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Particulate pollution transport episodes from Eurasia to a remote region of northeast Mediterranean



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ABSTRACT

Long-range transportation of air pollutants from industrial and urban environments can significantly affect the quality of the air in remote regions. In this study, we investigate episodes of particulate transport (PT) from Eurasia to the remote environment of Northeastern Mediterranean, i.e., the region of the North Aegean Sea (NAS), during the summer when the synoptic Etesian wind conditions prevail. A temporary monitoring station was set up at a remote region on the island of Lemnos, which is located at the center of the NAS at a distance of ca. 250 km from the continent. Measurements of the aerosol particle size distributions, the total number and mass concentrations, as well as the chemical composition of the particles were conducted from 27 August to 10 September 2011. During this period, the wind speeds were high (typically higher than 5.5 ms^{-1}) with a direction that mostly ranged from north to northeast (68% frequency). Winds having direction ranging from northwest to south were less frequent (7% frequency), while the rest of the cases were characterized as calm (i.e., wind speeds less than 1 m s^{-1} ; 25% frequency). Seven PT episodes were observed during the sampling period. When the wind direction was northeastern we observed up to a six-fold increase in particle number concentration of nucleation mode, while the peak size of the particles decreased from 100 to 20 nm. Interestingly, the nucleation-mode particles grew from ca. 15 to 25 nm with rates of ca. 9.0 nm h^{-1} , which are representative of polluted areas. Analysis of the chemical composition of particle samples collected on filters during the PT episodes shows that the concentration of sulfates and nitrates increased by ca. 60%, while the OC/EC ratio increased by ca. 22% compared to the rest of the sampling period. Back-trajectory analysis for the period during the episodes shows that the air masses arriving at the station passed over the greater Istanbul area and the Black Sea 9 to 12 h before reaching our station. These observations provide strong evidence that the air quality in the remote region of the NAS can be significantly affected by the transportation of particulate pollution during the summer period, having potentially important effects upon human health and climate in the region.

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1. Introduction

Long range air pollution transport can significantly affect air quality and therefore human health and climate in both urban (Kubilay et al., 2000; Karaca et al., 2009) and remote (Masclat et al., 1988; Kato et al., 2001) regions. For instance, the region of the Eastern Mediterranean, which is considered a climate hotspot, is strongly affected by transportation of air pollutants originating

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from Europe, Africa and Asia. While numerous studies have focused on the southern part of the Eastern Mediterranean (Kouvarakis et al., 2002; Smolík et al., 2003), its northern part, i.e., the Northern Aegean Sea (NAS), has largely been ignored.

The NAS is an important part of the Mediterranean region, located very close to big cities of Greece (Athens and Thessaloniki), Turkey (Istanbul, Izmir, and Bursa) and Bulgaria (Burgas and Plovdiv) that have organized industrial zones, including large factories, power plants, and mines (Thöni et al., 2011; Civan et al., 2011). The area is of particular interest since on the one hand the economic crisis in Greece has affected everyday human practices, which in turn have led to an increase in anthropogenic emissions of particles (Saffari et al., 2013; Paraskevopoulou et al., 2014) and a decrease of gaseous pollutants such as NO₂ and SO₂ (Vrekoussis et al., 2013), while on the other the expanding economy of Turkey has strongly affected regional air quality (Alyuz and Alp, 2014).

The seasonal pattern of air pollution transport is strongly affected by the annual variability of the wind patterns in NAS (Kallos et al., 2007). During summer and early autumn, the prevailing winds (i.e., the Etesians) have a northeasterly direction (Kallos et al., 1998; Poupkou et al., 2011). The Etesian winds are associated with horizontal and vertical transport of air masses to the region of the Aegean Sea and the Eastern Mediterranean (Tyrllis et al., 2012; Anagnostopoulou et al., 2013). This in turn perplexes the air pollution pathways between southern Balkans, Turkey and the NAS (Kotroni et al., 2001), and influences the quality of the air and the climate of the entire region (Bezantakos et al., 2013; Tombrou et al., 2013, 2015).

In this study we investigate the properties of atmospheric particles observed in the remote environment of the NAS during the summer period when Etesian winds prevail, in order to understand how cross-border air pollution transport affects local air quality. Measurements of the size distributions, the number and mass concentrations, as well as the chemical composition of the atmospheric particles observed in the region were conducted on Lemnos, a remote island located at the center of the NAS.

2. Experimental

Measurements were conducted from 27 August to 10 September 2011 (239–253 Day of Year; DOY) at a temporary monitoring station at Vigla; a remote region on the island of Lemnos (39° 58' N, 25° 04' E; 420 m a.s.l.), which is located at a distance of ca. 250 km from mainland Greece and Turkey at the center of the NAS (cf. Fig. 1). A Scanning Mobility Particle Sizer (SMPS; TSI Model 3034) was used to measure the size distribution of aerosol particles having diameters in the range from ca. 10 to 500 nm with a 3-min time resolution. In addition, a micro-orifice uniform deposit cascade impactor (MOUDI; MSP Model 110) was used to collect PM₁ samples on Teflon filters (Pall Life Sciences Zefluor Supported PTFE filter, 47 mm in diameter; Part No. P5PLO47) at a flow rate of 30 lpm. A separate sampler operated at 20 lpm was used to collect samples on pre-treated quartz filters (Pall Life Sciences Pallflex Tissuquartz, 47 mm in diameter; Part No.7202) for determining the mass of the total suspended particles (TSP). After gravitational analysis, the filter samples were analyzed for inorganic ions (i.e., NH₄⁺, Na⁺, K⁺, Ca²⁺, Cl⁻, NO₃⁻ and SO₄²⁻) using an ion chromatograph (Dionex Model DX-500) equipped with a conductivity detector (Dionex Model CD20) and a gradient pump (Dionex Model GP50). Organic and elemental carbon (OC and EC) were analyzed with the Thermal Optical Transmittance (TOT) technique (Birch and Cary, 1996), using a Sunset Laboratory OC/EC Analyzer (Sunset Laboratory Inc. Model 4L ECOC Lab Instrument). In particular, a punch of 1.5 cm² from each sample was analyzed using the EUSAAR-2 protocol (cf. Paraskevopoulou et al., 2014).

Meteorological data including wind speed, wind direction, mean hourly temperature and relative humidity were provided by the meteorological station located at the airport of the island. Ultraviolet radiation (UV) radiation data were obtained by the Greek UV monitoring network operated by the Laboratory of Atmospheric Physics of the Aristotle University of Thessaloniki. Back-trajectories (5-days long) of the air masses arriving at Vigla station were determined by the NOAA HYSPLIT model (Draxler and Hess, 1998; Draxler and Rolph, 2003). The trajectories were calculated at 12:00 UTC, which is around the starting time of the episodes. In addition, we investigated the vertical wind velocity motion (omega fields) using the NCEP/NCAR Reanalysis 1 by NOAA (Kalnay et al., 1996) on a daily basis during the entire campaign.

3. Results and discussion

The prevailing winds had north to northeast direction (68% frequency) with wind speeds ranging up to 11.3 m s⁻¹ (cf. Fig. 2). Calm conditions (i.e., average wind speed < 1 m s⁻¹) were observed ca. 25% of the entire sampling period with the majority (73%) being observed during nighttime (from 8:00 in the morning to 18:00 in the evening). In eight out of the fifteen days of the campaign (i.e., 239, 240, 241, 246, 247, 248, 249 and 250 DOY), the meteorological conditions were representative of the Etesian conditions as identified by Tyrllis and Lelieveld (2013).

Fig. 3a shows the evolution of the normalized particle number size distributions throughout the campaign. The mean particle size during the entire period was ca. 100 nm, with 47% of the size distributions exhibiting a single mode, 40% two modes and 13% more than two modes. The total particle number concentration ranged from 1×10^3 to 7×10^3 # cm⁻³, having a mean value of 2×10^3 # cm⁻³, which is in agreement with measurements conducted in the Eastern Mediterranean (Kalivitis et al., 2008; Pikridas et al., 2010) and other coastal areas in Europe (Weijers et al., 2004). As shown in Fig. 3b, the mean number concentration of nucleation-mode particles (i.e., particles having diameters smaller than 25 nm; $N_{d<25\text{nm}}$) was ca. 20 # cm⁻³ (average value for most of the sampling period), but increased to values higher than 1×10^2 # cm⁻³ during almost all the Etesian days (referred as particle transportation PT episode days hereinafter): i.e., on 27 (239 DOY), 28 (240 DOY), and 29 (241 DOY) August as well as on 5 (248 DOY), 6 (249 DOY), and 7 (250 DOY) September. The same pattern was observed for 10 September (253 DOY), which is not identified as an Etesian day.

All the PT episodes started around midday and lasted for ca. six hours, coinciding with the period of the day that the lower troposphere influences most the boundary layer. During this period, the vertical distributions and the transport of air masses is enhanced (Tyrllis and Lelieveld, 2013). Almost all the episodes exhibited $N_{d<25\text{nm}}$ values that reached up to ca. 5×10^2 # cm⁻³ and can therefore be characterized as moderate events (light shaded areas in Fig. 3b). The PT episode on 253 DOY was characterized as strong (dark shaded area in Fig. 3b) with $N_{d<25\text{nm}}$ reaching values of ca. 1.2×10^3 # cm⁻³. The particle number concentration of the Aitken mode during all the PT episodes exhibited a 100% increase (i.e., from 0.7×10^3 to 1.4×10^3 # cm⁻³), while that of the accumulation mode ca. 30% decrease (i.e., from 1.2×10^3 to 0.8×10^3 # cm⁻³) compared to the rest of the sampling period.

Generally, $N_{d<25\text{nm}}$ increased up to 1.2×10^3 # cm⁻³ (strong episodes) when strong north-northeastern winds (average speed 7.5 ± 1.9 m s⁻¹) prevailed during the PT episode days, while $N_{d<25\text{nm}}$ increased up to 5×10^2 # cm⁻³ (weak episodes) under weaker north-northeastern winds (average speed 4 m s⁻¹). Some episodes (e.g., on 249 and 250 DOY) were interrupted for several hours as indicated by the decreases of $N_{d<25\text{nm}}$ below 100 # cm⁻³ (Fig. 3b), coinciding with short and sudden changes in wind speed and

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