

Accepted refereed manuscript of:

Price H, Stahlmecke B, Arthur R, Kaminski H, Lindermann J, Dauber E, Asbach C, Kuhlbusch TAJ, BeruBe K & Jones T (2014) Comparison of instruments for particle number size distribution measurements in air quality monitoring, *Journal of Aerosol Science*, 76, pp. 48-55.

DOI: [10.1016/j.jaerosci.2014.05.001](https://doi.org/10.1016/j.jaerosci.2014.05.001)

© 2015, Elsevier. Licensed under the Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International <http://creativecommons.org/licenses/by-nc-nd/4.0/>

Final draft

1 **Comparison of instruments for particle number size distribution measurements**
2 **in air quality monitoring**

3

4 Price, H.D^{1*}, Stahlmecke, B^{2.}, Arthur, R.A^{3.}, Kaminski, H^{2.}, Lindermann, J^{2.},
5 Däuber, E^{2.}, Asbach, C^{2.}, Kuhlbusch., T.A.J.^{2.}, Bérubé, K.A^{4.}, Jones, T.P¹

6

7 ¹School of Earth and Ocean Science, Cardiff University, Park Place, Cardiff, CF10
8 3YE, UK.

9 ²Institute of Energy and Environmental Technology (IUTA e.V), Bliersheimer Strasse
10 58-60, 47229 Duisburg, Germany.

11 ³Centre for Health and Environment Research, Department of Primary Care and
12 Public Health, Neuadd Meirionydd, 4th Floor, Heath Park, Cardiff, CF14 4YS, Wales,
13 UK.

14 ⁴School of Biosciences, Cardiff University, Museum Avenue, Cardiff, CF10 3US,
15 UK.

16

17 ***Corresponding author (present address):**

18 Address: Geography and Environment, Shackleton Building 44, University of
19 Southampton, University Road, Southampton, SO17 1BJ, UK

20 Email: h.price@soton.ac.uk/heatherprice127@gmail.com

21

22

23

1 **Highlights**

- 2 • We compared number size distributions from ELPI, SMPS, FMPS and APS
- 3 • Results from four lab generated aerosols were compared in a wind tunnel
- 4 • Good correlation was found between instruments in their middle size ranges
- 5 • At the lower and upper particle diameters there were divergences
- 6 • Particle type (size and shape) affected the correlation between instruments

7

8

Final draft

1 **Abstract**

2 Number size distributions of airborne particles are relevant to fields including ambient
3 monitoring, pharmaceutical and automotive measurements. A number of
4 commercially available instruments can be used to determine particle number size
5 distributions including the Electrical Low Pressure Impactor (ELPI), Scanning
6 Mobility Particle Sizer (SMPS), Fast Mobility Particle Sizer (FMPS) and the
7 Aerodynamic Particle Sizer (APS). The comparability of the data provided by these
8 instruments has not been fully tested for different kinds of aerosols. This study
9 compared number size distributions of laboratory generated aerosols (TiO₂, NaCl,
10 fumed silica and soot) in a wind tunnel. Reasonable agreement was noted between the
11 different instruments, though there were divergences. For example the ELPI was
12 inconsistent at the upper and lower limits of its working size limits (at low
13 concentrations). Instruments responded variably to different particle types, which has
14 important implications for sampling heterogeneous particle mixtures such as those
15 found in urban air. This study highlights the need for caution when comparing data
16 obtained from different particle instruments, and demonstrates the requirement for
17 further comparison studies in controlled settings using an assortment of particle types
18 with the aim to standardise and harmonise particle sampling protocols.

19

20 **Key words**

21 ELPI, SMPS, FMPS, APS, number size distribution (NSD)

22

23

1 **1. Introduction**

2 Different particulate matter size distribution instruments are often used
3 interchangeably, or to extend the measured particle size range within a single study,
4 thus implying that the values they provide are comparable. Examples include studies
5 where a combination of an ELPI (Electrical Low Pressure Impactor) and multiple
6 SMPS (Scanning Mobility Particle Sizers) were used to analyse the vertical particle
7 profiles on either side of a motorway (Imhof et al., 2005), and an investigation in
8 which an APS (Aerodynamic Particle Sizer) was used to extend the SMPS sampling
9 size range in an urban atmospheric study (Harrison et al., 2000). Some studies have
10 illustrated that this is accurate, at least to an acceptable extent, for example in roadside
11 particle measurements in Birmingham, UK (Shi et al., 1999a), and a study of
12 particulate matter (PM) from motor exhausts (Ushakov et al., 2013), both of which
13 compared results from an SMPS and an ELPI. In contrast, other studies have not
14 identified such consistent comparability. These include a study where test aerosols
15 were used to analyse the variability in number and mass values provided by aerosol
16 collection equipment including an ELPI and two APS models (Pagels et al., 2005). In
17 that study the sub-micrometre scale values measured were found to be precise and
18 accurate, however for larger particles the ELPI and one APS model (3320) were
19 found to overestimate the particle concentration, while the second APS model (3321)
20 underestimated the concentration. In a study using an ELPI, APS and SMPS, while
21 comparability was identified between the ELPI and SMPS in the sub-micrometre
22 particle size range, the ELPI was found to overestimate the number concentrations of
23 larger particles (Nussbaumer et al., 2008).

24

1 There is therefore uncertainty regarding the accuracy and vigour of comparing results
2 from various instruments, especially those based on different measurement principles.
3 It is, however, essential that these results are comparable, as the associated advantages
4 and disadvantages of using the different particle counting techniques encourage the
5 use of different types of equipment in different studies (Table 1). The need for
6 standardisation of particle size distribution devices in order to provide confidence in
7 the comparison of particle data from different instruments was stated in 2001
8 (Dahmann et al., 2001). Ten years later, results from further investigations (Asbach et
9 al., 2009, Kumar et al., 2010) reaffirmed that this requirement still exists. Thus,
10 further comparisons are needed before standardising procedures can be implemented.

11
12 While a number of studies have investigated particle size distributions in a variety of
13 settings (e.g. atmospheric [Wehner et al., 2002], engine cycle [Shi et al., 1999b],
14 indoor air [Long et al., 2000] and occupational exposures [Stroszejn-Mrowca and
15 SzadKowska-Stańczyk, 2003]), studies focussing on the comparison between devices
16 which help to validate the results from these studies and support their conclusions are
17 more scarce. Some equipment comparison studies have dealt with near-spherical
18 particles including Dioctyl Sebacate (DOS; e.g. Keskinen et al., 1992; Marjamäki et
19 al., 2000) however the properties of these particles (i.e. spherical morphology and
20 liquid state) encourage improved detection by the instruments (Van Gulijk et al,
21 2004). Conversely, in situations where particle number size distributions (NSDs) are
22 of interest, for example in atmospheric studies, perfect spherical particles are less
23 common (Shi et al., 2001). Other studies have compared results from different
24 instruments with experimental methodologies which include sampling in urban
25 locations (Held et al., 2008) and workplaces (Brouwer et al., 2009). While this

1 provides useful comparison, these are changeable environments. By ensuring that all
2 devices are exposed to the same concentrations in a controlled laboratory setting, the
3 variability (including particle number concentration, morphologies and sizes) to
4 which the instruments are exposed to is reduced. Asbach et al. (2009) used such a
5 controlled environment but compared only instruments based on electrical mobility
6 analysis.

7

8 In this study four instruments for measuring airborne particles were compared; an
9 Electrical Low Pressure Impactor (ELPI, Dekati), Scanning Mobility Particle Sizer
10 (SMPS, TSI model 3936-L86), Fast Mobility Particle Sizer (FMPS, TSI model 3091)
11 and an Aerodynamic Particle Sizer (APS, TSI model 3321) which work on different
12 operational principles. The ELPI operates at a flow rate of 30 L/min and separates
13 particles onto impaction plates based on their inertia, a function of their size
14 (Keskinen et al., 1992). Particles are classified into twelve size fractions within the
15 range 7 nm – 10 µm dependent upon their aerodynamic diameter (Marjamäki et al.,
16 2000) with 1 s time resolution. Electrical current carried by the charged particles
17 (imparted by a unipolar charger on entry) in each size fraction is then converted to
18 particle number concentration. The SMPS comprises of an Electrostatic Classifier
19 (EC) for particle sorting, followed by a CPC for particle counting (Wang and Flagan,
20 1990). Sampled particles are bipolarly charged to a known charge distribution in a
21 ⁸⁵Kr neutralizer before they enter the EC. Particles then navigate an electrical field
22 where their ability to pass through (dependent upon their electrical mobility, and
23 therefore proportional to the ratio of particle charge to diameter) separates the
24 particles. The CPC counts the mobility-classified particles and along with the known
25 charge distribution thus allows for the calculation of the NSD. The SMPS was

1 operated with a long Differential Mobility Analyser (DMA) with an aerosol to sheath
2 flow ratio of 0.3/3 l/min, thus measuring in a particle size range from 14 to 750 nm
3 (electrical mobility diameter) with a potential resolution of 64 channels per size
4 decade. Time resolution of the SMPS was set to 240 s scan, 20 s retrace and 40 s wait
5 time, i.e. 5 minutes per scan. The FMPS uses the same separation principles as the
6 SMPS, however uses a unipolar charger and utilises an array of sensitive
7 electrometers for the simultaneous detection of the full mobility range (Crooks, 2011).
8 The FMPS measures in the size range from 5.6 to 560 nm (electrical mobility
9 diameter) with a resolution of 16 channels per decade and a time resolution of 1 s.
10 The APS works on the principle that once preliminarily accelerated, particles move at
11 speeds in proportion to their diameter (Shi et al., 2001). Particle profiles are identified
12 by measuring particle velocity between two laser beams, which is then converted to
13 particle diameter. The APS measures particle size distributions in the size range from
14 0.5 to 20 μm (aerodynamic particle diameter) in 32 channels per size decade and with
15 up to 1 s time resolution. In summary, the APS and ELPI determine the particle size
16 distribution based on the aerodynamic diameter while the SMPS and FMPS are based
17 on the electrical mobility diameter.

18
19 The aim of this study was to compare the outputs of the four aerosol sizing
20 instruments (ELPI, SMPS, APS, FMPS) with overlapping size ranges, when
21 challenged by four particle types with differing particle morphologies and size
22 distributions within a controlled atmosphere.

23

24 **2. Experimental**

25

1 **2.1 Aerosol generation**

2 Four particle types (TiO₂, NaCl, soot and fumed silica) representing a variety of
3 particle sizes, morphologies and chemical compositions were used to challenge the
4 instruments. The importance of using a variety of particle types, particularly those
5 found in urban air, or simulating particles found in urban air has been highlighted in
6 previous studies (e.g. Khlystov et al., 2001). Soot particles were generated using an
7 experimental soot generator (PALAS, Defined Soot Particle Generator, DSP 3000)
8 using Ethene (C₂H₄). TiO₂ (P25, Degussa), fumed silica (Cabot, UK) and NaCl
9 aerosols were generated using a TriJet atomizer (TriJet 3460, TSI inc.) by first
10 dispersing a specific amount of powder in 500 ml deionised water within an ultrasonic
11 bath for 20 minutes (TiO₂: 7.4 g/l; fumed silica: 3.8 g/l; NaCl: 10.0 g/l). After
12 generation, TiO₂, NaCl and fumed silica were dried using two PermaPure driers (50
13 Nafion membranes each). The aerosol was dried with these consecutive dryers to a
14 relative humidity of <20%. Soot particles were introduced directly before the flow
15 straightener (Figure 1), while TiO₂, NaCl and fumed silica were introduced into the
16 mixing chamber. Particles were not neutralized prior to injection into the wind tunnel.

17

18 **2.2 Measurement set-up**

19 Measurements were conducted in a wind tunnel at the Institute of Energy and
20 Environmental Technology (IUTA), Duisburg, Germany. Flow rate was set to
21 1000 m³/hr which corresponded to a speed of 1.3 m/s within the tunnel at the
22 sampling points. The instruments were connected to the wind tunnel through four
23 inlets with different diameters, each pre-calculated to allow isokinetic sampling
24 (Table 2). At the point of sampling, the wind tunnel had a width and height of 63 cm.
25 Sampling inlets were located in the centre of the wind tunnel with a spacing of 5 cm

1 (Figure 1). A hygrometer (temperature and humidity probe from Hygrosens), placed
2 after the sampling inlets 20 cm below the upper wall of the wind tunnel, was used to
3 monitor relative humidity and temperature during the measurements. The temperature
4 was nearly constant for all measurements with a value between 20° - 21° C for the
5 TiO₂, NaCl and soot measurements and of 18°C for the fumed silica measurements.
6 The relative humidity differed for the substances and the following values were
7 observed: TiO₂: 45% rH, NaCl: 41% rH, fumed silica: 36% rH. At the beginning of
8 the soot measurements the humidity had a value of 35 % rH which changed within 0.5
9 hours to a value of only 17 % rH. All instruments were exposed to identical rH
10 conditions during measurement.

11
12 Sampling inlets were located 2 m down-tunnel of a flow straightener, and therefore
13 laminar and uniform flow at the instrument inlets was assumed during sampling. After
14 the point of sampling a HEPA filter was used to efficiently remove particles. The
15 number concentration of particles during measurement was approximately 10⁴-10⁶
16 particles/cm³, which was suitable for the instruments included in the study. Particle
17 Number Concentrations (PNCs) were left to stabilise before the measurements took
18 place. This eliminates the effects of “influx events” which could affect the SMPS
19 measured concentrations (Wright, 2014). These “influx events” are short term
20 increases in PNC, such as may be found at the roadside and can affect instruments
21 with lower time resolution. The ELPI, FMPS, SMPS and APS had individual
22 sampling inlets. Due to different instruments requiring different flow rates, each inlet
23 had different inner diameters which were configured for the individual instrument to
24 provide isokinetic sampling (Table 2). The instruments were connected to the inlets
25 using flexible inert tubing which was as short as was practically possible. The

1 concentration of the particles in the wind tunnel was not accurately controlled because
2 the focus was a comparison between instrument responses rather than measuring
3 absolute values of PNC. Diffusion and other losses were not taken into account as in
4 previous studies (Leskinen et al., 2012).

5

6 **2.3 Sampling periods**

7 Each sampling period consisted of two hours of particle measurement at fairly stable
8 PNC (PNC varied by around 10% during the measurement period for TiO₂, silica and
9 soot, and by around 30% for NaCl in comparison to the mean concentration value).
10 The ELPI was cleaned and collection substrates replaced after each sampling period.
11 The instruments counted particles in different size bins and used different particle
12 sizing methodologies (electrical mobility/aerodynamic), making comparison more
13 difficult (Khlystov et al., 2001; Table 2). In this study comparison was made between
14 the NSDs based on the aerodynamic diameter of the particles as provided by the
15 different instruments. The ELPI and the APS directly measure the aerodynamic
16 diameter of airborne particles. The measurements of the SMPS and FMPS were
17 converted from electrical mobility diameter into aerodynamic diameter using an
18 effective density for each particle type. The effective density was chosen so that the
19 measured size distribution of the APS and SMPS gave a near-continuous NSD over
20 the whole measurement size range. Instrument model type, model description and
21 inlet details are provided for each of the devices used in the comparison in Table 2.
22 Further information on the handling and theory of operation for the different
23 instruments is available within the manuals provided by the manufacturers.

24

25

1 **3 Results**

2 The NSDs of the different substances are shown in Figure 2 as a function of
3 aerodynamic diameter. All distributions showed particles over the whole size range
4 with modal diameters of ranging from 25 nm for soot, 80 nm for NaCl, 105 nm for
5 TiO₂ and 110 nm for fumed silica.

6
7 Between 50 and 300 nm reasonable agreement was noted between the measurement
8 devices (particle number concentrations within 25 %; SMPS, FMPS and ELPI) for
9 TiO₂ (Figure 2a). Below 50 nm, the ELPI showed higher concentrations of particles in
10 contrast to the SMPS and FMPS. Above 1 μm, the ELPI measured higher
11 concentrations than the APS. Generally, the APS was shown to extend the sampling
12 range of the SMPS for TiO₂, with reasonable overlap. The standard deviations of the
13 results provided by a single instrument were generally small, indicating stable number
14 size distributions during the measuring period; however at the outer size limits of the
15 detection ranges of the ELPI and APS the variation was greater. In contrast, standard
16 deviations for the FMPS and SMPS were consistent throughout their sampling ranges.
17 While the SMPS, ELPI and APS correlated to a unimodal distribution, the FMPS data
18 showed a bimodal distribution.

19
20 For NaCl (Figure 2b), the SMPS, FMPS and APS showed satisfactory agreement
21 between distributions. Standard deviations were low, except for particles below 20 nm
22 measured with the FMPS, and some size fractions measured with the APS. In contrast
23 to the unimodal distribution shown by the SMPS, FMPS and APS, the ELPI displayed
24 a bimodal distribution, with a peak below 20 nm which was significantly greater than
25 measured by the SMPS and FMPS. A concentration minimum was noted at around

1 85 nm in contrast to the correlating SMPS and FMPS data. As for TiO₂, above 1 μm
2 the ELPI measured values higher than the APS; however the shape of the distribution
3 was comparable.

4
5 For soot particles (Figure 2c) there was agreement of a bimodal distribution with
6 peaks at 25 nm and 110 nm by the SMPS, FMPS and ELPI, though some deviation
7 was noted with regards to the FMPS following the second peak where values obtained
8 were lower than provided by the other instruments. There was discontinuity in the
9 overlapping portions of the SMPS and APS, with much lower concentrations
10 observed by the APS in comparison to the SMPS, FMPS and ELPI, and with a
11 differing distribution shape. Standard deviations for the SMPS and FMPS were small,
12 and this was also generally the case for the ELPI and APS.

13
14 While the instruments provided relatively comparable data for TiO₂, NaCl and soot, in
15 the case of fumed silica (Figure 2d), much more variability was noted between the
16 different particle size distributions. Agreement of data from the SMPS, FMPS and
17 ELPI was noted between 150 nm and 500 nm (within 15%), and this extended to
18 between 60 nm and 600 nm (within 8%) when considering only the SMPS and FMPS.
19 The ELPI showed higher concentrations of particles than the other instruments in
20 those size ranges below 100 nm and above 1 μm. Similarly to TiO₂, the particle
21 number size distribution for fumed silica provided by the FMPS was bimodal, in
22 contrast to the other samplers, which all provided a unimodal distribution. Again, the
23 ELPI and APS showed greater standard deviations in the upper sections of their
24 working ranges, with the FMPS also showing increased variability in the lower
25 section of its measuring range.

1

2 **4. Discussion**

3 **4.1 Calculation of effective density**

4 In order to make the NSDs measured by the different instruments comparable, the
5 data from the FMPS and SMPS were converted to aerodynamic diameter using an
6 assumed value of effective density for each of the substances (Equation 1), where D_{ae}
7 = aerodynamic diameter, D_m = mobility diameter, ρ_p = effective particle density, and
8 ρ_0 = density of water.

9

$$10 \quad D_{ae} = D_m \sqrt{\rho_p / \rho_0} \quad \text{Equation 1}$$

11 The effective density was chosen to obtain a smooth NSD over the whole size range
12 according to the SMPS/ APS data within the overlap region, and to compare to
13 literature density values. This method has been used in previous studies (e.g. Pitz et
14 al., 2011). The effective density used for NaCl particles was 2.164 g/cm³, nearly
15 identical to the bulk value of 2.165 g/cm³ (Lide, 2004). This very good agreement is
16 due to the fact that NaCl forms compact particles. The bulk density of TiO₂ particles
17 is approximately 4.000 g/cm³ (Dewalle et al., 2010), however in this study, to reflect
18 the agglomerated nature of the particles and to provide fit between APS and SMPS, an
19 effective density of 0.900 g/cm³ was used. This low effective density indicates that the
20 TiO₂ were loosely packed agglomerates. The effective density used for soot particles
21 was 1.700 g/cm³. This was within the density range of soot agglomerates identified in
22 previous studies of between 0.560 and 1.780 g/cm³ (Zhang et al., 2008, Evans et al.,
23 2003). In a previous study, for fumed silica particles of 10 nm size, an effective
24 density similar to the bulk density was identified of 2.200 g/cm³ (Keskinen et al.,

1 2011). For larger, loose agglomerates the effective density would be lower, as in this
2 study, and a density of 1.300 g/cm^3 provided continuity to SMPS/ APS data.

3

4 **4.2 Comparison of instrument performance**

5 **SMPS and FMPS**

6 For all four measured particle types, the SMPS was generally found to correlate well
7 with the FMPS (Figure 2). This was especially the case for NaCl and soot particles,
8 which was interesting since these particles possess very different morphologies
9 (Asbach et al., 2009), and in this study had differing modal particle diameters
10 (obtained from NSDs) of approximately 80 nm and 25 nm respectively. A previous
11 comparison of a SMPS and a FMPS in a laboratory study testing salt and gold
12 particles produced highly comparable results (Jeong and Evans, 2009), though
13 multimodal NSDs were identified from the FMPS. This effect was also seen in two of
14 the sampled particle types in this study; TiO_2 (Figure 2a) and fumed silica (Figure 2d).
15 A peak not identified in the SMPS data was noted at 15.7 nm aerodynamic diameter,
16 corresponding to 16.5 nm electrical mobility diameter in the TiO_2 and fumed silica
17 FMPS data. This is comparable to previous studies which have identified a small peak
18 at 10 nm (using salt particles; Asbach et al., 2009) and 10.7 nm (using salt and gold
19 particles; Jeong and Evans, 2009). It has been proposed that the erroneous FMPS
20 peaks were a result of the algorithm used in the conversion of raw data to a size
21 distribution in the FMPS (Jeong and Evans, 2009). This was only observed for SiO_2
22 and TiO_2 particles, and not for soot and NaCl. This may be due to the low
23 concentration of particles for the former two materials in comparison to the latter two
24 materials which had concentrations two and three orders of magnitude higher. The
25 FMPS is considered a less sensitive instrument than the SMPS (Morawska et al.,

1 2009), and there has been little research focused on the accuracy of data provided by
2 the FMPS and how it relates to other sampling instruments. In this study the FMPS
3 was generally found to correlate well with SMPS data, however there are clearly
4 divergences, specifically below 50 nm aerodynamic diameter, and it is currently
5 unknown whether this is a particle size or particle morphology effect (Asbach et al.,
6 2009), or the result of another aspect of the sampling procedure entirely.

7

8 **ELPI**

9 Particle NSDs measured by the ELPI generally showed agreement with the SMPS,
10 FMPS and APS, however this agreement was much poorer at the upper and lower
11 ends of the ELPI working range, especially for TiO₂, NaCl and fumed silica. This
12 general comparability acknowledged between the ELPI and the other instruments has
13 been identified in a number of studies investigating different particle types, including
14 monodisperse and polydisperse DOS (Hillamo et al., 2000), biomass combustion
15 (Nussbaumer et al., 2008), atmospheric urban studies (Shi et al., 1999a), and engine
16 test bed systems (Shi et al., 1999b). In the engine test bed system and atmospheric
17 study a filter stage was not used, meaning particles below 30 nm were not measured.
18 In this study, data from the filter stage (7-28 nm) was found to be significantly higher
19 than the other instruments for TiO₂, NaCl and fumed silica. The apparent
20 overestimation of particle counts in the nano-size range has been noted in previous
21 studies (Maricq et al., 1999; Held et al., 2008). This disparity has been attributed to
22 the differing sizing principles between the ELPI (aerodynamic diameter) and the
23 SMPS (electrical mobility diameter). Additionally, particle bounce (Marjamäki et al.,
24 2000) and particle deagglomeration (Brouwer et al., 2009) might play a role. In this
25 study, ELPI overestimation on the filter stage was found for not only agglomerates

1 but also compact particles, for example NaCl. This suggests that particle
2 deagglomeration was unlikely to be a predominant cause of overestimation. Particle
3 bounce has previously been identified to cause increases in PNC at the lower working
4 range of the ELPI (e.g. Leskinen et al., 2012). Due to the high impaction velocity,
5 particles which bounce have been found to collect on the filter stage of the ELPI
6 (Virtanen et al., 2010), thus increasing the current produced by this size bin.

7

8 The ELPI measured significantly higher levels of coarse particles than the APS for
9 TiO₂ and fumed silica, a finding which replicated previous studies (Hillamo et al.,
10 2000; Evans et al., 2003). This has been hypothesised by these authors to be a result
11 of the low number of particles in the coarse size fractions or the low total charge that
12 these particles carry which would be consistent with the small mean diameters of
13 particles generated in this study.

14

15 The length of sampling may have been an additional factor affecting the reliability of
16 the particle data at the upper and lower size limits of the ELPI. Previous laboratory
17 studies have shown that after sampling periods of 60 seconds (Van Gulijk et al., 2001)
18 and 20 minutes (Maricq and Xu, 2004) there can be a shift in the particle size
19 distribution. Particles build up on the substrate surface during the collection period
20 into “domes”. These have the potential to affect the cut off diameters for the different
21 stages and cause particle bounce, shifting the modal diameter towards more coarse
22 particles. In addition, the ELPI has a more coarse size resolution than the other
23 instruments, which means that part of the detail in the size distributions is missing.
24 This might account for a portion of the difference between size distributions.

25

1 **APS**

2 The APS was found to extend the sampling range of the SMPS for NaCl and TiO₂ in
3 this study, though standard deviations of observed values became large above 4 µm in
4 the case of TiO₂. This continuance of SMPS data was similar to a previous traffic
5 study (Harrison et al., 2000). However, the APS has been previously found not to
6 handle fluffy agglomerates well (Tsai et al., 2008). This was identified in this study in
7 the largest size fractions. The APS accelerates particles with a defined force in a
8 nozzle. ‘Fluffy’ or loose agglomerates may tend to deagglomerate in this nozzle and
9 this may lead to discontinuities when comparing values with other particle sizers. In
10 addition particle losses from the APS, potentially at the surface of the inner nozzle
11 due to inertial impaction, have previously been found (Tsai et al., 2008) which may
12 also affect the signal.

13

14 **4.3 Response to different particle types**

15 For the assessment of instrument comparability, four particle types (TiO₂, NaCl, soot
16 and fumed silica), which exhibit different sizes and morphologies were chosen. The
17 differing particle sizes, morphologies and particle compositions were intended to
18 simulate the variety of particle types encountered in the studies in which these
19 samplers are often used, including outdoor air, indoor air, exhaust studies and
20 occupational exposure (Asbach et al., 2009). Though there were some consistent
21 trends between particle types, for example the ELPI generally measuring higher
22 particle numbers than the other instruments at its upper and lower working size limits,
23 the instruments often behaved differently in reaction to different particle types. For
24 example, the FMPS successfully identified a unimodal NSD for NaCl, however in
25 contrast to the other instruments provided a bimodal distribution for TiO₂ and fumed

1 silica. This has important implications for heterogeneous aerosol sampling; as there is
2 potential for particles from complex mixtures, for example urban air, to be skewed
3 according to preferential counting and/ or collection. While the instruments have been
4 previously known to produce artifacts within the data, for example the FMPS (e.g.
5 Asbach et al., 2009) and ELPI (e.g. Evans et al., 2003), additionally their reaction to
6 differing particle types has significant sampling effects. Importantly, the properties of
7 the particles (for example size, morphology or composition), responsible for the
8 changing efficiencies of the instruments cannot be elucidated from studying only four
9 particle types. There is therefore a requirement for further investigations with these
10 instruments with a larger number of different particle types.

11

12 **4.4 Effect of differing collection principles**

13 The SMPS and FMPS size particles according to their electrical mobility diameter;
14 particles are separated in an electrical field. In contrast, the ELPI and APS size
15 particles based upon their aerodynamic diameter, with the ELPI separating particles
16 based upon inertial impaction and the APS separation related to single particle time of
17 flight between two laser beams. The aerodynamic diameter is the diameter of an
18 equivalent spherical particle with a density of 1 g/cm^3 that standardises for
19 morphology and density (Rostedt et al., 2009). The electrical mobility diameter
20 standardises differently for morphology and not for density; particles are classified
21 based upon their efficiency at crossing an electric field. These differing sizing
22 principles may have significant implications for derived NSDs, for example with soot
23 agglomerates (Van Gulijk et al., 2004). The differing sizing metrics employed by the
24 instruments has previously been cited as a potential source of error when comparing

1 data collected simultaneously by these instruments (e.g. Shi et al., 1999a; Evans et al.,
2 2003).

3

4 **5. Conclusion**

5 This study has compared the results from four instruments sampling from the same
6 airstream for four particle types. This comparison has shown that different
7 instruments provide generally similar results in a controlled sampling setting.
8 Divergences were generally noted at the lower and upper working size ranges of the
9 instruments and at low number concentration. Where differences were noted, these
10 could be a response to the different operating procedures used by the instruments
11 and/or the different particle types that were sampled. Further work should focus on
12 the complete characterisation of homogeneous aerosols in order that a clear analysis
13 of the effects of particle characteristics (e.g. morphology and size) on the different
14 instruments can be assessed. There is a requirement for standard protocols in aerosol
15 measurement and primary standards for particle number and size.

16

17 **Acknowledgements**

18 The authors would like to thank Frank Schmidt and Achim Breidenbach for their
19 assistance during the measurements.

20

21

1 **References**

2 Asbach, C., Kaminski, H., Fissan, H., Monz, C., Dahmann, D., Mülhopt, S.,
3 Paur, H.R., Kiesling, H.J., Herrmann, F., Voetz, M., & Kuhlbusch, T.A.J. (2009).
4 Comparison of four mobility particle sizers with different time resolution for
5 stationary exposure measurements. *Journal of Nanoparticle Research*, 11, 1593-1609.

6 Brouwer, D., Van Duuren-Stuurman, B., Berges, M., Jankowska, E., Bard, D.,
7 & Mark, D. (2009). From workplace air measurement results toward estimates of
8 exposure? Development of a strategy to assess exposure to manufactured nano-
9 objects. *Journal of Nanoparticle Research*, 11(8), 1867-1881.

10 Crooks, M. (2011). A particle sizer for real-time measurement of rapidly
11 changing aerosols. Available at: [http://www.envirotech-online.com/articles/air-](http://www.envirotech-online.com/articles/air-monitoring/6/mark_crooks/a_particle_sizer_for_realttime_measurement_of_rapidly_changing_aerosols_-_mark_crooks/913/)
12 [monitoring/6/mark_crooks/a_particle_sizer_for_realttime_measurement_of_rapidly_c](http://www.envirotech-online.com/articles/air-monitoring/6/mark_crooks/a_particle_sizer_for_realttime_measurement_of_rapidly_changing_aerosols_-_mark_crooks/913/)
13 [hanging_aerosols_-_mark_crooks/913/](http://www.envirotech-online.com/articles/air-monitoring/6/mark_crooks/a_particle_sizer_for_realttime_measurement_of_rapidly_changing_aerosols_-_mark_crooks/913/). Accessed on 10/10/2013.

14 Dahmann, D., Riediger, G., Schletter, J., Wiedensohler, A., Carli, S., Graff,
15 A., Grosser, M., Hojgr, M., Horn, H.G., Matter, U., Monz, C., Mosimann, T., Stein,
16 H., Wehner, B., & Wieser, U. (2001). Intercomparison of mobility particle sizers
17 (MPS). *Gefahrst Reinhalt Luft*, 61, 423-427.

18 Dewalle, P., Ouf, F.-X., Pontreau, S., Gensdarmes, F., Vendel, J., Weulersse,
19 J.-M., Hervé, P., & Decobert, G. (2010). Determination of the effective density of
20 particles in a heterogeneous aerosol. *IAC 2010 conference abstracts*.
21 (<http://www.atm.helsinki.fi/IAC2010/abstracts/pdf/588.pdf>). Accessed 06/07/2013.

22 Evans, D.E., Harrison, R.M., & Ayres, J. (2003). The generation and
23 characterisation of elemental carbon aerosols for human challenge studies. *Journal of*
24 *Aerosol Science*, 34(8), 1023-1041.

1 Harrison, R.M., Shi, J.P., Xi, S., Khan, A., Mark, D., Kinnersley, R., & Yin, J.
2 (2000). Measurement of number, mass and size distribution of particles in the
3 atmosphere. *Philosophical Transactions A*, 358, 2567-2580.

4 Held, A., Zerrath, A., McKeon, U., Fehrenbach, T., Niessner, R., Plass-
5 Dülmer, C., Kaminski, U., Berresheim, H., & Pöschl, U. (2008). Aerosol size
6 distributions measured in urban, rural and high-alpine air with an electrical low
7 pressure impactor (ELPI). *Atmospheric Environment*, 42 (36), 8502-8512.

8 Hillamo, R., Mäkelä, T., & Kerminen, V.-M., Keskinen, J., & Marjamäki, M.
9 (2000). Use of Electrical Low Pressure Impactor (ELPI) in Atmospheric Aerosol
10 Studies. *Poster, EUROTRAC-Symposium 2000*, 81-84.

11 Imhof, D., Weingartner, E., Vogt, U., Dreiseidler, A., Rosenbohm, E., Scheer,
12 V., Vogt, R., Nielsen, O.J., Kurtenbach, R., Corsmeier, U., Kohler, M., &
13 Baltensperger, U. (2005). Vertical distribution of aerosol particles and NO_x close to a
14 motorway. *Atmospheric Environment*, 39 (31), 5710-5721.

15 Jeong, C.H., & Evans, G.J. (2009). Inter-comparison of a fast mobility particle
16 sizer and a scanning mobility particle sizer incorporating an ultrafine water-based
17 condensation particle counter. *Aerosol Science and Technology*, 43, 364-373.

18 Kerminen, V.M., Pakkanen, T.A., Mäkelä, T., Hillamo, R.E., Sillanpää, M.,
19 Rönkkö, T., Virtanen, A., Keskinen, J., Pirjola, L., Hussein, T., & Hämeri, K. (2007).
20 Development of particle number size distribution near a major road in Helsinki during
21 an episodic inversion situation. *Atmospheric Environment*, 41 (8), 1759-1767.

22 Keskinen, J., Pietarinen, K., & Lehtimäki, M. (1992). Electrical low pressure
23 impactor. *Journal of Aerosol Science*, 23 (4), 353-360.

24 Keskinen, H., Romakkaniemi, S., Jaatinen, A., Miettinen, P., Saukko, E.,
25 Jorma, J., Mäkelä, M., Virtanen, A., Smith, J.N., & Laaksonen, A. (2011). On-Line

1 Characterization of Morphology and Water Adsorption on Fumed Silica
2 Nanoparticles. *Aerosol Science and Technology*, 45 (12), 1441-1447.

3 Khlystov, A., Kos, G.P.A., ten Brink, H.M., Mirme, A., Tuch, Th., Roth, Ch.,
4 & Kreyling, W.G. (2001). Comparability of three spectrometers for monitoring urban
5 aerosol. *Atmospheric Environment*, 35 (11), 2045-2051.

6 Kumar, P., Fennell, P., & Robins, A. (2010). Comparison of the behaviour of
7 manufactured and other airborne nanoparticles and the consequences for prioritising
8 research and regulation activities. *Journal of Nanoparticle Research*, 12, 1523-1530.

9 Leskinen, J., Joutsensaari, J., Lyyräinen, J., Koivisto, J., Ruusunen, J., Järvelä,
10 M., Tuomi, T., Hämeri, K., Auvinen, A., Jokiniemi, J. (2012). Comparison of
11 nanoparticle measurement instruments for occupational health applications. *Journal of*
12 *Nanoparticle Research*. 14 (718).

13 Lide, D.R., ed. (2004). CRC Handbook of Chemistry and Physics, 86th edition;
14 CRC Press, Florida.

15 Long, C.M., Suh, H.H., & Koutrakis, P. (2000). Characterization of indoor
16 particle sources using continuous mass and size monitors. *Journal of Air & Waste*
17 *Management Association*, 50 (7), 1236-1250.

18 Maricq, M.M., Podsiadlik, D.H., & Chase, R.E. (1999). Examination of the
19 Size-Resolved and Transient Nature of Motor Vehicle Particle Emissions.
20 *Environmental Science and Technology*, 33 (10), 1618-1626.

21 Maricq, M.M., & Xu, N. (2004). The effective density and fractal dimension
22 of soot particles from premixed flames and motor vehicle exhaust. *Journal of Aerosol*
23 *Science*, 35, 1251-1274.

1 Marjamäki, M., Keskinen, J., Chen, D. -R., & Pui, D.Y.H. (2000).
2 Performance evaluation of the electrical low-pressure impactor (ELPI). *Journal of*
3 *Aerosol Science*, 31, 249-261.

4 Morawska, L., Wang, H., Ristovski, Z., Jayaratne, E.R., Johnson, G., Cheung,
5 H.C., Ling, X., & He, C. (2009). Environmental monitoring of airborne nanoparticles.
6 *Journal of Environmental Monitoring*, 11, 1758-1773.

7 Nussbaumer, T., Czasch, C., Klippel, N., Johansson, L., & Tullin, C. (2008).
8 Particulate Emissions from Biomass Combustion in IEA Countries- Survey on
9 Measurements and Emission Factors. *International Energy Agency (IEA) Bioenergy*
10 *Task*, 32, 4-40.

11 Pagels, J., Gudmundsson, A., Gustavsson, E., Asking, L., & Bohgard, M.
12 (2005). Evaluation of aerodynamic particle sizer and electrical low-pressure impactor
13 for unimodal and bimodal mass-weighted size distributions. *Aerosol Science and*
14 *Technology*, 39 (9), 871-887.

15 Pakkanen, T., Mäkelä, T., Hillamo, R., Virtanen, A., Rönkkö, T., Keskinen, J.,
16 Pirjola, L., Parviainen, H., Hussein, T., & Hämeri, K. (2006). Monitoring of black
17 carbon and size-segregated particle number concentrations at 9-m and 65-m distances
18 from a major road in Helsinki. *Boreal Environmental Research*, 11, 295-309.

19 Pitz, M., Gu, J., Soentgen, J., Peters, A., & Cyrys, J. (2011). Particle size
20 distribution factor as an indicator for the impact of the Eyjafjallajökull ash plume at
21 ground level in Augsburg, Germany. *Atmospheric Chemistry and Physics*, 11, 9367-
22 9374.

23 Rostedt, A., Marjamäki, M., & Keskinen, J. (2009). Modification of the ELPI
24 to measure mean particle effective density in real-time. *Journal of Aerosol Science*, 40
25 (9), 823-831.

1 Shi, J.P., Khan, A.A., & Harrison, R.M. (1999a). Measurements of ultrafine
2 particle concentration and size distribution in the urban atmosphere. *Science of the*
3 *Total Environment*, 235, 51-64.

4 Shi, J.P., Harrison, R.M., & Brear, F. (1999b). Particle size distribution from a
5 modern heavy duty diesel engine. *Science of the Total Environment*, 235, 305-317.

6 Shi, J.P., Harrison, R.M., & Evans, D. (2001). Comparison of Ambient
7 Particle Surface Area Measurement by Epiphaniometer and SMPS/APS. *Atmospheric*
8 *Environment*, 35, 6193-6200.

9 Stroszejn-Mrowca, G., & Szadkowska-Stańczyk, I. (2003). Exposure to dust
10 and its particle size distribution in shoe manufacture and repair workplaces measured
11 with GRIMM laser dust monitor. *International Journal of Occupational Medicine and*
12 *Environmental Health*, 16 (4), 321-328.

13 Tsai, C-J., Wu, C-H., Leu, M-L., Chen, S-C., Huang, C-Y., Tsai, P-J., & Ko,
14 F-H. (2008). Dustiness test of nanopowders using a standard rotating drum with a
15 modified sampling train. *Journal of Nanoparticle Research*, 11, 121-131.

16 Ushakov, S., Valland, H., Nielsen, J.B., Hennie, E. (2013). Particle size
17 distributions from heavy-duty diesel engine operated on low-sulfur marine fuel. *Fuel*
18 *Processing Technology*, 106, 350-385.

19 Van Gulijk, C., Schouten, J.M., Marijnissen, J.C.M., Makkee, M., & Moulijn,
20 J.A. (2001). Restriction for the ELPI in diesel particulate measurements. *Journal of*
21 *Aerosol Science*, 32, 1117-1130.

22 Van Gulijk, C., Marijnissen, J.C.M., Makkee, M., Moulijn, J.A., & Schmidt-
23 Ott, A. (2004). Measuring diesel soot with a scanning mobility particle sizer and an
24 electrical low-pressure impactor: performance assessment with a model for fractal-
25 like agglomerates. *Journal of Aerosol Science*, 35 (5), 633-655.

1 Virtanen, A., Joutsensaari, J., Koop, T., Kannosto, J., Yli-Pirilä, P., Leskinen,
2 J., Mäkelä, J.M., Holopainen, J.K., Pöschl, U., Kulmala, M., Worsnop, D.R.,
3 Laaksonen, A. (2010). An amorphous solid state of biogenic secondary organic
4 aerosol particles. *Nature*, 467, 824-827.

5 Wang, S.C., & Flagan, R.C. (1990). Scanning electrical mobility spectrometer.
6 *Aerosol Science and Technology*, 13, 230-240.

7 Wehner, B., Birmilli, W., Gnauk, T., & Wiedensohler, A. (2002). Particle
8 number size distributions in a street canyon and their transformation into the urban-air
9 background: measurements and a simple model study. *Atmospheric Environment*, 36
10 (13), 2215-2223.

11 Wright, M.D. (2014). Errors in particle size distributions from Sequential
12 Mobility Particle Sizers due to varying number concentration at an urban roadside
13 location. *Journal of Aerosol Science*, 67, 1-12.

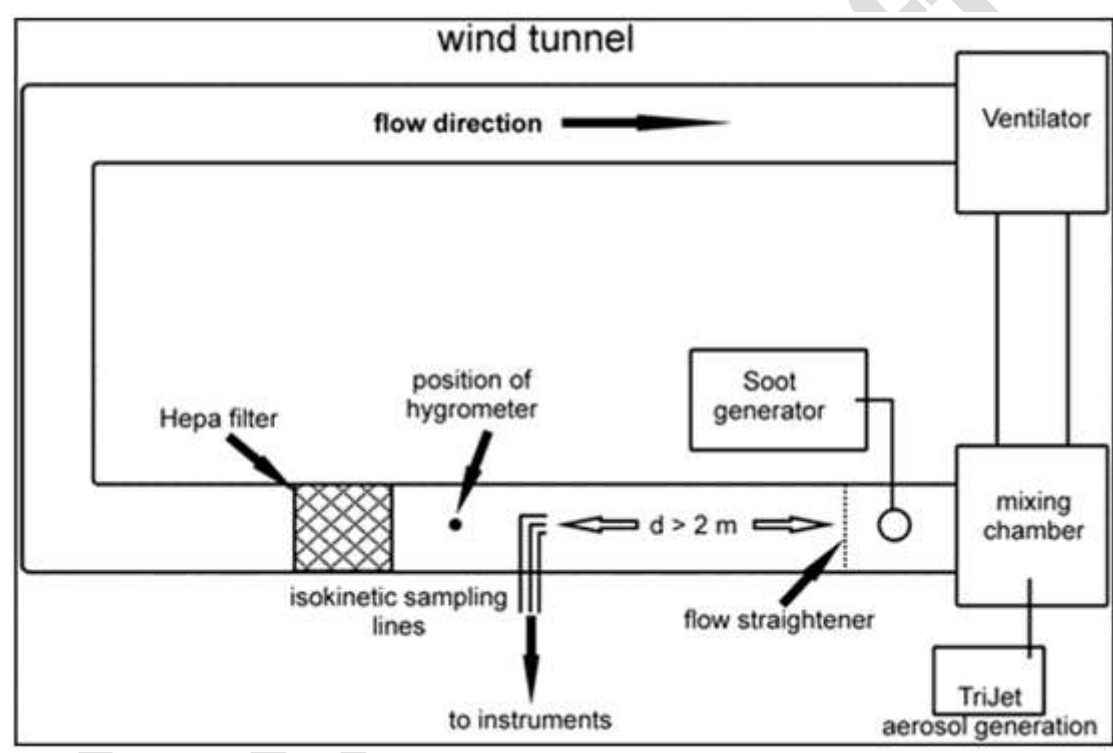
14 Zhang, R., Khalizov, A.F., Pagels, J., Zhang, D., Xue, H., & McMurry, P.H.
15 (2008). Variability in morphology, hygroscopicity, and optical properties of soot
16 aerosols during atmospheric processing. *Proceedings of the National Academy of*
17 *Sciences*, 105 (30), 10291-10296.

18

1

Figures

2 Figure 1: Schematic of experimental setup. After generation of the aerosol using the
3 trijet and introduction into the mixing chamber, the aerosol has to pass the flow
4 straightener. The distance of the flow straightener to the inlets of the different
5 measurement devices is above 2 m to provide a laminar flow profile. Isokinetic
6 sampling was ensured by different inlet diameters selected according to the wind
7 speed within the tunnel and flow rate of the different devices.



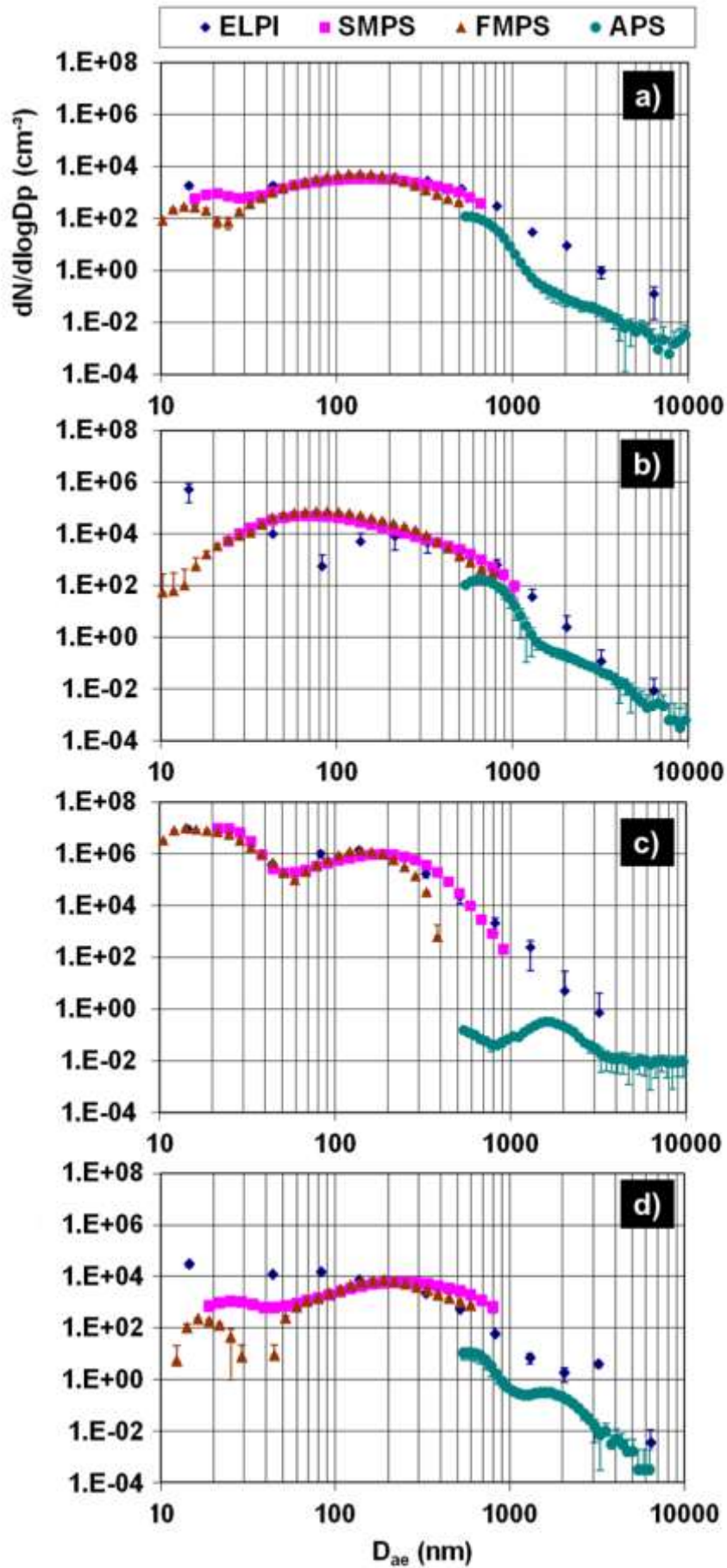
8

9

10

1 Figure 2: Comparison of the number size distributions (by aerodynamic diameter)
2 provided by the four instruments (ELPI, SMPS, FMPS and APS) for the four tested
3 substances. Error bars are shown for each data point (though these are sometimes
4 smaller than the data point itself) and = 1 standard deviation \pm mean of sampling
5 period. a) TiO₂ particles. An effective density of 0.900 g/cm³ was assumed for the
6 particles. b) NaCl particles. An effective density of 2.164 g/cm³ was assumed for the
7 particles. c) Soot particles. An effective density of 1.700 g/cm³ was assumed for the
8 particles. d) Fumed silica particles. An effective density of 1.300 g/cm³ was assumed
9 for the particles.

Final draft



1 **Tables**

2

3 **Table 1: Advantages and disadvantages of the sampling instruments used in this**

4 **study.**

Instrument (model used in this study)	Operating particle size range	Example type of study used in	Advantages	Disadvantages
ELPI	7 nm–10 µm	Atmospheric, occupational	Large size range, excellent time resolution	Classic drawbacks of cascade impactors e.g. particle bounce. Low size resolution
SMPS	14–730 nm	Atmospheric, occupational	High size resolution, ability to sample very small particles; applications in atmospheric studies	Slow time resolution
FMPS	5–550 nm	Occupational, fuel cycle, wood burning, emissions	High size resolution, fast scan time, ability to sample very small particles; applications in atmospheric studies	Less sensitive than SMPS
APS	0.5–20 µm	Atmospheric, occupational	Extends particle size range when used in combination with SMPS, high size resolution	Limited size range; unsuitable for atmospheric studies with the importance of nano-sized particles

5

6

1 **Table 2: Overview of the different instruments used for particle measurement.**

Instrument	Type/model	Inlet diameter (mm)	Flow rate (l/min)
FMPS	TSI, model 3091	15.5	9.8
SMPS	TSI, model 3080 with long DMA, model 3081	9.3	0.3
APS	TSI, model 3321	10.7	4.2
ELPI	DEKATI, outdoor air ELPI with filter stage	28.1	29.4

2

Final draft