Growth Properties of Submicron Aerosols during Cold Season in India

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Abstract

The growth properties of submicron aerosols were investigated on the basis of particle number size distributions measured in field experiments. An analysis of condensation sink, growth rate, concentration of condensable vapors and their source rate, and real and apparent nucleation rates as a function of particle sizes and their number concentrations in the submicron range are presented to quantify their effects during nucleation events. Higher number concentrations of newly formed particles of 0.013, 0.024, 0.075, and 0.133 μm around 0900 h indicate that photochemistry plays an important role in their formation, and that they grow principally by gas-to-particle conversion. The magnitudes of nucleation parameter η are indicative of the number concentration of newly formed particles by nucleation and their subsequent growth. The estimated value of η is around 10 nm for Aitken mode particles (0.013 and 0.024 μm) and 6 nm for accumulation mode particles ($D_p > 0.1$ μm). The formation rate of 0.013 and 0.024 μm particles is 2.33 $cm^{-3}s^{-1}$ and of intermediate particles (0.075 and 0.133 μm) is 1.33 $cm^{-3}s^{-1}$. The growth rate of 0.013 and 0.024 μm particles is 0.3 and 1 nm h^{-1} and the rate for 0.075 and 0.133 μm particles is 8 and 21 nm h^{-1}, indicating that high formation rate and rapid growth are generally found in semi-urban and urban areas. These results may be of interest for future study of nucleation processes in different environments.

Keywords: nucleation event, nucleated clusters, condensable vapor, fog – formation.

1. Introduction

Aerosols influence the behavior of the atmosphere by acting as cloud condensation nuclei, thus exerting a strong effect on the radiation properties of the atmosphere. In urban environments, aerosol particles can affect human health through their inhalation (Stieb et al., 2002). Aerosol generation

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starts with nucleation in a supersaturated vapor phase. After the nucleation period in the sub-microsecond range, the nuclei grow to detectable size. The most important heterogeneous nucleation processes in the atmosphere involve nucleation of vapor molecules on insoluble and soluble aerosol particles. Nucleation of water vapor on soluble particles is the process by which cloud and fog droplets form in the atmosphere. The driving force for both nucleation and particle growth is the concentration of 'non-volatile" vapors (Kulmala et al., 2004). Kulmala et al., (2004) suggested that potential candidates for such vapors are sulfuric acid (H_2SO_4)and some yet unidentified organic compounds, all formed by oxidation reactions involving suitable precursor gases (SO_2, volatile organic compounds). The growth condition for nucleated clusters changes from the kinetically controlled free molecule regime ($D_p < 0.02$ μm) to the transition regime ($0.02 < Dp < 0.6$ μm), and finally to the well-known diffusion-controlled continuum regime ($0.4 < D_p < 0.6$ μm). Aerosol of 0.01 to 1.0 μm diameter grow principally by gas-to-particle conversion, the process by which vapor molecules diffuse to the surface of a particle and then are incorporated into the particle (Seinfeld and Pandis, 1998). Also, particles in this size range are found in the atmosphere due to vehicular emissions in urban environments (e.g. Zhu et al., 2002).

The formation and growth of new aerosol particles are key processes in atmospheric aerosol particles as demonstrated by numerous aerosol number size distribution and number concentration measurements in several field experiments (Weber et al., 1997, Clark et al., 1999, Kulmula et al., 2001a, 2001b). Particle formation rates have been investigated by Weber et al. (1996; 1997). Kulmala et al. (2001b) have outlined the methodology to estimate the particle growth rate, the condensation sink and concentration of vapors responsible for the particle growth and its source rate based on data of particle size spectra measured in the field experiments. However, an evolution of atmospheric aerosol with nucleation growth and aging is difficult to measure. Monkkonen et al. (2004) reported the results of diurnal variation of number size distributions measured in field experiments and modal parameters for urban background aerosols at New Delhi (India). They estimated formation rate, growth rate, and condensation sink for submicron particles (0.003 to 0.8 μm). However, it is difficult to distinguish between new and pre-existing particles in urban locations, where background particle concentrations are higher (Alam et al., 2003).

Data on continuous measurements of size distribution of particle sizes ≥ 0.003 μm is also useful to determine particle formation and growth rates (Kulmala et al., 2004). Chate and Pranesha (2004) presented the results of field measurements of profiles of atmospheric temperature and particle size distributions for submicron particles during the cold season over Pune, India. In this study, particle number size distributions were measured continuously from 0300 to 1000 h at 1 m above the ground with an Electrical Aerosol Analyzer (EAA Model 3030, TSI Inc.). The smallest particle size observable with the EAA system was 0.013 μm. Generally particles start to be observed at the detection limit of an instrument, which means that they have already grown for some time from the size of a nucleated cluster (0.001 μm). Since the measurement site at Pune was fixed and observed wind speeds remained almost zero until morning, it is assumed that the aerosol must be quite

homogenous in an air mass. Also, Narasimha (1994) reported a temperature minimum in the air layer of 20 – 50 cm above the bare soil on calm, clear nights during the cold season at Pune. Nucleation events are likely to be possible in this kind of atmosphere in winter season at Pune. Kulmala et al. (2004) have reviewed the particle formation and growth rates of ultrafine atmospheric particles on the basis of existing field observational data representing more than 100 investigations in different environments. Based on this review, it is difficult to decide the most relevant nucleation mechanism in the atmosphere. The formation of new particles and their subsequent growth seem to occur almost everywhere. Ground observations in the Amazon rain forest (Zhou et al., 2002) report the regular occurrence of newly formed particles of relatively large diameters (about 0.03 μm), which could indicate an abundant vapor reservoir in that region. Nucleation parameters (growth rate, condensation sink, condensable vapor concentrations, and their source rates, etc.) depend upon the size of the particles. Estimation of these parameters and apparent and real nucleation rates at relatively larger particle sizes ($D_p \geq 0.013$ μm) based on measured size spectra during nucleation events are innovative and will be useful for a better understanding of the health effects. The concentration of condensable vapor and its source rate at different particle sizes is likely to interest global and regional modelers. Apparent and real nucleation rates for each size of the particles are also useful. Theories of nucleation mechanism suggest that nucleated particles are initially on the order of 0.001 μm in diameter, which is too small to measure with the existing measurements techniques. Therefore, in field measurements one actually measures the apparent nucleation rate at which aerosols appear at some larger size as a result of their growth by condensation (Kerminen and Kulmala, 2002). Because critical clusters formed by atmospheric nucleation events can not yet be measured, we were unable to measure the true atmospheric nucleation rate but rather the formation rate of particles of diameters $D_p \geq 0.013$ μm. Direct measurements of apparent nucleation rate and derivation of real nucleation rate as a function of particle size and their number distributions measured in the field experiment are sparse in the Indian region. We present here the particle growth rate, condensation sink, concentration of condensable vapor and its source rate, apparent and real nucleation rates as a function of size of the particles based on data of earlier field measurements (Chate and Pranesha, 2004).

2. Methodology

The measurement techniques, geographical details of the observational site, and Electrical Aerosol Analyzer (EAA Model 3030) system and its accuracy have been discussed elsewhere (Kamra et al., 2003, Chate and Pranesha, 2004). Parameters such as transitional correction factor (β_m) for the condensational mass flux and vapor concentration (C) of condensable vapor are derived for the range of particles between diameter 0.013 and 0.75 μm. Growth rate (GR), condensation sink (CS), and its source rate (Q) are estimated for these particles.

The transitional correction factor for the condensational mass flux – as given by Fuchs and Sutugin (1971) and adopted by Kerminin and Kulmala (2002) and Boy et al. (2004) – is expressed as

$$\beta_m = \frac{1+K_n}{1+0.377K_n+1.33K_n(1+K_n)/\alpha} \tag{1}$$

where α is the mass accommodation coefficient of the condensing vapor, and $K_n = 2\lambda/D_p$ is the Knudsen number with λ representing the vapor mean free path in the air.

The diffusion coefficient for water vapor in air between -40 and 40°C (Pruppacher and Klett, 2000) is expressed as

$$D_v = 0.211(T/T_0)^{1.94}(P_0/P) \tag{2}$$

with $T_0 = 273.15$°K, $P_0 = 1023.25$ mb, T = 293.15°K, P = 950 mb and D_v in cm^2s^{-1}.

Based on these parameters and assuming growth of particles at constant rate, the GR can be written (Kulmala, 1998, Kulmala et al., 2004) as

$$\frac{dD_p}{dt} = \frac{8m_v\beta_m D_v C}{D_p\rho_p} \tag{3}$$

where m_v is the molecular mass of condensable vapor, C is the concentration of vapors responsible for the particle growth, and ρ_p is particle density. Equation (3) can be integrated from D_{p0} to D_p to obtain vapor concentration (Kulmala et al., 2001a, 2004) as

$$C = \frac{\rho_p(D_p^{\,2}-D_{p0}^{\,2})}{32\Delta t D_v m_v} + (2/3\alpha - 0.312)\lambda\frac{(D_p-D_{p0})}{2} + 0.623\lambda^2\ln\frac{(4\lambda+D_p)}{(4\lambda+D_{po})} \tag{4}$$

Here α is assumed to be unity and Δt is the time difference between nucleated cluster formations and their growth from initial diameter D_{p0} to D_p. The GR can be obtained from equation (3) for the range of submicron particles.

The rate of aerosol CS strongly depends on the particle size spectra (Pirjola et al., 1999, Kulmala et al., 2001, 2004) and can be calculated from

$$CS = 4\pi D_v\int_0^\infty D_p\beta_m N(D_p)dD_p \tag{5}$$

where $N(D_p)$ is the particle size distribution function. Parameters of the measured particle number size distributions such as geometric standard deviation ($\sigma_g = 1.4$), count mean diameter (CMD =

0.092 μm) and diameter of the average size of the number concentration (D_{avg} = 0.102 μm) describe the size distribution at Pune over the period of observations (Chate and Pranesha, 2004).

Assuming condensable vapor molecules of some species, the time dependence of the vapor concentration can be expressed (Kulmala et al., 1998, 2004) as

$$\frac{dC}{dt} = Q - CS.C \tag{6}$$

Assuming a steady state vapor concentration, the source rate Q can be estimated from

$$Q = CS.C \tag{7}$$

Parameter η is a measure of number concentration of newly formed particles and can be expressed in terms of condensation sink at a mass accommodation coefficient α = 1 and nuclei growth at constant rate (Kerminen and Kulmala, 2002) as

$$\eta = \frac{\gamma CS_{\alpha=1}}{GR} \tag{8}$$

$$Where \quad CS_{\alpha=1} = \sum_i 0.5 D_{p,j} \beta_m N(D_p) \tag{9}$$

$$\text{and } \gamma = \gamma_0 \left[\frac{D_{nuc,ini}}{0.001\mu m}\right]^{0.2} \left[\frac{D_p}{0.003\mu m}\right]^{0.075} \left[\frac{D_{mean}}{0.1\mu m}\right] \left[\frac{\rho_{nuc}}{1000 kgm^{-3}}\right]^{-0.33} \left[\frac{T}{293K}\right]^{0.75} \tag{10}$$

where γ_0 is equal to 0.23 $nm^2 m^2 h^{-1}$, ρ_{nuc} is the nucleated cluster density and D_{mean} is the number mean diameter of the pre-existing particles' population. Thus, γ is a function of characteristics of aerosols and their size distribution and T is temperature.

In the field measurements of particle size spectra (Chate and Pranesha, 2004), it was possible to measure the particle size distributions of particles ≥ 0.013 μm. Number concentrations of smaller particles (D_p < 0.013 μm) can be estimated from

$$N(D_{nuc,ini}) = \frac{N(D_p)}{\exp(\eta / D_p - \eta / D_{nuc,ini})} \tag{11}$$

where $N(D_p)$ is aerosol number concentrations and $D_{nuc,ini}$ is initial nucleated cluster size.
Real nucleation rate (Kerminen and Kulmala, 2002) is written as

$$J(t)_{real} = \frac{N(D_{nuc,ini})}{\Delta t} \tag{12}$$

Apparent nucleation rate can be expressed as a function of real nucleation rate (Kerminen and Kulmala, 2002)

$$J(t)_{apparent} = J(t)_{real} \exp[\eta / D_p - \eta / D_{nuc,ini}] \tag{13}$$

Thus, apparent and real nucleation rates are obtained as a function of size and number concentrations of submicron particles through equations (8) to (13).

3. Results and discussion

Submicron aerosol distributions in the lowest 1-meter of the atmosphere were measured between 08 – 16 December 1997 and 24 – 28 December 1998 at Pune (Chate and Pranesha, 2004). Particle size distribution measured on 10 December 1997 at six levels (5, 15, 20, 40, 80 and 100 cm) is presented in Figure 1. During this period, particle number concentrations depleted from 0300 h onwards, attained a minimum value between 0600 to 0700 h and thereafter increased. Observed number density of particles of all sizes reached maximum around 0900 h. The minimum concentration of submicron particles during 0600 to 0700 h may be due to the removal of particles, which served as nuclei for the condensation of atmospheric water to form fog-droplets. Particles that were incorporated in the process of fog formation began evaporating after sunrise and were released into the atmosphere. However, estimated percentage contributions of particles in the process of fog formation during 0600 – 0700 h is approximately 1 % for the particles 0.013 and 0.024 μm; 6% for 0.042, 0.075 and 0.133 μm; 10 % for 0.24 and 0.42 μm; and 25 % for 0.75 μm. This indicates that observed excess number concentrations of particle at 0900 h are attributed to the formation of new particles during nucleation events, which always occur during daytime (Kulmala et al., 2004). Thus, photochemistry plays a central role in the formation of new particles after sunrise.

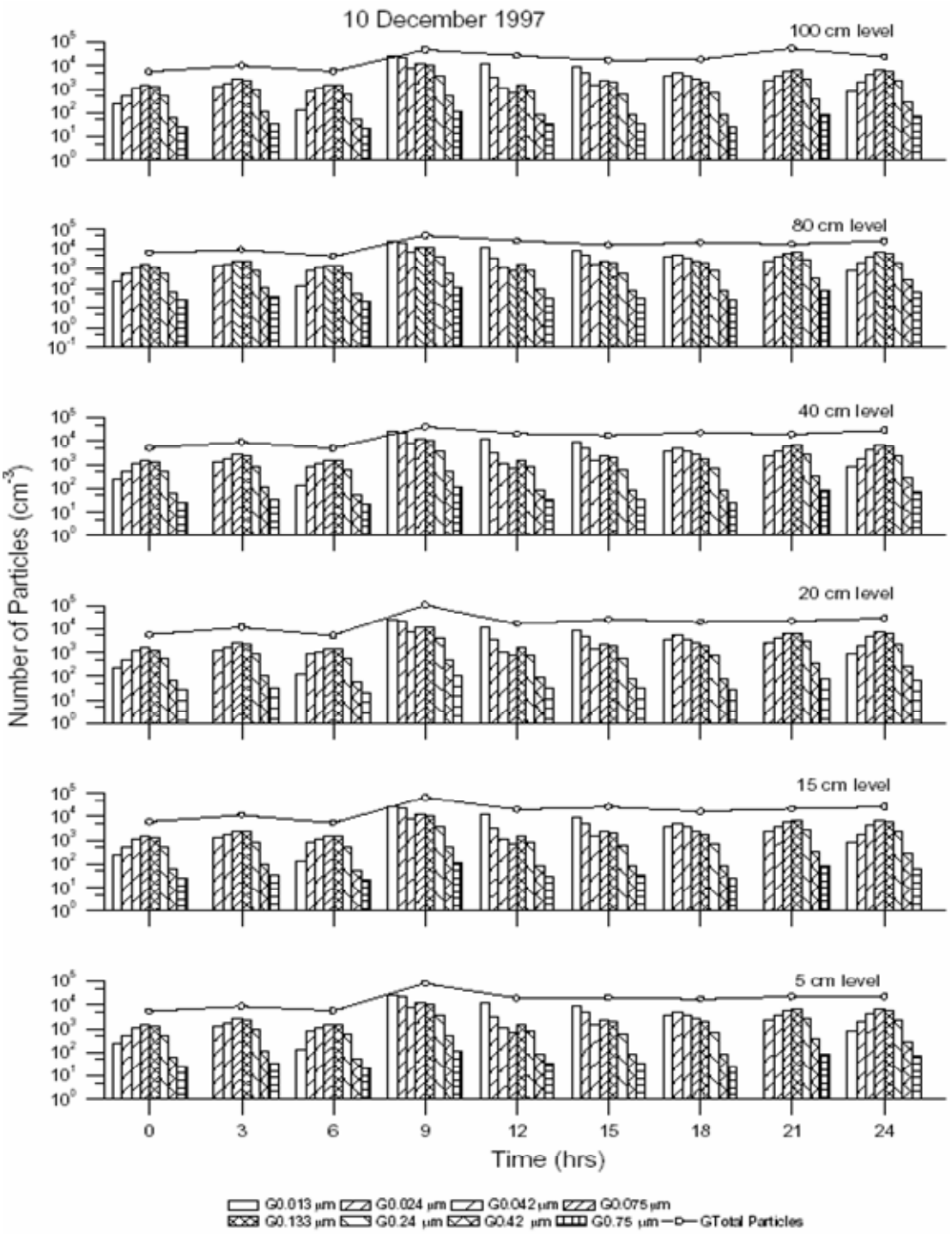

Figure 1. Selected diurnal number size distributions at six levels on 10 October 1997 in Pune.

Parameters of growth events such as CS at an accommodation coefficient equal to unity ($CS'_{\alpha=1}$), GR, and η as a function of particle size are plotted in Figure 2. The nuclei growth rate can be extracted from atmospheric measurements of particle size spectra. Factor η is estimated from $CS'_{\alpha=1}$, γ and GR as a function of particle size and their number distributions. For smaller particles (0.013 and 0.024 µm), $CS'_{\alpha=1}$ is about two orders higher than GR; thus, η approaches 10 nm. Factor η is approximately 6 nm for the particles of intermediate size (0.042, 0.075, 0.133 and 0.24 µm), and for

larger particles (0.42 and 0.75 μm) reduces to a lower value (0.32 and 0.06 nm) [Figure 2(a)]. Figure 2 (b) shows that η is directly proportional to the number density of the particles. Formation of new particles is significantly higher in the case of smaller particles and also for particles of intermediate size, so the value of η determines the number concentration of newly formed particles during nucleation episode. Kerninen and Kulmala (2002) also estimated η above 10 nm for an urban environment. $CS'_{\alpha=1}$ increases with particle size up to 0.24 μm and decreases thereafter, as shown in Figure 2 (c). GR, however, which is a function of concentration of condensable vapor, particle density and size, increases linearly as a function of particle size (Figure 2 (d)). Monkkonen et al., (2004) reported GR between 11.6 to 16 nm h^{-1} of 0.003 μm particle for urban location (New Delhi). We have estimated GR 0.3, 1, 3, 8 and 21 nm h^{-1} of 0.013, 0.024, 0.042, 0.075 and 0.133 μm particles. The discrepancy in GR may be due to large concentrations of pre-existing / background aerosols at a mega-city (New Delhi) compared to Pune (semi-urban), and the estimation of GR for smaller particles (0.003 μm). Other factors responsible for the discrepancy in GR are large vehicular traffic density, different atmospheric conditions, and the observational period at New Delhi.

The rate of aerosol CS determines how rapidly molecules will condense onto pre-existing aerosols and depends on the size of the particles (see Kulmala et al., 2001b). Nucleated clusters grow by condensation at a constant rate. Figure 3 illustrates the variability of the rate of condensation sink, concentration of condensable vapor (C) responsible for particle growth, and its source rate (Q) with particle size. CS is a measure of the rate by which the vapor condenses onto the whole particle population and thus is plotted as a function of number concentrations of the particles (Figure 3 c). As long as the number concentration is higher than 4000 cm^{-3}, CS increases rapidly from 0.64×10^{-2} to 27×10^{-2} s^{-1}, reaching maximum (27×10^{-2} s^{-1}) at a number density corresponding to particle size 0.24 μm and reduces with number concentrations at number density < 4000 cm^{-3} and particle size > 0.24 μm [Figure 3 (c)]. The number of newly formed particles (D_p > 0.24 μm) decreases as the value of CS > 27×10^{-2} s^{-1} because freshly formed aerosols are scavenged away more rapidly. Kerminen et al., (2004) reported CS around 10^{-2} s^{-1} for an urban aerosol system. Antila et al. (2004) pointed out that since the value of CS > 10^{-3} s^{-1} at continental boundary layers, diurnal variation of the temperature has only a minor effect on the time development of saturation ratio and consequently on particle formation taking place over continental areas. Concentration of condensable vapors is directly proportional to the size and inversely related to the number density of the particles. C increases linearly with particle size as the number concentration of newly formed particles reduces with size [Figure 3(d)]. Source rate of condensable vapor is a function of C, and CS assumes a steady state concentration. Figure 3 (b) shows that Q increases almost linearly with size for particles in the range 0.013 to 0.24 μm. Lower values of observed number density of newly formed larger particles ($D_p \geq$ 0.42 μm) and thus CS seem to be responsible for the constant variation of Q with particle size in spite of larger C for these particles. Monkkonen et al. (2004) reported CS between 5×10^{-2} to 7×10^{-2} s^{-1} and C between 15.8×10^{7} and 24.5×10^{7} cm^{-3} for New Delhi. We have estimated CS around 0.6×10^{-2}, 2×10^{-2} s^{-1} and 10×10^{-2} s^{-1} of 0.013, 0.024, 0.042 μm and 0.075 μm particles and C around 0.176×10^{7},

$0.6x10^7$, $18.5x10^7$ cm^{-3} for particle size 0.013, 0.024 and 0.133 μm, which are relatively less than those reported for urban environments (New Delhi).

It is assumed in an atmospheric nucleation event that new nucleated clusters of diameter around 0.001 μm are produced at a rate $J(t)_{real}$ (nuclei cm^{-3} s^{-1}). The relation of formation rate of critical clusters (real nucleation rate, $J(t)_{real}$) to the rate at which they appear at larger sizes (the apparent nucleation rate, $J(t)_{apparent}$) as a result of their growth by condensation determines real nucleation rate, which cannot be measured due to limitations of instruments and measurement techniques. Number concentrations of critical clusters, $J(t)_{apparent}$ and $J(t)_{real}$ derived from the factor η, and number density of newly formed particles and their size are plotted in Figure 4. When the nucleated clusters grow up to observable size by condensation and possibly by self-coagulation, their number density reduces significantly. Thus, the apparent nucleation rate is always smaller than the real nucleation rate [Figure 4 (a) (b)]. Formation rates $J(t)_{apparent}$ and $J(t)_{real}$ decreases with size and increases with number density of the particles as shown in Figure. 4 (a), (b). However, for the particles in accumulation range (0.075 – 0.2 μm), $J(t)_{apparent}$ and $J(t)_{real}$ maintains moderate values due to the higher number density of these particles compared to larger particles ($D_p > 0.2$ μm). Kulmala et al. (2004) have reported in their review paper the formation rates of 0.010 and 0.015 μm particles based on data of field observations conducted by other investigators. The formation rate for 0.015 μm particles worked out by Kulmala et al. (2004) and presented in Table 2 is from 0.5 to 16 cm^{-3} s^{-1}, based on measurements of particle size spectra in sub-tropic rural desert site at Socorro and mid-latitude coastal land at Penmarch (Brittany), respectively. Similarly, the formation rate for 0.010 μm particles estimated by Kulmala et al. (2004) varies from 1 to 1.5 cm^{-3} s^{-1} over mid-latitude coastal land at Preila and from 5 to 40 cm^{-3} s^{-1} in mid-latitude rural land at Hastings (Canada). Our estimated formation rate 2.33 $cm^{-3}s^{-1}$ of 0.013 μm particle seems comparable to those reported by Kulmala et al. (2004) for different environments. If the nuclei are able to grow initially by condensation without decreasing much in number, self-coagulation may be regarded as an important growth process. Potential environments which are significantly affected by self coagulation are certain urban areas characterized as a relatively low amount of background pollution together with high emissions of large nucleation mode particles (0.02 μm) by local traffic (Kerminen et al., 2004). Such atmospheric conditions likely existed at Pune during the field campaign to measure aerosol number size spectra and temperature profiles.

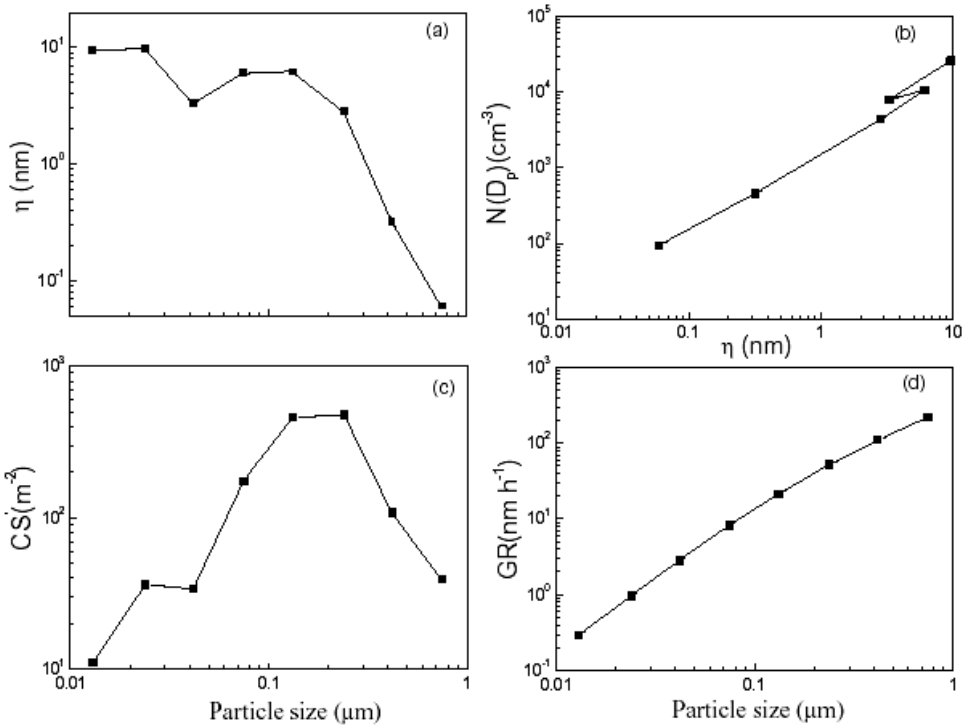

Figure 2. Variability of condensation sink (at $\alpha = 1$) and Growth rates (GR) as a function of particle size and nucleation parameter η with size and number distributions of particles.

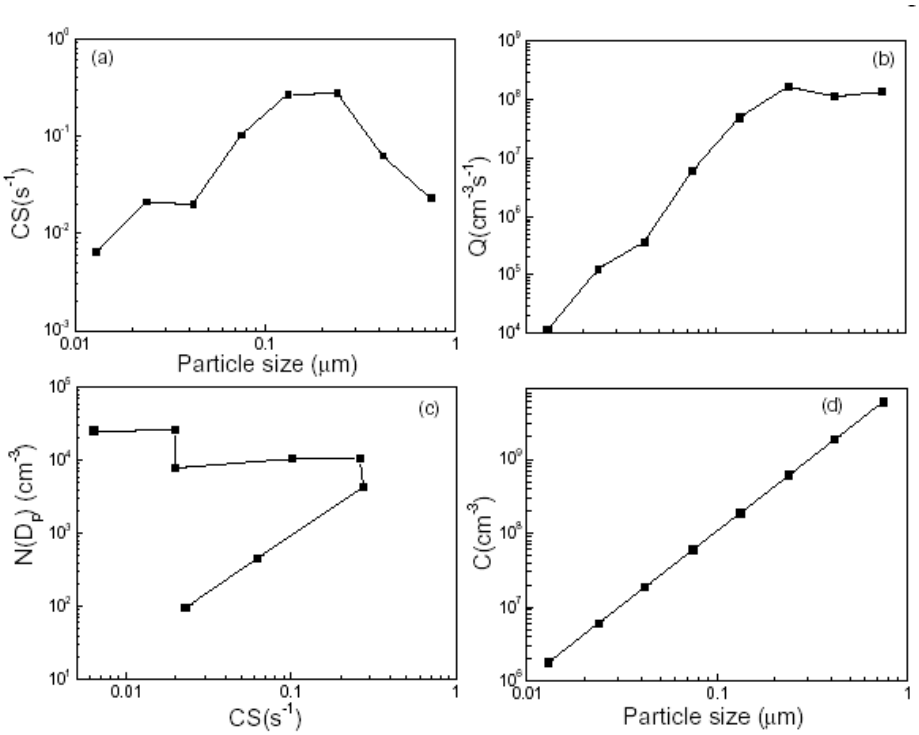

Figure 3. The rate of aerosol condensation sink as a function of size and number concentrations of particles and condensable vapors and its source rates with particles size.

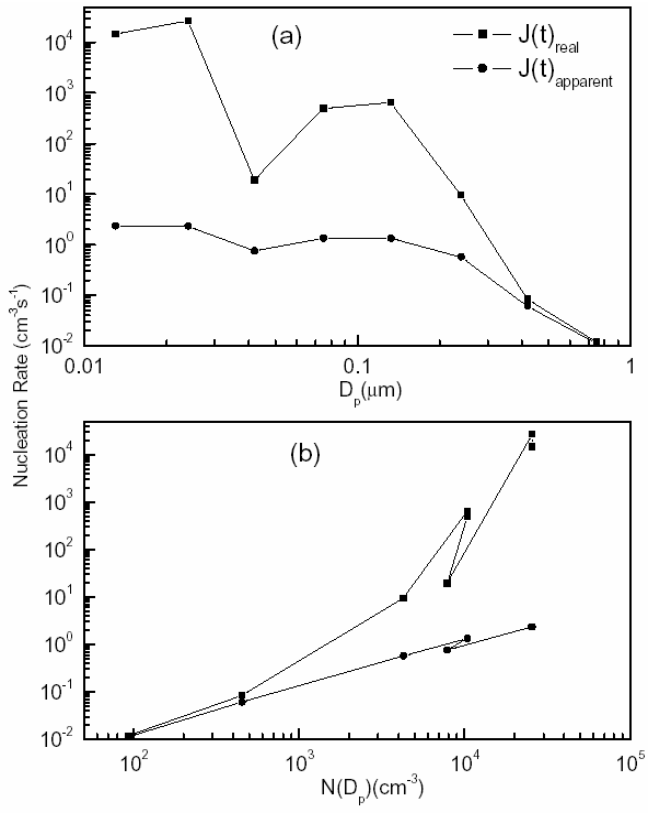

Figure 4. Apparent and real nucleation rates as function of particle size and number concentrations.

4. Conclusions

The production of new atmospheric particles either by homogeneous and heterogeneous and/or other nucleation mechanisms has received much attention from the research community in recent years. In the present study, the diurnal variation of the number size distribution (0.013 – 0.75 μm) at six levels above the ground surface, and nucleation parameters for submicron particles based on these observations are presented for the first time. Measurements of number size distributions were conducted during calm, clear nights when observed wind speeds were almost zero so that changes in number concentrations of particle size distribution could be expected due to nucleation mechanisms. Computations of growth rates, condensation sink, concentration of vapors responsible for particle growth and its source rate, real and apparent nucleation rates were performed for the submicron particles (0.013 – 0.75 μm). The formation rates of 2.33 cm^{-3} s^{-1} for smaller particles (0.013 and 0.024 μm), 1.33 cm^{-3} s^{-1} for intermediate particles (0.075 and 0.133 μm) and 0.06 and 0.01 $cm^{-3}s^{-1}$ for larger particles (0.42 and 0.74 μm) indicate rapid growth and high formation rate typical of semi-urban and close to urban locations. However, it must be cautioned that, results of nucleation parameters based on the preliminary measurements of particle size spectra at Pune may not be adequate because the measuring campaign was short. Further efforts are warranted to identify the nucleation events in the Indian region during cold season and sufficient data of particle size spectra with meteorological

parameters (relative humidity etc.) need to be examined to confirm the findings of this study. Relative humidity (RH) affects primarily the water uptake of clusters and larger particles. The pre-existing particle population absorbs more water as RH increases and they become a larger sink for condensing vapors, newly formed particles and nucleated clusters (Anttila et al., 2004). However, there are still number of practical problems in dealing with the aerosols associated with the atmospheric nucleation events. For example, the actual nucleation mechanism remains to be identified. Future work should include continuous observations of aerosol particles size distributions in different environments, measurements of gaseous compounds participating in nucleation and growth, and the determination of chemical composition of nucleated particles.

Acknowledgements

Authors express their gratitude to Dr. G. B. Pant, Director, IITM, Pune for encouragement. Thanks are also due to anonymous reviewers for their useful comments / suggestions and an American technical editor for editing the manuscript, which improved the scientific contents of the paper significantly.

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Received for review, March 16, 2005
Accepted, May 19, 2005