



Dust episodes in Beirut and their effect on the chemical composition of coarse and fine particulate matter



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HIGHLIGHTS

- An increase in particle volume for sizes between 2.25 and 5 μm
- Increase in crustal material in coarse and fine particles
- Biogenic enrichment of elements of anthropogenic origin like Ba and V
- Formation of organosulfates and organonitrates in fine PM

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ABSTRACT

Particles captured during dust episodes in Beirut originated from both the African and Arabian deserts. This particular air mixture showed an increase, over non-dust episodes, in particle volume distribution which was mostly noticed for particles ranging in sizes between 2.25 and 5 μm . It also resulted in an increase in average mass concentration by 48.5% and 14.6%, for the coarse and fine fractions, respectively. Chemical analysis of major aerosol components accounted for 93% of fine PM and 71% of coarse PM. Crustal material (CM) dominated the coarse PM fraction, contributing to 39 \pm 15% of the total mass. Sea salt (SS) (11 \pm 10%) and secondary ions (SI) (11 \pm 7%) were the second most abundant elements. In the fine fraction, SI (36 \pm 14%) were the most abundant PM constituent, followed by organic matter (OM) (33 \pm 7%) and CM (13 \pm 2%). Enrichment factors (EF) and correlation coefficients show that biogenic and anthropogenic sources contribute to the elemental composition of particles during dust episodes. This study emphasizes on the role played by the long-range transport of aerosols in changing the chemical composition of the organic and inorganic constituents of urban coarse and fine PM. The chemical reactions between aged urban and dust aerosols are enhanced during transport, leading to the formation of organo-nitrogenated and -sulfonated compounds. Their oligomeric morphologies are further confirmed by SEM–EDX measurements.

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1. Introduction

Dust episodes have been shown to affect particulate matter (PM) by increasing their mass loading to values above urban levels, and altering their chemical composition (i.e. (Bozlaker et al., 2013; Goudie and Middleton, 2001; Massoud et al., 2011; Rodríguez et al., 2001; Saliba et al., 2010; Saliba et al., 2007; Xie et al., 2005)). Furthermore, dust has been known to be transported thousands of kilometers reaching places as far as Northern Europe (Franzén et al., 1994) and Amazonia (Swap

et al., 1992). Even though dust storm episodes can vary in strength and duration (Gobbi et al., 2000), their remnants can remain in the atmosphere for a certain time after the storm ends (EU, 2011). This is also evident for African dust storms, where increases in PM₁₀ levels remains up to one to three days after the dust episode ends ((EU, 2011) and references therein). Indeed African air mass intrusions develop vertically (Alpert et al., 2004; Balis et al., 2004; De Tomasi et al., 2003; Pérez, 2005) and considering the speed and size of particles, they will need around 2–3 days to deposit (Zender et al., 2003).

The Eastern Mediterranean region is one of the top dust storm generation regions in the world (Idso, 1976), with studies showing an increase in the number of dusty days (Alpert et al., 2004; Goudie and Middleton,

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2001). Ganor et al. (2010) show that between 1958 and 2006, the number of dust episodes increased from 15 to 27, with a linear regression of 2.7 days per decade. This is of concern due to the association of morbidity with dust exposure, as documented by numerous epidemiological and biological studies (i.e. (Cheung et al., 2011; Perez et al., 2012; Tobias et al., 2011)).

Despite this, the number of reported studies in this region is still scarce (i.e. (Dada et al., 2013; Daher et al., 2013; Ganor et al., 2010; Goudie and Middleton, 2006; Herut et al., 2001; Marconi et al., 2013; Massoud et al., 2011; Saliba et al., 2007)), and an extrapolation from dust episode (of Saharan origin) studies in the Western Mediterranean would not be an accurate representation, mainly because the reported changes in PM composition remain specific to the local environmental and meteorological conditions (i.e. (Dayan et al., 1991; Ganor and Mamane, 1982; Griffin et al., 2007; Querol et al., 2009)). This study focuses on the chemical and physical effect of dust episodes on urban PM in Beirut, a city that lies on the eastern coast of the Mediterranean Sea, surrounded by semi-arid regions and periodically subjected to dust storms originating from North Africa and the Arabian Peninsula. In this study, coarse ($PM_{10-2.5}$), particles with diameter between 2.5 and 10 μm , accumulation ($PM_{2.5-0.25}$), particles with diameter between 2.5 and 0.25 μm , and quasi-ultrafine ($PM_{0.25}$), particles with diameter less than 0.25 μm PM samples are collected during dust and non-dust episodes in Beirut. The four dust episodes identified in this study vary between dust and dust remnant. Gravimetric analysis and several analytical methods were used to determine the mass, particle distribution, the chemical composition, and the morphology during these particular episodes. It is also important to note that the results highlight the reaction of urban and long-range transport of dust aerosols to produce organo-sulfates and organo-nitrates.

2. Methods

2.1. Sample collection

Size-segregated PM samples were collected at the rooftop of the Chemistry department at the American University of Beirut (AUB), an urban background site located at 33°90'N, 35°50'E at an elevation of 20 m above ground level. From the south this site is surrounded by a green area with the nearest street located about 150 m away. From the north, it overlooks the Mediterranean Sea, separated by a two-lane road which is congested during morning and afternoon rush hours. From the Eastern side resides the Beirut harbor within a distance of 2.5 km. Particle levels at this site are commonly influenced by sea and land breeze circulation (Baalbaki et al., 2013).

The sampling campaign was conducted on dust and non-dust episodes during summer and fall of 2012. Dust episodes were identified based on forecasting air mass trajectories using daily forecast-trajectories calculated by the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLOT) model (Draxler and Rolph, 2013) and relying on visual visibility limits and sky color. Those episodes were further confirmed using BSC-/DREAM dust maps (<http://www.bsc.es/projects/earthscience/DREAM/>) (Fig. S1). However, the residence time of transported dust particles is dependent upon the removal mechanisms, with residence times varying from few hours to 10 days (Goudie and Middleton, 2006; Kubilay et al., 2000; Papayannis et al., 2008). Moreover, if dust air masses are transported at high altitudes, they may affect ground level PM concentrations up to 2 days after an episode ends (EU, 2011). Therefore, in this study, dust episodes include days during which air mass trajectories were coming from the desert, and subsequent days where transported dust remnants possibly remain airborne. Moreover, the identity of these episodes was further confirmed using the Al data from the chemical analysis as a geo-chemical tracer. An average increase of 74% and 189% over non-dust episodes in the coarse and fine fractions, respectively, is reported. During the

campaign, four dust episodes were identified, for which six samples were collected (Table S1, Fig. S1).

Coarse ($PM_{10-2.5}$), accumulation ($PM_{2.5-0.25}$) and quasi-ultrafine ($PM_{0.25}$) size-fractionated particles were collected using three parallel Sioutas Personal Cascade Impactor Samplers (Sioutas PCIS, SKC Inc., Eighty Four, PA, USA, (Misra et al., 2002)) preceded by PM_{10} inlets (Chemcomb Model 3500 Speciation Sampling Cartridge) and operating at a flow rate of 9 $\text{L}\cdot\text{min}^{-1}$. Two PCISs were loaded with Teflon filters (Pall Life Sciences, Ann Arbor, MI) and were used for the analysis of: gravimetric mass, inorganic ions and total metals. The third PCIS was loaded with quartz filters (Whatman International Ltd.) and used for the analysis of water-soluble organic carbon (WSOC) as well as organic and elemental carbon (OC and EC, respectively).

Concurrently, 24 h PM samples were collected during dust episodes (November and December 2012) using a six stage cascade impactor (MPS – 6 Microanalysis Particle Sampler) of cut point sizes: 10, 5, 2.5, 1.0, 0.5 and 0.1 μm . Samples were collected either on 12 mm aluminum stubs coated with carbon adhesive tape or on ZnSe windows and used for SEM (Scanning Electron Microscope) imaging.

2.2. Sampling analysis

To determine PM mass concentration, prior to sectioning, Teflon filters were pre- and post-weighed using a UMX2 microbalance (Mettler Toledo GmbH, CH-8606 Greifensee, Switzerland), following 24 h equilibration under controlled temperature and relative humidity conditions (22–24 °C and 40–50%, respectively). The first set of Teflon filters was cut into 3 equal sections. The first section was analyzed for total elemental mass by means of a high resolution magnetic sector Inductively Coupled Plasma Mass Spectrometry (HR-ICP-MS Thermo-Finnigan Element 2). For the extraction, a microwave-aided Teflon bomb digestion protocol using a mixture of 1 mL of 16 M nitric acid, 0.25 mL of 12 M hydrochloric acid, and 0.10 mL of hydrofluoric acid was used (Herner et al., 2006). The digestion protocol was comprised of a 9 min ramp to 180 °C followed by a 10 min hold at 180 °C and 1 h of ventilation/cooling. Following cooling, digestates were diluted to 15 mL with high purity water then analyzed by HR-ICP-MS (Lough et al., 2005). The second section was extracted with high purity water and used to quantify PM concentrations of water-soluble inorganic ions using ion chromatography (Model 2020i, Dionex Corp.) The third section and second set of Teflon filters were used for a separate study.

Quartz filters, analyzed for Elemental Carbon, Organic Carbon and Water-Soluble Organic Carbon (EC, OC and WSOC, respectively), were prebaked at 550 °C for 12 h and stored in baked aluminum foil prior to sampling. Elemental and organic carbon contents of the filters were determined using the NIOSH Thermal Optical Transmission method (Birch and Cary, 1996). Water-soluble organic carbon content was quantified using a Sievers 900 Total Organic Carbon Analyzer, following water-extraction and filtration of the samples (Sullivan et al., 2004).

2.3. Chemical mass closure

For the purpose of chemical speciation, chemical components were classified into six categories: organic matter (OM), elemental carbon (EC), sea salt (SS), crustal material (CM), trace elements (TE) and secondary ions (SI). Organic matter was calculated by multiplying organic carbon (OC) by a factor of 2.1 in order to account for aged particles as recommended by several earlier studies (Gnauk et al., 2005; Sciare et al., 2005; Takahashi et al., 2008; Terzi et al., 2010). Sea salt was estimated as the sum of Na^+ concentration and sea-salt fractions of Cl^- , Mg^{2+} , K^+ , Ca^{2+} , and SO_4^{2-} concentrations, assuming standard sea-water composition (Seinfeld and Pandis, 2006). Crustal material includes typical crustal material elements (Al, K, Fe, Ca, Mg, Ti and Si) and was determined by summing the oxides of these metals using Eq. (1) with Ca and Mg representing their non-sea-salt fractions

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