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Particle number size distribution in the eastern Mediterranean: Formation and growth rates of ultrafine airborne atmospheric particles

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HIGHLIGHTS

- Particle number concentration was measured at Akrotiri research station (Greece).
- The data were analysed according to the origin of the air masses reaching the site.
- The characteristics of size distributions were analysed.
- New particle formation events were investigated.

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ABSTRACT

Particle number concentration was measured between June 2009 and June 2010 at Akrotiri research station in a rural/suburban region of western Crete (Greece). Overall, the available data covered 157 days during the aforementioned period of measurements. The objectives were to study the number size distribution characteristics of ambient aerosols and furthermore to identify new particle formation events and to evaluate particle formation rates and growth rates of the newborn particles. Aerosol particles with mobility diameters between 10 and 1100 nm were measured using a Scanning Mobility Particle Sizer (SMPS) system. Measurements were performed at ambient relative humidities. The median total particle number concentration was 525 #/cm^3 whereas the number concentration ranged between 130 #/cm^3 and 9597 #/cm^3. The average percentage of particles with diameters between 10 nm and 100 nm (N_{10-100}) to total particles was 53% during summer and spring, but reached 80% during winter. Maximum average contribution of nano-particles (10 nm $< D_p <$ 50 nm) to total particles was recorded also in winter and was attributed partly to the effect of local heating. Furthermore, back trajectories (HYSPLIT model) showed that different air mass origins are linked to different levels of particle number concentrations, with higher values associated with air masses passing from polluted areas before reaching the Akrotiri station. Modal analysis of the measured size distribution data revealed a strong nucleation mode during winter (15–25 nm), which can be correlated with emissions from local sources (domestic heating). The nucleation mode was observed also during the spring campaigns and was partly linked to new particle formation events. On the contrary, an accumulation mode (80–120 nm) prevailed in the measurements during summer campaigns, when the station area was influenced by polluted air masses arriving mainly from Eastern Europe. In total, 13 new particle formation events were recorded during the 157 days of measurements. Nucleation events were associated with low values of N_{100} particle number concentration and reduced coagulation sinks. Mean growth and formation rates were calculated and showed values equal to 6 nm hr^{-1} and 13 cm^{-3} s^{-1}, respectively.

1. Introduction

Atmospheric aerosols influence substantially life on Earth. Climate change, human health, visibility and air quality are affected by atmospheric particles. More precisely, particles with aerodynamic

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diameter below 1 μm are affecting the radiative budget in the troposphere (Seinfeld and Pandis, 2006) and penetrate deep into the human respiratory tract and affect pulmonary health (Housiadas and Lazaridis, 2010; Lazaridis et al., 2001). Moreover, particles with diameter lower than 20 nm (nucleation mode) can be generated in the atmosphere due to nucleation processes. New particle formation can be an important source of cloud condensation nuclei (CCN) and thus influence climate and water cycling (Seinfeld and Pandis, 2006; Sun and Ariya, 2006). The formation of new particles is followed by condensational growth up to the 50–200 nm size range and occurs in almost any part of the troposphere (Holmes, 2007; Kulmala et al., 2007b; Kulmala and Kerminen, 2008). Recent studies showed that nucleation events can occur during the whole year at the Mediterranean area. Such events were recorded both at urban and remote sites (Dall'Osto et al., 2012; Pikridas et al., 2012; Ždímal et al., 2010; Reche et al., 2011).

The atmospheric aerosol distribution depends on the type of a region (urban, rural continental, remote continental, maritime, polar, desert) and on processes occurring at the area and result to the emission, removal or size change of local aerosol particles. Each mode has homogeneous origin and derives from a specific process (e.g. nucleation, condensation, emission). Typically, the aerosol size spectra is characterized by two modes, but unimodal size distributions are also common. Three or four modes in a distribution are rare in the ambient atmosphere (Seinfeld and Pandis, 2006).

New particle formation bursts can occur on a large scale. Several mechanisms have been suggested to explain nucleation in the atmosphere (Jung et al., 2006). These include binary homogeneous nucleation of sulphuric acid – water (Vehkamäki et al., 2002), ternary homogeneous nucleation of sulphuric acid – water – ammonia (Kulmala et al., 2000; Lazaridis, 2001), ion-induced nucleation (Laakso et al., 2002; Lee et al., 2003; Winkler et al., 2008), nucleation of organic vapours (Zhang et al., 2004) and heterogeneous nucleation (nucleation on pre-existing particles) (Lazaridis and Kulmala, 1992; Lazaridis et al., 1991; Winkler et al., 2008). Furthermore, different particle formation mechanisms might be prevailing in different environmental conditions (Dal Maso et al., 2005; Lehtipalo et al., 2009; Rimnacova et al., 2011; Yoon et al., 2006; Vuollekoski et al., 2010).

The last 20 years researchers have managed to identify and reliably measure particles with electrical mobility diameters below 20 nm. Recently, new condensation particle counters (CPCs) have been developed, so clusters with diameter below 3 nm can be measured (Kulmala et al., 2007a; Sipilä et al., 2008), even though the cluster concentration does not seem to be the main critical factor determining whether a nucleation burst occurs (Lehtipalo et al., 2009). On the other hand, the concentrations of sulphuric acid, ammonia, water and organic compounds, precursors to particle nucleation and growth, are believed to be important factors (Boy et al., 2008; Jeong et al., 2010; Kulmala et al., 2005; Paasonen et al., 2010).

The current paper presents an analysis of measured particle number size distributions during several measurement campaigns conducted between June 2009 and June 2010, at Akrotiri station, Crete, Greece. The main objectives of this work were to identify the levels of particle number concentration in the region and correlate them to the possible particle sources, to study the average atmospheric particle size distributions for Nucleation, Aitken and Accumulation modes, to investigate the occurrence of nucleation events (NEs) and to evaluate particle formation and growth rates.

2. Materials and methods

2.1. Measurement site – campaigns

Measurements were conducted at Akrotiri station, a coastal site in the northwest part of Crete, Greece. Crete is located in the middle of east Mediterranean. Detailed description of the Akrotiri research station can be found in Lazaridis et al. (2008) and Kopanakis et al. (2012). The measurement site is approximately 5 Km from the city of Chania (108,000 inhabitants) and 140 Km from the city of Heraklion (173,000 inhabitants). A weather station is located at the measurement site and meteorological data for temperature, humidity, wind speed and wind direction, solar radiation and rainfall are recorded at a 10 min time interval. There are no major industrial facilities in the area, except of a power plant placed within 10 Km from the station. The weather in the area is characterized by hot dry summers and mild winters with low precipitation (401–662 mm yearly average values for the last decade). Clear sky conditions prevail most of the days while mean temperatures for winter and summer are 12 ± 3.1 °C and 26 ± 3.59 °C respectively. The yearly average relative humidity is 65 ± 12% with minimum average values during summer (56 ± 14%) and maximum average values during winter (71 ± 11%).

Particle number size distributions for aerosols with mobility diameters between 10 and 1100 nm (scanned range) have been measured during several campaigns between June 2009 and June 2010. A Scanning Mobility Particle Sizer (SMPS) system, consisting of a Condensation Particle Counter (CPC) and a Differential Mobility Analyzer (DMA) (SMPS, Model 5.401, Grimm Inc., Germany), was used to measure the particle size distributions. Particles sizes were scanned every 406 s into 43 consecutive size bins. The entire system is automated and is equipped with customized software, provided by Grimm, which was used for the inversion of the raw size distributions and for exporting the data to other applications. The inlet of the system was placed 2.2 m above the ground and a cover was used to protect it from the rain. The SMPS system was located inside a laboratory container, which was air conditioned. The indoor temperature was 22 °C during all measurements campaigns. Sheath air was circulated in a closed loop through a drying system, designed to keep relative humidity below 20%. Aerosol measurements were conducted at ambient relative humidities. All the collected data were examined for flawed measurements due to instrumentation malfunctions, which were removed from the data set. Diffusional losses were calculated according to Hinds (Hinds, 1999) and the penetration efficiency was 96% for particles with diameters below 30 nm, 97% for particles with diameters between 30 nm and 50 nm and more than 97% for bigger particles. On the whole, the collected data covered a period of 54 summer days (38 days between 23 June and 18 August, 2009 and 16 between 2 June and 26 June, 2010), 41 autumn days (between 9 October and 30 November, 2009), 21 winter days (between 12 January and 12 February 2010) and 51 spring days (between 3 April and 31 May, 2010).

2.2. Back trajectories

Back trajectories were studied to reveal the origin of air mass reaching the study area. The trajectories were obtained using the HYSPLIT4 Model (HYbrid Single-Particle Lagrangian Integrated Trajectory), developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2003). The 3-dimensional trajectories were computed for the Akrotiri station (coordinates 35.53 N, 24.06 E). The trajectories were 120 h (5 days) long.

2.3. Data processing – the AMANpsd algorithm

An aerosol algorithm (AMANpsd algorithm) was used to evaluate the modal structure of the measured particle number size distribution data (Ondracek et al., 2009). The algorithm can be used 1) for merging data derived from instruments measuring particle number concentrations in different size ranges or operating under