

Volatile properties of atmospheric aerosols during nucleation events at Pune, India

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Continuous measurements of aerosol size distributions in the mid-point diameter range 20.5–500 nm were made from October 2005 to March 2006 at Pune (18°32'N, 73°51'E), India using Scanning Mobility Particle Sizer (SMPS). Volatilities of atmospheric aerosols were also measured at 40°, 125°, 175°, 300° and 350°C temperatures with Thermodenuder–SMPS coupled system to determine aerosol volatile fractions. Aerosols in nucleated, CCN and accumulated modes are characterized from the measured percentage of particles volatilized at 40°, 125°, 175°, 300° and 350°C temperatures. Averaged monthly aerosol concentration is at its maximum in November and gradually decreases to its minimum at the end of March. The diurnal variations of aerosol concentrations gradually decrease in the night and in early morning hours (0400–0800 hr). However, concentration attains minimum in its variations in the noon (1400–1600 hr) due to higher ventilation factor (product of mixing height and wind speed). The half an hour averaged diurnal variation of aerosol number concentration shows about 5 to 10-fold increase despite the ventilation factor at higher side before 1200 hr. This sudden increase in aerosol concentrations is linked with prevailing conditions for nucleation bursts. The measurement of volatile fraction of ambient aerosols reveals that there are large number of highly volatile particles in the Aitken mode in the morning hours and these volatile fractions of aerosols at temperatures <150°C are of ammonium chloride and ammonium sulfate, acetic and formic acids.

1. Introduction

Nucleation is a gas-to-particle conversion process in which solid or liquid aerosol particles nucleate directly *via* gas phase species. In recent years, this process is considered to be a significant source of atmospheric aerosols in the global troposphere (Spracklen *et al* 2006). It is also an important process in the chain reactions that lead to cloud formation, but the mechanisms are poorly known because of the involvement of various species. For instance, current nucleation mechanisms are involving species like water and sulphuric acid

(Vehkamäki *et al* 2002), water-sulphuric acid-ammonia (Merikanto *et al* 2007), ion-induced nucleation (Modgil *et al* 2005; Yu 2006, 2010), combined ion-induced and sulfate aerosols (Kazil and Lovejoy 2007), and sulphuric-acid vapour (Riipinen *et al* 2007; Kuang *et al* 2008). Therefore, it is not yet possible to know with certainty which species are most involved in nucleation bursts and what will be the compositions of nucleated particles (Weber *et al* 1997; Kulmala *et al* 2004; McMurry *et al* 2005). Species responsible for the growth of nucleated particles may be different from those that participate in nucleation. Also,

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nucleation processes were assessed by number concentration because that correlates with the prevalence of nucleation mode particles.

The volatile property of atmospheric aerosols is important especially in the nucleation process and significant fractions of atmospheric aerosol numbers are of highly volatile and semi-volatile compounds. These compounds exist in large amount in both the gas and particulate phases (Kanakidou *et al* 2005). The most important semi-volatile inorganic aerosol compounds are ammonium nitrate and ammonium chloride whose atmospheric behaviour is thought to be well documented (Seinfeld and Pandis 2006). Volatility is a distinct characteristic of chemical compounds which can be utilized to infer the chemical compositions of ambient aerosols (Kalberer *et al* 2004; Philippin *et al* 2004). Measurements of the volatility of atmospheric aerosols suggest that thermal analysis can be used to examine the chemical composition of nucleated particles, cloud condensation nuclei (CCN) and accumulation mode particles (Wehner *et al* 2004; Kuhn *et al* 2005; Huffman *et al* 2008, 2009).

To the authors' knowledge, there are no observations related to chemical characterizations of ambient aerosols in nucleation, CCN and accumulation modes associated with nucleation bursts in India. In this paper, an attempt has been made to infer compositions of nucleation and Aitken mode particles on the basis of volatility measurements. Pune being an urban site with moderate levels of precursor gases and incoming solar radiations in the morning hours which are essential for

photochemically driven nucleation and growth of nucleated particles, made it possible to infer chemistry of associated particles during nucleation events (Murugavel and Chate 2009). We analyse the measured aerosol size distributions and their volatilities at various temperatures using Thermodenuder–SMPS coupled system. An experimental investigation by inferring composition of nucleation, Aitken and accumulation mode particles from measurements of volatility of size-classified aerosols may be useful to understand the nature of species involved in nucleation and growth processes. Also, characterization of total aerosols are made from comparisons of measured volatilities at temperatures $\leq 150^\circ\text{C}$ as highly volatile, $150^\circ\text{--}300^\circ\text{C}$ as moderately volatile and $\geq 300^\circ\text{C}$ corresponding to less volatile aerosol species.

2. Experimental setup and methodology

A thermodenuder (TSI Inc., USA, model 3065) coupled with a Scanning Mobility Particle Sizer (SMPS, model 3936 of TSI Inc, USA) system have been used for measurements of size resolved thermal properties of aerosol number concentrations. SMPS with Long Differential Mobility Analyser (LDMA) was operated in low flow mode to measure the aerosol number concentration in the mid-point diameter range of 20.5–500 nm. Thermodenuder is used to remove the volatile portions of the aerosols. It consists of a heating tube and a gas adsorption tube (figure 1a). The heating tube is equipped with a microcontroller to adjust the

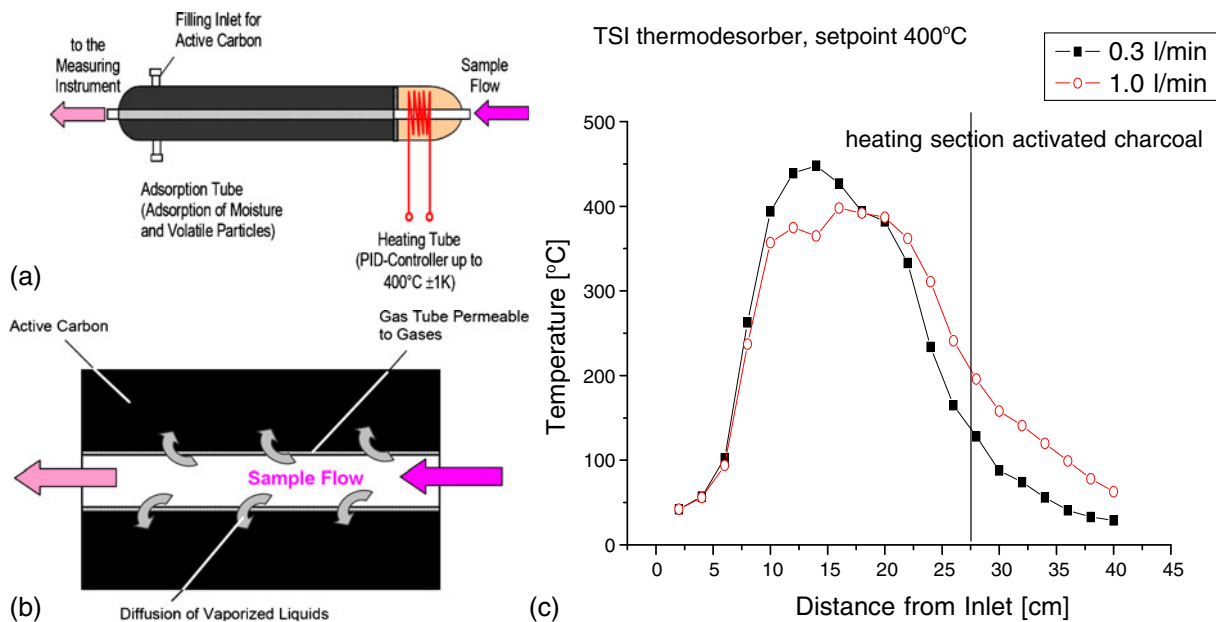


Figure 1. Thermodenuder (TD) system: (a) schematic of TD tube, (b) diffusion of vapour in to active carbon after passing the air sample heated by heating section, and (c) temperature profile inside the TD tube (these figures are reproduced with permission from TSI, Inc, USA (a, b) and from Dr Martin Fierz (c) through personal communication).

temperature from ambient to 400°C with $\pm 1^\circ\text{C}$ accuracy. The gas adsorption tube is made up of a smooth glass tube permeable to moisture/volatile vapours with an outer glass tube filled with active carbon. Air-sample is heated to preset temperature in the heating section and then the heated gas flows through the adsorption tube. The vapourized materials in this heated gas are removed by means of diffusive transportation processes (figure 1b) with the active carbon filled inside the adsorption tube. Since there is no direct contact with the adsorptive material, the primary aerosol is not affected inside the adsorption tube. Sampling conditions with glass sections within the thermodenuder may cause particle loss for charged particles and this may lead to a minor source of error in the measurements. Figure 1(c) shows the temperature profile inside the glass tube with respect to the distance from heating section for different flow rates. The vapourized gas is absorbed by the activated carbon as it passes in the adsorption tube and the air sample cools to ambient temperature at ~ 40 cm from the heating section. This ensures that the air sample fed in to SMPS after removing the volatile portion is at ambient temperature. The residence time inside heating tube for TSI-thermodenuder varies with flow rate. A laboratory thermodenuder can be operated for a wide range of residence time and temperature (Woo *et al* 2007).

SMPS, thermodenuder and a data logging PC are housed inside the observatory room. The inlet for air sampling is taken with a 2 m low-loss silica tube passing through the wall of the observatory and connected to an outside point which is 2 m away from the wall and 1.2 m above the ground surface. The silica tube was regularly cleaned with alcohol to avoid any contamination. The flow rates of sample air, sheath air and total air were adjusted to the desired values at the time of warm-up mode run. The SMPS automatically stops/reports if the flow rates and temperature ranges deviate beyond the tolerance limit.

Two sets of experiments are conducted in this study. (1) Continuous measurements of aerosol size distributions were made from October 2005 to March 2006 using SMPS system with time resolution of 3 minutes a sample. Such measurements are essential to estimate nucleation and growth rates. (2) Volatility of aerosols is also measured at ambient, 40°, 125°, 175°, 300° and 350°C temperatures at 0830, 1130, 1430, 1730 and 2030 hr, using Thermodenuder–SMPS setup. The reduction in aerosol number concentrations while exposing the air sample to higher temperatures is calculated from these measurements in order to quantify their volatile fractions. Since only one set of SMPS and thermodenuder is available with us, experiments (1) and (2) were not made simultaneously. To

complete one set of volatile measurement at all the temperatures, it takes 35–40 min and any change in the air mass over this period is assumed to be uniform.

3. Topography and meteorology of the experimental station

The experimental station, Pune ($18^\circ 32' \text{N}$, $73^\circ 51' \text{E}$) is situated on the lee side of Western Ghats and is about 100 km inland from the west coast of India, which is also a fast growing tropical urban city. The experimental site is located at an elevation of about 573 m above mean sea-level (AMSL) and is surrounded by hillocks as high as 760 m AMSL, forming a valley-type configuration. The transport and dispersion of pollutants, particularly those in the lower levels of atmosphere, are believed to be affected by the circulation pattern associated with this terrain. The geographical details of this observational site, frequency of measurements, details of instrument setup and its accuracy have been discussed elsewhere (Murugavel and Chate 2009). Westerly flow weakens in the lower troposphere and the easterly flow sets in during the winter season. Daily minimum temperature starts falling by the end of October and continental airmasses, rich in nuclei of continental origin, passes over this region. Fair-weather conditions, with clear skies and very low relative humidity in day time, prevail during the winter season. Daily surface minimum temperatures go down to $3^\circ\text{--}4^\circ\text{C}$ and light surface winds prevail, low-level inversions occur during morning and evening hours. The possible aerosols type present over the station is a mixture of water-soluble, dust and soot aerosols. Formation of aerosols in the accumulation mode is considered to be due to gas-to-particle conversion processes. We have recorded diurnal wind speeds during December 2005 to March 2006. Monthly averaged wind speeds with local standard time are presented in figure 2 and it is seen from the diurnal patterns in this figure that winds were low during night and it is almost absent during 0700–0900 hr. Light wind ($0.4\text{--}0.8\text{ ms}^{-1}$) prevails during 0915–1000 hr and increases to 2.5 ms^{-1} between 1200 and 1400 hr. It is worth to note that the same diurnal wind pattern is observed during the entire winter season. Similar diurnal wind cycle was reported during winter season (December) at this site (Chate and Pranesha 2004). Since there is no combustion source nearby, observed wind patterns in figure 2 during 0600–1000 hr suggest that any sudden increase in the concentration of smaller aerosol size and eventual rapid growth of them to the larger size during this period of time could be due to nucleation bursts at this site.

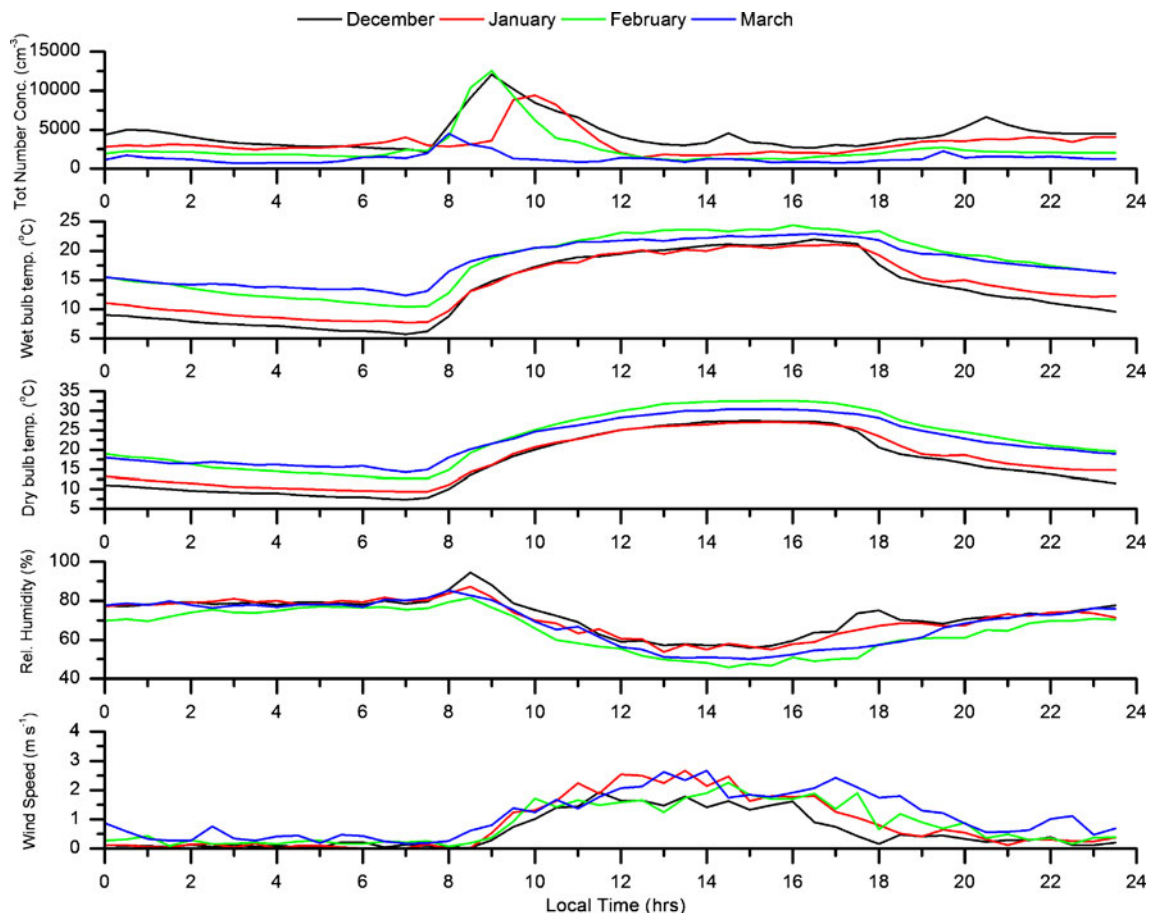


Figure 2. Monthly averaged diurnal variation of meteorological parameters and total aerosol number concentration.

4. Results

Dataset of diurnal variations in aerosol size distributions at Pune is comprised of 75 days of measurements at every half-an-hour interval from October 2005 to March 2006 and their analysis was performed on a day-to-day basis. Such a fine time resolution is useful to study growth properties of aerosols during nucleation events. For example, half-an-hour averaged diurnal variation of aerosol number concentration in 13 different size bins are shown in figure 3. It is clearly seen in this figure that there is a steep increase in aerosol number concentration after the sunrise in all diameter sizes on December 27, 2005. Start of these events varies between 0700 and 0900 hr local time and once started to increase, it takes ~ 2 h to reach the maximum, it sustains at the maximum for about an hour and then it gradually decreases to its background value within ~ 2 h accounting total duration of about 5–6 h. The increase is about 5 to 10-fold and it has been observed frequently after the sunrise at this site (Chate and Pranisha 2004; Chate and Devara 2005; Murugavel and Chate 2009).

Furthermore, measured total aerosol concentration at ambient, 40° , 125° , 175° , 300° and 350°C

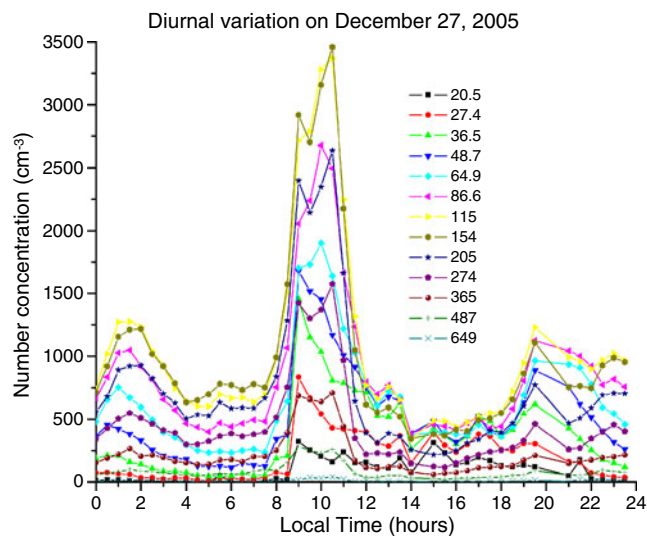


Figure 3. Diurnal variation aerosol number concentration in each size from 20–500 nm on December 27, 2005.

temperatures during typical nucleation events on 24, 25 October and 17 December, 2005 are shown in figure 4. Each group of bars show the total number concentration at given time and individual bars for different temperatures as given in the

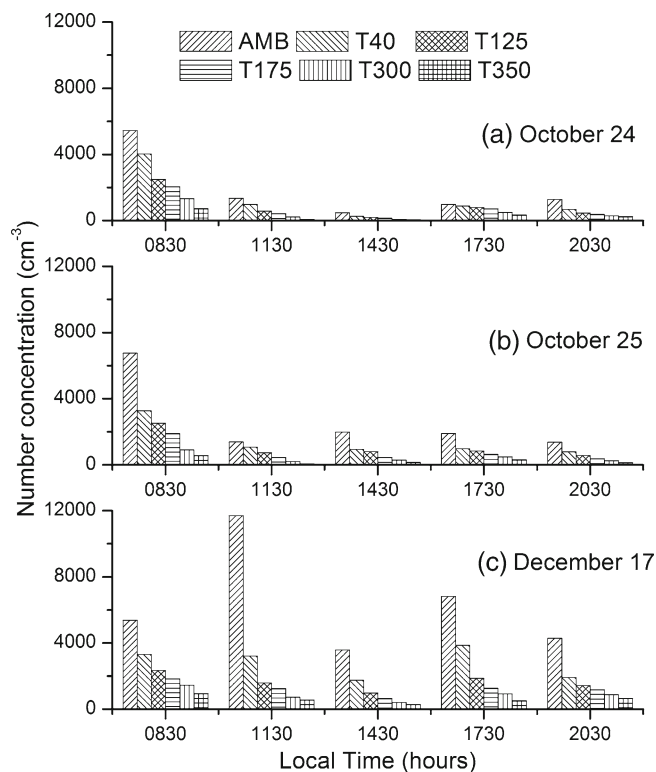


Figure 4. Volatile measurements on typical days. The bars show total number concentration measured using TD set at specific temperature. The aerosol at ambient temperature gradually reduces as the air sample is exposed to higher temperatures. The volatile portions are calculated from the amount of reduction in aerosols while exposing to higher temperatures. The x axis shows the time of the day. TA, T40, T125, T175, T300, T350 and T > 350 refer to ambient temperature, 40°, 125°, 175°, 300°, and >350°C, respectively.

legend. The reduction in concentration while treating the air sample at higher temperature can be seen in this figure. Volatile portions can be estimated by measuring the reduction in concentration while evaporating ambient aerosols to a desired volatile temperature levels. These measurements take some time to obtain complete dataset at these temperatures and since single set of instrument was available with us, type of measurements shown in figure 3 could not be simultaneously made possible during volatility measurements. Figure 4 shows that total aerosol numbers at ambient temperature between 0830 and 1130 hr are higher as compared to those at rest of the day period. The number concentration apparently dominant after sunrise (0830 hr) may be due to large increase in nucleation mode particles by photochemically driven nucleation bursts and subsequent condensation growth at this site. Percentages of particles evaporated out of total number concentrations at various temperatures are tabulated in table 1. As shown in this table, the major fraction of total aerosols evaporated at temperatures below 150°C. For example, though it varied at different timings of the day, on an average, it is about 51, 56 and 71% of total particles evaporated at temperatures below 150°C on 24, 25 October and 17 December, respectively. The corresponding fractions of volatile particles on these days are 23%, 25% and 13.3% at temperature 150–300°C and 9.3%, 7.8% and 5% at temperature 300–350°C. Also, the averages of remaining particles which are not evaporated

Table 1. The percentages of particles evaporated out of total number concentrations at various temperatures and time of the day.

Observations	N	% evaporated <150°C	% evaporated between 150° and 300°C	% evaporated between 300° and 350°C	% remaining at 350°C
October 24					
0830	5447	54.36	21.25	10.98	13.41
1130	1354	57.06	26.18	11.81	4.95
1430	482	61.53	23.31	2.75	12.41
1730	973	18.26	30.96	16.54	34.24
2030	1284	64.20	13.26	4.47	18.07
October 25					
0830	6755	62.70	23.77	5.09	8.45
1130	1398	47.37	37.70	9.99	4.94
1430	1987	59.11	26.39	6.50	8.00
1730	1900	55.56	18.62	9.45	16.37
2030	1381	58.31	23.12	7.95	10.63
December 17					
0830	5373	56.48	16.61	9.41	17.51
1130	11703	86.43	7.30	1.45	4.82
1430	3573	72.82	15.86	3.42	7.90
1730	6817	72.60	13.78	6.11	7.51
2030	4276	66.73	12.74	5.25	15.28

even after heating them to 350°C are 16.6%, 9.7% and 10%.

The period of this measurement is followed after the Indian summer monsoon season. The prevailing south-west low level winds during monsoon turn into north-east during September and continues to be north-easterly during the period of measurement. The automatic weather system (AWS) was not operational during October and November, 2005 at this site. The half-an-hour monthly averaged meteorological parameters measured over the site along with total aerosol number concentration are shown in figure 2. The monthly averaged temperature is at its minimum in December and gradually increases thereafter. The diurnal minimum temperature occurs between 0700 and 0730 hr. The humidity remains in the range 70–80% during night and shoots up more than 95% between 0800 and 0830 hr. The wind speed is below 0.8 ms⁻¹ during night and gradually increases after 0830 hr. The maximum wind speed is observed between 1200 and 1400 hr. The overall wind speed is at its minimum during December.

The total aerosol concentration data have been averaged over half-an-hour on day-to-day basis and these values are used to calculate monthly averages. To have a net assessment on aerosol concentrations, monthly averaged values are plotted in

figure 5. The vertical bars are standard deviations from the mean. The monthly averaged concentration is at its maximum in November as compared to other months and standard deviation bars indicate large variations in number concentration in this month. The aerosol concentration systematically increases up to December and gradually decreases to minimum in March.

The concentration is found to increase after sunrise in all mid-point diameters (figure 3) and on the monthly scale this event occurs before 1200 hr. To show the difference in composition during this event and after this event, the volatility measurements made at different hours are averaged for those before 1200 hr and for those after 1200 hr and the resulting size distributions are presented in figure 6. The volatile portions of number concentrations at each temperature for each size are apparently higher before 1200 hr as compared to those after 1200 hr. Figure 6 is reproduced in figure 7 to show the total number of volatile particles at different temperatures. Apparently the volatile portions are higher during morning hours than evening hours. The evaporated fraction of particles below 150°C corresponds to volatile organic carbon (VOC) and sulphuric acid (Burtscher *et al* 2001; Huffman *et al* 2009 and references therein). It is noteworthy that the volatile portion together

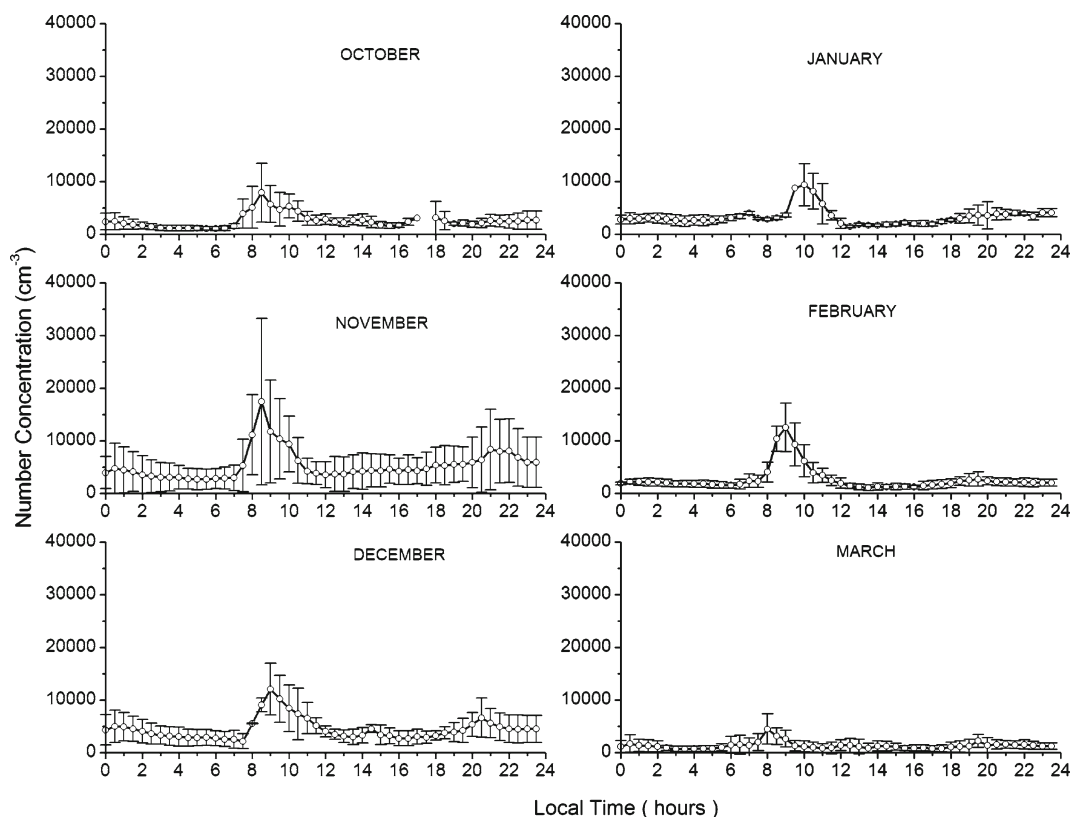


Figure 5. Monthly averaged total number concentration.

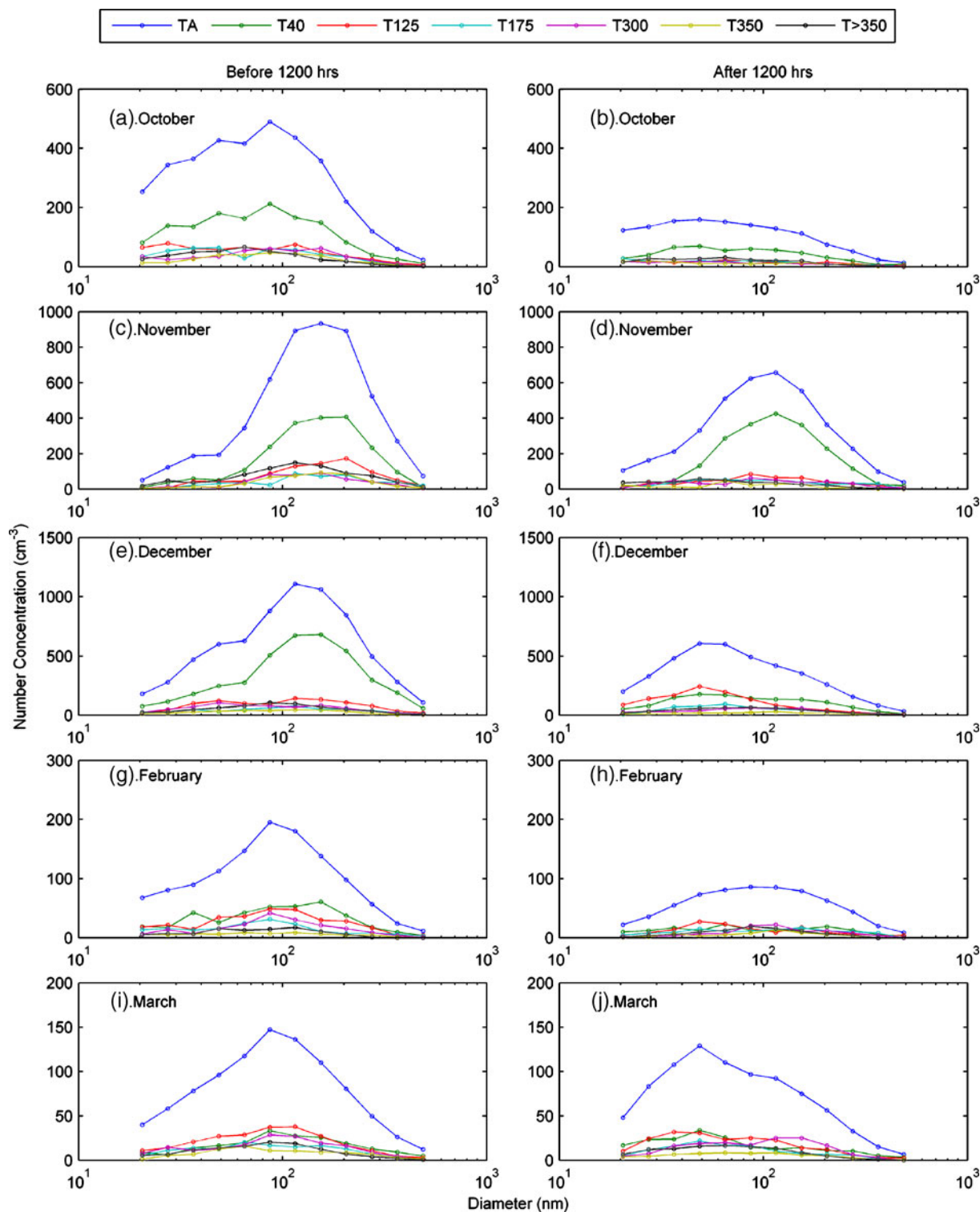


Figure 6. Monthly averaged size-distribution of volatile components at different temperatures. Left pane shows before 1200 hr and right pane shows after 1200 hr for the particular month. TA, T40, T125, T175, T300, T350 and T > 350 for ambient temperature, 40°, 125°, 175°, 300°, and >350°C, respectively.

at 40° and 125°C is the largest among all as shown in figure 6 and this feature is more evident in figure 7.

The measured volatilities of standard species at temperature $\leq 150^\circ\text{C}$ (highly volatile particles), in the temperature range 150°–300°C (moderately

volatile) and at temperature $\geq 300^\circ\text{C}$ (less volatile) are presented in Ishizaka and Adhikari (2003) with appropriate references. Similar characterization is presented in O'Dowd *et al* (2000) and Burtscher *et al* (2001). The standard species whose volatile temperature is $\leq 150^\circ\text{C}$ corresponds to evaporation

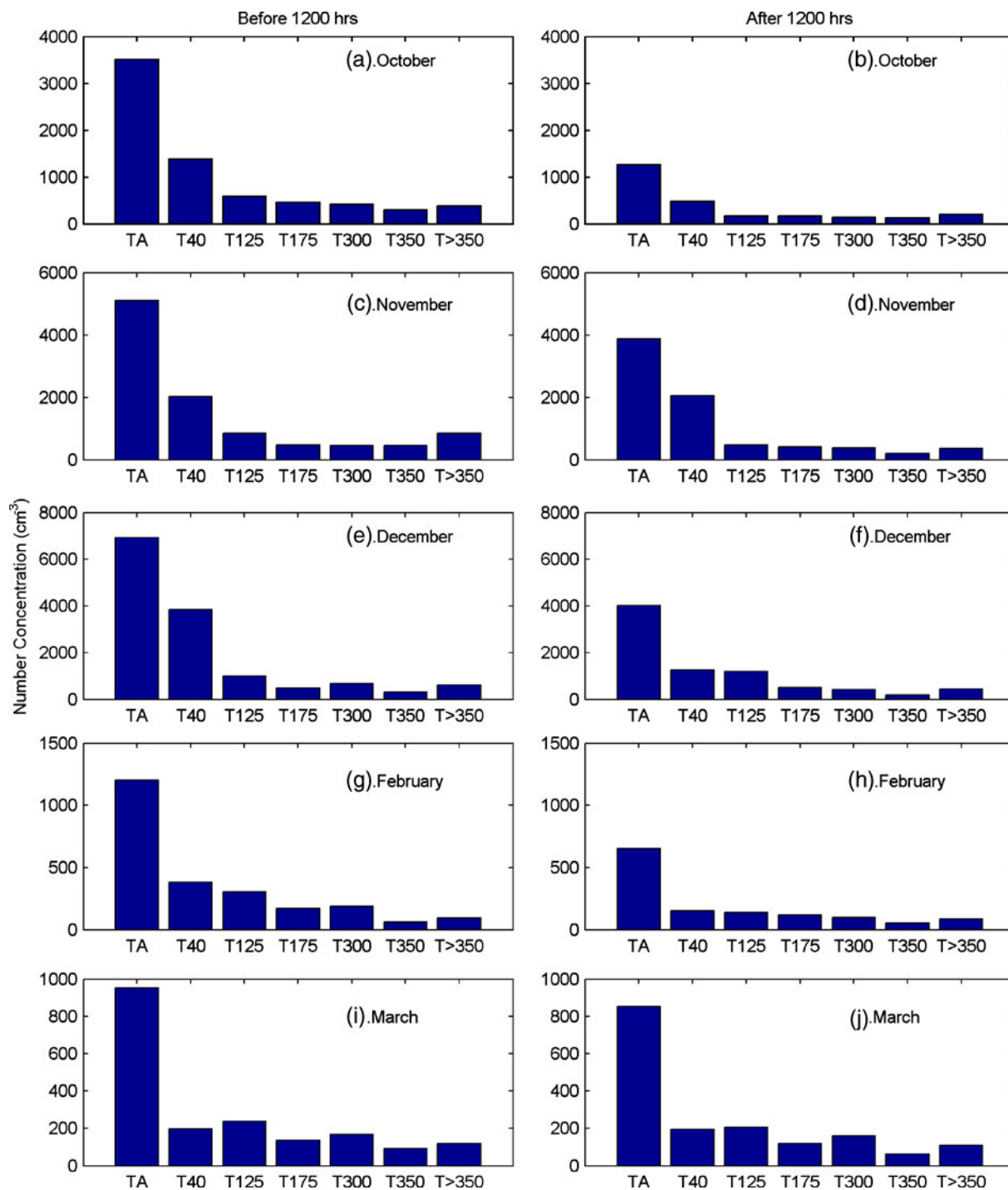


Figure 7. The monthly averaged total number concentration of volatile particles. The x-axis shows TA, T40, T125, T175, T300, T350 and T > 350 for ambient, 40°, 125°, 175°, 300°, and >350°C, respectively. Left pane shows averages before 1200 hr data and right pane shows averages after 1200 hr data for the particular month.

of ammonium chloride and ammonium sulfate and sulfuric, acetic and formic acids. The volatile temperature range 150°–300°C corresponds to evaporations of ammonium sulfate, ammonium bisulfate and ammonium nitrate and organic carbon. The volatilities at higher temperature ($\geq 300^\circ\text{C}$) correspond to soot carbon, calcium carbonate, sea-salt particles and polymerized organic

compounds (Ishizaka and Adhikari 2003 and references therein). Variations in percentage of particles evaporated with the mid-point diameters as a function of temperature are plotted for November 2005 in figure 8. At temperature $\leq 150^\circ\text{C}$, the percentage of evaporated particles out of ambient aerosols in their respective sizes varied between 32–65% and 18–76%, respectively in the morning and evening

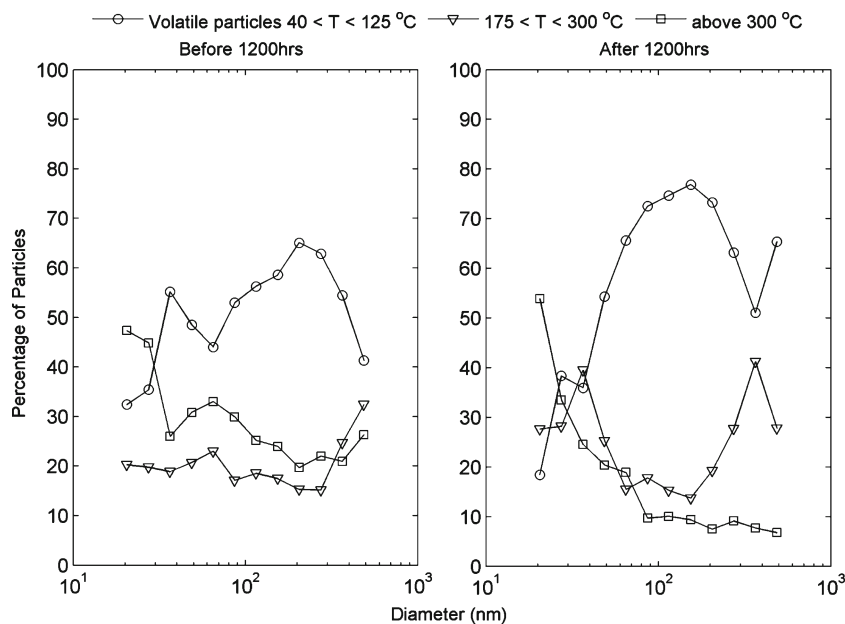


Figure 8. Percentage of volatile particles out of ambient concentration in their respective sizes.

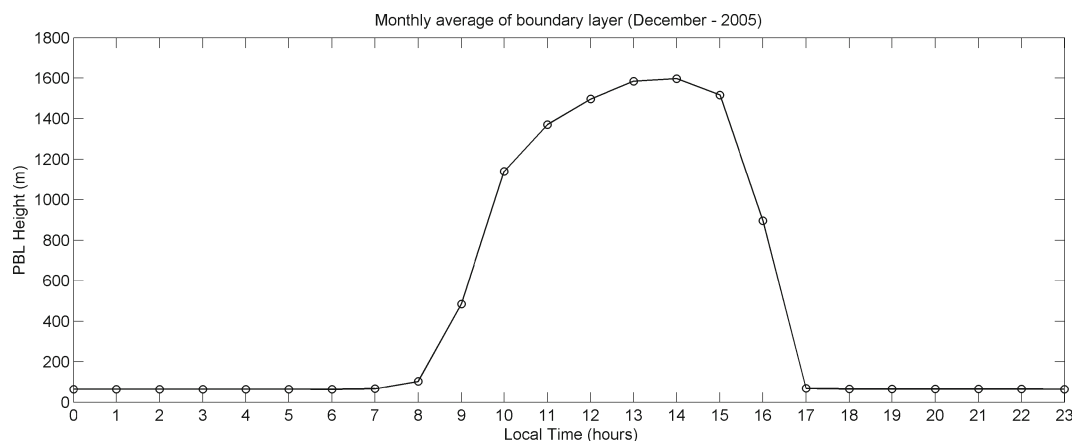


Figure 9. Diurnal variation of planetary boundary layer height for December 2005.

hours. These particles are of ammonium chloride and ammonium sulfate including those of sulfuric, acetic and formic acids (Ishizaka and Adhikari 2003). About 18–32% and 13–41% are evaporated in the morning and evening hours respectively which are of ammonium sulfate, ammonium bisulfate and ammonium nitrate and organic carbon. The particles remained above 300°C are 19–47% and 6–53% in the morning and evening hours respectively and these particles are of soot carbon, calcium carbonate, sea-salt particles and polymerized organic compounds. It is also noteworthy that the highly volatile particles (<150°C) in smaller size (≤ 36 nm) at morning hours (before 1200 hours) are higher than that of evening hours (in the same size). For example, smaller particles of 20.5, 27.4 and 36.5 nm are volatilized 32.4%, 35.4%

and 55% at temperature <50°C, in the morning hours as compared to 18%, 38% and 35% in the evening hours, respectively. It means that the total volatile number concentration is always higher in the morning hours than the evening hours (figure 7).

The temporal variation of concentration depends on the diurnal pattern of atmospheric boundary layer (ABL) height and ventilation factor (defined as the product of ABL height and wind speed, Hsu 2003). To examine diurnal variations of aerosol concentration with the ABL heights, monthly mean data for December 2005 retrieved from the Modern Era Retrospective-analysis for Research and Applications (MERRA) (<http://gmao.gsfc.nasa.gov/research/merra/intro.php>) are plotted in figure 9. The ABL height remains shallow (50 m) and almost no winds till

0900 hr (figures 9 and 2) results in too low ventilation factor. Lower ventilation factor should confine ambient aerosols at the Earth surface and manifest as an increase in the aerosols population in the early morning hours to 0900 hr, however, lower concentrations of aerosols are evidently observed in the figures 3 and 5. Once the solar heating of the land increases resulting in more convective activity leading to sharp increase in ABL height from 500–1400 m between 0900 and 1100 hr as well as wind speed about 2 ms^{-1} (figure 2) resulting in the higher ventilation factor. The sharp increase in ventilation factors should lead to a faster dispersion or dilution of ambient of aerosols over continental areas as reported in the literature (Safai *et al* 2002, 2004; Pillai *et al* 2002; Pey *et al* 2009). However, as seen in figures 3 and 5, aerosol concentrations are at their maximum levels despite the ventilation factor at higher side during 0900 to 1100 hr caused by high winds ($>2 \text{ ms}^{-1}$, figure 2) and ABL heights (1400 m, figure 9), which contradicts the routinely observed temporal variations of aerosol levels with the diurnal pattern of ABL heights (dispersion and ventilation effects). Similar trends are reflected in figures 6 and 7 for volatile portion of aerosols due to nucleation bursts during morning hours. Aerosol nucleation strongly depends on concentrations of sulphuric acid and other condensable vapours of low volatility. Also, solar radiation, atmospheric mixing processes, levels of precursors and chemistry of aerosols affect the nucleation processes. Photochemically driven nucleation, coagulation of nuclei onto pre-existing particles and also coalescence among freshly produced nucleated clusters resulted into the higher percentage of volatile particles in the smaller size in the morning hours at this location.

5. Conclusion

Averaged monthly aerosol concentration is at its maximum in November and gradually decreases to its minimum at the end of March. The results presented for half-an-hour averaged diurnal variation of aerosol number concentration gradually decreases in the night and in the early morning hours (0400–0800 hr) and at its minimum in the noon (1400–1700 hr). About 5 to 10-fold increase in number concentration before 1200 hr relative to those after 1200 hr in all particle diameters are in agreement with the results of aerosol measurement studies conducted at this site. The prevailing meteorological conditions such as minimum temperature, low wind, high relative humidity, levels of precursors and chemistry of aerosols during nucleation events as discussed in this study, coincides with the sudden increase in aerosol concentration. We

further support these nucleation bursts using the volatile composition of the aerosols measured at this site. The volatile fraction of ambient aerosols reveals that large number of highly volatile particles in the Aitken mode are present in the morning hours relative to the evening hours. These volatile portions of aerosols at temperatures $<150^\circ\text{C}$ and between 150° and 300°C are of ammonium chloride and ammonium sulfate, acetic and formic acids, ammonium nitrate and organic carbon. The inferred compositions of nucleation and Aitken mode particles can be linked to photochemical processes driven by typical solar spectra over this site. It is possible to support these measurements in near future from new online aerosol instrumentation, in particular aerosol mass spectrometers which are capable of measuring real-time aerosol compositions during nucleation events (Douglas *et al* 2003).

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