



Bipolar charge distribution of a soft X-ray diffusion charger



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ABSTRACT

The conditioning of the aerosol particle population into a bipolar charge equilibrium is an essential prerequisite to calculate the particle number size distribution using mobility particle size spectrometers. This is commonly realized by diffusion charging of bipolar air ions generated by e.g. a ⁸⁵Kr source. Because of strict legal regulations on radioactive sources in several countries, soft-X-ray (SXR) appears as a suitable alternative. However, multiple measurements showed a systematic and significant difference between the particle charge distribution delivered by a radioactive source and an SXR charger, respectively. In this investigation, a calibrated particle charge distribution, suitable for the SXR chargers, was calculated based on the Fuchs model. An approximation analogous to the commonly used Wiedensohler approximation formula (Wiedensohler, 1988) was computed. The use of the new SXR approximation of the bipolar charge equilibrium for the inversion of an electrical mobility distribution to a particle number size distribution improves the comparability of these results, compared to measurements involving a ⁸⁵Kr charger or to bipolar chargers using radioactive material in general. A systematic error in case of using the SXR charger could be eliminated and hence the root mean square deviation could be reduced from 13% using the common parameters for both charger types to 7% using the new SXR approximation for the SXR bipolar charger.

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1. Introduction

Mobility particle size spectrometers are commonly used to determine the number size distribution of aerosol particles in the submicrometer size range from a few nanometers up to one micrometer. Besides a differential mobility analyzer (DMA) and a condensation particle counter (CPC), an essential part of mobility particle size spectrometers is the bipolar diffusion charger. The main advantage to use a bipolar diffusion charger is to bring the whole aerosol particle population into a known bipolar charge equilibrium. To be able to reach this goal, a high concentration of positive and negative air ions is needed, which has to be in excess to the particle number concentration. In a bipolar diffusion charger, high concentrations of air ions are traditionally produced by a radioactive material, which is used to ionize the aerosol carrier gas. Common radioactive nuclides used in bipolar diffusion chargers are ⁸⁵Kr, ²⁴¹Am, ²¹⁰Po and ⁶³Ni. It is believed that the use of these nuclides produces ions with similar properties leading to a kind of ‘universal bipolar charge equilibrium’ unless the aerosol

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is highly pre-charged. This bipolar charge equilibrium can be predicted using Fuchs' (1963) model. Adjustment of the values of the mean ion mass and electrical mobility of positive and negative air ions allows for the simulation of experimentally determined charging probabilities of singly and multiply, negatively and positively charged particles using Fuchs' model. Since the Fuchs model does not provide an analytical solution, Wiedensohler (1988) provided a polynomial, i.e. analytical equation which approximates the numerically obtained Fuchs model very well. The common bipolar charge equilibrium shows a distinct asymmetry between negatively and positively charged particles towards higher fractions of negatively charged particles. In practice, particle number size distributions are calculated from electrical mobility distributions measured by a mobility particle size spectrometer employing the size-dependent bipolar charge distribution in an inversion routine. Comparisons between a directly measured total particle number concentrations, i.e. concentration measurement without any classification, and the integral of the particle number size distribution show in most cases an agreement within around 10% under laboratory conditions for ambient air as shown in Wiedensohler et al. (2012). Presently, the bipolar charge distribution by Wiedensohler (1988) is commonly used in all commercial instruments and described in ISO 15900.

The residual deviation can be attributed to the variation of relevant input parameters for the Fuchs model namely positive and negative ion properties in ambient air. In Tigges et al. (2015) the sensitivity of the charge distribution to these input parameters has been discussed extensively.

The usage of a radioactive source to charge aerosol particles is however often limited by national safety standards. Hence, in recent years there has been an increasing interest for alternative solutions. Recently, even unipolar chargers have been suggested to condition the aerosol upstream of the differential analysis (e.g. Qi et al., 2009). With a unipolar charger, e.g. a corona charger, the number of charges on the particles is much higher and consequently the 'multiple charge correction' is much more relevant in this case. Therefore, larger size intervals are required and this unfortunately leads to a more smoothed particle size distribution curve, i.e. reduced resolution in particle size. Also Kaminski et al. (2013) discussed that the Cunningham (1910) slip correction factor becomes dominant in sizing of unipolarly charged particles. However, for larger particle sizes, this correction factor becomes less depending on particle diameter. Consequently, the sensitivity of electrical mobility sizing of unipolar diffusion charged particles is continuously decreasing for increasing particle sizes.

There are a number of instruments, which use the bipolar chargers, on the market: TSI FMPS (model 3091), TSI UFP monitor (model 3031), TSI nanoScan, Grimm miniWRAS, Kanomax PAMS and ELPI. However, these instruments do not have the same requirements in high resolution compared to the common SMPS.

Also a couple of alternative bipolar chargers have been designed and characterized. Stommel & Riebel (2004, 2005) and Alguacil & Alonso (2006) analyzed corona chargers. Kwon et al. (2006) presented an aerosol charger based on the dielectric barrier discharge (DBD). However, these bipolar charger types may produce ozone in operation.

Furthermore, a number of publications about SXR chargers have been published by Shimada et al. (2002), Han et al. (2003), Lee et al. (2005), Yun et al. (2009), Modesto-Lopez et al. (2011), Gälli et al. (2010) and Kallinger et al. (2012). Yun et al. (2009) pointed out that this charger type produces particles in case of poor construction. However, the commercial devices AAN-TSI (Advanced aerosol neutralizer, 9.5 keV, Mod. 3087, TSI inc., USA) and SXR-HCT (soft X-ray bipolar charger, 9.5 keV, Mod. 4530, HCT, Korea) did not show any particle production. Hence, these devices represent a suitable alternative to radioactive sources.

But even under standardized conditions (temperature, humidity and artificial gas composition), the charge distribution is depending on the charger type. A couple of comparison investigations between SXR and radioactive sources showed a systematic and significant divergence. SXR showed a more balanced charge distribution between negatively and positively charged particles compared to chargers using a radioactive material. This qualitative trend could be confirmed by multiple authors (Han et al., 2003; Kallinger et al., 2012; Lee et al., 2005; Yun et al., 2009), and own measurements (see Fig. 3). Hence, this systematic difference is significant enough to be confirmed under various environmental influences. Nevertheless, no quantitative correction to this systematic deviation is available until now. Therefore, up to now commercial instruments provide a data inversion which is based on one set of input parameters (chosen by Wiedensohler, 1988) irrespective of the used charger type. Hence, mobility particle size spectrometer results, measured with different chargers, become less comparable.

Consequently, the aim of this work is to provide adapted values of the ion properties to determine a reasonable charge distribution that provides quantitatively comparable results without systematic deviation between SXR and radioactive sources. Furthermore, a new polynomial fit of this charge distribution is given as well in order to facilitate the application of the newly proposed charge distribution.

The physical background for different charge distributions in dependency of the charger type is still highly controversial. Lee et al. (2005) argued that a variation of just the radioactive intensity from an α -ray charger results in a significant difference. According to Lee, the ionization energy is the relevant factor. Flagan (2005) associated the differences to the ionization rate (the nt-product). As discussed in Tigges et al. (2015), ion aging (that is a time dependent formation of ion clusters) takes place in the charger. Hence, the live-time of the ions is very important for their properties. This effect is depending on the following parameters:

- flow rate;
- charger geometry:
 - volume,

- turbulence effects;
- gas components (due to their affinity to form ion clusters);
- ion concentration (due to recombination probability).

Anyway, based on experience out of numerous workshops within the World Calibration Center for Aerosol Physics (Leipzig, Germany) there is some indication that ^{85}Kr , ^{241}Am , and ^{63}Ni chargers give comparable particle size distribution under identical conditions and consequently the chargers are supposed to give comparable bipolar charge distributions for ambient aerosols. However, this is only valid provided the radioactive activity is high enough.

2. Aim and approach

The aim of this investigation was to improve the comparability of particle number size distribution measurements using radioactive chargers and SXR bipolar diffusion chargers.

To calculate the bipolar charge distribution using the Fuchs (1963) model most realistically, in principle a two-dimensional ion mobility–mass distribution would be required. However, those data are typically not available, since they are barely measurable with a reasonable effort. Hence, the simplification of using fixed mean ion properties is almost inevitable for a practical application as widely used and also accepted.

The intention of this study was to obtain a new set of input parameters (mean ion mobility and mass) required to invert electrical mobility distributions measured by a mobility particle size spectrometer using an SXR charger. Analogous to the Wiedensohler approximation, a polynomial fit is generated subsequently.

This method comprised the following conditions:

- the bipolar particle charge distribution were calculated by the Fuchs (1963) theory using the correction of the α -parameters by Hoppel & Frick (1986),
- approximation of the charge fractions for singly and doubly charged particles by a 5th order polynomial fit function:

$$f(N) = 10 \sum_{i=0}^5 a_i(N) \cdot (\log D_p/nm)^i \quad (1)$$

- the charge fraction in excess of two charges per particles were calculated based on Gunn & Woessner (1956).

With the newly determined approximation coefficients, an implementation of the modified bipolar charge distribution in existing inversion routines is easily possible.

It should be noted that the differences between the two chargers may be not constant for any measurement conditions, because the charger type is just one of multiple and partly interdependent influences (Tigges et al., 2015), hence a comparison is inherently case specific. Especially, a comparison may differ for technical or laboratory-generated aerosols, which are often highly pre-charged or may consist of carrier gas components, which can significantly differ from ambient air. For a highly pre-charged aerosol it is likely that the particle population does not reach the bipolar charge equilibrium in the usual residence time in the charger (see also supporting informations). Hence, this kind of aerosols is not so well suited for our purposes.

Therefore outdoor ambient air was chosen as the test aerosol for the further investigation (see Fig. 1), because it is pre-charged naturally in a bipolar atmosphere due to ambient air ions. Consequently, ambient aerosols can be considered to be typically close to charge equilibrium. Furthermore, ambient aerosols are commonly characterized by mobility particle size spectrometer measurements. Hence, this calibration is particularly valid for measurements of particle number size

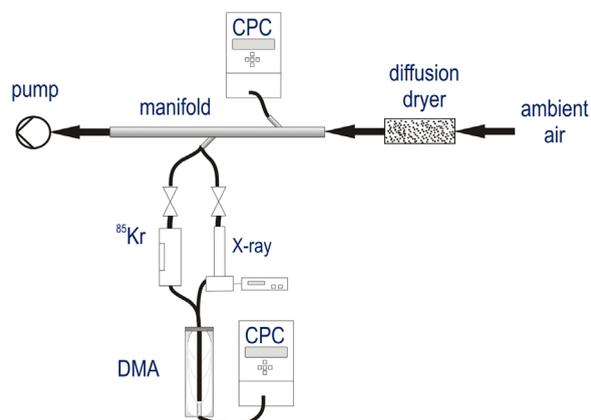


Fig. 1. Experimental setup.

distributions of atmospheric aerosols in the considered size spectrum (compare Fig. 1). It should be noted, that Fuchs (1963) theory is strictly only valid for spherical particles. Nevertheless, application to real aerosols is quite common. However, this could be one reason for the residual impreciseness (see Section 4).

3. Procedure

In our experimental set-up, a mobility particle size spectrometer and two bipolar diffusion charger were employed. The two chargers are placed in parallel lines and operated alternately. This setting was preferred instead of two separated measurement devices to avoid errors due to differences between the measurements devices (see Kaminski et al., 2013). Hence, for one series of measurements just one system was used. To verify the new set of parameters, two different systems were used consecutively. First a TSI system with following components and adjustments was used: electrostatic classier 3080, long DMA 3081, CPC 3775 with a flowrate of 0.3 l/min, scan time of 240 s, impactor size 0.0508 cm; second a Tropos system including a TSI CPC 3772 with a flowrate of 1 l/min, scan time of 285 s (see Table 1). The aerosol was dried with a diffusion dryer to a relative humidity lower than 40%.

The particle electrical mobility distribution was measured in sequences of five minutes. The valves in front of the chargers were switched on/off all 10 min, i.e. after every second scan. The DMA was thus fed either by the aerosol flow from the SXR charger (Advanced aerosol neutralizer, Mod. 3087, TSI inc., USA) or from the ^{85}Kr (TROPOS, 370 MBq) charger (see Fig. 1). Data were collected over several hours for one experimental run (see Table 1). Hence, it was measured at different time (day and night, workday and weekend) and changing meteorological conditions. All electrical mobility distributions were then grouped by charger type and mean particle mobility distributions for both chargers were calculated. Subsequently the respective mean particle number size distributions were calculated. Since one complete experiment took several hours, it might represent an average over various ambient air conditions occurring during the day.

For the calibration to determine a new set of mean ion property values, four experimental series (ES) were performed. For a validation of the new parameter set 10 additional experimental series were done.

Figure 2 shows the mean particle size distributions for each experimental series measured by the ^{85}Kr charger. Those data have been inverted by a data inversion technique using a charge distribution according to Wiedensohler (1988). It can be clearly recognized that despite the considerable averaging time really different particle size distributions have been assessed during these experiments. This is favorable in terms of adapted ion properties being representative in general.

On the other hand, mobility data measured using the SXR charger were inverted using diverse charge distributions. In a first step, Wiedensohler's (1988) approximation of the bipolar charge distribution was used to highlight the systematical divergence. Figure 3 shows the particle size-dependent ratio between the particle number concentration measured using the ^{85}Kr charger and the SXR charger.

For a negative DMA polarity, hence positively charged particles, the concentration measured by the SXR charger is always higher than that for the ^{85}Kr charger. For the positive DMA polarity, the opposite trend is observed, i.e. we get less negatively charged particles and more positively charged particles compared to radioactive neutralizers. However, the resulting charge distribution is still skewed to negatively charged particles as will be shown in Figs. 6 and 7.

The second step was the calibration (see Fig. 4). For this purpose, mobility data measured with the SXR charger were inverted based on a large number (5050) of potential bipolar charge distributions, which were calculated using the Fuchs model. To achieve the input parameters suitable for the SXR charger, the ion mobilities have been varied in a reasonable range of 1–2 $\text{cm}^2/\text{V s}$ in steps of 0.01 $\text{cm}^2/\text{V s}$. Here, we assumed the mean electrical mobility of positive ions to be always lower than the mean electrical mobility of negative ions, as confirmed by measured values (see Table 1 in Tigges et al. (2015)). The related ion masses were calculated by the Kilpatrick (1971) relation, because this is believed to be an adequate mobility–mass relation for ambient air (Tigges et al., 2015). For each parameter set, the particle electrical mobility data produced by the SXR charger were averaged over time and inverted in order to calculate the averaged particle number size distribution for each experimental series. The resulting size-resolved particle number concentration were divided by the respective concentrations measured by the ^{85}Kr charger, which were inverted by the common input parameters.

For the calibration, the size range from 20 to 400 nm was considered only, since outside this range the measured particle number concentrations are relatively low (see Fig. 2). Due to poor statistics, significant fluctuations in the concentration ratios were observed there (see Fig. 3). Hence, the accuracy of the calibration outside this range could not be verified.

Table 1
Experimental runs.

Process	Number ES	Type of SMPS	DMA polarity	Time duration	Number of scans
Calibration	1	TROPOS	Neg.	7 h 20 min	88
	2	TROPOS	Pos.	65 h 30 min	786
	3	TSI model 3080	Pos.	21 h 50 min	262
	4	TSI model 3080	Neg.	47 h	564
Validation	V 1–10	TROPOS or TSI model 3080	Pos. or Neg.	∅ 26 h	∅ 312

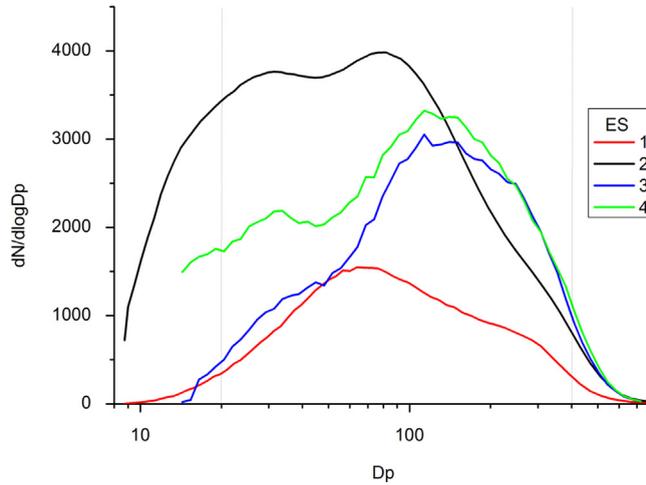


Fig. 2. Measured mean particle size distributions during 4 experimental series used for calibration of ion properties.

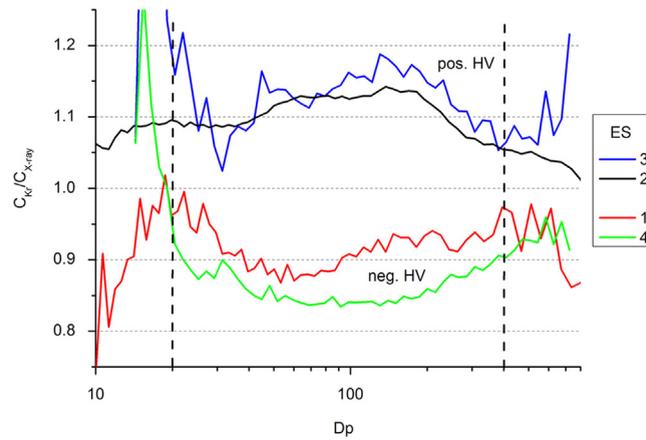


Fig. 3. Particle size dependent concentration ratios obtained from SXR and Kr chargers using standard charge distribution for inversion.

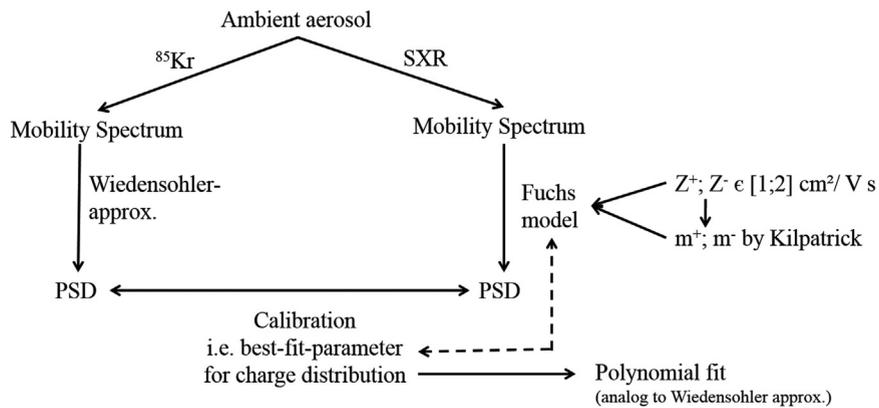


Fig. 4. Overview approach.

For each parameter set the root mean square deviation (RMSD) of the size-dependent distance of the concentration ratio to 1 was calculated for each experimental series:

$$RMSD_{ES,PS} = \sqrt{\frac{\sum_{i=1}^{i_{max}} (1 - C_{i,Kr}/C_{i,X-ray})^2}{i_{max}}} \quad (2)$$

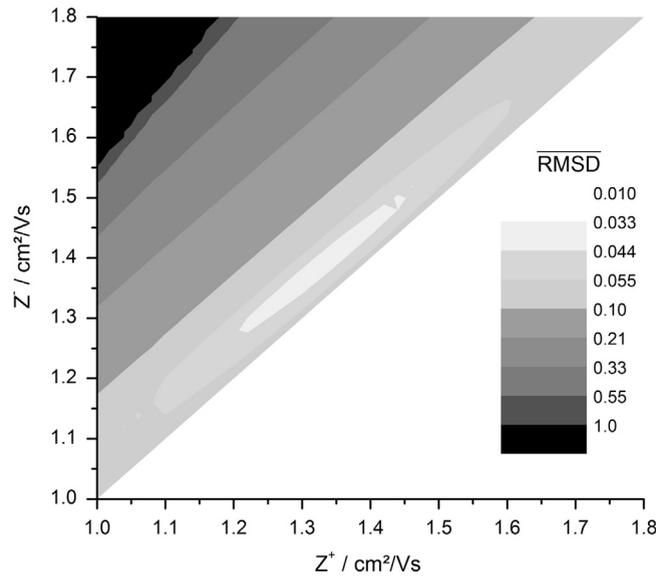


Fig. 5. Root mean square deviation of four experimental series with varying ion mobilities in case of using SXR charger.

Table 2
Best fit parameters for ion properties using SXR charger.

Polarity	Ion mobility ($\text{cm}^2/\text{V s}$)	Ion mass (amu)
Neg.	1.4	177
Pos.	1.34	204

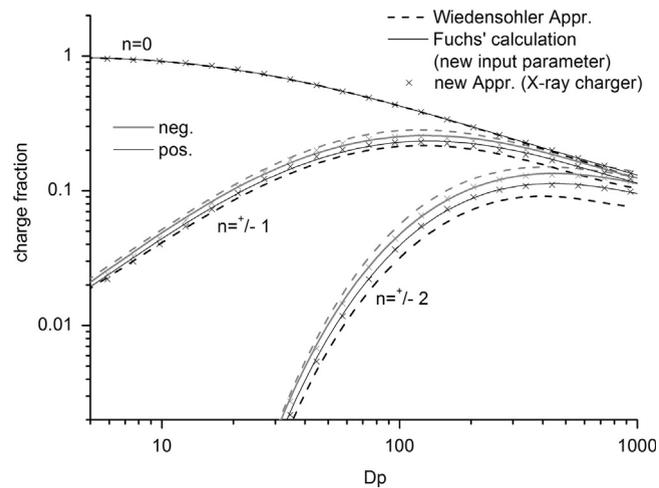


Fig. 6. Comparison of charge fractions obtained using Fuchs model and approximation.

Subsequently, the mean value of RMSD for all four experimental series (\overline{RMSD}) is calculated and plotted with respect to the ion mobilities (Z^+ , Z^-) in the considered range. The minimum of \overline{RMSD} finally leads to the best fit parameter set of ion properties.

4. Results

4.1. Calibration

A corresponding plot of the \overline{RMSD} value in the considered range of ion properties is given in Fig. 5. The minimum of \overline{RMSD} for all four experimental series is 0.04, i.e. 4%. In comparison, a use of the common standard parameters for both chargers leads to a \overline{RMSD} of 0.112 (i.e. 11.2%) for the four experimental series. Figure 5 shows that for a constant difference in

Table 3
Approximation coefficients for polynomial fit of charge fractions.

$a_i(N)$	N				
	-2	-1	0	+1	+2
a_0	-30.61558	-2.33509	0.00163	-2.35889	-27.25320
a_1	46.33885	0.43635	-0.11384	0.45169	38.47963
a_2	-31.18191	1.08654	0.33393	0.99798	-24.27128
a_3	11.39070	-0.55679	-0.35714	-0.48173	8.44162
a_4	-2.22028	0.04981	0.10770	0.02631	-1.60589
a_5	0.17935	0.00551	-0.01082	0.00804	0.12917

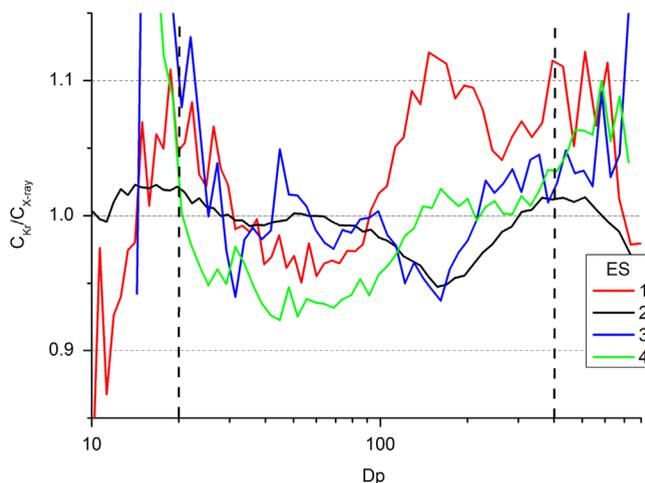


Fig. 7. Ratio of particle concentrations obtained using the adapted values for SXR measurements.

ion mobilities, the \overline{RMSD} values are almost constant in a large range of values as indicated by the nearly diagonal bands. Furthermore one can recognize that the \overline{RMSD} value is strongly increasing for large differences of positive and negative ion mobilities. Since the minimum \overline{RMSD} value appears to be well within the range of assessed ion mobilities, it becomes evident that this range has been chosen adequately.

The corresponding best fit parameters are listed in Table 2.

The ratio of ion mobility Z_{I+}/Z_{I-} required for the method based on Gunn & Woessner (1956) is consequently 0.957. The values for the negative and positive ion mobilities are closer together compared to the values chosen by Wiedensohler (1988). As expected, the bipolar charge distribution is more symmetric for the SXR charger, i.e. the differences in the fraction of positively and negatively charged particles become smaller (see Fig. 6).

These parameters will lead to the following approximation coefficients for Eq. (1).

The resulting size-dependent fraction of neutral, singly and doubly charged particles is then approximated by a 5th order polynomial according to Eq. (1). The coefficients obtained this way are given in Table 3. As shown in Fig. 6, the approximation gives an almost perfect fit to the Fuchs calculation.

In Fig. 7, the size-dependent concentration ratio is plotted again for all experimental series using the new approximation coefficients and the new ion mobility ratio for the inversion. The figure shows clearly that there is no systematic deviation any more. Although there are still fluctuations, they are however below 10% in most cases. Hence, the remaining fluctuations are within the tolerance commonly accepted for ambient aerosol measurements (Wiedensohler et al. 2012) since they appear to be unavoidable.

Figure 8 shows the time-averaged particle number size distributions for all four experimental series. In all four experimental series the solid line (SXR inverted with the newly determined charge distribution) shows good agreement with the dashed line (^{85}Kr inverted with the common charge distribution) in contrast to the dotted line (SXR inverted with the common bipolar charge distribution). Hence, the calibration shows a significant improvement of the comparability of measurements with the two chargers.

4.2. Validation

For the validation, 10 more experimental series in a later time period were realized (see Table 4). Data measured by the ^{85}Kr were inverted as usual. The data measured by the SXR charger were inverted equally and based on the developed

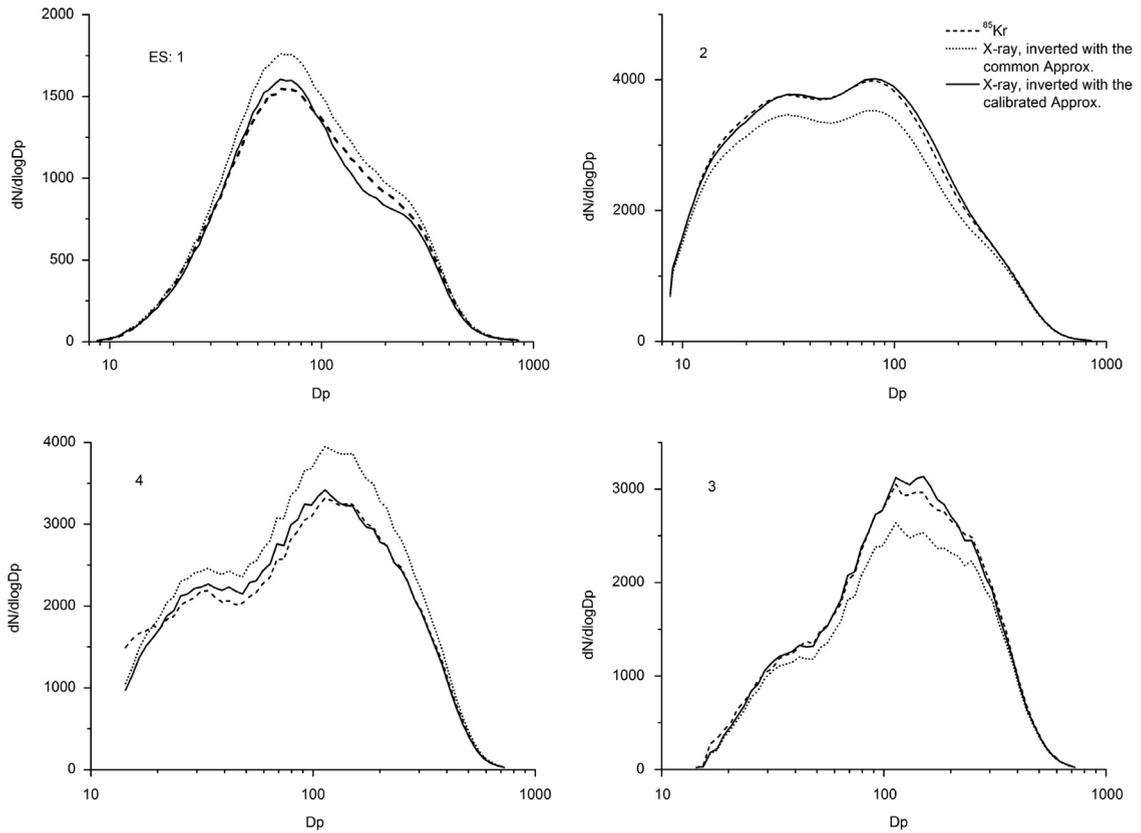


Fig. 8. Particle size distributions for all 4 experimental series obtained using standard and adapted ion properties.

Table 4

Results of 10 experimental series for validation of new SXR charge distribution.

Nr.	Type of SMPS	DMA polarity	RMSD before cal. (%)	RMSD after cal. (%)	Over- or under-cor.	High fluctuation edge zone
V1	TSI model 3080	Pos.	20.4	8.7	Over	Yes
V2	TSI model 3080	Neg.	15.6	5.5	Over	No
V3	TROPOS	Pos.	2.1	9.1	Under	No
V4	TSI model 3080	Pos.	11.6	9.4	Under	Yes
V5	TSI model 3080	Neg.	12.4	8.5	–	Yes
V6	TSI model 3080	Neg.	8.9	6.2	–	Yes
V7	TSI model 3080	Pos.	11.7	6.6	Over	No
V8	TROPOS	Pos.	17.3	9.1	Under	Yes
V9	TROPOS	Pos.	9.7	4.3	–	No
V10	TROPOS	Pos.	18.6	2.6	–	No
			∅ 12.8	∅ 7.0	Balanced	

approximation as outlined in the previous section. The aim was to investigate the comparability between measurements with the two different chargers.

In case of using standard parameters, all 10 experimental series show the systematic divergence between the chargers in dependency of the polarity as already shown in Fig. 3.

An overview of the evaluation results using the newly adapted parameter set is listed in Table 4. In 9 out of 10 experimental series the new fit provides a substantially better result with a significantly reduced RMSD (see column ‘RMSD’) value. On average the RMSD were reduced from 12.8% to 7.0% by using the new approximation. Even though the systematic and significant difference between those chargers can be eliminated by the new SXR bipolar charge distribution, the RMSD values after the calibration indicate that there are still some deviations and that these values are partly still considerable (9.4% in maximum). Nevertheless, this has to be attributed to commonly accepted tolerances and hence to other influencing factors as outlined in Tigges et al. (2015).

If the particle size distribution of the ^{85}Kr is not overlapping exactly with the calibrated particle size distribution of the SXR charger, an over- or under-correction is possible. This is described in the sixth column. In total the validation showed as many over- as under-corrections, hence there is no systematic deviation any more.

In V1, V4, V5, V6 and V8 an additional effect leads to a high RMSD. As shown in Fig. 3, a low particle concentration leads to a high fluctuation in the edge zone. Therefore the further data evaluation was limited to the particle size range from 20 to 400 nm. This was a suitable range for these four experimental series used for the calibration. However, the listed experimental series showed high fluctuations in the edge zone even within this range. In consequence, this leads to a higher RMSD.

A further validation was performed with a X-ray charger produced by HCT (type 4530, anode voltage 9.5 kV DC). These measurements show very good agreement with the new set of parameters (see supporting informations). Hence, we consider the approximation coefficients to be valid for X-ray chargers of comparable types.

5. Conclusion

Multiple measurements showed a systematic and significant difference between the bipolar charge distributions delivered by a ^{85}Kr and an SXR charger, respectively. An optimized fit for calibration experiments leads to a new, modified bipolar charge distribution for SXR chargers with an anode voltage of 9.5 kVDC. This newly developed, calibrated bipolar charge distribution can be used to increase the comparability to measurements with a ^{85}Kr charger or to bipolar chargers using radioactive material in general. It should be noted, however, that the values present just a relative adaptation to a radioactive source: As discussed in Tigges et al. (2015), changes in the gas composition generally affect the ion properties. This is commonly not considered for the inversion of a particle electrical mobility distribution. Up to now, the inversion software considers just one set of input parameters independent of the measurement conditions. This uncertainty factor is commonly accepted. It is not clear whether the different chargers respond similar to changes in the gas compositions. If not, the comparability would be compromised even with the newly calibrated charge distribution. Hence, the validity is so far only ensured for atmospheric aerosols measurements in which the aerosol is dried below 40% RH.

In conclusion, the presented SXR bipolar charge distribution appears to be a good mean, valid for ambient air to improve comparability between radioactive sources and SXR bipolar diffusion chargers.

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Appendix A. Supplementary data

Supplementary data associated with this paper can be found in the online version at <http://dx.doi.org/10.1016/j.jaerosci.2015.07.002>.

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