et al., 2011). During the Sahel dry season, dust is transported southward to areas where biomass burning occurs (Capes et al., 2008). When dust encounters gaseous and aerosol pollutants, several interactions may occur. Reactions of sulfuric and nitric acids results in the coating of dust particles (Dall'Osto et al., 2010). Dust may absorb smaller ammonium-sulfate or soot particles onto their surface (Alastuey et al., 2005).
- Mixing state and homogenization. When dust encounters other aerosol population (e.g. of industrial origin, including ammonium-sulfate), they originally present an ‘external mixing’ state (different particles have different chemical composition). Over time, these populations tend to evolve to a more ‘internal mixing’ degree, in which most of particles tend to have a similar composition as a function of their size (Raes et al., 2000). Condensation of gases onto dust and in-cloud dust processing also contribute to homogenize the aerosol dust population (Raes et al., 2000; Baker and Croot, 2010).
- Size distribution. The preferential deposition of larger particles during atmospheric transport modifies the original size distribution, enhancing the relative contribution of smaller dust particles (Maring et al., 2003). This may also influence on the aerosol dust mineralogy (e.g. small clays versus large quartz; Glaccum and Prospero, 1980).

The so-called ‘dust processing’ or ‘dust aging’ modifies the properties of the surface of ‘aerosol dust’ with respect to the original ‘soil dust’. This has some important implications. Bauer et al. (2007) found that heterogeneous dust-sulfate–nitrate chemistry and their mixing state (external vs internal mixing) influences the radiative forcing of dusty air masses. Dust processing with acids enhances iron solubility, and hence its bioavailability when deposited in oceans (Baker and Croot, 2010). Herich et al. (2009) found that pure clays are less hygroscopic than natural desert dust, and that the absorption of soluble compounds onto dust surface may enhance dust ability to act as Cloud Condensation and Ice nucleii. Thus, measurements of other none-dust components present

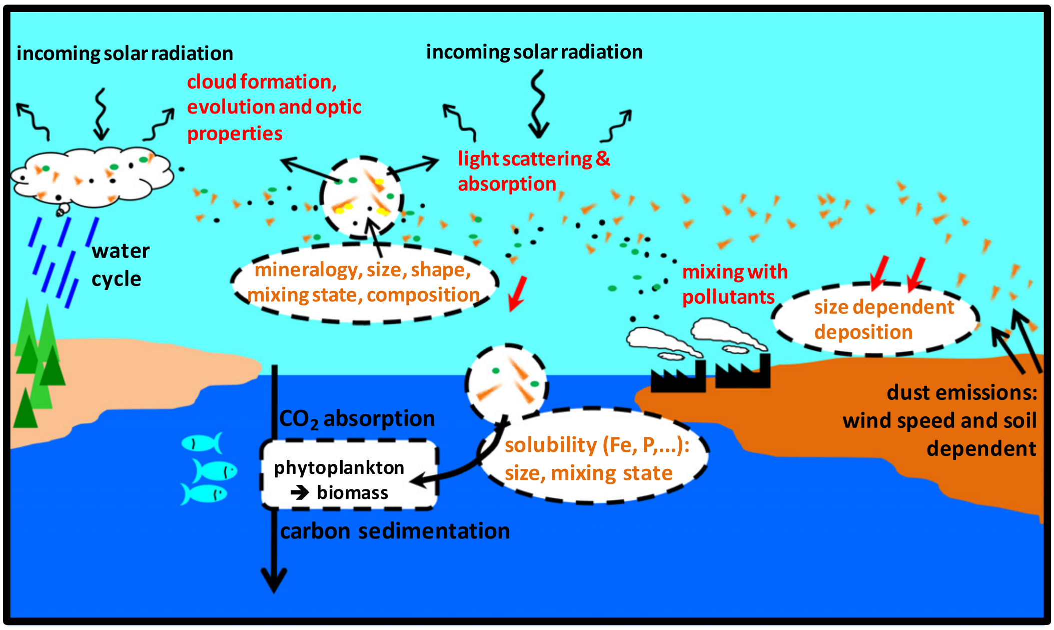

Fig. 1. Schematic illustration how microphysical and chemical properties of aerosol dust particles (usually measured by in situ techniques) participate in processes affecting climate.

in the dusty air mass needs to be included in the aerosol dust observations programs.

The increasing number of review articles on dust testifies to the upsurge of interest in the atmospheric sciences (Monks et al., 2009). Kohfeld and Tegen (2007) reviewed the past to present evolution of dust and introduced the concept of the dust cycle. Mahowald et al. (2009) and Baker and Croot (2010) studied dust iron deposition, whereas Maher et al. (2010) focused on the link between dust, climate and biogeochemistry. Shao et al. (2011) explored the relationship between dust, energy and carbon cycles, and their feedbacks. Schulz et al. (submitted for publication) highlighted the need for improvements in modeling and experimental data to better understand the impact of dust on ocean biogeochemistry.

In this article we focused on the in situ observations of aerosol dust. During the last two decades there has been an important increase in the capacities and in the number of models (Nickovic et al., 2001; Ginoux et al., 2004; Huneeus et al., 2010), satellite (Aerosol Optical Depth/AVHRR, Husar et al., 1997; UV - Aerosol Index, Herman et al., 1997; SeaWiFS, Borde and Verdebout, 2003; MODIS, Kaufman et al., 2005; CALIPSO, Liu et al., 2008) and land based remote sensing networks (AERONET, Holben et al., 1998; EARLINET, Müller et al., 2003) for atmospheric dust research. These tools have improved our understanding of how dust is transported and distributed throughout the atmosphere. However, many of the dust properties that influence Earth systems must be measured by in situ techniques. Most of these in situ measurements have been performed in short periods or campaigns (weeks-to-months) whereas long term (multi-decadal) monitoring of aerosol dust properties has been conducted only at a few sites. These long term measurements are crucial for a number of reasons. For example, the in situ aerosol dust observations performed by J.M. Prospero for more than 40 years in the Caribbean have played a key role in identifying the processes affecting multi-decadal evolution of dust emissions and transport (Prospero and Lamb, 2003; Chiapello et al., 2005), assessing the consistency of remote sensing data (Chiapello et al., 1999; Smirnov et al., 2000) and model validations (Ginoux et al., 2004; Huneeus et al., 2010).

In this paper we review the programs and techniques that are being or have been used for in situ characterization of aerosol dust. Accuracy, sources of errors and features of dust measurements are reviewed. Issues that, in our opinion, should be considered are also discussed.

2. Long term programs of in situ dust measurements

We review the programs which have focused on long term monitoring of aerosol dust by in situ techniques for at least 4 years (Table 1 and Fig. 2). Bulk chemical composition is the most commonly used technique for identifying and quantifying the aerosol dust concentrations. In the Atlantic, dust observation started in 1965 at Barbados, in 1972 at Miami and in 1987 at Izaña. In the Pacific, a number of sites were active during the 1980s and 1990s, and some even until 2000s (Midway and America Samoa). In the Mediterranean, at least two programs are active since the early 2000s (Montseny and Finokalia). In some of these sites, complementary aerosol measurements such as size distribution, optical properties or chemical composition of none-dust species are also performed (Table 1).

Below we describe the techniques that are widely used for in situ aerosol dust characterization. Techniques that allow us to measure dust mixing degree and other none-dust aerosols present in the dusty air mass are also described. Some online techniques enable us to obtain continuous records for many years (e.g. number size distribution, optical properties or bulk aerosol concentration). Other techniques require the collection of samples on filter for further offline analysis in laboratory (e.g. bulk chemistry or isotopic characterization). Some recently developed techniques are not still designed for continuous long term use (e.g. mass spectrometers, or automatic elemental composition analysis); nevertheless, they could be incorporated into long term programs for performing measurements in specific periods every year (e.g. dust season).

In addition to the techniques used in the programs listed in Table 1, dust measurements performed in field campaigns (usually lasting from weeks-to-months) were also considered: PRIDE in the Caribbean (Reid et al., 2003a,b; Journal of Geophysical Research special issue, vol. 108), SHADE in Cape Verde (Formenti et al., 2003), MINATROC in the Mediterranean and the Canary Islands

Table 1
Observatories where long term dust characterization has been performed during at least 5 years. Elements: elemental composition. Starting date is highlighted in black. See acronyms list.

Observatory	Parameter	Size fraction	Period	Parameter	Technique	Reference
Atlantic Ocean						
Barbados, BAR 13.17°N,59.43°W	Bulk chemistry	Total	1965-on going 48 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: Ash, INAA, IC, FAA, AC	Prospero and Lamb (2003)
Miami, MIA 25.75°N,80.25°W	Bulk chemistry	Total	1972-on going 39 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: Ash, INAA, IC, FAA, AC	Prospero (1999)
Izaña, IZO 28.30°N,16.50°W	Bulk chemistry		1987-on going26 years			
		Total	1987–1999	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: Ash, INAA, IC, FAA, AC	Prospero et al. (1995)
		Total	2002-on going	Elements, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), NH_4^{+}, (OC, EC)	Filter: (ICP-AES, ICP-MS), IC, ISE, TOR	Rodríguez et al. (2011)
		<10 μm	2005-on going	“	“	“
		<2.5 μm	2002-on going	“	“	“
		<1 μm	2009-on going	“	“	
	Size distribution	10–400 nm	2008-on going	dN/dlogD	SMPS	Rodríguez et al. (2009)
		0.5–20 μm	2007-on going	dN/dlogD	APSTOFS	Rodríguez et al. (2011)
	optical properties	<10 μm	2008-on going	Total and back scattering (3λ	Integrating nephelometer	Andrews et al. (2010)
		<10 μm	2007-on going	Absorption (1λ)	Absorption photometry	Andrews et al. (2010)
Bermudas, BER 32.27°N, 64.87°W	Bulk chemistry	Total	1988-1998 11 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: Ash, INAA, IC, FAA, AC	Prospero (personal communication)
Mace Head, MHD 53.32°N, 9.85°W	Bulk chemistry	Total	1988-1994 7 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: Ash, INAA, IC, FAA, AC	Prospero (personal communication)
Cape Verde, CVO 16.45°N,22.57°W	Bulk chemistry	Total	1991-1994 4 years	Major elements (Si, Al, Ca, K)	Filter: XRF	Chiapello et al. (1995, 1997a)
Kumasi, KUM 6.40°N,1.34°W	Size distribution	0.5–25 μm	1996-2005 10 years	Number size distribution	OPC	Sunnu et al. (2008)
Mediterranean						
Montseny, MSY 41.76°N, 2.58°E	Bulk chemistry		2003-on going 10 years			
		<10 μm	2003-on going	Elements, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), NH_4^{+}, (OC, EC)	Filter: (ICP-AES, ICP-MS), IC, ISE, TOR	Querol et al. (2009a,b)
		<2.5 μm	2002-on going	“	“	“
		<1 μm	2009-on going	“	“	“
	Size distribution	10–800 nm	2010-on going	dN/dlogD	SMPS	
	Optical properties	<10 μm	2008-on going	Total and back scattering (3λ	Integrating nephelometer	Pandolfi et al. (2011)
		<10 μm	2009-on going	Absorption (1λ)	Absorption photometry	“
Finokalia, FKL 35.20°N,25.40°E	Bulk chemistry		2004-on going 9 years		Filter: ICP-MS, (IC), TOR	Koulouri et al. (2008)
		<10 μm	2004-on going	Elements, ($SO_4^{=}$, NO_3^{-}, Cl^{-}, NH_4^{+}, K^{+}, Mg^{+2}, Ca^{+2}), (OC, EC)		
		<1.3 μm	2004-on going	“	“	“
	Size distribution	18–800 nm	2004-on going	dN/dlogD	SMPS	Kalivitis et al. (2008)
	optical properties	<10 μm	2001-on going	Total and back scattering (1λ	Nephelometer	Kalivitis et al. (2011)
		<10 μm	2004-on going	Absorption (3λ)	Absorption photometry	“
Pacific Ocean						
Mauna Loa, MLO 19.54°N,155.58°W	Bulk chemistry		1979-1997 19 years			
		Total	1979–1991	Elements	Filter: INAA	Holmes and Zoller (1996)
		<2.5 μm	1989–1997	Elements	Filter: PIXE,XRF,PESA	Perry et al. (1999)
	Optical properties	<10 μm	1974-on going	Total and back scattering (3λ	Integrating nephelometer	GAWSYS, Andrews et al. (2010)
		<10 & < 1 μm	1990-on going	Absorption (3λ)	Absorption photometry	GAWSYS, Andrews et al. (2010)
Midway, MID 28.22°N,177.35°W	Bulk chemistry	Total	1981-2001 21 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero et al. (2003)
Oahu Hawaii, OAH 21.33°N,157.70°W	Bulk chemistry	Total	1981-1995 15 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Enewetak Atoll, ENE 11.33°N,162.33°E	Bulk chemistry	Total	1981-1987 15 years	Bulk dust, Al, ($SO_4^{=}$, NO_3^{-}, Cl^{-}), Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)

Table 1 (*continued*)

Observatory	Parameter	Size fraction	Period	Parameter	Technique	Reference
Fanning Island, FAN 3.92°N, 159.33°W	Bulk chemistry	Total	1981-1986 6 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
A. Samoa, SMO 14.25°S,170.58°W	Bulk chemistry	Total	1983-on going 29 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Norfolk Island, NOR 29.08°S,167.98°E	Bulk chemistry	Total	1983-1997 15 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Cook Islands, COO 21.25°S,159.75°W	Bulk chemistry	Total	1983– 199412 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
NAURU, NAU 0.53°S,166.95°E	Bulk chemistry	Total	1983-1988 6 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}	Filter: INAA, IC	Prospero (personal communication)
TUVALU, TUV 8.50°S,179.20°W	Bulk chemistry	Total	1983-1987 5 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}	Filter: INAA, IC	Prospero (personal communication)
Cheju, CHE 33.52°N,126.48°E	Bulk chemistry	Total	1991-1995 5 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Hedo, HDO 26.92°N,128.95°E	Bulk chemistry	Total	1991-1994 4 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Indian Ocean						
CAPE GRIM, CGO 40.68°S,144.68°E	Bulk chemistry	Total	1983-1996 14 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Antartida						
Mawson, MAW 64.60°S,62.50°E	Bulk chemistry	Total	1987-1995 9 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)
Palmer Station, PAL 64.77°S,64.05°W	Bulk chemistry	Total	1990-1996 7 years	Bulk dust, Al, $(SO_4^{=}, NO_3^{-}, Cl^{-})$, Na^{+}, NH_4^{+}	Filter: INAA, IC	Prospero (personal communication)

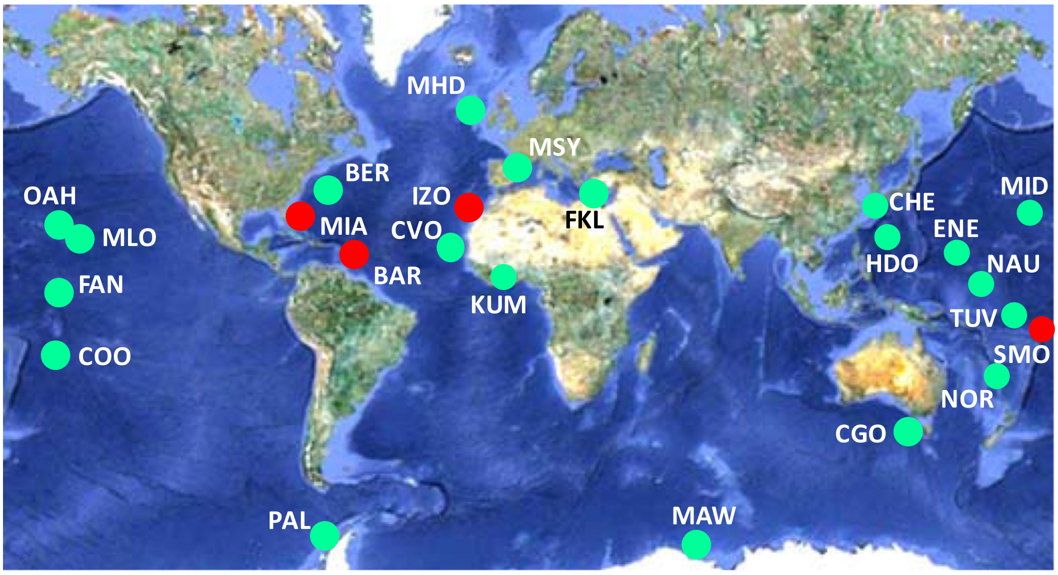

Fig. 2. Location of the atmospheric remote/background observatories where long term in situ measurements of aerosol dust have been performed during at least years (Table 1). Red circle: sites active during at least the last two decades.

(Putaud et al., 2004; Van Dingenen et al., 2005; Alastuey et al., 2005; Atmospheric Chemistry and Physics Discussion special issue MINATROC), SAMUM (Kandler et al., 2007; Tellus B special issues 61B and 63B) and AMMA in Niger (Formenti et al., 2008; Journal of Geophysical Research special issue, vol. 113).

3. Techniques for in situ characterization of dust

3.1. Size distribution

Dust size distribution has been measured with instruments that use different principles of measurement, i.e. inertial impaction, time of flight and intensity of light scattered. Measurement of size distribution of coarse particles (>1 µm) plays a key role in dust studies. However, fine (0.1–1 µm) and ultrafine (<0.1 µm) particles are also of interest. The use of equivalent diameters is sometimes required to merge data collected with different instruments (see details in Shen et al., 2002; Hand and Kreidenweis, 2002; Khlystov et al., 2004).

3.1.1. Cascade impactors

Cascade impactors allow the collection of dust samples segregated in ranges of aerodynamic diameter using the inertial impaction as the principle of operation. Particle size may range from somewhat less than 0.05 μm to more than 30 μm.

The aerodynamic diameter of a given particle is defined as the diameter of a spherical particle with density 1 g/cm^3 that settles at the same velocity as the given particle. Settling velocity, lung deposition, airborne movement and impaction depend on the aerodynamic diameter. Because its density is >1 g/cm^3, dust particles have an aerodynamic diameter larger than a geometric diameter (Hinds, 1982):

$$D_{\mathrm{p,a}} = D_{\mathrm{p}}\sqrt{\frac{Cs(D_{\mathrm{p}})\cdot\rho_{\mathrm{p}}}{Cs(D_{\mathrm{p,a}})\cdot\rho_{\mathrm{o}}}} \quad (1)$$

where, $D_{\mathrm{p,a}}$ is the aerodynamic diameter, D_{p} is the geometric diameter, ρ_{p} particle density, ρ_{o} = 1 g/cm^3 and Cs Cunningham slip correction factor.

Particle selection is performed by passing the sample through a nozzle. Particles coarser than a given size impact and remain adhered to a surface located in front of the nozzle. Smaller particles follow the streamlines of the airflow. A series of stages, with successive smaller cut points are used to obtain size distribution. Samples are analyzed offline for obtaining data on the size distribution of dust mass, chemical composition, mineralogy or mixing state.

Cascade impactors are regularly used for collecting dust samples in short field campaigns or for complementing (in some periods, e.g. dust season) the reference sampling techniques used in long term dust programs. They have been used for studying Asian dust (Mouri et al., 2003; Park et al., 2004), Australian dust (Radhi et al., 2010; 0.056–18 μm) and Saharan dust transported to the Mediterranean (Putaud et al., 2004; diameter range 0.05–10 μm), Sahel (Mallet et al., 2008; 0.04–10 μm) and to the Caribbean (Reid et al., 2003a; 0.07–5 μm and 0.4–22 μm). The hourly evolution of elemental composition of Saharan dust (0.1–12 μm) was studied with a Davis Rotating-drum Universal-size-cut Monitoring (DRUM; Reid et al., 2003b). Reid et al. (2003a) concluded that the dust mass size distributions collected with these devices tend to have a median mass diameter equal to 4.5 ± 1.3 μm.

Impactors are usually an offline instrument. However, the Electrical Low Pressure Impactor is able to provide online number size distributions in the range 0.01–10 μm (Keskinen et al., 1992). The particles are detected in the stages of the impactor by measuring the electric current rate across the sample after charging them with a unipolar corona charger. Because of the low total charge of coarse particles, the sensitivity of the impactor may be lower for coarse (>1 μm) than for fine (0.1–1 μm) and ultrafine (<0.1 μm) particles. After assessing their specific response to dust, these devices could be useful for long term monitoring (Sellegri et al., 2003; Held et al., 2008).

In cascade impactors, inaccuracies in particle size may occur because of airflow deviations, aspiration losses, non sharp or ideal cut size, particle bounce and particle blow off. High dust concentrations may result in the overload of the sampling substrates, and in subsequent particle bounce and/or blow off downstream. Intercomparisons between cascade impactors are scarce (e.g. Howell et al., 1998; Reid et al., 2003a), and standards for multistage impactors for research networks do not exist. Reid et al. (2003a) suggested that the breaking of dust agglomerates (with usual sizes 5–40 μm) in the upper stage of the impactors could result in a cascade of smaller dust particles downstream and in a consequent underestimation and overestimation of large and small particles, respectively.

3.1.2. Aerodynamic particle size – Time of flight spectrometers

An Aerodynamic Particle Size – Time Of Flight Spectrometer (APSTOFS) measures the number concentration and size of particles by measuring the time required by each particle to flight between two points in an accelerating airflow (Wilson and Liu, 1980). Size may range from 0.5 to 30 μm aerodynamic diameter (Baron et al., 2001).

These measurements are being performed at least in the long term dust monitoring programs of the Izaña Observatory (Table 1). Moreover, APSTOFSs have been used in Saharan dust field campaigns in Morocco (Kandler et al., 2009), Cape Verde (Kandler et al., 2011) and the North Atlantic (Maring et al., 2000; Reid et al., 2003a; Rodríguez et al., 2011).

In an APSTOFS, particles are accelerated through a nozzle. This acceleration is produced by applying sheath airflow to the aerosol flow. At the exit of this nozzle, small particles tend to follow the air motion, whereas large particles lag behind, causing an increase in the relative velocity between air and particles. This relative velocity increases with size and density of the particles and consequently is related to the aerodynamic diameter. Velocity is determined by measuring the time of flight of particles between two laser beams located at the exit of the nozzle. Time of flight spectrum is converted to aerodynamic size spectrum using a calibration curve previously obtained typically with monodisperse PLS (density 1.05 g/cm^3).

Several models of the most commonly used commercial instrument have been available for the last three decades: Aerodynamic Particle Sizer (APS) of TSI Inc (Shoreview, Minnesota, US), models 33, 3300, 3310, 3320 and 3321. Issues affecting accuracy, such as 'coincidence of particles in the sensing zone (coincidence error)', 'detection of a particle by only one sensor (phantom particles)', 'artificial coarse particles due to re-circulation in the sensing zone ('anomalous particles') have been partially or totally solved in the version 3321 (Heitbrink and Baron, 1991, 1992; Armendariz and Leith, 2002; Stein et al., 2002; Peters and Leith, 2003; Volckens and Peters, 2005).

Sizing accuracy has been measured for different models. Sizing errors due to deformation of droplets during the acceleration have been detected (Armendariz and Leith, 2002; Baron, 1986; Bartley et al., 2000; Chen et al., 1990; Kinney and Pui, 1995; Marshall et al., 1990; Peters and Leith, 2003; Tsai et al., 2004). The model 3321 has been characterized using monodisperse solid PLS with the following observations:

- a counting efficiency of 85% at 0.8 μm, 99% at 3.0 μm, 99% at 5.1 μm and 90% at 9.4 μm (Volckens and Peters, 2005).
- a sizing accuracy of 2 and 3% when measuring spheres of 0.65 μm and 0.96 μm diameter, respectively (Peters and Leith, 2003).
- a 15% instrument-to-instrument variability when sizing 1 μm spheres (Volckens and Peters, 2005).

However, to our knowledge, no sizing accuracy assessment has been performed with monodisperse mineral dust probably because of the difficulties in obtaining high concentrations of this material. Peters et al. (2006) suggested that breaking of >10 μm dust agglomerates may occur in the inner nozzle. Given that particles flow at super-Stokian velocities through these instruments, Marshall et al. (1991) suggested that APSTOFSs could undersize irregular shaped dust particles because of the deviation of the actual from the calibration velocities (e.g. ~25% undersizing for particles with shape factors of 1.2). Reid et al. (2003a) suggested that this could have an influence when comparing data collected with an APSTOFS and with cascade impactors. When using these instruments for long term monitoring of dust we should consider the following: (1) inaccuracies in flow rate will result in biases in determining size and concentrations of particles, and (2) high dust concentrations (e.g. during several dust storms) may result in 'coincidence errors'. At 10^3 cm^{-3} coincidence errors are ~5% for 0.5 μm and ~10% for 10 μm solid particles in some models.

APSTOFS data can be used for deriving mass size distributions:

$$\frac{dM}{d\log D_{p,a}} = \rho_p \frac{\pi}{6} D^3_{p,ve} \frac{dN}{d\log D_{p,a}} \quad (2)$$

$$D_{p,ve} = D_{p,a}\sqrt{\chi \frac{Cs(D_{p,a}) \cdot \rho_o}{Cs(D_{p,a}) \cdot \rho_p}} \quad (3)$$

where, $D_{p,a}$ is the aerodynamic diameter, $D_{p,ve}$ volume equivalent diameter, ρ_o = 1 g/cm^3, Cs Cunningham slip correction factor, χ dynamic shape factor (values 1.2–1.8 for dust). Density of dust particles, ρ_p, is a weighted average of density of dust components, e.g. some irregular shaped minerals: silicon oxide (2.1–2.3 g·cm^{-3}), illite/muscovite (2.7–3.1 g·cm^{-3}), montmorillonite (2.2–2.7 g·cm^{-3}) and quartz (2.65 g cm^{-3}). Because of the third moment dependence ($D_p{}^3$), small errors in the sizing or counting of coarse particles result in large errors when calculating the calculated mass concentrations.

3.1.3. Optical particle counters

Optical Particle Counters (OPCs) determine the number concentration and size of the particles by measuring the amount of light scattered by each individual particle when this is illuminated by a focused beam of light (Gebhart, 2001). A fraction of such scattered light is directed to a detector and converted to electrical pulses. Number concentration is determined from the count rate of pulses, whereas size of particles is calculated from the height or area of each pulse. Size may range between 0.1 and 100 μm.

These devices have been used in the long term dust program of Kumasi–Sahel (Table 1) and in field campaigns in the Caribbean (Reid et al., 2003b), the Mediterranean (Van Dingenen et al., 2005), Sahel (Rajot et al., 2008), Morocco (Petzold et al., 2009; Weinzierl et al., 2009) and Asia (Murayama et al., 2001; Sugimoto et al., 2003; Kim et al., 2004; Park and Kim, 2006).

The disadvantage of using OPCs for atmospheric aerosol measurements is that the key particle properties necessary for determining their size from pulse height are unknown: refractive index and shape. Because these instruments are regularly calibrated using monodisperse PLS, they determine the optical equivalent diameter when used for atmospheric measurements. Their accuracy decreases when increasing the differences between the calibration spheres and atmospheric aerosols. In general, pulse height does not show a monotonic increase with particle diameter (Gerbhart et al., 1976). There is a wide spectrum of commercially available OPCs with a variety of designs, the Passive Cavity Aerosol Spectrometer Probe – PCASP (Droplet Measurement Technology) and the Forward Scattering Spectrometer Probe – FSSP (Particle Measuring Systems) being the most commonly used in dust studies.

Peters et al. (2006) observed that the particle sizing with commercial OPC and APSTOFS showed differences within the range 10–15% when measuring PLS and differences within the range 40–45% when measuring Arizona mineral dust. Friehmelt and Heidenreicht (1999) demonstrated that the irregular shape of quartz dust particles enhanced the light scattering when compared with theoretical calculations based on spherical particles. These authors proposed a method for calibrating the OPCs with APSTOFSs. Similarly, Reid et al. (2003b) concluded that some OPCs oversized the Saharan dust particles transported to the Caribbean by a factor >2 when compared with data collected with APSTOFS, cascade impactor and a bulk aerosol mass monitors. OPC calibrations for aerodynamic diameter determinations have also been proposed using pre-selectors such as impactors (Marple and Rubow, 1976) and cyclones (Büttner, 1990).

Some potential sources of inaccuracies should be considered for long term monitoring of dust with these instruments: (1) Inaccuracies in the airflow will affect the number concentration, but not the size of particles (in contrast to APSTOFS), and (2) Coincidence of two or more particles in the illuminating sensing volume may result in the detection of false coarser particles. These 'coincidence errors' do not occur below specific concentrations which are usually provided by manufacturer (e.g. 2·10^3 cm^{-3} in some OPCs).

3.1.4. Electrical mobility analysers

Electrical mobility analysers (EMA) are normally used to measure the number size distribution of particles within the range 3 nanometres (nm) to 1 micron (1 μm). The fraction of dust in the fine mode (0.1–1 μm) is low. Measurements with these instruments allow the identification of other aerosols externally mixed with dust. Detailed descriptions of EMAs and other devices for measuring fine and ultrafine particles are given by McMurry (2000), Morawska et al. (2009) and Wiedensohler et al. (2012).

In EMAs, particles are exposed to a bipolar cloud of ions which gives a Boltzman charge equilibrium to the sample. The aerosol population is then conveyed to a Differential Mobility Analyzer (DMA) where particles of a given size are selected and transferred to a detector by applying a given voltage. The complete particle size distribution is obtained by scanning the voltage applied to the DMA. A Condensation Particle Counter is the most commonly used detector (Hermann et al., 2007). Wiedensohler et al. (2012) proposed a harmonization of technical standards to facilitate high quality long term monitoring of particle number size distributions from 20 to 200 nm with an accuracy of 10%. This harmonization has already been performed in a set of background/rural sites in Europe (Asmi et al., 2011).

Measurements of number size distributions in the ultrafine and fine ranges have been performed at a few sites where long term dust monitoring is undertaken (Table 1). During ACE-2 campaign, Raes et al. (1997) identified the presence of ultrafine particles externally mixed with dust in the Saharan Air Layer. The bimodal size distribution of the Saharan Air Layer particles have been attributed to the presence of coarse dust and fine and ultrafine ammonium-sulfate linked to North African industrial emissions (Rodríguez et al., 2011). Measurements with OPCs, APSTOFSs and EMAs also enabled to identify dust and pollutant mixings in Morocco (Müller et al., 2009c; Kandler et al., 2009; Weinzierl et al., 2009).

3.2. Mass concentration

3.2.1. Bulk aerosol mass concentrations

Measurements of bulk mass concentrations of particles smaller than given diameter (TSP, PM$_{10}$, PM$_{2.5}$ or PM$_1$) do not allow us to perform an exclusive characterization of dust. However, these measurements are useful for complementing dust measurements and chemical mass closure (dust, sea salt, biomass burning or urban/industrial compounds). At atmospheric observatories that usually record a low aerosol background and it is known that high aerosol mass events are caused by dust episodes, bulk aerosol mass measurements represent a relatively cheap approximate method for long term dust observation.

3.2.1.1. Gravimetry of sampling filters. This is considered a reference method for air quality and some research networks. It is used in the observatories where long term dust chemical characterization is performed (Table 1). The ambient air is passed through a filter, where particles are collected. Filters are weighted before and after sampling at controlled temperature and relative humidity. Mass concentrations are determined by dividing the increase in the filter mass (due to sample collection) by the volume of sampled air.

In a filter, the particles <0.1 μm are collected by diffusion and those >0.5 μm by impaction and interception. Particles in the 0.1–0.5 μm range are the most penetrating (Lee and Mukund, 2001). Collection efficiencies may be >99%. The type (fibber, membrane, granular bed and nucleopore) and composition of the filter should be selected according to the chemical or mineralogical characterization to be performed. A review of filters for aerosol chemical characterization was performed by Chow (1995).

Reference gravimetric methods used in air quality networks facilitate data comparability between different observatories (e.g. European EN-12341 for PM_{10} and EN-14907 for $PM_{2.5}$, or their US equivalent). For example, EN-14907 describes procedures (flow rate precision, weighting and filter handle procedures, sampler features, etc.) that result in estimations of the aerosol mass concentrations with uncertainties of 8% with respect to the reference method for concentrations within the range 1–120 $\mu g/m^3$. Before weighting, filters are conditioned to a relative humidity (usually 30 or 50%) lower than that of the ambient air during sampling. This requirement is difficult to meet when sampling in very dry environments (e.g. Deserts or free troposphere, where $RH < 20\%$). Positive artifacts (absorption of vapors on the filter), negative artifacts (evaporation of semi-volatile compounds from the filter) or filter contamination during transport and handling may occur (Vechhi et al., 2009). These should not affect dust components but semi-volatile compounds mixed with dust. The collection of blank field filters is recommended.

3.2.1.2. Real time measurements. Three techniques are described here. Details and other available methods were discussed by Baltensperger et al. (2001). These techniques have been used in some long term programs (Table 1) and in field campaigns, such as AMMA in Sahel (Rajot et al., 2008), PRIDE in the Caribbean (Reid et al., 2003a) and in the laboratory (Alfaro et al., 2004). Air quality impairment due to dust transport has also been studied using data collected with these devices in air quality networks (e.g. Viana et al., 2002; Escudero et al., 2007; Lazaridis et al., 2008).

3.2.1.2.1. Tapered element oscillating microbalance (TEOM). The mass of the particles collected on a substrate that vibrates at constant amplitude is determined as a function of the decreasing frequency prompted by an increase in particle mass through time Patashnick and Rupprecht (1983).

$$m = k_o\left(\frac{1}{f_a^2} - \frac{1}{f_b^2}\right) \tag{4}$$

Where, m is the aerosol mass, K_o is the spring constant unique for each instrument and f_b and f_a are the oscillating frequencies before and after the sample collection. The sampling substrate is mounted on a tapered tube free to vibrate horizontally. Its lowest part is mounted on a rigid base. An optical system measures the oscillating frequency. The instrument has a detection limit <2 $\mu g/m^3$ for 24-h means, and a resolution of ±5 $\mu g/m^3$ for 10 min sampling. The only required calibration is the determination of the constant K_o. Its value is provided by the manufacturer and can be checked along time by adding a known mass to the tapered element. In order to measure the dry particle mass (at $RH < 40\%$), these devices heat the sample and this may evaporate the semi-volatile species potentially mixed with dust (Ayers et al., 1999; Salter and Parsons, 1999). Alternatively, diffusion driers may be used, but care should be taken with potential losses of sample (Eatough et al., 1999). In these devices, the sampling substrate should be changed manually. This is a drawback when measuring high dust concentrations which could result in overload or clogging (>10^5 $\mu g/m^3$ may occur close to dust sources; Kandler et al., 2009). Marticorena et al. (2010) used TEOMs for studying the temporal and spatial variability of dust in the Sahel region from 2006 to 2008.

3.2.1.2.2. Beta gauge. The number of beta particles transmitted across a filter decreases when increasing the sample load (Evans, 1955):

$$I = I_o e^{-\mu_\beta \cdot x} \tag{5}$$

where μ_β is the mass absorption coefficient for beta radiation, x mass thickness of the sample, I_o incident flux and I transmitted flux. Attenuation is caused by the scattering of beta particles by the atomic electrons (of the sample and filter) and is therefore dependent on the areal density. The mass absorption efficiency depends on the atomic number to atomic mass ratio (Z/A). These instruments are calibrated with a 'standard foil' whose composition (Z/A ratio) is close to that of atmospheric aerosols. Because the Z/A ratio for typical elements in the aerosol dust varies in a narrow range (0.47–0.50 for C, Si, Al, Ca, Fe, Mg, K, Cl, Na, N, O and S) the error produced by using a given number to mass atomic ratio is of about 10%. These instruments may detect down to 5–25 $\mu g/cm^2$ of material deposited on the filter (McMurry, 2000). Commercial instruments, with detection limits of 1 $\mu g/m^3$ (24-h average) and 4 $\mu g/m^3$ (1-h average), resolution of 0.1 $\mu g/m^3$ and precision ±2 $\mu g/m^3$ are available. In order to measure the dry particle mass ($RH < 40\%$), heating or drying systems may be required depending on the relative humidity of the ambient air. These devices perform the filter change automatically, and this is an advantage during events of high dust concentrations.

3.2.1.2.3. Bulk mass determination based on number size distribution measurements. Size distributions measured with OPCs, APS-TOFS and EMAs may be used to determined bulk volume and then mass concentrations of particles:

$$\frac{dV}{d\log D_p} = \frac{\pi}{6} D_p^3 \frac{dN}{d\log D_p} \tag{6}$$

$$\frac{dM}{d\log D_p} = \rho_p \frac{dV}{d\log D_p} \tag{7}$$

$$PMx = \sum\nolimits_{p=d_1}^{p=d_2} \frac{\Delta M}{\Delta \log D_p} \Delta \log D_p \tag{8}$$

where, D_p is the diameter, ρ_p particle density, d_1 and d_2 are the smallest and largest particle measured diameters and PM_x refer to TSP, PM_{10}, $PM_{2.5}$ or PM_1. Dp may refer to the equivalent diameter. The use of this technique with APSTOFS was described by Shen et al. (2002) and Peters (2006). Effective particle density may be obtained experimentally by performing some simultaneous measurements of mass concentrations in certain periods, e.g. PM_x sampling several days per month.

These three methods are suitable for long term measurements. Instrument maintenance includes calibration of airflow and sensors and checking of impactors.

3.2.1.3. Consistency of mass concentration data. Gravimetry of sampled filters, the only direct measurement of the aerosol mass, is considered as the reference method. Data obtained with other instruments should be referred to this standard. Mass concentrations determined with automatic instruments (TEOM, beta gauge or size spectrometers) may differ from those obtained with manual filter sampling for a number of reasons (e.g. heating temperature, volatilization of semi-volatile species, inlet losses, deliquescent point, etc.). These intercomparison exercises enables us to convert such data to 'gravimetric equivalent' (EU Guideline, 2002). Some commercial instruments have demonstrated their equivalence with gravimetric reference methods (e.g. EN-12341 for PM_{10} and EN-14907 for $PM_{2.5}$).

3.2.2. Bulk dust mass concentrations

The two most commonly used methods for estimating bulk dust concentrations are weighting of filter ash and tracer analysis.

3.2.2.1. Filter ash. This method has been used by J.M. Prospero and coworkers since the mid 1960s in Atlantic, Pacific and Indian sites (Table 1 and Fig. 2). Samples are collected in glass fiber filters at about 60 m^3/h flow rate. Filters are extracted with de-ionized water and the extracts are analyzed for major soluble inorganic ions: Na^+ by flame atomic absorption; Cl^-, NO_3^- and $SO_4^=$ by suppressed ion chromatography and NH_4^+ by automated colorimetry. Then, non sea salt sulfate is calculated using the $SO_4^=/Na^+$ ratio in bulk sea water (0.2517). The extracted filters are then placed in a muffle furnace for 14-h (overnight) at 500 °C. The ash residue weight divided by the sampled volume is considered the mineral dust concentration. Standard error is considered ±0.1 μg/m^3 for concentrations <1 μg/m^3, and about 10% for higher concentrations. This technique underestimates dust concentrations because of the loss of soluble minerals (carbonates, halides). The scatter plot of Al versus dust (analyzed with this technique) shows a slope of 10.4%, which is somewhat higher than the mean content of Al in soils (8%). A correction factor of 1.3 is therefore used (Arimoto et al., 1995). The longest records of dust concentrations have been obtained with this method: in Barbados since 1965, in Miami since 1974, in Midway from 1983 to 2001, in American Samoa since 1983 and in Izaña since 1987 and for 12 years (Table 1; Fig. 2).

3.2.2.2. Tracer analysis. Dust samples are also collected on a filter at a given flow rate. Then, the concentrations of soil tracers are determined by the techniques described below. Subsequently, the mean content of the selected tracers in soil is used to estimate bulk dust composition. In a first approach, a single tracer may be used, the most common being Si and Al, which account for 33% and 8% of mean soil composition, respectively (Bowen, 1966; Taylor and McLennan, 1985). Chiapello et al. (1995) used Si to estimate total dust concentrations in Cape Verde, whereas Al was employed by Kubilay et al. (2000) in the Eastern Mediterranean and by Arimoto et al. (1995) in the North Atlantic. In a second approach, several tracers may be used. Dust concentrations may be estimated assuming that Si, Al, Fe and Ti are present in their normal oxide forms as reported by Perry et al. (1999) for Asian dust at the Mauna Loa observatory. The relationship between tracers and their mean content in soil varies in accordance with dust particle size (Moreno et al., 2006).

3.3. Chemical characterization

Bulk chemical characterization is the most widely used technique for identifying and quantifying the presence of dust (Table 1). Variability in dust composition is often used to identify dust sources. Typical mineral elements are Si, Al, Ca, K, Na, Mg, Fe, K and P (as major elements) and Ti, V, Cr, Mn, Co, Ni, Cu, Zn, As, Sr, Cd, Sn, Sb and Pb as trace elements. The analysis of compounds other than dust is also of interest to assess the degree of mixing with pollutants. Thus, characterization may include compounds potentially linked to anthropogenic emissions, such as sulfate, nitrate, ammonium and organic and elemental carbon or some of the cited elements (As, Cd, Cu, Cr, Ni, Pb, V and Zn). Modern analytical instruments allow us to study the mixing state between dust and pollutants such as sulfate or nitrate.

3.3.1. Offline methods

Collection of samples on filter and subsequent chemical analysis is the most commonly used method for long term monitoring of dust composition (Table 1).

3.3.1.1. Filter media. All kinds of filters are used depending on the research purposes. Given the relatively low concentrations of some key elements in atmospheric aerosols, it is essential to use high grade filters with low contents of impurities for chemical analysis. Blank filters that correspond to the same lot as the filters sampled must be analyzed in the same batch in order to correct final concentrations. Selection of the filter will depend on the determinations to be carried out and on the type of sampler used (with different flow and inlet diameter).

Basically there are three main types of filter: cellulose fiber, glass and quartz fiber and membrane. The general characteristics, sampling and analysis utility are described elsewhere (e.g. Chow, 1995). Briefly, the following considerations have to be taken into account for dust measurements:

- For gravimetric determination of dust mass concentrations, the most adequate filters are those made of glass and quartz microfiber and membrane filters. Glass fiber filters are not suitable for chemical determinations given their high impurity content.
- For X-ray fluorescence (XRF) analysis, Teflon filters are highly recommended to reduce the XRF response of the filter media.
- When wet chemistry is used for the analysis of the bulk dust composition, quartz microfiber, cellulose fiber and membrane filters are commonly used.
- Cellulose fiber filters and membrane filters (such as cellulose, Teflon, polycarbonate) are usually made of carbon based materials. These materials are unsuitable for organic and elemental carbon determination by thermo-optical methods. Therefore quartz microfiber filters are recommended if the carbonaceous fraction is analyzed.

3.3.1.2. Sample treatment. There are different analytical methods for chemical characterization of aerosol dust samples collected on a filter. The technique used will determine the method employed for the treatment of the filter. Some techniques, such as X-ray Fluorescence (XRF), Proton Induced X-ray Emissions (PIXE) and Instrumental Neutron Activation Analysis (INAA), have a distinct advantage since these allow us to directly analyze the sample without any pre-treatment.

Other off-line techniques are based on the analysis of a liquid solution, and therefore the dust sample plus filter substrate must be dissolved. A key issue is the methodology used for the filter treatment. A large number of studies on air quality have not focused on dust, and consequently soft acidic digestion, acid or neutral leaching procedures have been applied to analyze major and trace elements, anions and cations. Thus, the US-EPA methods for metal extraction based on microwave digestion and hot acid extraction used mixtures of nitric and hydrochloric acids, thereby avoiding hydrofluoric acid (US-EPA, 1997). These methods enable us to dissolve most of the easily soluble compounds, such as the secondary inorganic aerosols, marine aerosols, hydrophilic organic compounds and a fraction of mineral compounds, but do not allow total-bulk dissolution of an important fraction of mineral dust, mainly silicate and aluminum–silicate species. Therefore, dust concentrations have been underestimated in many air quality studies.

A suitable method for dissolution of mineral dust should include the dissolution of aluminum silicates. Based on previous experience of the analysis of geological materials, Querol et al. (2001) devised a method for the bulk dissolution of the dust samples based on the combination of HNO_3 (for dissolution of carbonate, oxide species and most of organic fraction), HF (for dissolution of aluminum–silicates), and $HClO_4$ (for organics). This is a four step method which consists of:

[1] Addition of 2.5 ml HNO_3^- and 5 ml HF to a ¼ filter of a 15 cm diameter quartz microfiber filter in a 60 ml PFA reactor (75 PSI).
[2] Introduction of the closed reactors into a laboratory furnace at 90 °C for 6 h.

[3] After ambient cooling of the reactors, addition of 2.5 ml $HClO_4$ to the reactors, placing them on a hot plate at 250 °C until total dryness.

[4] Re-dissolution of the dry residue with 2.5 ml HNO_3, making up 50 ml volume with milli Q grade de-ionized water to obtain a 5% nitric acid solution.

The third step is of paramount importance given that HF in the solution may spoil the glass and metal parts of the analytical instruments. Furthermore, if Cl^- (from $HClO_4$) is present in the solution, a number of key trace elements (such as As) may undergo spectral interferences in the subsequent analysis. The above acid volumes are used for quartz microfiber filters. If other filters are used (such as Teflon and nitrate acetate cellulose) the amount of HF may be considerably reduced.

3.3.1.3. Analytical techniques. The 5% HNO_3^- solution obtained from the above digestion procedure may be used for subsequent analysis, including the combination of Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) without major matrix problems for interferences. This combination allows the determination of the bulk concentrations of more than 50 major and trace elements. ICP-AES is a suitable technique for analysis of major elements (Al, Ca, Fe, K, Mg, Na, P, and S) and some trace elements (Sr, Mn, Ti, V, among others). By contrast, ICP-MS is a powerful technique for precise and fast determination of concentrations of most trace elements (Li, Be, P, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Cd, Sn, Sb, Ba, La, Ce, Tl, Pb, Bi, Th, U, among others). ICP analysis is currently been used in the long term dust programs of Izaña, Montseny and Finokalia (Table 1). Moreover, it was used during dust campaigns in Barbados and Miami (Trapp et al., 2010) and the Saharan-Sahel dust corridor (Moreno et al., 2006).

Major disadvantages include the following: sample treatment is time consuming, manipulation of dangerous acids, and the impossibility of determining Si levels. The last disadvantage is due to the use of quartz filters and/or the volatilization of Si and B when evaporating HF on the hotplate. Si concentrations may be estimated from Al data, since the Si/Al mass ratio tends to show a relatively low variability. For example, the Si/Al ratio in the Saharan dust exported to the North Atlantic is within the range 1.8–2.5 depending on the dust source region (Caquineau et al., 1998).

Another fraction of the filter (i.e. ¼ of the filter) may be water leached for determination of concentrations of soluble anions and cations. EMEP (1996) provides reference methods for leaching fractions of the sampled filters for the subsequent analysis of SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, K^+, Ca^{2+}, and other ions. The solutions can be analyzed by means of Ion Chromatography, ICP-AES, ICP-MS, selective electrodes and colorimetry, among others.

Another fraction of the quartz filter (1 cm^2, approx.) may be employed for the determination of the organic carbon (OC) and elemental carbon (EC) contents using thermo-optical methods with US-NIOSH or EUSAAR2 (European) protocols among others (Chow et al., 2001; Cavalli et al., 2010).

Finally another fraction of the quartz filter may be used for the analysis of the organic material mixed with dust using organic chromatography or mass spectrometry (Van Drooge et al., 2010).

The use of high volume samplers, quartz filters and the above protocol allow us to determine around 60 aerosol components in a single sample, accounting for about 75% of the aerosol mass load, the remaining unaccounted mass being attributed to water molecules (the filters cannot be heated since semi-volatile matter may be lost). Another important advantage is the very low detection limits reached for a large number of elements.

The chemical characterization described here, which includes elemental composition by ICP-AES and ICP-MS, IC and selective electrodes for ions, and thermo optical analysis of OC and EC is currently being used for long term observations of dust at Izaña and Montseny (Alastuey et al., 2005; Pey et al., 2009; Rodríguez et al., 2011).

Uncertainties linked to these analytical techniques have been estimated (Escrig Vidal, 2009): 2–4% for most elements and ions determined with ICP and IC, respectively. These may reach 20% for Zr and Hf, 10–15% for F^- and NH_4^+ and 20% for other elements which may have high blank concentrations in the filter, such as Mo, Zn, Ba and Na. Errors in airflow are typically <5%. Thus, uncertainties in concentration are considered to be about 8% for most elements, 25% for the metals that are present in significant amounts in the blank filter, 30% for Zr and Hf, and 25% for F^- and NH_4^+.

XRF, PIXE or INAA analyses of dust samples collected on filter are also powerful analytical tools for dust studies, since major elements, such as Al, Ca, Fe, K, Mg, and Si are detected without serious problems with the above techniques (Chiapello et al., 1997b; Calzolai et al., 2008; Lucarelli et al., 2010; Alemón et al., 2004). Examples of specific applications of these analytical tools can be found in D'Alessandro et al. (2003), Richard et al. (2010) and Weckwerth (2010). These techniques have been widely used for long dust measurements (Table 1). An advantage over the method described above is that Si levels may be determined when using appropriate filters. The possible disadvantages are the relatively high detection limits for some elements, and the impossibility of analyzing a number of light elements.

Other analytical techniques are available. Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) coupled with Energy Dispersive X-ray analysis (EDX) allows individual particle characterization for size, morphology, chemical and mineral composition. This type of analysis: a) enables us to identify the size of each dust component; b) shows the composition of single particles; and c) may provide information about the source of aerosols (even for a small number of cases). These techniques have been used in PRIDE (Reid et al. (2003b), MINATROC (Alastuey et al., 2005) and SAMUM (Kandler et al., 2009) field campaigns.

Some impactors that allow sample collection with hourly resolution have been designed for specific analytical techniques. This is the case of DRUM and streaker impactors. Samples collected by the DRUM impactor in eight size stages within the range 0.1–12 μm are analyzed with synchrotron XRF (Reid et al., 2003b; Bukowiecki et al., 2005; Richard et al., 2010). In the streaker, fine (<2.5 μm) and coarse (2.5–10 μm) particles are collected on Nuclepore (0.4 μm pores) and Kapton foil, respectively. Analyses of the filter (fine particles) and impactor deposits (coarse particles) are carried out by PIXE in an external beam of a particle accelerator (Lucarelli et al., 2010; D'Alessandro et al., 2003).

3.3.1.4. Isotopic characterization. Isotope geochemical analyses may also be carried out using dust samples obtained with offline methods. Isotope patterns of specific elements are very good fingerprints of emission sources. The most common isotope analyses performed in aerosol samples includes stable and radiogenic isotopes.

Among the application of the stable isotopes studies we should highlight the source tracing studies performed using Sr, Nd, Sm, Pb, Zn, Cu, Fe, S, O and N isotopes to distinguish and quantify contributions from different anthropogenic and natural emission sources. A number of authors, e.g. Goldstein et al. (1984), Nakai et al. (1992), Nakano et al. (2005), Grousset and Biscaye (2005), Yang et al. (2009) and Lupker et al. (2010), have analyzed Sr, Nd, Sm and Pb stable isotopes in an effort to investigate source types and areas of aerosol dust. Stable isotopes of Pb, Hg, Cu, Zn and Fe (heavy stable isotopes) in atmospheric aerosols may be used to trace different pollution sources (Kylander et al., 2010; Majestic et al., 2009).

Levels of stable isotopes of S, N and O have been measured in atmospheric aerosols with the aim of investigating the source origin of sulfate, nitrate and ammonium present in atmospheric aerosols (McArdle and Liss, 1995; Querol et al., 2000; Yeatman et al., 2001; Bindeman et al., 2007). In the case of desert dust, this issue is of high climatic interest since sulfate and nitrate salts may change the hygroscopicity of dust, with consequent implications for altering the dust properties for cloud and ice nucleation. The origin of such salts is very varied, e.g. resuspension of evaporitic salts from sabkha-like deposits, interaction during air mass transport of dust and anthropogenic pollutants (such as SO_2, H_2SO_4, NO_2 or HNO_3), influence of sea salt sulfate, interaction of long range transport of dust with local anthropogenic pollutants or volcanic origin, among others. In the case of dust tracing and heavy stable isotopes the samples are analyzed by means of Thermal Ionization Mass Spectrometry (TIMS), Sector Field High Resolution Mass Spectrometry (HR-ICP-MS) or Multi Collector-Inductively Coupled plasma Mass Spectrometry (MC-ICP-MS). By contrast in the case of S, O and N, specific separation lines and mass spectrometers for stable isotopes (with overall reproducibility of isotope ratios of around 0.1 per mil) are required.

The analysis of activity levels of radionuclides usually focuses on the following issues:

(a) The evaluation of the radioactive spectra of the sample to determine the concentrations of Be, Cs, U, Th and other radionuclides to trace the contribution of specific radioactive sources or air mass origins. Thus, activity levels of various artificial radionuclides (^{90}Sr, ^{137}Cs, uranium, thorium and plutonium isotopes, ^{241}Am) have been measured to evaluate their contribution from African dust to several regions, such as France and the Canary Islands (Hernández et al., 2005; Massona et al., 2010) since the testing of nuclear weapons in the Sahara in the 1960s.

(b) ^{7}Be has a cosmogenic origin and it is considered a tracer for intrusions of air masses into the troposphere (Jasiulionis and Wershofen, 2005). The variability of levels of ^{7}Be in dust transported from northern Africa to the Canary Islands has been also evaluated (Prospero et al., 1995; Hernández et al., 2008). Radiometric measurements for ^{7}Be and the above radionuclides are usually performed by low-level gamma spectrometry, alpha-particle spectrometry, gas-proportional counting (GPC) or liquid scintillation spectrometry (LSS) depending on the radionuclides.

(c) Radiocarbon (^{14}C) analysis allows the distinction between modern and fossil sources of carbonaceous aerosols (Gustafsson et al., 2009). Separation of OC and EC fractions can be done before the ^{14}C analysis (Szidat et al., 2004). Organic carbon (OC), elemental carbon (EC) or total carbon (TC) is oxidized in a stream of pure oxygen; evolving CO_2 is trapped cryogenically and sealed in ampoules for subsequent ^{14}C measurement with an Accelerator Mass Spectrometer.

3.3.2. Online methods

Given the high cost and duration of offline analytical methods, new instruments for online chemical characterization of aerosols have been developed during the last decade. These techniques are mostly focused on secondary and carbonaceous species and are still limited for bulk dust characterization. New online analytical techniques are not intended for long term monitoring. However, they may be useful for performing characterization during some periods (e.g. dust season) and then may be used to complement long term observations programs based on offline methods.

3.3.2.1. Online XRF analysis. Semi-real time XRF analyzers based on the particle collection on a filter tape during a given time period, and the subsequent analysis by XRF are currently available in the market (e.g. Xact manufactured by Cooper Environmental Services LLC, Portland, Oregon, US). Yadav et al. (2010) used this technique for determining the content of metals such as V, As, Ba, Cd, Cr, Mn, Ni, Se, Ti, Zn, Ca, Fe and K in atmospheric particles with a relative uncertainty better than 10%. This technique has limitations for determining some key dust elements such as Si, Al or P (Petterson et al., 2010). These analyses may be of interest when dust is mixed with trace anthropogenic metals as observed by Trapp et al. (2010).

3.3.2.2. Online analysis of aerosol ions and gaseous precursors. Semi-real time analyzers of soluble ions and specific aerosol gaseous precursors have been developed. These instruments are based on denuder and systems to trap specific gaseous pollutants and aerosol nebulisation into a water leaching system to dissolve aerosol components. The leachate is then conveyed towards the analyzer. The components trapped in the denuder are also water leached and conducted to the analyzer(s). The detection and quantification of soluble ions from aerosols and denuders are carried out by means of Ion Chromatography (IC). The complete aerosol system may be operated remotely and may work as a semi-real time aerosol monitoring instrument. It may be used for time-resolved direct measurements of anion particulate nitrate, sulfate, nitrite, phosphate and chloride as well as for ammonium, sodium, calcium, potassium and magnesium if an additional detector is available (ten Brink et al., 2007; Weber et al., 2001). If denuders are available time-resolved direct measurements of gaseous precursors such as NH_3, HCl, NO_2, HNO_3, HONO and SO_2 may be carried out. Commercial versions of this technique are available, e.g. Ambient Ion Monitor (AIM, URG), Monitor for AeRosoles and Gases in Air (MARGA, Applikon analytical) and Particle into Liquid Sampler with IC (PILS-IC, Metrohm BMI).

3.3.2.3. Semi-real time analyzers based on thermal decomposition processes. Instruments based on the accumulation of a given amount of sample during a variable sampling period, in some cases preceeded by a denuder system to trap gaseous pollutants, and followed by different thermal decomposition processes and detector systems, have been developed. These devices are useful for monitoring non dust components mixed with dust. Three main types of such instruments can be distinguished:

(a) Semi-real time sulfate analyzers. A denuder trapps SO_2 before a filter medium accumulates aerosol sample for a given period. Subsequently, the filter is exposed at high temperatures to volatilize sulfate into gaseous sulfur dioxide, whose levels are determined by a conventional SO_2 analyzer (Allen, 2006).

(b) Semi-real time nitrate analyzers. A denuder trapps NO_2 before a filter medium accumulates aerosol sample for a given period. Subsequently, the filter is exposed to high temperatures to volatilize nitrate into gaseous nitrogen oxide, whose levels are determined by a conventional NO_2 analyzer (Gómez-Moreno et al., 2007).

(c) Semi-real time OC and EC analyzers. A filter medium accumulates aerosol sample for a given period. Subsequently, the filter is exposed to sequential heating under different gaseous mixtures. This gives rise to the combustion of OC followed by the combustion of EC. Simultaneously with the measurement of EC and OC concentrations, the levels of the CO_2 resulting from these combustion stages are determined by a conventional CO_2 analyzer (Plaza et al., 2006). Correction for charring is performed in a thermo optical transmittance or reflectance scheme.

3.3.2.4. Aerosol mass spectrometer. The aerosol mass spectrometer (AMS) provides quantitative size and chemical composition in real-time for non-refractory submicron aerosol particles. Particles are focused on a beam, and non-refractory compounds are thermally evaporated at adjustable temperatures from 200 to 900 °C, and then detected by electron impact ionization quadrupole mass spectrometry (Jayne et al., 2000; Canagaratna et al., 2007). This system permits direct linear detection of sulfate, nitrate, ammonium, chloride and organic aerosol species. This instrument determines size distribution within the range 40–1000 nm, aerodynamic diameters being determined by a particle beam chopping technique (Jimenez et al., 2003). The instrument has a high time resolution, 1–10 s, data being reported in 1–5 min intervals.

Depending on the mass spectrometer detector used, there are several AMS versions: quadruple (Q-AMS), compact time of flight (C-ToF-AMS) and high resolution time of flight (HR-ToF-AMS) aerosol mass spectrometers. The detection limit will depend on the chemical species and on the mass spectrometer option. The ToF systems have increased sensitivity by around two orders of magnitude (Drewnick et al., 2005; DeCarlo et al., 2006). The HR-ToF (DeCarlo et al., 2006) improves the separation of components. These instruments may be coupled with additional modules such as black carbon (BC) detector or light scattering module, thermal denuder system or negative ion detection for ToF AMS. BC is the only refractory component that may be detected by the AMS. The principal advantage of the instrument is the size resolved chemical composition for submicron particles.

Recently, a smaller aerosol spectrometer, the Aerosol Chemical Speciation Monitor (ACSM, Ng et al., 2011), has been developed. This instrument, devised for routine monitoring, is based on the AMS system, with a similar principle of operation but without size distribution. In this case, particles are thermally vaporized at 600 °C and, as in the case of AMS, detected by impact ionization and mass spectrometer. The ACSM provides composition information for particulate ammonium, sulfate, nitrate, chloride and organic compounds for sub micrometer particles (40–1000 nm) but does not report aerosol size resolved data. Time resolution is also lower than for AMS, usually reporting data in 15–50 min intervals. This instrument requires a minimal maintenance and can be remotely controlled.

Because AMS does not allow the analysis of refractory particles, it is not suitable for the characterization of dust. However, it may prove very useful for studying pollutants externally mixed with dust, such as ammonium-sulfate, ammonium-nitrate, or organics.

3.3.2.5. Aerosol Time Of Flight Mass Spectrometer. The Aerosol Time Of Flight Mass Spectrometer (ATOFMS) instruments determine the chemical composition of individual particles as well as the size in real-time. The ATOFMS collects bipolar mass spectra of individual aerosol particles from 0.1 to 3 μm (Gard et al., 1997). Ambient aerosol is focused into a narrow particle beam in the above range of sizes. Using a 2-laser velocimeter, aerodynamic particle size is determined from particle velocity after acceleration into the vacuum. In addition, the light scattered by the particles is used to trigger a pulsed high power desorption and ionization laser (λ = 266 nm, about 1 mJ/pulse), which evaporates and ionizes the particle in the centre of the ion source of a bipolar reflectron ToF-MS. Thus, positive and negative ion spectrums of a single particle are obtained. The mass spectrum is qualitative in that the intensities of the mass spectral peaks are not directly proportional to the component mass but are dependent on the particle matrix, on the coupling between the laser and the particle and on the shot to shot variability of the laser. However, the ATOFMS can supply quantitative information on particle number as a function of composition, providing measurements of all the particle components (including OC, EC, sulfate, nitrate, dust and sea salt), and can also be used to assess the mixing state (chemical heterogeneity of single particles) of the aerosol (Gard et al., 1997).

This instrument has proved useful for studying the mixing state of dust with pollutants. Thus, the application of this technique has demonstrated that Saharan dust particles collected in Cape Verde contain internally mixed nitrate but no sulfate (Dall'Osto et al., 2010), whereas Saharan dust particles collected on the coast of Ireland (Dall'Osto et al., 2004) show a very high degree of internal mixture with secondary species, including nitrate, sulfate and methanesulphonate.

The main advantage of ATOFMS is the size resolved chemical characterization of individual particles for both refractory and non-refractory material. The main limitations are the qualitative character of the chemical determinations, the relatively high cost and the relatively complex instrumental control and data treatment.

3.4. Optical properties

Dust particles scatter and absorb radiation. The fact that a significant fraction of dust particles is coarse determines the way in which the light is scattered. Light absorption is mainly due to the hematite and goethite minerals. In a recent review of published data, Redmond et al. (2010) found that the refractive index of dust (VIS to near IR: 400 – 1000 nm) has a relatively constant real part (~1.5). By contrast its imaginary part tends to decrease with wavelength (e.g. $1.5{\cdot}10^{-3}$ at 1000 nm and $3.5{\cdot}10^{-3}$ at 400 nm).

3.4.1. Light scattering

The integrating nephelometer (Beuttell and Brewer, 1949) is the most commonly used instrument for measuring the total light scattered by aerosol particles (Heitzenberg and Charlson, 1996). Ideally, integration angles attempt to cover forward to backward scattering: 0–180°. The scattering of air is quantified by alternating measurements of ambient aerosols with measurements of filtered (particles free) air. The aerosol scattering is calculated by the difference between the two measurements. These instruments are calibrated by filling them with gases of a given scattering coefficient, usually air and CO_2 (Bodhaine, 1979). Thus, an instrument designed for measuring the Mie scattering of aerosols is calibrated by measuring Rayleigh scattering. Commercial instruments may measure total and back scattering in 3 wavelengths, with a detection limit of 0.1 Mm^{-1}.

Integrating nephelometers have been used in the field campaigns of AMMA (Mallet et al., 2008) and SAMUM (Müller et al., 2009c; Schladitz et al., 2009) to derive dust optical properties. Some of the long term dust monitoring programs (~ a few) include integrating nephelometer measurements (Table 1).

There are two main sources of errors concerning dust measurements:

(1) Non-ideal illumination and truncation. The first refers to the fact that the angular intensity distribution provided by the opal glass diffuser may differ from the cosine (lambiertian) function, whereas truncation error refers to the fact that near forward scattering cannot be directly measured due to the optic design (from 0° to 7–10° depending on the instrument). Because forward scattering increases with particle size, truncation error may be significant when measuring coarse dust (in contrast to smaller aerosols). Anderson and Ogren (1998) developed an empirical equation for correcting these two effects in the commercial TSI integrating nephelometer (model 3563, Inc, St Paul, MN, USA). This is a mean correction equation for aerosols with a refractive

index within the range 1.4–1.52 for the real part and 0–0.01 i for the imaginary parts, and with a bimodal size distribution with mean volume diameters within the ranges 0.2–0.4 μm and 2–4 μm for the fine and coarse modes, respectively. The size dependence correction involves the inclusion of the angstrom exponent in the correction equation. Periodical exclusion of coarse particles from the sample (e.g. with an impactor) is recommended for assessing the accuracy in the measurements. If correction is not applied, the total scattering is underestimated by between 5–15% for submicron particles and by 40–60% for coarse particles (Ensor and Waggoner, 1970; Moosmüller and Arnott, 2003; Heintzenberg et al., 2006). Back scattering correction is considered negligible. The angstrom dependent correction equation has been tested in other commercial nephelometers (Radiance Research, Seattle, WA, USA; Ecotech, knoxfield, VIC, Australia) and it has been concluded that, although the Anderson and Ogren (1998) equation is a good approach, the size distribution measurement is desirable for a proper correction (Müller et al., 2009a). Anderson and Ogren's equation may introduce an uncertainty of up to 30% in aerosols that reflect more than dust (e.g. refractive index 1.7–0.3*i*; Massoli et al., 2009) and of up to 5% in aerosols that are much more absorbent than dust (e.g. $n = 1.45–1.52$ and $k = 0–0.2i$; Bond et al., 2009).

(2) High relative humidity (RH) increases the scattering of dust, especially that of soluble minerals (e.g. carbonates, halides, gypsum/anhydrite). Measurements of dry particle scattering (RH < 40% in the instrument) are recommended. Heating of the sample for decreasing RH may result in evaporation of semi-volatile species mixed with dust (Bergin et al., 1997). Alternatively, diffusion driers may be used. Li-Jones et al. (1998) used a nephelometer to study the effects of relative humidity on light scattering by dust.

Anderson et al. (1996) reviewed the uncertainties linked to these instruments. Some recommendations for long term monitoring include routine automatic checks with calibration gases for assessing the accuracy, drift and need of new calibration (Anderson and Ogren, 1998). Differences between measured and theoretical scattering coefficient in CO_2 calibration checks are typically 2–3% (Heintzenberg et al., 2006; Müller et al., 2009a). Instrument to instrument variability has been reported to be of ~7% for TSI nephelometers (Heintzenberg et al., 2006) and of 3% for Optec NGN-2 (Saxena et al., 1996). Uncertainty in field measurements is estimated at ~10% (Heizenberg et al., 2006).

3.4.2. Light absorption

Measurements of aerosol light absorption are subject to uncertainties that are higher than for scattering. Traditionally long term monitoring of aerosol light absorption has been performed with filter based photometers, which can use 1–7 wavelengths. Recent methods based on in situ measurements (i.e. not based on accumulation of sample on filter; e.g. photo-acoustic; Arnott et al., 1999; Lack et al., 2006; Lewis et al., 2008) could play a key role in future long term measurements. A review of available techniques has been performed by Moosmüller et al. (2009).

In the filter based instruments ambient air with dust particles is drawn at a constant volumetric airflow rate through a filter. The optical transmission across the filter is continuously monitored using a source light and a detector. When particles accumulate on the filter, transmission decreases and less light reaches the detector. The derivative in time of the light attenuation or transmission signal is measured, and it is then used for estimating the aerosol light absorption. A reference (blank) filter is used for zero and for detecting potential changes in the source emission. In practice, the relationship between light transmission and absorption coefficient is affected by other factors, and corrections are necessary. The two most relevant corrections (Hänel, 1987) are due to back scattering of the filter + sample system (back scattering correction) and to the decrease in the optical path in a non-linear way owing to particle accumulation on the filter (loading correction). These and other corrections are implemented in different ways depending on the instrument design.

The *Particle Soot Absorption Photometer* (PSAP; Radiance Research, Seattle, WA, USA; Bond et al., 1999) requires correction for back scattering, filter loading, flow and spot size. In practice, flow and spot size differ from those used by the internal software in the instrument (Bond et al., 1999). At least two adjustment schemes have been proposed, including differentiation between instrument models (Bond et al., 1999; Virkkula et al., 2005; Virkkula, 2010; Ogren, 2010). Correction equations require data of scattering coefficient (usually measured with a nephelometer working in parallel). As shown by Müller et al. (2011) the correction schemes proposed by Virkkula et al. (2005) may result in absorption coefficients of up to 6% higher than those obtained with the scheme devised by Bond et al. (1999). This equipment requires a manual filter change; this is a drawback in long term programs.

The *Aethalometer* (Magee Scientific, Berkeley, CA, USA; Hansen et al., 1984) has a principle of measurement very similar to that of PSAP, although in these devices the filter change is performed automatically. Several correction schemes have been proposed (Weingartner et al., 2003; Arnott et al., 2002; Schmid et al., 2006; Virkkula et al., 2007; Collaud Coen et al., 2010). Weingartner et al. (2003) argued that a constant $C = 2.14$ is necessary for correcting measurements due to multiple light scattering effects in the filter fibber when calculating the absorption coefficient from attenuation. Collaud Coen et al. (2010) performed an intercomparison between five correction schemes and concluded that the constant C may reach values between 2.9 and 4.3.

The *Multi-Angle Absorption Photometer* (MAAP; Thermo Scientific, Waltham, USA; Petzold and Schönlinner, 2004) includes additional sensors for measuring the back scattering of the filter plus sample system at two angles. This minimizes the artifact of back scattering in such a way that corrections are not considered necessary (Petzold et al., 2005). A good agreement between measurements of MAAP and the in situ photo-acoustic method was observed, slope = 0.99 ± 0.01 (Sheridan et al., 2005). However, Müller et al. (2011) observed that the light source of MAAP emits at 637 nm instead of at 670 nm specified by the manufacturer. As a consequence, MAAP measurements should be adjusted to 637 nm and the absorption coefficient multiplied by 1.05.

A detailed description of how to perform corrections for these three instruments was performed by Müller et al. (2011). Some recommendations include operation at low transmittance decay (>70%) to minimize load artifacts and frequent checks of the airflow and spot size. Because of the lack of a generally accepted calibration standard (including dust), uncertainties are obtained in intercomparisons (Saathoff et al., 2003; Sheridan et al., 2005; Park et al., 2006; Slowik et al., 2007; Lack et al., 2009; Nakayama et al., 2010; Müller et al., 2011). Unit to unit variability of 30% for PSAPs, 30% for Aethalometers and 5% for MAAPs have been observed (Müller et al., 2011). Uncertainties in field measurements are considered to be of 30% for the PSAP (Schladitz et al., 2009) and 10% for the Aethalometer (Mallet et al., 2008) and the MAAP (Schladitz et al., 2009).

Absorption photometers were used in the field campaigns of PRIDE (Reid et al., 2003b), AMMA (Mallet et al., 2008) and SAMUM (Müller et al., 2009c; Petzold et al., 2009; Schladitz et al., 2009). Some of the long term dust monitoring programs include absorption photometer measurements (Table 1). Multi-wavelength photometers are recommended for distinguishing dust from other absorbing aerosols (e.g. soot black carbon; Fialho et al., 2005, 2006). Multi-wavelength models of PSAPs and Aethalometers are currently available; however, MAAP is still manufactured with a single wavelength.

3.4.3. Dust properties inferred from optical measurements

Scattering and absorption coefficients are used for determining key radiative properties (e.g. refractive index; Hand and Kreidenweis, 2002; Petzold et al., 2009; Schladitz et al., 2009). Some of these properties help us to identify the presence of dust in ambient air (Andrews et al., 2011), and to perform dust climatology (e.g. frequency of dust events, long term changes of dust properties, etc...):

- The 'aerosol mass scattering efficiency' (scattering to mass ratio) is typically lower for dust (0.2–1.0 m^2/g; Maring et al., 2000; Formenti et al., 2001; Andreae et al., 2002; Alfaro et al., 2003; Vrekoussis et al., 2005) than for other aerosols, e.g. biomass burning (2.5–5.0 m^2/g; IPCC, 2001; Trentmann et al., 2002; Adam et al., 2004; Reid et al., 2005) or urban/industrial (3.0–6.0 m^2/g; Bergin et al., 2001; IPCC, 2001; Carrico et al., 2003; Pereira et al., 2008) aerosols. Its value is higher for fine particles than for coarse ones (Hand and Malm, 2007).
- The 'aerosol mass absorption efficiency' (absorption to mass ratio) is also lower for dust (0.01–0.02 m^2/g; Alfaro et al., 2004), than for other aerosols, such as humic organics (0.02–0.04 m^2/g; Hoffer et al., 2006) and soot (5–25 m^2/g; Bond and Bergstrom, 2006).
- The increase of single scattering albedo (scattering to extinction ratio) with wavelength is a distinctive feature of dust (Redmond et al., 2010). This property was used for performing a climatology of dust transport to the Alps (Collaud Coen et al., 2004).
- The Angström scattering exponent decreases with particle size, with typical values < 0.5 when dust is present (Delene and Ogren, 2002; Collaud Coen et al., 2007; Schladitz et al., 2009; Andrews et al., 2011).
- The Angström absorption coefficient variability (3–7) has been related to the content of iron oxides in dust (Petzold et al., 2009; Müller et al., 2009b).
- Back-scatter fraction increases with the amount of small particles (Collaud Coen et al., 2007). It typically shows values of 0.09–0.10 for dust and within 0.12–0.20 for populations dominated by Aitken particles (Schladitz et al., 2009; Andrews et al., 2011).
- Asymmetry parameter (a measurement of the hemispheric distribution of light scattering in the Mie range, i.e. forward versus back scatter; Andrews et al., 2006) is positive when forward scattering of coarse dust particles dominates (0.65–0.75; El-Metwally et al., 2008; Osborne et al., 2008), and negative when back scattering prevails.

3.5. Considerations for long term dust monitoring programs

General considerations for aerosol measurements (Hinds, 1982) should also be taken into account in long term dust programs.

All instruments are usually connected to a single (principal) sampling inlet. The coarse nature of dust should be taken into account when designing inlet or selecting sampling flow rates. Because inertia increases with size and density of particles, isokinetic sampling becomes more important for dust than for other smaller aerosols. Wind faced inlets may be used in windy observatories. Deviations in the face velocity of the sampling tube of each instrument connected to the general sampling inlet gives rise to considerably higher errors in the measurements of coarse particles than of fine ones; e.g. three times higher face velocity in a sampling tube than in the general inlet may result in losses of 50%, 23% and 8% of 10, 5 and 2.5 μm dust particles, respectively. Losses in bent tubes are also higher for coarse particles than for fine ones (e.g. dust vs. ammonium-sulfate).

Dry particle measurements are recommended. The Global Atmospheric Watch program recommends measurements with a sample RH <40%. These RH values may be reached using diffusion driers, dry air dilutors or heaters. Size dependent losses should be characterized in diffusion driers. Dilutors may result in data below detection limit (under low sample conditions). Heaters may evaporate semi-volatile species mixed with dust. High RH enhances dust scattering, increases particle size and mass concentrations.

The methods for assessing long term drift depend on the instrument used. Sizing accuracy for particle size spectrometers (OPCs, APSTOFS or EMA) are usually checked with PLS (Wiedensohler et al., 2012). Gases of a given scattering coefficient are used for nephelometers (Anderson and Ogren, 1998). Reference materials (e.g. certified fly ashes) are available for elemental composition of dust. Regular checks of sampling flow rate and measurements of instrumental noise (placing an absolute filter in the inlet) are recommended for dust and other types of aerosols.

4. Summary and discussion

To better understand the long term evolution (multi-decadal) of dust and its interactions with climate and biogeochemistry, it is necessary to obtain data on physico chemical properties of dust by in situ measurements. This type of dust observations has mostly been performed in campaigns lasting from weeks-to-months. Long term dust observations have only been made at a few sites.

Collection of samples on filter and offline analysis of bulk dust or bulk major elemental composition (Si, Al, Fe, Ca, K, Mn, Na and Mg) is the most reliable method for long term observations of dust concentrations. Despite some advances in online bulk elemental composition analysis in the last decade, these methods are not yet been implemented for long term use. Because dust is frequently mixed with other compounds, analysis of potential anthropogenic species (sulfate, nitrate, ammonium and trace elements such as As, V, Ni, Zn, Cd, Pb or Cr), sea salt or organic matter tends to be included in long term aerosol dust monitoring programs. Simultaneous measurements of bulk aerosol mass concentrations with automatic methods are useful for chemical mass closure dust studies (dust vs other species) and for long term data consistency assessment.

Size segregated long term dust programs are recommended (e.g. simultaneous total or <10 μm and <1, 2.5 μm dust measurements). Available long term observations have shown that there is a significant year-to-year variability in total dust (all sizes particles) concentrations (Prospero and Lamb, 2003). Dust mass is considerably influenced by particle size, i.e. a few coarse particles may dominate mass concentrations. However, some processes seem to be influenced by fine dust particles. For example, fine dust particles may exhibit an enhanced solubility in sea water with respect to coarse dust particles (Baker and Croot, 2010). Interaction of radiation with coarse and fine dust particles also displays different properties (Otto et al., 2007).

Bulk chemical characterization used in long term dust programs may be complemented by analytical techniques which are not necessarily designed for long term use. Two examples are provided. The use of online mass spectrometers in certain periods (e.g. dust season) over years would enable us to obtain quantitative data on the long term evolution of the mixing state of dust with anthropogenic aerosols. The isotopic characterization of samples collected during the dust season every year would provide data on potential changes in the sources of some key minerals (evaporites) or compounds mixed with dust (e.g. sulfate, nitrate, or metals). Future evolution of anthropogenic emissions of aerosol precursors or changes in the strength of dust sources may prompt changes in the degree and type of dust mixings.

Physical and optical dust properties are also included in long term programs. Techniques for atmospheric aerosol measurements have improved significantly in the last decade and it is expected that this evolution will benefit aerosol dust measurements as well. Some of the instruments used for dust characterizations are calibrated with synthetic aerosols whose properties differ from those of dust particles. In the case of optical properties, the availability of mineral dust standards with different scattering and absorption spectrum would enable us to perform quantitative assessments on the instrument response. Particle sizers are regularly calibrated with standards of PLS and this has implications when measuring dust particles. Reid et al. (2003b) found reasonable agreement for size distributions measured with different methods (cascade impactors, ASPTOFS and OPC). However, such relatively low variability in particle sizing introduced greater uncertainties in the radiative transfer algorithms. The availability of monodisperse dust particle standards would improve instrument calibration.

There is a clear deficit of long term dust observations. Only a few sites located downwind of North African and Asian deserts (Fig. 2) that have been active in the last 20–30 years continue operative nowadays.

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