



Contribution of particulate water to the measured aerosol optical properties of aged aerosol



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HIGHLIGHTS

- We show that aerosol scattering is caused almost entirely by PM₁.
- Supersaturation is a cause for the existence of particulate water at low RH.
- We develop an equation to estimate the volume growth factor from B_{sp} measurements.

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ABSTRACT

Measurements of aged aerosol scattering in the Eastern Mediterranean at ambient and low relative humidity (RH) are combined with measurements of its size composition distribution and its aerosol water content to gain insights about its hygroscopic and optical properties. The particle water uptake above 60% RH can be explained by the water absorption by the inorganic particulate matter in this sulphate rich environment. However, the particles were supersaturated aqueous solutions at low RH and this water cannot be explained just by the water associated with their inorganic components in a metastable state. We estimate that approximately 20% of this water may be contributed by the aged organic particulate matter. A semi-empirical equation is derived allowing the estimation of the aged aerosol Volume Growth Factor (VGF) from the ratios of scattering at ambient and low RH. The predictions of the equation are in good agreement with the measurement dataset obtained during the study.

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1. Introduction

Atmospheric aerosols affect climate directly by scattering and absorbing radiation and indirectly by modifying the lifetime and the radiative properties of clouds (Pilinis et al., 1995; Haywood and Boucher, 2000; Segikuchi et al., 2003; Yu et al., 2006; IPCC, 2007; Spichtinger and Cziczo, 2008; Ramanathan and Xu, 2010). Understanding the impact of aerosols on climate is challenging, since their concentration and chemical composition are quite variable in space and time (Chin, 2009). Estimation of their radiative forcing requires knowledge of their size, chemical composition and microphysical state (Pilinis and Li, 1998). Atmospheric aerosols consist of various inorganic and organic compounds and water.

Absorption of water by particles is a strong function of the ambient relative humidity (RH) and the particle composition (Nenes et al., 1998; Kreidenweis et al., 2008). Hygroscopic growth alters the size and the refractive index of atmospheric particles, affecting the magnitude of radiative forcing (Nemesure et al., 1995; Pilinis et al., 1995; Koloutsou-Vakakis et al., 1998; Haywood and Boucher, 2000; Malm and Day, 2001; Yu et al., 2006; Garland et al., 2007; Jimenez et al., 2009; Zieger et al., 2010).

Although water is usually assumed to be mainly associated with the inorganic fraction of particulate matter, atmospheric aerosols are complex mixtures of both organic and inorganic species (Ansari and Pandis, 2000; Engelhart et al., 2011; Smith et al., 2012). Organic Aerosol (OA) is a major component of PM_{2.5} (Chow et al., 1994; Turpin et al., 2000; Viidanoja et al., 2002; Sillanpää et al., 2006; Zamora et al., 2011) but while an important fraction of the organic mass is water-soluble (Saxena and Hildemann, 1996; Zappoli et al., 1999; Mayol-Bracero et al., 2002; Decesari et al.,

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2005; Hallquist et al., 2009) the OA hygroscopic properties and its interactions with the inorganic aerosol components are not well understood. This limited knowledge is due to the fact that organic PM consists of thousands of compounds (Goldstein and Galbally, 2007). These compounds continue to react after their emission in the atmosphere and are converted to oxygenated organic aerosol (OOA) (Zhang et al., 2007). Evidence suggests that fresh primary OA together with the fresh secondary OA are converted in the atmosphere to OOA through a series of chemical processes collectively called chemical ageing. Eastern Mediterranean is an excellent natural laboratory to study the OOA properties because during the late spring, summer, and early fall practically all the OA is in the form of highly oxidized OOA (Hildebrandt et al., 2010).

Most of our knowledge of the water uptake of OA is based on laboratory studies with model systems or fresh secondary OA formed during the oxidation of a single precursor. Despite the simplicity of the examined systems these studies have revealed quite complex interactions among the organic and inorganic particle components and water. For example, the presence of organics was found to decrease the water uptake of NaCl (Choi and Chan, 2002), while the presence of dicarboxylic acids resulted in the suppression of the efflorescence behaviour of ammonium sulphate (Prenni et al., 2003; Parsons et al., 2006), and the increase of its aerosol water uptake (Choi and Chan, 2002). In another study the presence of solid succinic acid in supersaturated supermicron particles of ammonium nitrate was found to facilitate its efflorescence (Lightstone et al., 2000). In the study of Andrews and Larson (1993), the presence of a surfactant organic film decreased the Deliquescence RH (DRH) of NaCl particles. This was attributed to the fact that certain organic aerosol species, which are surface-active, can reduce aerosol surface tension due to film formation and thus decreasing the particle's DRH (Li et al., 2011). Brooks et al. (2003) reported increased water uptake of mixed ammonium sulphate/maleic acid particles, lowering of the ammonium sulphate DRH while its Efflorescence RH (ERH) was practically unaffected. In other cases only a slight or no change of DRH was observed (Lightstone et al., 2000; Prenni et al., 2003; Dennis-Smith et al., 2012). Complex mixtures of dicarboxylic acids and inorganic salts resulted in lower DRH than the individual components (Marcolli et al., 2004). These results suggest that small concentrations of organic species can prevent tropospheric aerosols from becoming fully solid at lower RHs. In a recent study by Bertram et al. (2011) a variety of highly oxygenated organics – ammonium sulphate binary systems exhibited decreasing DRH and ERH values with increasing organic to sulphate mass ratios. Extrapolation of these results to realistic ambient particles especially for aged OA remains a challenge.

The aerosol scattering coefficient is quite sensitive to the water uptake by the particles. In the present work we attempt to take advantage of this fact, in order to quantify the effect of aged organic aerosol on the water uptake and optical properties of ambient particulate matter. To do so we use measurements of aerosol chemical composition and mass size distributions, at the Finokalia measurement site in Greece. In the following sections we give a brief description of the site and the related measurements, along with the modelling methodology we used. We test a variety of approaches for the estimation of the aerosol scattering and a number of hypotheses about the role of organics. Finally we derive a simple parameterization for the estimation of water uptake (expressed as volume growth factor) from the change in scattering for aged aerosol.

2. Site description and measurements

All measurements were conducted during May 2008 during the Finokalia Aerosol Measurement Experiment 2008 (FAME-08)

campaign (Pikridas et al., 2010), part of the European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions (EUCAARI). Finokalia is a coastal site (35°20'N, 25°40'E) situated in the southeast of the Mediterranean Sea on the island of Crete, Greece (Fig. 1). The island of Crete is a crossroad of three continents, thus measurements are influenced by air masses from Europe, Asia and Africa (Mihalopoulos et al., 1997). The Finokalia station is located at the top of a hill (230 m) at 1.3 km from the coast, facing the sea. A number of measuring campaigns have been conducted in this site during the last decade (Vrekoussis et al., 2005; Liakakou et al., 2009; Violaki and Mihalopoulos, 2010; Lee et al., 2010; Pikridas et al., 2010; Engelhart et al., 2011). The nearest urban area is Heraklion located 50 km away from the measuring site, thus Finokalia is rather isolated and an ideal place to measure air pollution in the Eastern Mediterranean. During this study the weather was sunny, relatively warm, with winds mainly from the North, representing quite nicely the annual mean meteorological conditions at Finokalia (Mihalopoulos et al., 1997). Therefore while the analysis described in the following sections, relies on the May 2008 data, the conclusions are representative for most of the year in Finokalia (with the potential exception of a few months in the winter). Even more importantly, they are applicable for a mixture of highly aged organic aerosol and sulphates that is quite relevant for a number of areas around the world.

A thorough description of FAME-08, onsite instrumentation and measurements are provided in Pikridas et al. (2010). The source region analysis of FAME-08 campaign presented by Pikridas et al. (2010) revealed that the air masses arriving at the site during the period of interest were either of marine or continental origin (Greece, Balkans and Turkey), sporadically affected by the major city of Athens. The site was also affected by African air masses with increased dust levels from May 19 to May 21, 2008.

During the campaign the aerosol optical properties, as well as its size distribution and chemical composition were continuously

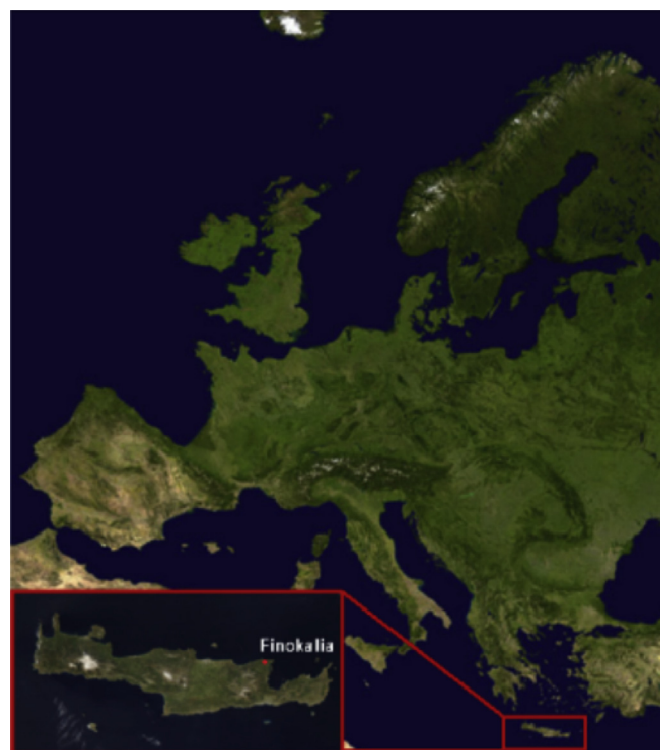


Fig. 1. Site of the Finokalia Station (NASA satellite).

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