



On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments

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On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments

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Abstract

The growth properties of nucleation mode particles were investigated. The variation of source rates of condensable vapors in different locations and environmental conditions was analyzed. The measurements were performed in background stations in Antarctica and in Finnish Lapland and Boreal Forest stations (SMEAR I and SMEAR II) as well as in polluted urban sites in Athens, Marseille and New Delhi. Taking advantage of only the measured aerosol particles spectral evolution as a function of time the formation and growth properties of nucleation mode aerosols have been evaluated. The diameter growth-rate and condensation sink have been obtained from the measured size distribution dynamics. Using this growth rate and condensation sink, the concentration of condensable vapours and their source rate have been estimated. The growth rates and condensation sinks were between $0.3\text{--}20\text{ nm h}^{-1}$ and $10^{-4}\text{--}0.07\text{ s}^{-1}$, respectively. The corresponding source rate of condensable vapors varied more than 4 orders of magnitude from 10^3 to over $10^7\text{ cm}^{-1}\text{ s}^{-1}$. The highest condensation sink and source rate values were observed in New Delhi and the smallest values in Antarctica.

1. Introduction

Aerosol particles are ubiquitous in the Earth's atmosphere and affect our quality of life in many different ways. In polluted urban environments, aerosol emissions can affect human health through their inhalation (e.g. Donaldson et al., 1998), whilst globally, aerosols are thought to contribute to climate change patterns (e.g. Charlson et al., 1987). In recent years, considerable effort has been devoted to understanding how aerosols directly affect the Earth's radiation budget by scattering and absorbing incoming solar radiation. Aerosols also affect the radiation budget indirectly by modifying many cloud properties such as their albedo and lifetime. It is generally thought that increases in aerosol concentrations will lead to brighter and more sustained clouds, thus providing additional planetary cooling.

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In order to be able to better understand the health and climatic effects of atmospheric aerosols, the formation and growth processes of atmospheric aerosols should also be better understood (Kulmala, 2003). Nucleation, the formation of ultrafine particles detected at a few nm, and subsequent growth to ~100 nm in 1–2 days, has been observed frequently in the continental boundary layer. Such observations span from northern-most sub-arctic Lapland (Vehkamäki et al., 2004), over the remote boreal forest (Mäkelä et al., 1997; Kulmala et al., 1998, 2001b) and suburban Helsinki (Väkevä et al., 2000), to industrialised agricultural regions in Germany (Birmili et al., 2001) and also to coastal environments around Europe (O'Dowd et al., 1999). The atmospheric new particle formation rates have also been investigated by Weber et al. (1996) and Weber et al. (1997), and the biogenic aerosol formation by Kavouras et al. (1998). A recent overview summarised the formation and growth properties in a global point of view (Kulmala et al., 2004), quantifying especially the formation and growth rates of nucleation events, where available.

The main purpose of this paper is to analyse particle formation and growth events in detail in order to quantify the concentration of condensable vapours as well as their source rates. Some estimations on these quantities are already available (Kulmala et al., 2001a) for the Finnish boreal forest. However, since such a multitude of studies has appeared on particle formation events all over the world (Kulmala et al., 2004), a more thorough analysis on the range of vapour concentrations and source rates is likely to be of interest also, especially for the needs of regional and global modelers.

2. Analytical expressions

The observed nucleation mode growth, the concentration of condensable vapour and its source rate during the nucleation and growth events are analysed using two equations describing the rate of change of vapour concentration and particle growth (see Kulmala et al., 2001a). Considering condensable vapour molecules of some species, the time dependence of the vapour concentration (C) can be expressed (see also Kul-

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mala et al., 1998) by

$$\frac{dC}{dt} = Q - CS \cdot C, \quad (1)$$

where Q is the source rate of the vapour and CS is its condensation sink (see Eq. 4) to the pre-existing aerosol. The growth-rate can be expressed as (Kulmala, 1988)

$$\frac{dd_p}{dt} = \frac{4m_v\beta_m DC}{d_p\rho}. \quad (2)$$

Here d_p is particle radius, m_v is molecular mass of condensable vapour, D is diffusion coefficient, and ρ is particle density. For the transitional correction factor for the mass flux β_M we use the Fuchs-Sutugin expression (Fuchs and Sutugin, 1971). The equilibrium vapour pressure of the condensing species is assumed to be negligible. Equation 2 can be integrated from $d_{p,0}$ to d_p to obtain (see Kulmala et al., 2001a):

$$C = \frac{\rho}{\Delta t D m_v} \left(\frac{d_p^2 - d_{p,0}^2}{8} + \left(\frac{2}{3\alpha} - 0.312 \right) \lambda (d_p - d_{p,0}) + 0.623 \lambda^2 \ln \frac{2\lambda + d_p}{2\lambda + d_{p,0}} \right). \quad (3)$$

Here α is the mass accommodation coefficient (i.e. sticking probability, here assumed to be unity) and λ is the mean free path. The growth rate dd_p/dt and condensation sink CS can be obtained directly from the size distribution evolution measurements.

The aerosol condensation sink determines, how rapidly molecules will condense onto pre-existing aerosols and depends strongly on the shape of the size distribution (see e.g. Pirjola et al., 1999; Kulmala et al., 2001a). The condensation sink CS is obtained by integrating over the aerosol size distribution:

$$CS = 2\pi D \int_0^\infty d_p \beta_M(d_p) n(d_p) dd_p = 2\pi D \sum_i \beta_M d_{p,i} N_i, \quad (4)$$

where $n(dp)$ is the particle size distribution function and N_i is the concentration of particles in the size section i . Now, with the condensable vapor concentration C from

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the Eq. (2) and the condensation sink CS directly from the size distribution evolution measurements, by assuming a steady state vapor concentration one can estimate the source rate Q from

$$Q = CS \cdot C. \quad (5)$$

3. Aerosol size distribution measurements

3.1. Instrumentation

Sub-micron aerosol size distribution measurements are a necessary requirement for condensable vapor and its' source rate estimations. In this study, calculations are based on similar instrumentation operated campaignwise or continuously in various locations. These measurements were conducted in the course of a few years. Due to this and the fact that during the campaigns also the sites themselves imposed demands on the instrumentation setups, differences in the detailed description of the measurement devices arised. However, in all places, a differential mobility particle sizer (DMPS) with a closed loop flow arrangement (Jokinen and Mäkelä, 1997; Aalto et al., 2001) was utilized to obtain size distributions. The time resolution was typically 10 min in which time aerosol from 3 nm in diameter (In Hyytiälä, Athens, Marseille, New Delhi and Antarctica) was sampled. The system used in Värriö had a slightly larger cutsize of 8 nm in diameter. The upper size limit varied between 600 and 1000 nm.

More specifically, a DMPS systems consisted of two parallel devices. The first was used to characterize the the size distrubution of ultra-fine particles (typically 3–20 nm), where the size classification with respect to electrical mobility equivalent diameter was obtained with a short (0.109 m) Vienna-type (Winklmayr et al., 1991) Differential Mobility Analyzer (DMA). The particles were counted with a TSI 3025 Ultra-fine Condensation Particle Counter (Stolzenburg and McMurry, 1991). The other half of the DMPS-system was dedicated to Aitken and accumulation particles (typically 20–700 nm). It

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comprised of a longer (0.28 m) Vienna-type DMA and a TSI 3010 Condensation Particle Counter (Quant et al., 1992). Prior to the size classification, aerosol particles were exposed to a radioactive β -source, which ensured the Boltzmann charge equilibrium in the aerosol population and enabled particle classification based on their electrical mobility. Total sub-micron aerosol number concentrations were obtained from the integrated size distributions. Sheath flows in the DMAs adjusted at each site to yield the most useful measurement size range and time resolution. Typically the sheath flows varied between 5 to 10, and 10 to 25 l min⁻¹ for long and short DMA, respectively.

The aerosol sampling inlet of the DMPS systems was located at 2 m height above ground at Hyytiälä and Värriö stations. In the campaigns, the inlet structure extended up to 3 m height above the surface (Marseille and Athens). The DMPS system in New Delhi was placed in the 5th floor next to a window. The inlet tube was placed outside of the window so that the inlet was 15 m above the ground level and 0.5 m from the wall of the building. In Antarctica, the inlet was placed 2.5 m above the container roof, which was about 3 m above the ground. The sample air was led through a vertically-placed, 60 mm steel tube with a total flow of 26.5 l min⁻¹. The sample was taken from the main flow and led to the DMPS system through a 30 cm long stainless steel tube having a diameter of 6 mm.

The aerosol size distribution was obtained from the raw data through an inversion method, which included charging probabilities according to Wiedensohler (1988) and DMA transfer functions by Stolzenburg (1988) as well as counting efficiencies of the particle counters based on Mertes et al. (1995) for CPC 3010 and laboratory calibrations for CPC 3025 (Aalto, 2004). Losses in the inlet tube were estimated based on laminar flow diffusion theory (e.g. Brockmann, 2001). In the Marseille and Athens campaign an external permapure drier was used to remove excess water from the aerosol particles. This ensured that the sampled aerosol population went through the sizing system as dry particles. This, in turn, increased losses in the sampling lines. The effect of the drier was not taken into account in the Athens data analysis.

During the Marseille and Athens intensive field campaigns, supermicron aerosol size

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distributions were measured with an Aerodynamic Particle Sizer (APS-3320, TSI Inc, USA). This instrument extended the measurement range up to approximately 10 μm (Armendariz and Leith, 2002). The APS, however, used aerodynamic size as a basis of the classification whereas the DMPS used the electrical mobility equivalent diameter.

5 They are related to each other through an unknown density of the particles. Combined size distributions from these two instruments was obtained through fitting the DMPS and the APS size distributions in a least-squares sense so that the largest sizes of the DMPS matched the lower boundary of the APS size distributions. The APS was not used in other locations, which therefore narrowed the observed size range. However, 10 the effect of larger particles to the condensable vapor concentrations and source rate estimations were typically less than 5% in Hyytiälä and coastal Mace Head site in Ireland (Dal Maso et al., 2002).

3.2. Measurement sites

The aerosol size distribution measurements related to this study with DMPS-systems 15 were conducted at six different locations: from Finnish Arctic (Värriö) to Boreal forest (Hyytiälä) and further south to the Mediterranean (Athens and Marseille) and India (New Delhi). The southernmost measurement site was located in Antarctica (Finnish research station Aboa). Such variety in studied environments enabled us to achieve maximal variability both in the condensable vapour concentrations and source rates 20 of these vapours. Data was collected either campaignwise (Athens, Marseille, New Delhi and Antarctica) leading to datasets of 3–4 weeks or utilizing stable measurement facilities (Hyytiälä and Värriö), where the amount of accumulated data is several years.

The SMEAR I station is located in Värriö (67°46' N, 29°35' E), 250 km north of the Arctic circle in Eastern Lapland, less than 10 km from the Finnish-Russian border. The 25 DMPS measurements were done on top of a hill 390 m above sea level (a.s.l.). There are no towns or industry close by, and thus practically no local pollution. The nearest major pollution sources are Montschegorsk located 150 km east and Nikel located 190 km north of the station. The measured air is representative of the arctic boreal

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forest background when the air is not coming from these source areas.

The SMEAR II -station is located in Hyytiälä, Southern Finland (61°51' N, 24°17' E, 181 m a.s.l.). The terrain around the station is representative of the boreal coniferous forest. The 40-year old Scots pine (*Pinus sylvestris* L.) dominated stand is homogenous for about 200 m in all directions, extending to the north for about 1.2 km. The largest city near the station is Tampere, ca. 60 km S-SW of the measurement site. The terrain is subject to modest height variations. A more detailed discussion and evaluation on the aerosol instrumentation used in the SMEAR II station is given in [Aalto et al. \(2001\)](#); [Kulmala et al. \(2001a\)](#), and in the SMEAR I station by [Vehkamäki et al. \(2004\)](#).

The measurement site in Athens was located in Thrakomakedones (38°8'37" N, 23°45'29" E, 550 m a.s.l.). The site was located approximately 20 km north from the city centre of Athens, Greece in the foothills of mountain Parnitha. It is surrounded by suburban areas in the south and forests in the north. Due to the proximity of urban areas, the site can be categorized as urban background station. The measurement campaign in the Athens area was conducted between 10 and 26 July 2003.

The Marseille dataset was obtained through a campaign held in Plan d'Aups village (43°19' N, 5°42' E, 700 m a.s.l.) approximately 40 km northeast from the city centre of Marseille, France. The measurements were conducted between 1 and 19 July 2002.

In India the field experiment was performed at India Habitat Centre (IHC/TERI) (28°35' N, 77°12' E, 218 m a.s.l.) in New Delhi from 26 October to 9 November 2002. The measuring site was located next to a traffic line in a residential area a few kilometers south from one of the city centers (Connaught Place). For more details, see [Mönkkönen et al. \(2004\)](#).

In Antarctica measurements were conducted at Finnish Antarctic station, Aboa (73°3' S, 13°25' W), between 5 and 22 January 2000 and from 1 to 26 January 2001. Aboa is located on nunatak Basen some 130 km from the coast line of Antarctica and 480 m above the sea level. The measurement site is described in more detail by [Koponen et al. \(2003\)](#).

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4. Results and discussion

Four aerosol formation event days during measurement campaigns in Antarctica, Athens, Marseille and New Delhi as well as two days from continuous measurements performed at SMEAR I and II are presented in Figs. 1–6. In all these days, the sub-micron size distribution data showed a clear increase in the small-particle (<10 nm) concentration during the late morning, followed by the subsequent growth of these particles into Aitken and accumulation mode sizes throughout the afternoon and evening. The evolution of the size spectra illustrates thereby the growth of the nucleation mode up to sizes of the order of 50–100 nm over periods of about 10 h. For each event period, the start and end times of the event, the nucleation mode growth rate and condensation sinks were determined from the experimental data.

During the observed particle formation events, the condensation sink was usually higher in more polluted areas. The highest value of CS ($5\text{--}7\cdot 10^{-2}\text{ s}^{-1}$) was encountered in New Delhi, while in the European cities Athens and Marseille CS was significantly (5–10 times) lower (Table 1). In non-polluted areas like Värriö (SMEAR I) and Antarctica, the values of CS were some 50–100 times lower than those observed in New Delhi. Condensation sinks measured during the Hyytiälä (SMEAR II) events were on average somewhat larger than those during the Värriö (SMEAR I) events.

The typical submicron DMPS data correspond to “dried” dehydrated aerosol sizes, while the supermicron distributions were taken at ambient humidity corresponding to ambient “wet” aerosol sizes. Consequently, the response of the ambient aerosol to humidity-induced growth will also be reflected in the calculated condensation sink. The effect of particle hygroscopic growth factors on the calculated condensation sink has been investigated by Kulmala et al. (2001a). In the present study we have used dry condensation sinks, resulting in some uncertainty is involved in our estimations. However, the typical overall underestimation of CS is between 5 and 50%, which is minor considering the large differences in its magnitude between the clean and polluted environments.

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Using Eq. (2) we can estimate the vapor concentrations needed to explain the observed condensational growth and, furthermore, the source rate of the condensable vapor. These estimations depend, however, on the assumptions made for the properties of this condensable vapour, such as its molecular mass, its gas-phase diffusion coefficient and its mass accommodation coefficient on particle surfaces (see Kulmala et al., 2001a, for details). Luckily, the needed vapor concentration is only sensitive to the mass accommodation coefficient, since the product of the diffusion coefficient and mass of condensing molecules is practically almost constant. In any case, if proper rigorous condensation theory is used like in our analysis, the mass accommodation coefficient of unity can be used. This is recently experimentally shown by Winkler et al. (2004). However, even the effect of mass accommodation on source rate (Q) is negligible (Kulmala et al., 2001a).

New particle formation events have been observed in both clean and polluted environments (Kulmala et al., 2004). In practise, new particle formation events cannot be observed unless the source rate of condensable vapours is high enough to induce a sufficiently large growth rate for the smallest nucleation mode particles. According to our analysis, the required source rates of $>10^7 \text{ cm}^{-3} \text{ s}^{-1}$ in the most polluted environment (New Delhi) are 10–100 times larger than those in the European cities (Athens and Marseille) and roughly four orders of magnitude larger than those for our cleanest environments (Aboa and SMEAR I).

A question that immediately arises is, whether the very large difference in the vapor source rates between the clean and polluted environments is realistic. Potential candidates for the condensable vapours responsible for the growth of nucleation mode particles are sulphuric acid and various organic vapours of low volatility. In the lower troposphere, gaseous sulphuric acid is produced mainly by the reaction of sulphur dioxide (SO_2) with the OH radical. The concentration of SO_2 varies by a more than three orders of magnitude between the very clean and polluted environments (Rotstain and Lohmann, 2002; Carmichael et al., 2003), whereas the concentration of the OH radical depends mainly on the intensity of solar radiation. By combining these things together,

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the source rate for gaseous sulphuric acid might well differ by the required four orders of magnitude between New Delhi and our cleanest environments.

5 The source rates of condensable organic vapours are very difficult to estimate since a large number of precursors for these vapours exist, the relevant precursors probably differ between the different environments, and the formation yields of condensable vapours from their precursors are likely to vary with time and location. Globally the production of secondary organic aerosols is likely to be dominated by biogenic precursors (Griffin et al., 1999; Tsigaridis and Kanakidou, 2003; Lack et al., 2004). Based on this it seems very likely that the source rate of condensable organic vapours does not
10 decrease as fast as the source rate of gaseous sulfuric acid, when going away from the polluted environments. If this turns out to be true, our results would indicate that the contribution of sulphuric acid to the particle growth might be substantially larger in urban environments than in most clean locations. Some support for this view is obtained by comparing the recent results by Boy et al. (2004) and Stanier et al. (2004), showing
15 that sulphuric acid is responsible of 10% and almost 100% of the particle growth in remote forested and polluted environments, respectively.

Observed growth rates of nucleation mode particles were surprisingly similar between the different measurement sites, differing on average by less than an order of magnitude. This can be explained by the close interdependence between Q , CS and emissions. Namely, sources that emit both gaseous (precursors for condensable vapours) and particulate pollutants result in simultaneous increases in both Q and CS .
20 Sources emitting only gaseous pollutants increase Q , which then leads to larger CS via an enhanced gas-to-particle transfer. The fact that Q and CS usually change in concert with each other balances variations in the condensable vapour concentration (Eq. 5) and thereby in the particle growth rate.
25

One interesting feature in our observations is that the growth rate of nucleation mode particles always exceeds 10 nm h^{-1} in New Delhi, and that growth rates $< 1 \text{ nm h}^{-1}$ can only be observed in the very clean environments. The probable reason for this is the strong interplay between the nuclei growth and their loss by coagulation: the larger the

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degree of pollution (larger CS), the faster small nuclei must grow to survive the coag-
ulational scavenging onto larger pre-existing particles (Kerminen et al., 2001; Kulmala
et al., 2004). This feature also explains the larger average growth rates observed in
the cleanest environments.

5. Conclusions

In this study, the formation and growth of aerosol particles in the nucleation mode have
been investigated using novel analytical tools. From the size spectra the diameter
growth-rates and condensation sinks can be calculated. With this information, the
concentration of condensable vapours and their source rate can be estimated. The
analysis has been applied to analysis of aerosol formation events in six different sites,
representing very different air pollution conditions.

The estimated source rate of condensable vapours was found to vary by up to four
orders of magnitude between the most polluted (New Delhi) and the cleanest sites
(Northern Finland and Antarctica) considered here. These high variations were nec-
essary to explain the formation and subsequent growth of new particles in these sites.
Preliminary indications were obtained on that the relative role of sulfuric acid and con-
densable organic vapours in the growth of nucleation mode particles in different envi-
ronments, the sulfuric acid being more important in polluted areas.

Despite large variations in vapour source rates, the growth rate of nucleation mode
particles did not vary by more than 1–2 orders of magnitude between the different sites.
The primary reason for this is that large vapour source rates usually appear in concert
with large condensation sinks, the overall effect of which is to balance the condensable
vapour concentrations. The average particle growth rates increased, however, with
the level of pollution. This can be explained by the fact the low particle growth rates
cannot be observed in very polluted environments due to the effective scavenging of
the smallest growing particles by the pre-existing aerosol population.

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The estimation of vapor source rates and concentrations using this quite straightforward technique agrees well with detailed box model studies (Kulmala et al., 1998, 2000), illustrating the usefulness of this approach in analyzing the particle size distribution data. The difference between polluted and clean environments needed for condensation sinks to obtain proper source rate of vapor molecules and growth rates of nucleation mode particles are also in agreement with detailed aerosol dynamic simulations.

Current estimates on the magnitude of regional and global secondary aerosol formation rely almost entirely on modeling and are subject to large uncertainties. The approach introduced here provides a simple yet reasonably accurate way of calculating condensable vapour source rates based on field data and in very different environments. As such, our approach might be very useful for closure studies in which the vapour source rate is calculated independently using either aerosol measurements or modeled atmospheric chemistry. By combining our approach with suitable chemical measurements, we could ultimately estimate the relative contribution of condensable vapors – particularly of sulfuric acid – to the formation of secondary aerosols in different atmospheric environments.

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Table 1. Observation sites, number of formation and growth events, minimum and maximum of condensation sink (*CS*), growth rate (*GR*), vapor concentration (*C*) and source rate of vapor (*Q*).

| Site | # events | <i>CS</i> [s^{-1}] | <i>GR</i> [nm h^{-1}] | <i>C</i> [cm^{-3}] | <i>Q</i> [$\text{cm}^{-3} \text{s}^{-1}$] |
|---|----------|--|----------------------------------|--|---|
| New Delhi 26 Oct.–9 Nov. 2002 | 8 | $5 \cdot 10^{-2}$ – $7 \cdot 10^{-2}$ | 11.6– 16.0 | $15.8 \cdot 10^7$ – $24.6 \cdot 10^7$ | $0.9 \cdot 10^7$ – $1.4 \cdot 10^7$ |
| Marseille 1–19 July 2002 | 10 | $3.2 \cdot 10^{-3}$ – $1.5 \cdot 10^{-2}$ | 1.1– 8.1 | $1.5 \cdot 10^7$ – $11.1 \cdot 10^7$ | $8.7 \cdot 10^4$ – $1.3 \cdot 10^6$ |
| Athens 10–26 June 2003 | 7 | $5.8 \cdot 10^{-3}$ – $1.3 \cdot 10^{-2}$ | 2.3– 11.8 | $3.1 \cdot 10^7$ – $16.2 \cdot 10^7$ | $2.6 \cdot 10^5$ – $1.6 \cdot 10^6$ |
| Antactica Jan. 2000/2001 | 10 | $2.4 \cdot 10^{-4}$ – $9.6 \cdot 10^{-4}$ | 0.3– 2.7 | $0.4 \cdot 10^7$ – $3.7 \cdot 10^7$ | $0.9 \cdot 10^3$ – $2.0 \cdot 10^4$ |
| SMEAR I Värriö 1998–2002 | 147 | $0.6 \cdot 10^{-4}$ – $3.6 \cdot 10^{-3}$ | 0.8– 10.6 | $1.1 \cdot 10^7$ – $14.7 \cdot 10^7$ | $1.9 \cdot 10^3$ – $4.8 \cdot 10^5$ |
| SMEAR II Hyytiälä winters 1997–2001 | 34 | $2 \cdot 10^{-4}$ – $7 \cdot 10^{-3}$ | 1.3– 5 | $2.1 \cdot 10^7$ – $8.2 \cdot 10^7$ | $5.0 \cdot 10^3$ – $6.9 \cdot 10^5$ |

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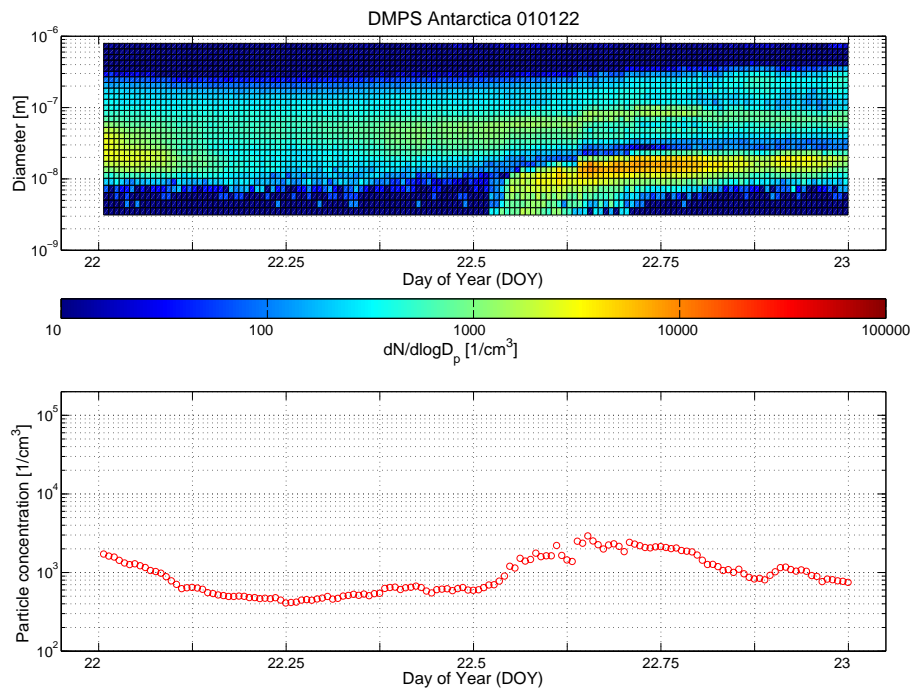


Fig. 1. Diurnal variation of Antarctica size distribution and total number concentration.

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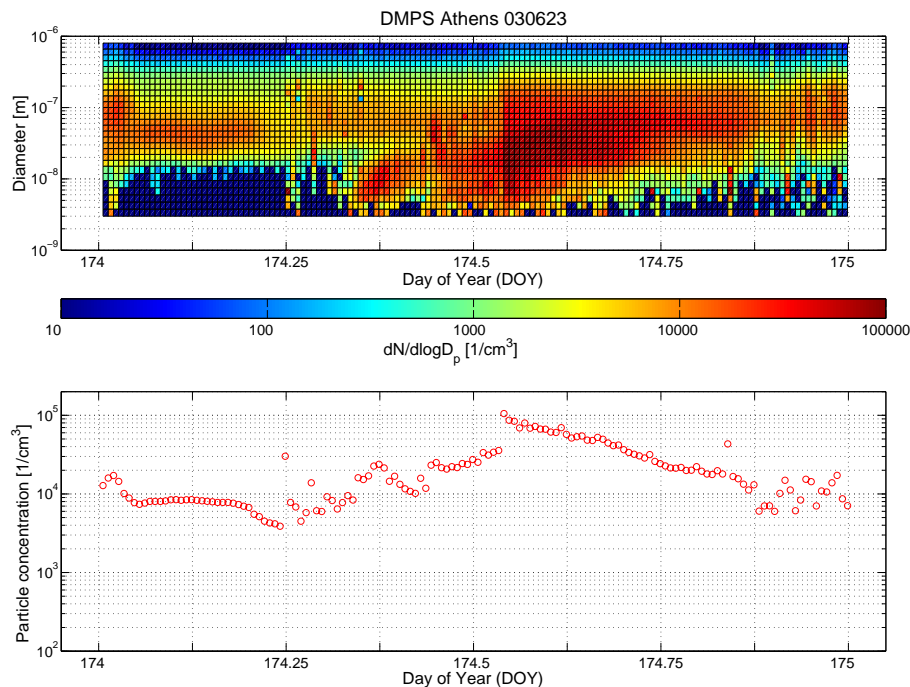


Fig. 2. Diurnal variation of Athens size distribution and total number concentration.

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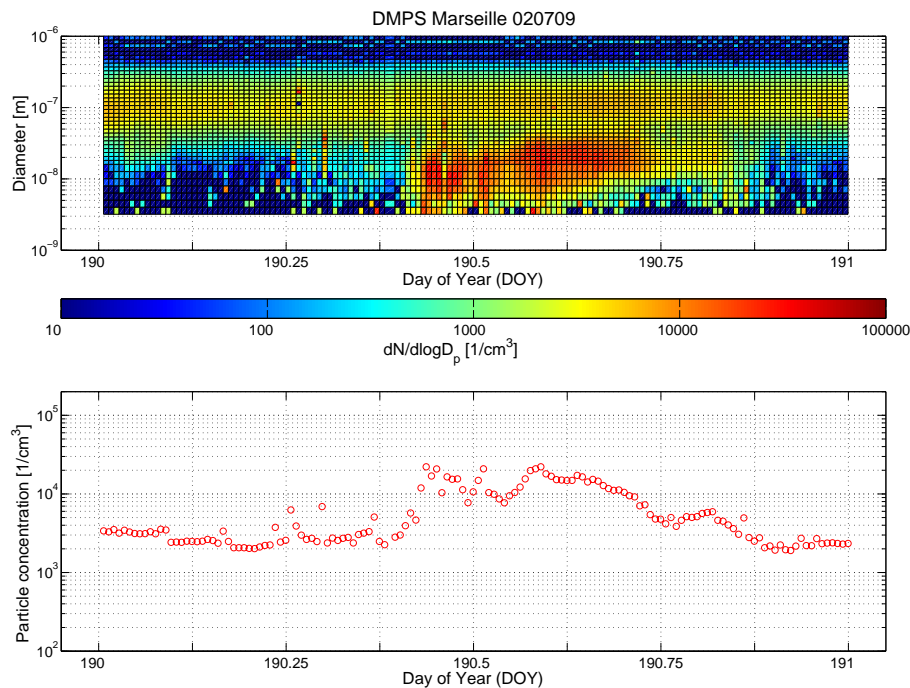


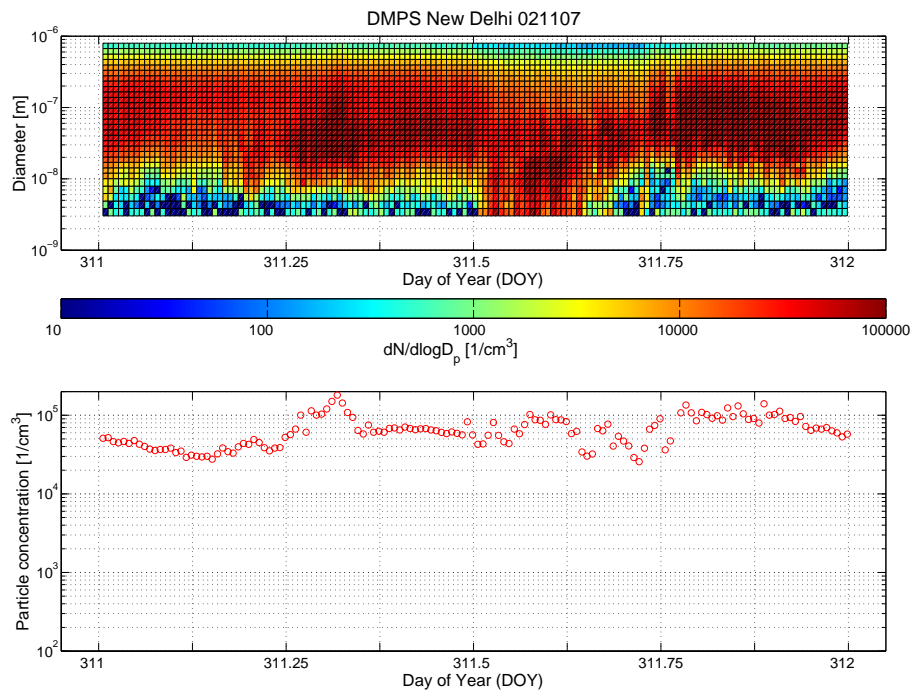
Fig. 3. Diurnal variation of Marseille size distribution and total number concentration.

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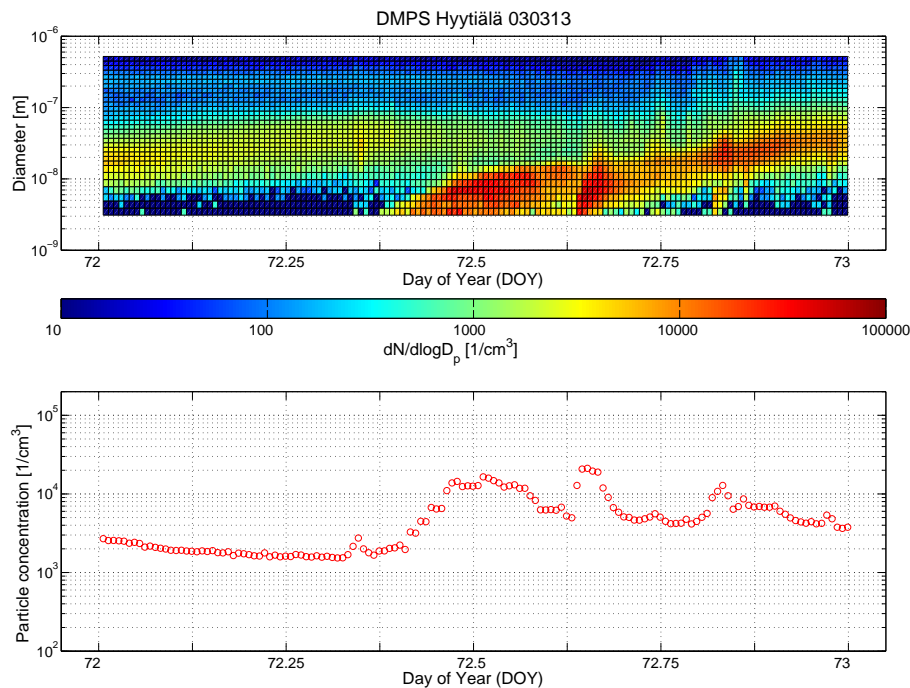
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**Fig. 4.** Diurnal variation of New Delhi size distribution and total number concentration.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

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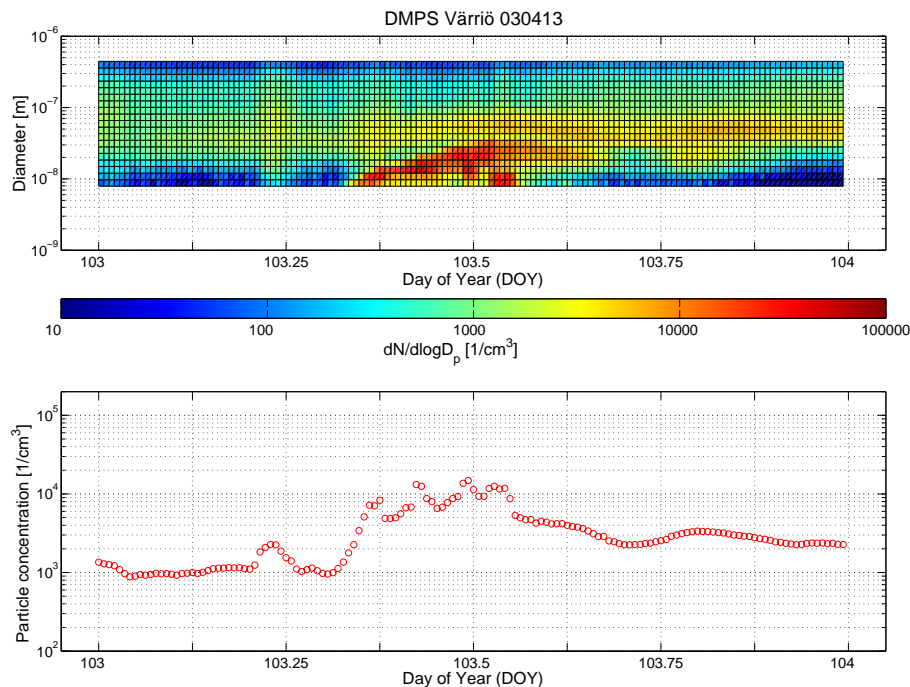


Fig. 6. Diurnal variation of Värriö size distribution and total number concentration.

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