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Aerosol Number Size Distribution Measurements at Hanle, a Free Tropospheric High-Altitude Site in Western Himalayas

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Abstract: Aerosol characteristics over the high altitude stations, akin to pristine and free conditions are important to understand the background aerosol features against which polluted and urban environments could be compared. In addition it is also important to monitor the changes brought about by the large-scale processes, which result in lofting and transporting of aerosols from different source regions to higher altitude levels. Moreover, these aerosols have a significant role in modifying clouds especially cold clouds. The availability of significant amount of solar UV radiation at these altitudes, along with water vapour and OH, if present, makes these regions conducive for formation of new particles from gas phase reaction products involving precursors of natural or anthropogenic origin. Such processes play a significant role in modulating the size distribution of free tropospheric aerosols and hence their radiative impacts. In this work, aerosol number size distribution measurements carried out from a high altitude free tropospheric Himalayan location, Hanle (32.78^{o}N, 78.96^{o}E, 4530 m amsl) during the period from May through December 2010 are examined. The monthly mean total number concentration (N_t) increased from May (614 ± 188 cm^{-3}) to October (1498 ± 792 cm^{-3}) and decreased slightly through December (1158 ± 470 cm^{-3}). Fine mode aerosols (size < 100nm) contributed mostly to the total number concentration. The fractional contribution of the fine mode aerosols to the total number concentration, showed a clear increasing trend from May (~ 0.57) to December (~ 0.81). The number size distribution, which remained unimodal in May, June months with a mode in accumulation size range (around 100nm), changed to a bimodal distribution subsequently with a mode in the nucleation size range (< 25nm), indicating the possibility of new particle formation. The results are discussed in light of the possible association between the variation of the aerosol number concentration to long-range transport and thermally driven mesoscale processes such as mountain/valley winds.

Introduction

Aerosols affect the global climate with their direct and indirect effects and act as cloud condensation nuclei (for e.g., Charlson et al., 1995). The spatio-temporal heterogeneity in the aerosol properties, their sources and scavenging mechanisms make characterization of the aerosol radiative forcing highly difficult. One of the important variables that are essential to constrain the aerosol radiative effects is their number size distribution, which helps in identification of the sources of aerosols. In this regard the aerosol measurements from remote and highly pristine environments are vital in identifying the background aerosol features against which the polluted and urban environments could be compared. Since high altitude locations akin to pristine and free conditions are not influenced by local pollution, the number size distribution measurements from such locations provide insight into the particle formation, growth and transport mechanisms that are driving and influencing the particle concentration in different size regimes in regional and global scales. Moreover, high altitude aerosols have a significant role in modifying clouds

especially cold clouds. The availability of significant amount of solar UV radiation at these altitudes, along with water vapor and OH, if present, makes these regions conducive for formation of new particles from gas phase reaction products involving precursors of natural or anthropogenic origin. The development of orographic buoyant upward flows during daylight results in an increase of precursors such as water vapor, SO_2 and NO_y concentrations. Such processes play a significant role in modulating the size distribution of free tropospheric aerosols and hence their radiative impacts. At this juncture, with aerosol characterization in the saddle, the unique discovery of elevated aerosol layers over India and the northward gradient in the height and lower atmospheric warming due to these elevated aerosols (Satheesh et al., 2008; 2009) led to the formation of a field experiment 'Regional Aerosol Warming Experiment (RAWEX)' under Aerosol Radiative Forcing over India(ARFI) of Indian Space Research Organization-Geosphere Biosphere Program(ISRO-GBP).As part of RAWEX experiment a high altitude aerosol observatory (~ 4520 m above msl) has been established at the location, Hanle (32.78° N, 78.95° E) in the western Indian Himalayas. In this article we present the preliminary results from aerosol number size distribution measurements made at this location.

Site description and database

Hanle (valley) is located at 32.78^{o}N, 78.96^{o}E (Fig. 1) in the western Himalayas with an altitude of ~ 4300 m amsl, the actual measurement site being located in an igloo shaped hut on the top of Mt. Saraswati of altitude ~ 4520 m amsl. With a total population in around the mountain ~100 and with no major anthropogenic activities this site may be considered as a remote free troposphere (FT) background site. The surrounding valley areas maintain its natural environment of very sparse vegetation and arid regions.

The meteorological conditions that prevailed at the site during the observational period are shown in Figure 2. The monthly mean temperatures varied from ~ 4 $^{\circ}$C in May to a maximum in August (~12°C) which decreased in subsequent winter months to as low as~ -8°C. The relative humidity (RH) was mostly ≤ 30% except for the months of July, August. During august RH values reached as high as >80% at the times with slight drizzling. Notably the sky was cloudy during most of the days between May and August months with rather clear sky prevailing from September onwards. Generally the rainfall was scanty with a minimal precipitation of < 10 cm in a year. Due to the above dry and cold meteorological conditions, Hanle and its surrounding valley areas are termed as 'cold desert'. The monthly mean wind speeds varied in between 3-8 m/s with prevailing winds being mostly southerly to southwesterly.

Figure 1: Geographical location of Hanle (marked by the green balloon) superimposed on the digital elevation map of the Indian sub-continent (courtesy: Google maps)

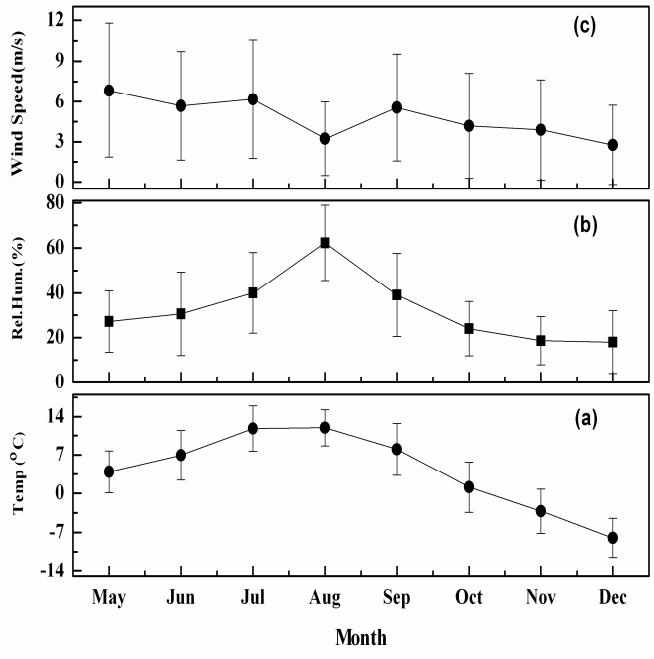

Figure 2: Monthly mean Meteorological parameters over Hanle-year 2010

Aerosol number size distribution measurements were carried out using a Sequential Mobility Particle Sizer (SMPS) of GRIMM, Germany (Helsper et al., 2008) during the period May to December 2010 were used in the present study. The SMPS system extracts a small electrical

mobility increment from the sample air using a Differential Mobility Analyzer (DMA) classifier and transfers it to a Condensation Particle Counter (GRIMM 5.400) for counting. During the study period, data has been collected using two DMA classifiers (Long-DMA and Short-DMA). But there was no time overlap of the data collected by LDMA and SDMA due to logistic reasons. LDMA covers the particle size range from 15.9 to 1363.6 nm, while SDMA covers 8.9 to 552 nm, both of them segregates the size distribution into 44 size classes.

Results and Discussion

Generally aerosol number size distribution is divided according to particles diameter (D_p) into a Nucleation mode (≤25nm), the particles representative of recent new particle formation, Aitkin mode ($25<D_p\leq100$nm) and accumulation mode ($100\text{nm} <D_p\leq1\mu$m) representing more aged particles. The monthly mean particle number concentrations are shown in Figure 3.

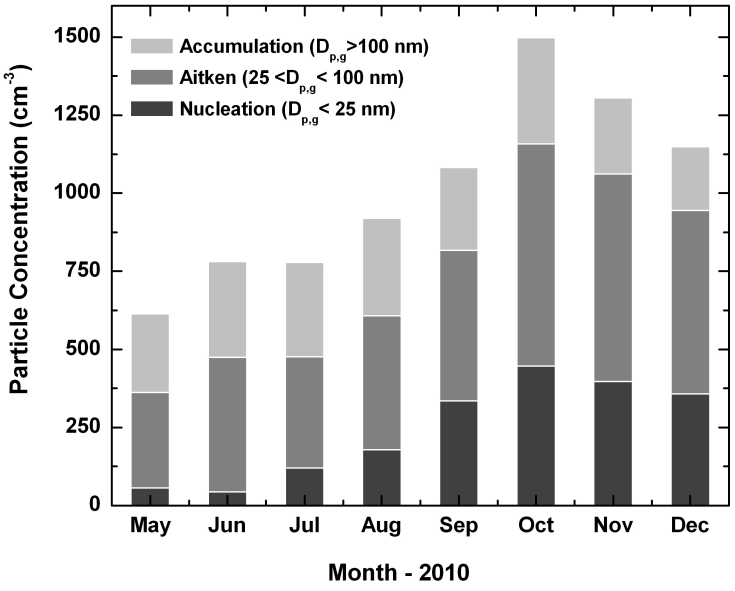

Figure 3: Monthly mean aerosol number concentrations at Hanle

As clearly evident from the above Fig. 3, the total number concentration is dominated by the fine particles with size <100nm (Nucleation +Aitkin mode). The fine fraction, fractional contribution of the fine particles to the to the total number concentration, showed a clear increasing trend from May (~0.57) to December (~0.81), which further reflected, in the variation of geometric mean diameter (g_d) for the study period. The monthly mean total number concentration (N_t) increased from May (614 ± 188 cm^{-3}) to October (1498 ± 792 cm^{-3}) and decreased slightly through December (1158 ± 470 cm^{-3}). The monthly mean total number concentration (N_t), Geometric mean diameter (G_d) and Fine fraction are tabulated in Table 1.

Table 1: Monthly mean total particle number concentration (Nt), Geometric Mean Diameter (G_d) and Fine fraction over Hanle during the study period

Month	**Total particle number Conentration (N_t)**	**Geometric Mean Diameter (G_d)**	**Fine fraction**
May	614 ± 188	87 ±22	0.57
June	781 ± 155	85 ±14	0.60
July	790 ±397	82 ±13	0.58
August	935 ±566	92 ±57	0.60
September	1097±951	64 ±13	0.70
October	1498±792	60 ±14	0.75
November	1306±774	54±9	0.80
December	1158±471	51 ±9	0.81

At this point of time it would be interesting to examine the monthly mean number size distribution pattern, which is shown in the figure 4.The nature of the size distribution shows clear variation from May to December. In the months of May and June the number size distribution is uni-modal with a mode in the accumulation size range (around and above 100nm). From July onwards a distinct mode in the nucleation size range is also seen apart from the accumulation mode. Thus the average monthly mean size distribution at Hanle is significantly driven by the occurrence of possible ultra fine particle events. The Nucleation mode of the number size distribution became much steeper from august and subsequent months, which became significantly noticeable in October and November months along with another mode in Aitkin mode size range.

Incidentally, more clear skies existed from September onwards which is more favorable condition for the new particle formation (NPF) events when compared to cloudy sky that were witnessed till August at the study location. NPF occurs by the nucleation of non or low volatile gas phase species emitted from either biogenic or anthropogenic sources followed by growth into small particles. Even though solar irradiance is not the sole driving factor of the NPF but it is also one of the important factors along with other meso-scale processes that contribute to increased photochemistry resulting in high frequent NPF (Venzac et al., 2008; Nishita et al., 2008; Weber et al., 1995; Weingartner et al., 1999). Recently Hamed et al., (2011) examined the anti-correlation between relative humidity (RH) and NPF. High RH results in increased low level cloud cover, diminished solar radiation, leading to diminished gas phase chemistry and increased condensation sink of condensable gases due to hygroscopic growth of the preexisting particles. Also, the characteristic thermally induced mountain-valley wind systems influence considerably the transport of air pollutants and precursor gases for NPF from Valley to the mountain (Nishita et al., 2008). Above discussion suggests that the increased ultra fine particle concentration is reflected in the nature of the size distribution and drives the total number concentration.

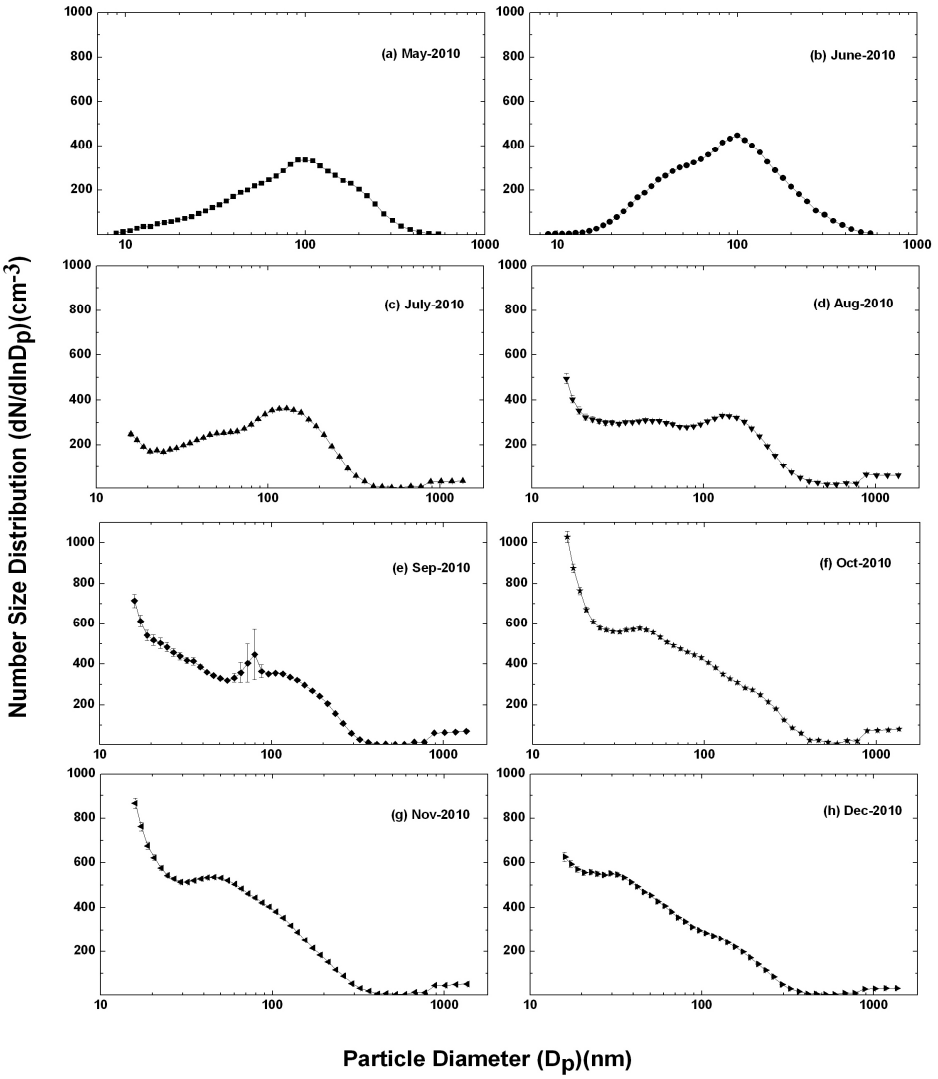

Fig 4: Monthly mean number Size Distributions, showing the significant increase in the nucleation mode abundance from July onwards

Conclusions

Aerosol number size distribution measurements from high altitude Himalayan background location Hanle were examined.

- The fine mode aerosols are dominant in the total aerosol number concentration during the study period with fine fraction increasing from May (0.57) to December (0.81).
- The size distribution was unimodal in May, June months. From July onwards an additional mode in the nucleation size range was also seen indicating new particle formation.

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