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Morawska, Lidia and Johnson, Graham R. and Ristovski, Zoran and Agranovski, Victoria (1999) Relation between particle mass and number for submicrometer airborne particles. *Atmospheric Environment* 33(13):pp. 1983-1990.

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RELATION BETWEEN PARTICLE MASS AND NUMBER FOR
SUBMICROMETER AIRBORNE PARTICLES

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Abstract- The relationship between particle mass and the number of ambient air particles for the submicrometer size range was examined using a Tapered Element Oscillating Microbalance to determine the mass concentration, and a Scanning Mobility Particle Sizer to determine the volume concentration and total number of particles. After validating the techniques through their application to the estimation of submicrometer particle density for two laboratory generated aerosols of known bulk density (Sodium Chloride and Di-2-ethylhexyl-sebacate), the submicrometer fraction of laboratory generated Environmental Tobacco Smoke and ambient air were examined and an estimate of the average submicrometer particle densities for these aerosols found to be 1.18 g cm^{-3} and 1.7 g cm^{-3} respectively.

Key word index. aerosol, density, ambient, tobacco, mobility

INTRODUCTION

In recent years the demand for knowledge of various parameters of airborne particulate matter has increased with the realisation of the potential health and environmental health effects caused by the particulates (Dockery, et al., 1993; Pope,

Schwartz & Ransom, 1992; Schwartz & Dockery, 1992). Unlike many other pollutants, which are characterised mainly by their total concentrations, airborne particulate matter can be characterised by a range of different parameters, including: total mass or number concentration, mass or number size distribution, modality of size distribution, and count median diameter of the distribution or of the modes in the distribution. Usually different instruments have to be used to measure the different particle characteristics in different size ranges, making comparisons between data sets or between individual characteristics within the same data set very complicated. In many cases, the relationship between particle number and mass concentrations is of particular importance. Current air quality standards are expressed in terms of particle mass concentration as PM₁₀ or PM_{2.5}, which are mass concentrations of particles with diameters smaller than 10 or 2.5 μm , respectively (United States Environmental Protection Agency, 1997). One major limitation of using particle mass standards is that the values of concentrations are heavily biased towards larger particles; the mass of a small number of large particles can be significantly larger than the mass of a large number of small particles. In ambient environments concentrations of small particles are usually significantly higher than concentrations of larger particles (Morawska, Thomas, Bofinger, Wainwright & Neale, 1998), and it thus appears that for health risk assessments, knowledge of particle number concentrations could be more important than knowledge of particle mass (Oberdorster, Ferin & Lehnert, 1994; Seaton, MacNee, Donaldson & Godden, 1995; Li, Gilmour, Donaldson & MacNee, 1996). New research efforts are now often directed towards experimental characterisation of particle number distribution however, in order to relate the results to the current standards, particle number concentrations must be recalculated to mass concentrations. With the increasing use of electrical mobility analysers and optical

particle counters and the ease and speed with which they allow particle concentration versus equivalent diameter data to be acquired, the instruments appear to offer a possible alternative to gravimetric and equivalent techniques for estimating mass concentrations within a specific particle size range. This is of particular interest for real time submicrometer measurements where the mass concentrations involved may be considerably lower than $5 \mu\text{g m}^{-3}$, requiring long integration times for the Tapered Element Oscillating Microbalance (TEOM) to achieve a meaningful result. A non-trivial question to answer is that of the validity of re-calculating particle number to mass given the uncertainties resulting from the assumptions necessary to conduct the re-calculations.

The main problems in relating ambient particle number to mass result from: (i) the need to make assumptions about particle density, (ii) variations in the density of ambient aerosols with time, (iii) differences in size ranges in which any pair of mass and number measuring instruments used for experimental comparison operate, and (iv) the need to make assumptions about the spherical shape of the particles. In an ideal situation of two instruments operating in the same size range, with the same efficiency through the whole range, sampling aerosol of stable characteristics, a simple correlation coefficient could be obtained which could then be used for relating particle number to mass for similar conditions. However, in real situations, conditions will vary with time and location, and at best, the coefficient can be used for estimations only, otherwise a number of different coefficients should be used.

The aim of this work was to explore the uncertainties in relating particle number and mass concentrations in the submicrometer size range for urban ambient particulate

matter and for environmental tobacco smoke (ETS). Before applying the experimental method developed for the study to the environmental aerosols, the method was first tested on laboratory generated aerosols of known density: polydisperse Di-2-ethylhexyl-sebacate (DEHS) ($\rho = 0.912 \text{ g cm}^{-3}$), and dry NaCl ($\rho = 2.165 \text{ g cm}^{-3}$). The paper presents the results of the relation between particle number and mass for the aerosols investigated, as well as a discussion of the uncertainties of the relation in application to environmental aerosols.

Comment [g1]:

EXPERIMENTAL METHODS AND TECHNIQUES

The experimental technique was based on the application of a Scanning Mobility Particle Sizer (SMPS) for direct measurements of particle number concentration and a TEOM operating in more or less the same size range, for particle mass concentration measurements. The TEOM provided the gravimetric equivalent means of measuring the overall mass concentration in all of the experiments while the SMPS was used to determine the particle volume concentration. Each instrument was restricted to the submicrometer particle size range by an input conditioning device. In addition, examination of the SMPS spectrum was conducted to ensure the aerosol volume concentration did not contain a significant fraction of the total in the region immediately below the impactor cut-point diameter.

Aerosol Measurement Instrumentation

Tapered Element Oscillating Microbalance. The TEOM is a real time device for particulate mass concentration measurements for airborne particles. The TEOM draws

the aerosol through a filter attached to a tapered oscillating glass rod at a constant flow rate and measures the accumulation of mass on the filter through the effect of this mass on the oscillation frequency. Patashnick and Rupprecht provide a detailed description of the TEOM system (Patashnick & Rupprecht, 1991). In this study the TEOM (Rupprecht & Patashnick model 1400a) was fitted with a cyclone aerosol conditioner (URG 1 micrometer cyclone) operated at its design flow rate of 16.7 L min⁻¹ to remove particles of aerodynamic diameter greater than 1 µm. The standard filter flow rate of 3 L min⁻¹ was used and supplemented by an auxiliary flow bypassing the filter to achieve the design flow rate for the cyclone. A standard filter temperature of 50 °C was set for the measurements. Calibration factors of $A = 3.0 \mu\text{g m}^{-3}$ and $B = 1.03$ are normally used to achieve results ($Y = A + BX$), comparable to PM-10 reference methods. Since the instrument was not used as a PM-10 monitor these factors were changed to $A = 0 \mu\text{g m}^{-3}$ and $B = 1.0$ so that the measured mass concentrations remain unaltered.

Scanning Mobility Particle Sizer. The TSI Incorporated Model 3934 Scanning Mobility Particle Sizer (SMPS) system was used. The SMPS consists of an electrostatic classifier (EC) (TSI model 3071A), a condensation particle counter (CPC) (TSI model 3010) and associated software and hardware for system control and data analysis and recording. The EC classifies particles according to their electrical mobility, allowing only particles of a selected mobility to pass to the CPC where they are counted after being sufficiently enlarged by vapour condensation. The SMPS system software carries out the mobility data collection and analysis (Wang & Flagan, 1990). The instrument is fitted with an impactor ($Stk_{50} = 0.23$, nozzle diameter = 0.508 mm) to reduce the influence of large, multiply charged particles on

the mobility spectrum. The SMPS is capable of measuring number concentrations as low as 1 cm^{-3} at diameters of the order of 10 nm corresponding to a mass concentration of the order of $10^{-6} \mu\text{g m}^{-3}$ for particles of density 1 g cm^{-3} . All measurements were carried out using a sheath flow rate of 2.5 L min^{-1} , an aerosol flow rate of 0.25 L min^{-1} and a scan time of 250 seconds. The working particle size range under these conditions is approximately 15 - 930 nm while the maximum aerodynamic diameter is limited by the impactor cut-point diameter to 893 nm.

Monodisperse aerosol generator. A commercially available aerosol generator (TSI model 3475) was used for generation of DEHS and NaCl particles. This generator is of the Sinclair-La Mer type and produces a monodisperse DEHS aerosol by passing an atomised salt solution through a diffusion drier and bubbling the resulting fine, dry salt aerosol through a heated aerosol material (DEHS). Vapour condenses on the salt particles to form much larger particles whose size can be controlled by altering the salt nuclei concentration (by diverting some of the salt aerosol through a filter), flow rate and saturator temperature. Operating the generator with the seeding solution removed allows homogenous condensation of the DEHS vapour to form a broad-spectrum fine particle aerosol. Aerosols generated in this way have a highly reproducible size distribution (Ristovski, Morawska & Bofinger, 1998) and consist of spherical droplets that have a constant density. The saturator temperatures used were in the range 80 to 120 °C while the flow rate through the saturator was fixed at 4.17 L min^{-1} . Pure NaCl aerosols were generated by collecting particles directly from the output of the diffusion drier of the generator. The salt solution concentration used was 2.0 g L^{-1} while the flow rate used was 4.17 L min^{-1} .

ETS Aerosol Generation. Morawska, et al. have described the ETS generator used (Morawska, Jamriska & Bofinger, 1997). The generator is designed to simulate the frequency, period and volume of inhalations and the deposition of mainstream (directly inhaled from the cigarette) smoke in the lungs of the smoker, but not the chemical and physical changes occurring in the mainstream smoke whilst in the smoker's lungs. The experiment used only the side stream component from the ETS generator and thus represents the aerosol products of a smouldering cigarette without the component exhaled by the smoker.

Aerosol generation and sampling

DEHS Aerosol. The DEHS aerosol was introduced at a constant rate into a 0.125 m³ cubic perspex sample chamber via a dilutor where it was mixed with filtered air introduced at a controlled rate. Mixing of the introduced aerosol with the aerosol in the chamber relied on the internal air circulation generated by the flow of aerosol into and out of the chamber. The aerosol in the chamber was sampled through various sampling points on the walls of the chamber. In order to prevent contamination of the aerosol by ambient air, a slight positive pressure was maintained in the chamber throughout the measurements by ensuring that the combined flow rate of the diluted aerosol into the chamber exceeded the outflow drawn by the instruments. The excess diluted aerosol was vented via small holes in the chamber. Aerosol concentration was adjusted by controlling the dilution air flow rate and the saturator temperature.

NaCl and ETS. For the salt and ETS aerosol measurements, the aerosol was introduced to a larger (3 m^3) sealed experimental chamber. The aerosol concentration was then allowed to decay slowly as the various instruments drew the aerosol from the chamber and it was replaced by clean air drawn through a HEPA filter or from the surrounding laboratory. Mixing of the aerosol with the clean air was assisted through the use of a fan on the floor of the chamber. The aerosol was sampled from points at equal heights above the floor of the chamber. Temperature and relative humidity in the chamber during the NaCl and ETS measurements were in the range $16.8\text{ }^{\circ}\text{C}$ to $20.9\text{ }^{\circ}\text{C}$ and 42.9% to 59.3% respectively.

Ambient Air Aerosols. Ambient air was sampled directly from the outdoor environment at regular intervals over a period of 12 days to obtain data over a range of concentrations.

Experimental Procedure

DEHS Measurements. The small chamber was initially purged using filtered dilution air until the TEOM reading was stabilised. Aerosol mixed with filtered dilution air was then injected into the chamber at a constant flow rate with the instruments running until stable readings were established for both the SMPS and TEOM for a period of 30 minutes. The dilution flow rate and the saturator temperature were varied to control the aerosol concentration, count median diameter (CMD) and geometric standard deviation (GSD).

NaCl Measurements. The experimental chamber was first purged with laboratory air passing through a HEPA filter. Throughout the remainder of the experiment, laboratory air passing through a HEPA filter was allowed to enter the chamber through a small opening in the roof to replace the aerosol drawn by the instruments. The instrument readings were allowed to stabilise while drawing air from the chamber, after which data on the initial background conditions were collected for a period of approximately 30 minutes. The NaCl aerosol was then introduced to the chamber over a period of 2 hours. The instruments continued to sample from the chamber and log the data until the aerosol mass concentration had fallen to the minimum levels measurable by the instruments.

ETS Measurements. The procedure for the ETS measurements was similar to the salt measurements with the two exceptions that no HEPA filter was used for the purging and replacement air and the aerosol was introduced to the chamber over a shorter period of 15 minutes. The HEPA filter was not used during these measurements, as ETS is by definition a mixture of the introduced tobacco smoke with any ambient aerosols already present in the environment. Measurements while the aerosol was being introduced to the chamber are not included in the results as the rate of change in concentration exceeded the ability of the instruments to respond.

Ambient Air Measurements. The ambient air measurements were conducted in summer during a period of high humidity. Ambient outdoor air was sampled directly by the two instruments from the same point. The TEOM sampled and logged the data at 15 minute intervals over the 12 day period while the SMPS recorded the mass spectrum every 4 hours.

Although some comparisons of the TEOM with gravimetric instruments for PM₁₀ air pollution levels have revealed discrepancies as high as 40% at high mass concentrations, the discrepancies have been attributed to the evaporation of volatile components when the TEOM heats the aerosol above the ambient air temperature (Allen, et al., 1997). Low volatility aerosols such as dry NaCl and DEHS are therefore expected to be measured accurately by the TEOM even at high concentrations. Comparison of TEOM and gravimetric methods for respirable suspended particles of ETS show a maximum discrepancy of 14.6% and an average of 5.3% at concentrations of around 1500 $\mu\text{g m}^{-3}$ (Nelson, et al., 1997).

EXPERIMENTAL RESULTS

DEHS Measurements. The data averaging period for the SMPS was 5 minutes. The resulting DEHS mass and number concentrations calculated from the SMPS spectra are plotted against the TEOM measurements in Figure 1. Assuming a linear relationship between the mass concentration values, the R^2 value for the regression is 0.92, indicating that the assumed relationship accounted for 92% of the variance in the data. Although the particle density is constant, the relationship between the number concentration and the TEOM measured mass concentration is complicated by the variation of the particle size distribution with saturator temperature. The R^2 value for the regression, assuming a linear relationship between the number concentration and the mass concentration measured by the TEOM, is 0.53. The estimated density corresponding to the measured mass and volume concentrations can be expressed as the ratio of the TEOM measured mass concentration values to the corresponding

SMPS derived volume concentration values. The density values have an average of 0.95 g cm^{-3} and a standard deviation of 0.15 g cm^{-3} while the bulk density of DEHS is 0.912 g cm^{-3} (*CRC Handbook of Chemistry and Physics, 68th Edition, 1988*). The inaccuracy of the DEHS results in comparison with the NaCl and ETS experiments discussed below is due to the relatively fast rate of change in concentration, which at times exceeded 15% per minute.

NaCl Measurements. The NaCl aerosol measurements were carried out with a slowly changing mass concentration (0.5% per minute) and a 5 minute data averaging interval. In order to overcome difficulties encountered with synchronising the measurements because of differences in the acquisition times of the two instruments, spline functions passing through the data points recorded by each instrument were used as the basis for deriving synchronised data pairs in Figure 2. The NaCl aerosol mass and number concentrations calculated from the SMPS spectra are plotted against the TEOM measurements in Figure 2. The relationship between the TEOM and SMPS derived mass concentration measurements is linear and highly correlated ($R^2 = 0.9981$). The number concentrations also correlate well ($R^2 = 0.9996$) with the TEOM mass concentration measurements due to the fact that neither the particle density nor size distribution varied significantly during the experiment. The average value for the density estimates is 2.22 g cm^{-3} and the standard deviation is 0.11 g cm^{-3} which is very close to the bulk density (*CRC Handbook of Chemistry and Physics, 68th Edition, 1988*) of NaCl at 2.165 g cm^{-3} .

ETS Measurements. The ETS measurements were also carried out with a slowly changing mass concentration (0.5% per minute) and a 20 minute sliding averaging for

the TEOM. Each SMPS sample took 5 minutes to acquire. The ETS aerosol mass and number concentration values calculated from the SMPS spectra are plotted against the TEOM measurements in Figure 3. The number concentration versus TEOM measured mass concentration is linear and highly correlated ($R^2 = 0.9997$) for this aerosol, indicating a constant particle density and size distribution throughout the experiment. The results are consistent with a density of 1.18 g cm^{-3} for the submicrometer component of ETS which compares well with published ETS particles densities of 1.12 g cm^{-3} (Lipowicz, 1988; Chen, Namenyi, Yeh, Mauderly & Cuddihy, 1990).

Ambient Air Measurements. The mass and number concentration values derived from the SMPS measurements are plotted against the mass concentration measurements from the TEOM in Figure 4. In both cases less than 25 % of the variance is accounted for by assuming a linear relationship. The density estimates for the ambient air measurements are summarised in Figure 5. The TEOM mass concentration and SMPS volume concentration display a variation with time of day, probably due to changes in human activity including traffic on the nearby freeway and the half daily alternation of the sea and land breezes (Queensland Department of Environment, 1997). The average particle densities also depended on the time of day, with the averages for different times ranging from 1.2 to 1.8 g cm^{-3} . The overall average submicrometer particle density was found to be 1.7 g cm^{-3} with a standard deviation of 1.0 g cm^{-3} .

A summary of particle densities obtained from these measurements for each aerosol is presented in Table 1.

Table 1. Summary of particle densities obtained for all the aerosols investigated.

Aerosol	Aerosol Material Density (g cm^{-3})	Submicrometer Particle Density $\rho_p(\text{g cm}^{-3})$	$\rho_p(\text{g cm}^{-3})$ [mass concentration < $7.7 \mu\text{g m}^{-3}$]
DEHS	0.912 ¹	0.95±0.15	
NaCl	2.165 ¹	2.22±0.11	^a 2.41±0.28
ETS	1.12 ²	1.18±0.06	
Ambient		1.7±1.0	^b 1.8±1.0

^a average mass concentration = $5.6 \mu\text{g m}^{-3}$, ^b average mass concentration = $4.7 \mu\text{g m}^{-3}$

Comparison of the ambient air density estimates with those for NaCl within the same concentration range as that found for the ambient measurements confirms that the uncertainty in the particle density estimate is much larger in the ambient air case (64%) than for NaCl (12%), and is therefore not due to operation near the TEOM sensitivity threshold. The effects of input cut-point and size range differences between the instruments were reduced by extrapolating the SMPS cumulative volume distribution to confirm that the contribution of the uncounted portion of the true distribution from the impactor cut-point (for the initial estimate of the average particle density) to $1 \mu\text{m}$ was less than 10%. The most likely cause of the greater uncertainty in the density estimates for the ambient particles appears therefore to be that the average particle density fluctuated (with a standard deviation of 1.0 g cm^{-3} about a mean of 1.7 g cm^{-3}) due to changes in the proportions of particles from various sources present in the atmosphere.

¹ (CRC Handbook of Chemistry and Physics, 68th Edition, 1988)

² (Lipowicz, 1988; Chen, et al., 1990)

DISCUSSION AND CONCLUSIONS

The method of experimental correlation of particle mass and number using a TEOM and an SMPS operating in the submicrometer size range was developed and validated using two types of aerosols of known density. For single mode polydisperse DEHS aerosols in the region of 100 nm, densities corresponding to calculated volume concentrations derived from SMPS size distributions and TEOM mass concentration measurements were within 14% of the density of bulk DEHS at standard temperature and pressure. For the NaCl aerosols in the region of 150 nm, densities found by this method were within 5% of the density of bulk NaCl at standard temperature and pressure for aerosols ranging in concentration from 10 to 1400 $\mu\text{g m}^{-3}$ and ranging in CMD and GSD from 191 to 197 nm and 1.5 to 1.6 nm respectively. The greater error in the DEHS measurements is largely attributable to the relatively fast rate of change in the aerosol concentration and differences in the rate of response of the various instruments.

The method was then applied to two types of environmental aerosols: ETS and ambient air particles. For the ETS aerosol the SMPS derived mass concentration values when compared with the TEOM values indicate a sub-micrometer particle density of $1.18 \pm 0.06 \text{ g cm}^{-3}$. Densities published for ETS have included $1.12 \pm 0.02 \text{ g cm}^{-3}$ (Lipowicz, 1988) and similarly 1.12 g cm^{-3} (Chen, et al., 1990). For ambient air the derived density was $1.7 \pm 1.0 \text{ g cm}^{-3}$.

The results of the investigations indicate that when particle density is constant the correlation between the SMPS determined submicrometer volume concentration and

the TEOM determined submicrometer mass concentration is strong. However, the same procedure used for ambient air demonstrates that such aerosols show considerable variation in average density, which is indicated by a much weaker correlation between TEOM and SMPS results. Any correlations between SMPS calculated and TEOM measured mass would need to be treated on a case by case basis with conditions for constant average density being established before determining an average density to be assumed in future measurements.

Due to the uncertainty in the density of the ambient air submicrometer particles, it would not be possible to use SMPS data alone to give an estimate of the submicrometer mass concentration with better than 60% uncertainty based on an assumed density value. However it may be possible to ensure that an average density can be assumed with greater accuracy by choosing more narrowly defined conditions of wind direction, wind speed, and time of day. Ambient air dominated by nearby sources such as those found downwind of fine particle sources should also show more reliable average submicrometer particle densities.

The use of the TEOM and SMPS was found to be one possible method for determining the submicrometer density of an aerosol. While the method used has the advantage that the instruments are used in an unmodified form, it permits a mismatch between the cut-point diameter for the input conditioning of the aerosol to the two instruments and a shortfall of 70 nm in the range of the SMPS compared with the range used for the TEOM. For a free-ranging aerosol such as ambient air, it is necessary to test each particle distribution measurement for evidence of a significant fraction of the submicrometer particle volume lying in the region between the upper

range boundaries of the SMPS and the TEOM. Several approaches can be suggested for improving the accuracy of the technique and overcoming these problems. A common cut-point might be achieved through adjustments to the parameters of either or both input conditioning devices, however a preferable scheme would involve the use of a separate input conditioning device affecting the common flow to both instruments adjusted via an auxiliary flow to match the required SMPS cut-point.

While a fixed average particle density was assumed in the calculation of mass concentration from the particle size distribution of the aerosol at the time of measurement, the calculation of number concentration from mass concentration further assumes a fixed size distribution. The fact that for ambient air, the correlation between measured mass concentration and number concentration is of similar strength to that obtained for mass concentration and volume concentration implies that the main sources of error in converting from submicrometer mass concentration to submicrometer number concentration for ambient air is variation in the average particle density.

For indoor aerosols of known origin, the correlation between number concentration and mass concentration is strong and suitable factors for converting between the mass and number can be estimated.

Acknowledgments- This research was supported by the Queensland Department of Environment and the ARC Collaborative Grant C295301023.

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³ (CRC Handbook of Chemistry and Physics, 68th Edition, 1988)

⁴ (Lipowicz, 1988; Chen, et al., 1990)