FINE PARTICLE MASS, NUMBER, SURFACE AREA CONCENTRATION METRICS AND THEIR CORRELATION IN OUTDOOR ENVIRONMENTS

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ABSTRACT

Exposure to fine particles is measured using metrics such as mass concentration (MC), number concentration (NC) and surface area concentration (SAC). This study aims to find correlation between the three metrics in outdoor environments of a city – city periphery, city centre and on-road. Simultaneous real-time mass, number and surface area concentration measurements were conducted in these environments. Arithmetic means, peak concentration values, and regression coefficients were determined to find relation between the three metrics. Results indicate remarkable high indoor number, mass and surface area concentrations. For ambient sampling sites, good correlation coefficient $r^2=0.80$ between MC and NC, and $r^2=0.69$ between NC and SAC metrics were observed. However, traffic-influenced on-road measurements showed little weaker correlations of $r^2=0.45$ and $r^2=0.30$ respectively.

INTRODUCTION

Aerosols are chemically, and physically diverse substances recognized also as Particulate Matter (PM). Aerosols are generated in various ways, by natural processes such as soil, dust, sea sprays, volcanic eruptions, or as byproduct of anthropogenic processes such as combustion of fuels, agricultural practices, industrial practices, transportation etc. They undergo changes through various processes such as condensation, nucleation etc. Outdoor aerosols are mainly due to vehicle emissions, outdoor combustion soot, windblown road and soil dust etc (USEPA, 1987). Burning fuels such as gasoline, oil, diesel or wood produce most of the $PM_{2.5}$ (particulate matter whose aerodynamic diameter is less than 2.5 µm or 2500 nm) pollution found in outdoor air. Studies show specific respiratory illnesses such as TB, cardio-vascular disease, chronic obstructive pulmonary disease etc could be attributed to continued exposure to particulate matter pollution in air (Bruce, et al., 2000; Smith, 2000).

For most health – related studies in recent years, mass concentration (MC) has been predominantly used as the exposure metric to assess the impact on health. But recently other metrics such as number concentration (NC) and surface area concentration (SAC) are considered important from studies on cell cultures and animal respiratory system (Brown, et al., 2001, Oberdörster, 2000, Tran, et al., 2000; US Environmental Protection Agency (USEPA), 987). Considering this, MC, NC, and SAC are the three important metrics measured in the study. In general, active surface area concentration may be a concentration metric that is distinct from particle number concentration and respirable mass concentration. For future health

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effects or toxicological studies involving nano-materials or ultrafine aerosols, this finding needs to be considered, as exposure metrics may influence data interpretation (Heitbrink, et al., 2009). NC and SAC metrics are significantly distinct from MC in characterizing exposure to airborne nanoparticles (Zou, et al., 2015).

One can by simple analysis, understand the relation of MC, NC and SAC for a cloud of uniform aerosols. Smaller the particles, greater the surface area relative to their mass. In other words, for a given mass, surface area increases as 1/diameter. Particles with a diameter of 25 nm when inhaled at same mass concentration have a number concentration that is approximately 6 orders of magnitude higher than for a 2500 nm diameter particle, and particle surface area is also greatly increased. Weighting a distribution by number, surface-area or mass can lead to visually very different distributions. With increased attention given to the smaller particles than PM_{10}, particle number and surface area become potentially important metrics to replace or complement the traditional importance given to particle mass.

In the present study, we want to simultaneously measure aerosol exposure in terms of these three metrics and find out correlation between different metrics of the same aerosol cloud. We hope the study gives an insight into the dependence of this correlation on the sampling environment; and also focuses on the possibility of proper and reliable estimations of other metrics from known metric values.

It may be expected or hypothesised that metric values of aerosols differ in their correlation with each other considerably for different nearby environments. This can be understood as due to the difference in density, particle shape and size range of PM generated by different activities. At other times, when there are no activities, they should correlate well, assuming uniform aerosol distribution attained over a period of time. In the present paper, I am going to present good correlation obtained between the metrics in the ambient outdoor environments. Slightly lesser degree of correlation was found in the sampling of traffic-emission affected on-road PM. This gives us positive hope on the estimation of one metric from other known metric values.

EXPERIMENTAL METHODS

a) Instrumentation

First part of the process of aerosol measurement is separating a small sample from the bulk aerosol for analysis. Sampled particles are then transported from the sampling inlet to the measurement instrument. The particles may be collected for future analysis (gravimetric sampling), or may be analyzed in real-time (dynamic measurement). Light scattering and extinction from a large volume is used to measure an integral scattering function that averages the signal over particle type and size distribution. This technique provides an instantaneous readout device that is calibrated to give particle mass if the aerosol does not change significantly with time.

If an aerosol is illuminated with a narrow beam of light, particles passing through it will scatter light out of the beam – detecting the scattered light then provides a simple basis for counting the number of particles passing through the beam. Detection efficiency falls off rapidly below around 0.3 μm as the intensity of the scattered light decreases as d_p^6. This problem is solved by allowing particles to grow through condensation. If the relative intensity of the scattered light is measured accurately, it could be used to provide information on the particle size. HHPC-6 (ARTI, Inc.) instrument works on this principle.

LQ1-DC directly measure active or Fuchs surface area by the method of Diffusion charging. The instrument uses the method of finding probability of deposition of ions on particles for measuring the active particle surface area. It measures the integral attachment cross section of ions. It is the fraction of geometrical surface, which is directly accessible from the outside. For these reasons, the term 'active surface' or 'Fuchs surface area' is used. The measurement does not take place on a single particle basis, only the total active surface area within a defined aerosol volume is given.

The specifications and initial settings of each instrument used in the present study are explained here.

(i) Laser photometer (model 8520, DustTrak, TSI Inc.):

DustTrak is a portable, battery operated laser photometer. Time-interval for data logging at both outdoor and traffic measurements was set at 5 sec. The data stored is the average over 5 sec, and is stored every 5 sec.

Particle size range: 100 nm to 2500 nm.

Resolution: ± 0.001 mg/m³, or 0.1%, whichever is greater.

(ii) Diffusion charger (LQ1-DC, Matter Engineering, AG):

LQ1-DC is the instrument used for the measurement of surface area on the principle of diffusion charging. Data logging time-interval for Indoor, outdoor and traffic measurements was set to 10 sec. Before each sampling, electrical zero is to be adjusted using zero adjustment potentiometer to show zero on the digital display. After a small swing of few digits caused by the air stream, it will return to 0 ± 2 digits.

Response time: few seconds.

Particle size range: few nm to 2000 nm.

Resolution: $1 \ \mu m^2/cm^3$

Gas flow rate: approx. 1.5 l/min

(iii) Optical Particle Sizing (HHPC-6, ARTI Inc.):

The effects of particle refractive index and scattering angle, and to a certain extent, particle shape can be reduced by detecting the scattered light over a large solid angle. This is usually achieved using a parabolic mirror to reflect the scattered light over a wide range of angles onto a detector (Baron and Willeke, 2001). Based on this principle, HHPC-6 measures number concentration of particles in six continuous channels: 300 nm - 500 nm, 500 – 700 nm, 700 – 1000 nm, 1000 – 2000 nm, 2000 – 5000 nm and > 5000 nm. We shall use this to compute the number concentration (NC1) between 300 nm to 2000 nm.

(iv) CPC (model 3007, TSI Inc.):

CPC consumes high-purity (99.5 %) isopropyl alcohol at a rate of about 1 ml per hour. Before each sampling, alcohol is supplied by taking out the alcohol cartridge from cartridge cavity and soaking it in isopropyl alcohol for about 3-4 minutes. After that, alcohol cartridge is replaced as it is within the cavity. CPC runs for approximately 5 hrs for one charging of the cartridge. Then, the cartridge will have to be charged again for further sampling. Data logging time-interval for indoor, outdoor and traffic measurements was set at 5 sec.

CPC data have coincidence error. CPC displayed concentration range is 0 to 500,000 particles/cc. For particle concentration up to 100,000 particles/cc, coincidence is low and no correction is necessary. At higher concentrations, correction of coincidence is required.

Particle size range: 10 nm to 1000 nm.

Gas flow rate: approx. 0.7 l/min.

We shall use this instrument to compute the number concentration (NC2) between 10 nm to 1000 nm.

b) Sampling

Outdoor samplings were carried out at places in city peripheral zones and city centre zones comprising different parts of the city. Also, on-road traffic measurements were carried out on a moving vehicle traversing from borderline region of the city through city central region and to the borderline region on the opposite side. Simultaneous real-time measurements of ambient particulate MC, NC, SAC and, also gravimetric $PM_{2.5}$ measurements were carried out at each sampling location during September to December. Table 1 gives information about measuring sites and periods.

For sampling, all the above said instruments were taken to the sampling location and were run by battery for 24 hr. Air was sucked through plastic tubing connected to inlets of the instruments. For outdoor measurements, plastic tubings were kept hanging into the ambient air outside the buildings at a distance of about 20 cm from the building wall. And traffic measurements were conducted by taking the instruments running on a moving auto, sampling on-road air through the plastic tubings.

c) Data analysis

Since, different instruments stored the data at different time intervals, all the data had to be merged into a single excel file with a common time-base, like 1-min or 5-min interval. This was done by time-aligning the data after interpolation. For example, the DustTrak stored the data every 5 sec, which is the average concentration for 5 sec. First, moving average was taken for all the data in a minute to remove some of the extreme variations, to obtain the 1-min moving average data. Then a list of required time points (with 1-minute or 5-minute time interval) is prepared. In the 1-min moving average data, two data points above and below the required time–point were noted. For this, INDEX and MATCH worksheet functions were used. From two data points got in this way,

Table 1. Measurement periods at different indoor and outdoor locations

Outdoor	City Periphery	**12-12 Sept**
		12-13 Oct
		20-21 Oct
		18-20 Dec
		3-11 Jan
	City centre	23-24 Oct
		28-29 Oct
		22-23 Nov

data for the required time-point is calculated by the linear interpolation method as follows:

$$M = M_1 + (T - T_1) * \frac{(M_2 - M_1)}{(T_2 - T_1)}$$

where,

T: Time at which the metric concentration is needed;

T_1: Time immediately before T in the metric data

T_2: Time immediately after T in the metric data

M_1: Metric concentration at time T_1

M_2: Metric concentration at time T_2

M: Required metric concentration at time T

Thus, all the data of indoor, outdoor sampling sites, and traffic zone data were converted into such time-aligned files. From such time-aligned files, it was easy to compare and relate any two metric values at any time point and also associate them with the recorded activities.

STATISTICAL ANALYSIS

Statistical analysis of each such time-aligned data set was performed in Excel.

Regression analysis of the data was also done, to know the correlation factor r. For this DustTrak and HHPC –data were used which had same size ranges. From regression analysis, regression equation is obtained, by which data of one metric can be estimated from the known data of the other metric. Estimated data is used to compare with the original data.

RESULTS AND DISCUSSION

Correlation between metrics during the entire sampling period

Correlation analysis for the entire sampling period at a site was done for MC metric (100 – 2500 nm) with corresponding NC1 (300 - 2000 nm) and SAC metrics (10 nm – 2000 nm). A reasonably good correlation was found between the metrics over a day. Table 2 gives statistics of r^2 values obtained for measurements in the two categories which spanned above 22 hrs. Outdoor data show good correlation values while the traffic data show a weaker correlation. This is possibly due to the fine particle emissions from many frequent and different types of sources such as vehicles, and consequent non-uniform mixing-up of aerosols.

Also, it is observed that the correlation with SAC is considerably lower than correlation between the other two metrics. Values of r^2 for correlation with SAC are obviously a little smaller due to the

ultrafine size range involved with SAC data. SAC data is over size range 10-2000 nm whereas MC is over size range 100-2500 nm. This indicates the concentration of ultrafine particles (10 – 100 nm) in these environments.

Table 3 shows correlation coefficients (r^2) between MC (100 – 2500 nm) and NC1 (300 - 2000 nm) for all outdoor measurements that lasted more than 22 hours. On an average, both metric values show good correlation of 0.83 ± 0.15, with a variance of about 18.18% to each other. (Fig. 1 and 2) show regression analysis plots between the metrics.

Comparison of results of present study with few other studies conducted in specific environments will be discussed now. (Zou, et al., 2015 conducted similar studies in some workplaces. Results of that study indicated the order of correlation coefficients between $NC_{20-1000 \ nm}$, $SAC_{10-1000 \ nm}$, and respirable $MC_{100-1000 \ nm}$ was: $R_{SAC \ and \ NC} > R_{SAC \ and \ MC} > R_{NC \ and \ MC}$. One more such study Heitbrink et al, 2009 found good correlation between active surface area concentration and ultrafine particle number concentration but found a weaker correlation between SAC and respirable MC. Correlation between number and active surface area concentration was stronger during winter (R^2=0.6) than in the summer (R^2=0.38). Stronger correlation in winter was attributed to the use of direct-fire gas fired heaters that produced substantial numbers of

Table 2. Correlation values (r^2) between measurement metrics

Outdoor measurements N = 7	r^2 for MC and NC1 (*Above 22 hrs*)	
Metrics for correlation	Average r^2 values for different categories	
	Outdoor	Traffic
$MC_{100 - 2500 \ nm}$ – $NC1_{300-2000nm}$	0.83 ± 0.15 (n =7)	0.45 ± 0.13 (n =7)
$NC1_{300-2000nm}$ - $SAC_{10 \ nm – 2000 \ nm}$	0.69 ± 0.11 (n =6)	0.30 ± 0.18 (n =5)

Table 3. Correlation values (r^2) between MC (100 – 2500 nm) and NC1 (300 - 2000 nm) at all Outdoor sites.

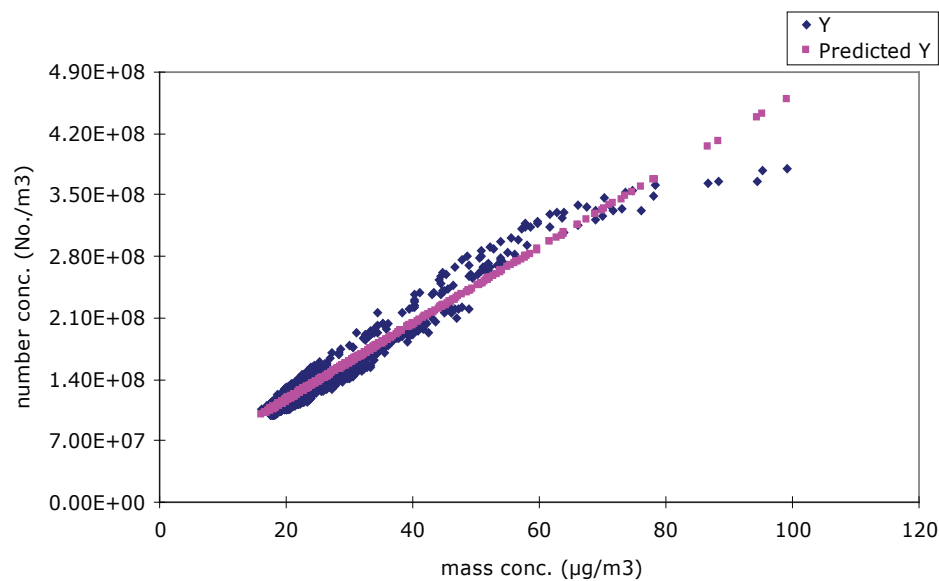|	
1	0.59
2	0.93
3	0.63
4	0.91
5	0.88
6	0.92
7	0.95
A.M.	0.83
S.D.	0.15
Variance (%)	18.18%

Fig. 1 Regression analysis plot for MC (100 – 2500 nm) and NC1 (300 - 2000 nm) at a city peripheral outdoor site. $r^2 = 0.94$, Regression equation: Y= (4.34E+06) × + 2.89E + 07.
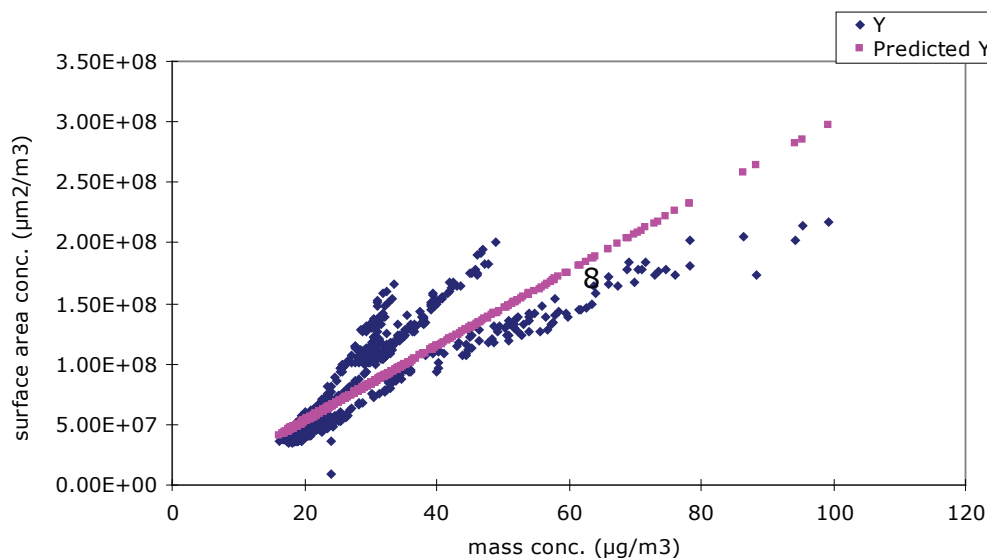

Fig. 2 Regression analysis plot for MC (100 – 2500 nm) and SAC (10 nm – 2000 nm) at a city peripheral outdoor site. Interestingly, three clear distinct relations can be seen. $r^2 = 0.77$, Regression equation: Y= (3.09E+6) × –(0.97E+07).

ultrafine particles with a modal diameter between 7 and 23 nm.

DustTrak and HHPC-6 data which match well with size ranges show good correlation with r=0.83 ± 0.15 at outdoor sites. For indoor studies conducted in the same city, correlation found for the above two metrics was also good at r=0.77 ± 0.12 [10].

For traffic related data, however, correlation between MC and NC1 is found to be weaker with r=0.45 ± 0.13. This may be because of non-uniform mixing up of aerosols generated by many different

traffic sources. Between NC1 and SAC, correlation gets further lower at r=0.30 ± 0.18, possibly related to the presence of ultrafine particles in the vehicle emissions.

Concentration levels of the metrics

Table 4 shows DustTrak correction factors obtained for each outdoor measurement. Correction factor is found to be around 0.3 for each measurement. For city peripheral zone, gravimetric sampling was conducted at only one site. Assuming that there was only slight difference in the data from different sites in this zone, single correction factor

obtained was used for DustTrak calibration for all measurements in this zone.

Background values were calculated for each site taking the average of MC during no-activity time, 12pm – 4am. Outdoor measurements were made at places comprising city peripheral zones, city centre zones for 24-hrs. Table 5 gives summary of metric values for outdoor measurements. Background value for MC is nearly the same as 24-hr MC for city peripheral region. Table 5 also gives ratio of concentrations for each metric found in the two categories of outdoor sites – city periphery and city centre. There is a moderate difference found in geometric mean (GM) ratios between the two categories for different metrics. The ratio is fond to vary from around 1.9 to 2.5. This may be inferred as due to the dominance of fine particles in busy zones of the city resulting from vehicular pollution. NC1 shows a high ratio of 2.43 for the two zones.

24-hr $PM_{2.5}$ average outdoor MC in city centre zones were on an average found to be about 1.96 times higher than 24-hr $PM_{2.5}$ average MC for city peripheral locations. NC1 and SAC showed little higher ratios (about 2.43 and 2.19 respectively) for the two zones. 24-hr average NC1 and SAC from multiple sites in urban zones were found to reach values around 2.00 E ± 08 particles/m³ and 1.50

E ± 08 µm²/m³ respectively. This appears to be associated with increased activity and vehicular emissions at city centre zones.

Table 6 shows the DustTrak correction factors obtained for each traffic measurement.

Table 7 gives the average concentration levels for traffic measurements while driving through the city on the trajectory to and fro. The average mass concentration is very high (206 µg/m³) than the average ambient levels (44 µg/m³) found in the Table 5. The same is true for other metrics also. Also, interestingly, here it can be observed that average on-road concentrations are higher than the 24-hr indoor average concentrations (Monkkonen, et al., 2005, Andresen, et al., 2005). From the table it is clear that number concentration for the size range 0.01 – 2.0 µm is about 10^3 times higher than for the size range 0.3 – 2.0 µm. However, for ambient measurements, such big difference was not found between the two as can be seen from Table 5. As other studies also indicated, this is because fine particulates of the order of nm are associated with the traffic related emissions.

For a new 24-hr $PM_{2.5}$ standard, range considered is 20-65 µg/m³, CASAC Review (McClellan and Miller, 1997). City peripheral locations were found to be having low mass concentration averages at 28 ± 17 µg/m³. City centre locations also showed relatively lower 24–hr average of 44 µg/m³. However, traffic on-road measurements through the city showed average of 206 ± 52 µg/m³, which is about 4 times the concentrations at ambient city centre sites, This traffic on-road level is also about 2 times the indoor 24–hr average found for kerosene using houses (Monkonen, et al., 2005). For on-road measurements, NC2 for the size range 10 – 1000 nm is about 10^3 times higher than NC1 for the size range 300 – 2000 nm. However, for ambient measurements, such a large difference was not found between the two. This indicates the

Table 4. Dust Trak correction factors obtained from gravimetric masses for outdoor measurements

Variables	Outdoor site and measurement date	DustTrak correction factor
City Peripheral sites	1 (12 Sept)	0.333
	2 (12 Oct)	
	3 (20 Oct)	
	4 (18 Dec)	
	5 (3 Jan)	
City centre sites	1 (23 Oct)	0.329
	2 (28 Oct)	0.327
	3 (16 Nov)	0.360

Table 5. Outdoor MC, NC1, NC2 and SAC measurements. Background outdoor pollution: 15 ± 10 µg/m³

24-hr (approx.) Arithmetic mean values	City peripheral Zone (n = 5)	City centre zone (n = 3)	City Centre / City Periphery ratio: range and G.M.
MC (100 – 2500 nm)	24 ± 17 µg/m³	44 ± 6 µg/m³	1.111 to 7.4 GM = 2.960
NC1 (300 - 2000 nm)	8.57 ± 5.52 E ± 07 No./m³	1.98 ± 0.10 E ± 08 No./m³	1.47 to 8.39 GM = 3.53
NC2 (10 – 2000 nm)	9.13 ± 5.52 E ± 09 No./m³	1.73 ± 0.10 E ± 10 No./m³	1.37 to 3.79 GM = 2.30
SAC (10 - 2000 nm)	8.35 ± 7.11 E ± 07 µm²/m³	1.47 ± 0.45 E ± 08 µm²/m³	0.9 to 6.37 GM = 2.97

influence of traffic related emissions for dominant presence of ultrafine particles, as it is known that fine particulates of the order of few nanometers get released into air by traffic related emissions.

Table 8 gives 24 –hr average values at each outdoor site. For MC, ratio of 24-hr mean value to background value is found to vary from 0.33 to 3.20 with an average of 2.11. The ratio of 5-min average peak value to 24-hr mean value for each site is found to vary from 2.5 to 7.17 with an average of 4.28. City peripheral zones and city centre zones were found to be statistically different at 5% level for NC1 and NC2, but not for MC and SAC. Comparing this with Monkonen et al. (2005) study for the same city of Mysore shows indoor 24–hr average concentrations are much higher. Indoor mass concentrations, for example, ranged from 41-240 $\mu g/m^3$. This shows the importance of workplace-specific studies for health effects of PM pollution (Fig. 3-5).

Similar work conducted by Zou et al. (2015) studied the ratio of cumulative percentage by number (APN) and cumulative percentage by mass (APM) and it

Table 6. Dust Trak correction factors obtained from gravimetric masses for traffic measurements

Measurements	D.T. correction factor
Traffic1(22 Aug)	0.546
Traffic2(3 Dec)	0.464

Table 7. 24-hr average levels for different metrics and their comparison at different outdoor sites

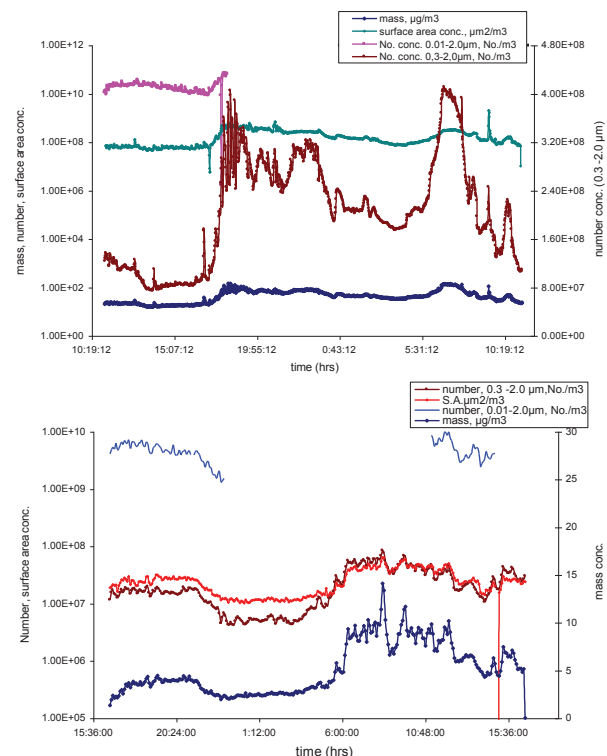| Arithmetic mean values	On Road measurement
Mass Conc. (100 – 2500 nm)	206 ± 52 $\mu g/m^3$
Number Conc. NC1 (300 - 2000 nm)	(2.51 ± 0.30) E ± 08 No./m^3
Surface area Conc. (10 - 2000 nm)	(5.50 ± 1.96) E ± 08 $\mu m^2/m^3$
Number conc. NC2 (10 - 2000 nm)	(2.24 ± 0.88) E ± 11 No./m^3

Fig. 3 Plot showing concentration levels over a day at outdoor city peripheral sites: NC data is sparse due to the weakening of alcohol cartridge in CPC.
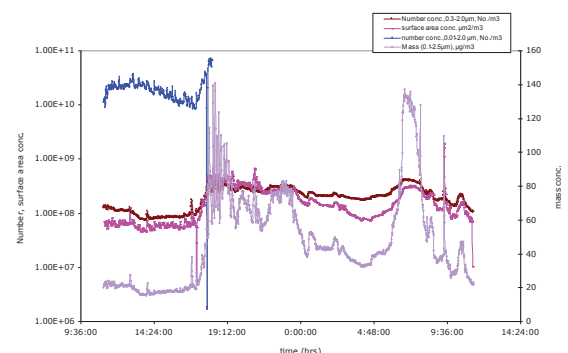

Fig. 4 Plot showing concentration levels over a day at an outdoor city centre site: CPC data is sparse due to the weakening of alcohol cartridge, CPC data showing a sudden dip due to the weakening of cartridge.
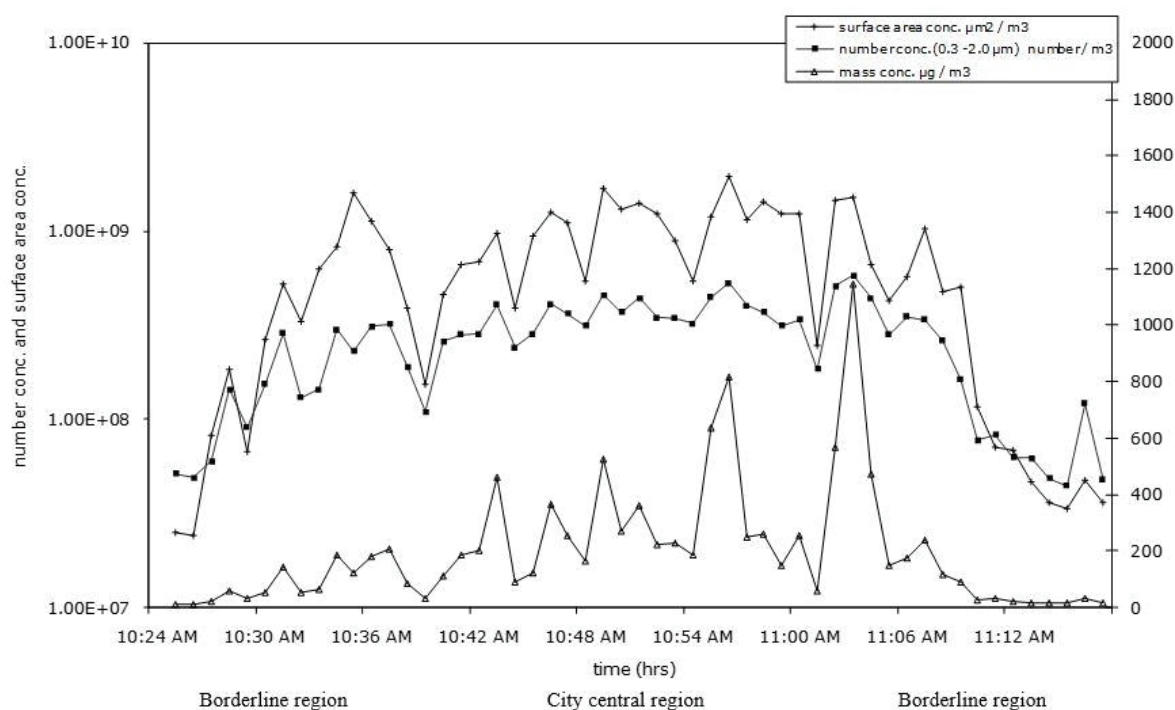
Table 8. 24-hr average levels for different metrics and their comparison at different outdoor sites

Sampling site	MC ($\mu g/m^3$)	NC1 (No./m^3)	SAC ($\mu m^2/m^3$)	NC2 (No./m^3)	NC2/ NC1	MC/ Background (15 $\mu g/m^3$)	Peak for MC. ($\mu g/m^3$)	peak/ 24-hr avg forMC.
CC-1	46	2.01E ± 08		1.53E ± 10	76	3.07	332	7.17
CC-2	48	2.06E ± 08	1.79E ± 08	2.05E ± 10	186	3.20	140	2.91
CC-3	37	1.87E ± 08	1.15E ± 08	1.61E ± 10	86	2.47	215	5.85
CP-1	38	9.32E ± 07	9.75E ± 07	NA	NA	2.53	168	4.42
CP-2	44	1.40E ± 08	1.99E ± 08	1.50E ± 10	84	2.93	147	3.34
CP-3	5	2.23E ± 07	2.51E ± 07	4.77E ± 09	267	0.33	14	2.80
CP-4	25	1.38E ± 08	6.81E ± 07	1.27E ± 10	98	1.67	87	3.48
CP-5	10	3.54E ± 07	2.81E ± 07	4.04E ± 09	98	0.67	25	2.50
CC – City Centre Zone; CP –City Peripheral Zone; NA – Not Available.								
For a new 24-hr PM$_{2.5}$ standard, the range considered is 20-65 $\mu g/m^3$, CASAC Review, (Mcclellan and Miller, 1997)								

Fig. 5 A plot of mass, number, surface area concentrations traversing through the city.

was analyzed to know whether nanoparticle number is predominant, as compared with nanoparticle mass. The study found the ratios of APN and APM for nano-Al_2O_3 and grinding-wheel particles (less than 100 nm) at the same work site were 2.03 and 1.65 respectively.

CONCLUSION

There is an overall good correlation between the metrics, indicating the possibility of estimating one metric value from the other known ones. However, for satisfactory estimation of one metric data from another known metric data, peaks during activities need to be studied as to the association with particle size range and density of particle. In future, this can simplify the task of measuring all the metrics. (Maynard, et al., 2003) study has estimated SAC from a lognormal size distribution derived from MC and NC levels measured indoor in households during combustion activities using kerosene and LPG fuels. It found a linear association between the measured and estimated surface area, although the gradient differed significantly from one in both the fuel types. The study showed that SAC may be estimated from MC and NC measurements to within a factor of 1.4 at concentrations above 500 m^2/cm^3.

These correlations support findings obtained through theoretical analysis (Heitbrink et al, 2009). Such analysis predicts that the active surface area of the aerosol increasingly underestimates the geometric surface area with increasing particle

size, particularly for particles larger than 100 nm. Thus, a stronger correlation between particle number concentration and active surface area concentration is expected in the presence of high concentrations of ultrafine particles.

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