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Errors in particle size distributions from Sequential Mobility Particle Sizers due to varying number concentration at an urban roadside location

Matthew D. Wright*,[1]

H. H. Wills Laboratory, School of Physics, University of Bristol, Tyndall Avenue, Bristol BS8 1TL UK

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ABSTRACT

Scanning or Sequential Mobility Particle Sizers (SMPS) are commonly used to obtain number size distributions (NSD) for submicron aerosols. In SMPS data inversion, correction for particle multiple-charging typically utilises data obtained at earlier times, therefore there is potential for error if concentration varies over the cycle time. We examine this error by using roadside experimental data to define simulated SMPS response distributions, including a time-varying transient peak due to a particle influx event, for input to inversion procedures. Because this distribution is specified for all times, the correct multiple-charging contributions can be used in data inversion. The results are compared with NSD obtained using a standard algorithm. We show that the net effect is to underestimate the true concentration, especially for diameters 80–200 nm (for the instrumentation studied here). For parameters typical of observed roadside transient events, errors in total concentration are ~2%, with up to ~8% error for concentration at individual diameters, and in all but the most extreme cases are less than typical experimental uncertainty estimates for SMPS instruments at roadside sites. Many previously published roadside size distributions may be affected, and for only a modest improvement in time resolution the effect may be exacerbated. The error introduced for given particle influx characteristics (which can be determined experimentally in future work and may be estimated for previous works) and for different instruments can be easily evaluated.

1. Introduction

Characterisation of aerosol particle properties and concentration is important for the determination of their potential health impacts (Pope et al., 2002; Russell & Brunekreef, 2009). In urban areas, the predominant source of particulate matter is often vehicular exhaust, especially from diesel and heavy-duty types. Of increasing interest in terms of health effects is the submicron and especially the ultrafine particle (UFP, <100 nm) diameter range (Donaldson et al., 1998; Wichmann et al., 2000; Delfino et al., 2005; Schulz et al., 2005), produced abundantly by vehicles and present in large numbers at roadsides (Hitchins et al., 2000; Kittelson et al., 2004; Zhu et al., 2009). At roadside locations, particle number concentration (PNC) is often highly variable, as individual vehicles emit particles in their immediate vicinity (Kurniawan & Schmidt-Ott, 2006;

* Tel.: +44 117 9288765; fax: +44 117 9252990.
E-mail address: matthew.wright@bristol.ac.uk
[1] Present address: Atmospheric Chemistry Research Group (ACRG), School of Chemistry, University of Bristol, Cantock's Close, Bristol BS8 1TS, UK.

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Buzzard et al., 2009) or where traffic conditions are variable leading to vehicle acceleration/deceleration patterns, such as at signals or during congestion (Lingard et al., 2006; Tsang et al., 2008; Minoura et al., 2009). A subsequent drop in the number concentration at the roadside site, due mainly to dilution processes, is then observed in the absence of further particle influxes from vehicles (Shi et al., 1999; Ketzel & Berkowicz, 2004; Zhang & Wexler, 2004). Therefore ideally, high temporal resolution is required for the study of particle emissions at roadside sites.

Many studies of submicron particle size distributions in urban areas and at roadsides (e.g. Charron & Harrison, 2003; Birmili et al., 2009; Davison et al., 2009; Zhu et al., 2009) have used Sequential or Scanning Mobility Particle Sizers (SMPS) comprising a Differential Mobility Analyser (DMA) for electrical mobility selection followed by a Condensation Particle Counter (CPC) or other instrumentation for particle counting (Knutson & Whitby, 1975; Wang & Flagan, 1990; Winklmayr et al., 1991; Heim et al., 2004). These often require a period from 30 s up to several minutes (per DMA voltage cycle) to obtain a size distribution over the full range of operation. During this time, if the particle concentration changes significantly, inaccuracies may be introduced in the calculation of particle size distribution, concentration and mean diameter. Essentially, the problem arises because the contribution of multiply-charged, larger particles must be removed from the system response to obtain a size distribution. To achieve this, data collected at earlier times in the voltage cycle must be used, which may be inaccurate at later times.

Some studies have used DMA+CPC systems at fixed voltage (Maricq et al., 1999; Bowker et al., 2007) to improve time resolution of measurements, at the cost of losing information across a wider size range. If particle production is repeatable from run to run, multiple sizes can be selected over multiple measurements (Maricq et al., 1999) but at roadside locations this is clearly not the case. 'Fast MPS' systems have been developed in recent years (e.g. Tammet et al., 2002; Shah & Cocker, 2005) and are now commercially available (FMPS, Model 3091, TSI Inc., Shoreview, MN, USA; FAPES, Model #5.600, Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany; DMS500 Mk.II, Cambustion Ltd., Cambridge, UK). However, their response to various particle types appears to differ from SMPS systems, and the reasons for this are not yet fully understood (Jeong & Evans, 2009; Asbach et al., 2009). In order to prolong the use of existing low time-resolution SMPS systems for this type of measurement, it is beneficial to determine in which circumstances they can still reliably be used.

This study explores the effect of sudden, short-lived increases in particle concentration, termed 'influx events', on the calculation of size distributions from SMPS. Previous work has examined the effect of varying particle concentration on short-term data sets (Yao et al., 2006a, b). Here, the effect on long-term measurements is examined by specifying the system response to a simulated 'influx event' for all times during the DMA voltage cycle. This allows data inversion to proceed using multiple-charging contributions from concentrations at the relevant time of measurement, rather than those obtained from earlier in the cycle. Comparison of size distributions retrieved from each inversion procedure reveals the extent of the error introduced by the influx event. The method is applied to experimental data collected at two urban roadside sites in central Bristol, UK, to determine typical influx event characteristics at these locations, in order to estimate the likely magnitude of this error in this case. We discuss how different influx event characteristics, influenced by a variety of factors, may influence this error at other locations and for different meteorological and traffic conditions. Although this work focuses on one commercially available 'sequential' or voltage-stepping system, the Sequential Mobility Particle Sizer and Counter (SMPS+C, Grimm Aerosol Technik GmbH & Co. KG) in certain operating conditions, similar principles apply to other instruments, including 'scanning' systems.

2. Background and theory

2.1. Data inversion procedure

Several inversion procedures exist for obtaining aerosol size distributions from SMPS data (e.g. Knutson, 1976; Hoppel, 1978; Alofs & Balakumar, 1982; Collins et al., 2002). A key aspect is the removal of the contribution of multiply-charged, larger particles to the system response for each electrical mobility sampled. The following discussion is based on the work of Reischl (1991), which itself follows the Knutson (1976) linear inversion method, accounting for multiply-charged particles by solving a system of equations using measurements made at larger particle sizes as inputs to later calculations. Briefly, the response of the CPC for a given DMA voltage V, $R(V)$, is given by

$$R(V) = \frac{Q_a^2}{Q_{sh}} \sum_{q=1}^{\infty} E(d)\Psi(d)\alpha(d,q)\frac{dN}{d\ln d}\bigg|_{d=d_q}, \tag{1}$$

where d is particle diameter, Q_a and Q_{sh} are, respectively, the volumetric flow rates of the sample and sheath air flows through the DMA, q is the number of electronic charges on the particle, $E(d)$ is the size-dependent counting efficiency of the CPC (assumed here to be 1), $\alpha(d,q)$ is the charging probability for particles of diameter d to carry q charges and $(dN/d\ln d)|_{d=d_q}$ is the input size distribution evaluated at mobility-equivalent diameter d_q. The function

$$\Psi(d) = -((d(d)/d)/(dZ/Z)) = (C_c(d)/d^2)/(\partial(C_c/d)/\partial d), \tag{2}$$

where Z is electrical mobility, represents the influence of slip correction, C_c, on the diameter resolution of the DMA, where (Hinds, 1982)

$$C_c = 1 + \frac{\lambda}{d}[2.514 + 0.800\exp(-0.55(d/\lambda))], \tag{3}$$