Particle Charging and Transmission Efficiencies of Aerosol Charge Neutralizers

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ABSTRACT. Diffusion losses and charging efficiency were measured for three types of charge neutralizers commonly used in aerosol research: two with ^{85}Kr and one with ^{210}Po as radiation sources. The diffusion losses were characterized at flows of 0.5–6 l min^{-1} typically used in atmospheric aerosol physics measurements. All of the neutralizers tested exhibited high transmission efficiencies, with losses up to 25% at the smallest tested size of 3 nm, varying with size and flow in general agreement with diffusion loss theory. Charging efficiency was measured for a singly charged, monodisperse aerosol at the same flows and at concentrations of 10^3–10^4 particles cm^{-3}. Neither of the ^{85}Kr chargers brought the charge distribution close to equilibrium at 2 l min^{-1}, except at concentrations $\leq 10^3$ cm^{-3}. The ^{210}Po charger produced the theoretically expected fraction of singly charged particles within the uncertainty of the experiment. AEROSOL SCIENCE AND TECHNOLOGY 27:206–214 (1997) © 1997 American Association for Aerosol Research

INTRODUCTION

Much current work in aerosol physics, particularly the size dependent measurement and characterization of particles, relies on the use of an aerosol of known charge distribution. Several designs of bipolar diffusion chargers or neutralizers have been used to apply an equilibrium charge distribution to an aerosol of initially unknown charge distribution. In these units, radioactive sources of alpha or beta particles produce high concentrations of air ions which allow the particles to approach equilibrium charge. However, not much is known about how well these neutralizers perform compared to the theoretically predicted equilibrium charge distribution described by Fuchs (1963) and further elaborated and parameterized by Adachi et al. (1985) and Wiedensohler (1988). Modeling of charging efficiency depends greatly on effective radiation source strength, geometry, and ion production rate of the individual unit which are not well characterized. Therefore, direct measurement provides an expedient means of establishing the charging characteristics of such neutralizers. While alpha sources have high specific ion pair production rates and are likely to achieve more complete equilibrium charging, they have a disadvantage of a potential for particle production and, in the case of some isotopes, for radioactive contamination.

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Since charge neutralizers are frequently used in-line in particle generation and measurement systems, it is important to characterize particle losses which occur during transmission. In order to provide high air ion concentrations and adequate residence times, the inner diameters of neutralizers range from 2 to 5 cm. This large inner diameter (compared to the inlet and outlet tubing) and irregular geometry around the radiation sources results in an ill-defined flow profile and can result in the development of eddies, dead volume, and enhanced diffusion losses. These losses are functions of the geometry and the Reynolds number and require detailed fluid mechanical modeling to estimate the particle losses to the walls of the neutralizer housing. Again, direct measurement is needed to determine the transmission characteristics of charge neutralizers.

We have made measurements to determine two central aspects of neutralizer performance: the extent to which they apply an equilibrium charge distribution and their particle transport efficiency. These measurements were intended to characterize the neutralizers in advance of the International Global Atmospheric Chemistry Project, Southern Hemisphere Marine Aerosol Characterization Experiment (ACE1) in which aerosol physics measurements were made at many sites for intercomparison. The flows, particle concentrations, and particle sizes chosen were not exhaustive but rather focus on conditions and limits anticipated for this field experiment. The tests were done at a pressure of ca. 1000 hPa and a temperature of 298 K.

NEUTRALIZER DESIGNS

We employed three neutralizer designs with radioactive sources in this study. The first is commercially available (model 3077, TSI Inc., St. Paul, MN; designated TSI) and uses 2 mCi of ^{85}Kr as a source of beta particles. Due to the 10.8 year half-life of this isotope, these neutralizers are effective and may be used for a period of 10 years after manufacture. It has an inner diameter of 2 cm, an internal volume of 100 cm^3, and 0.4 cm inner diameter inlet tubing connecting directly to the inner volume. It is designed for flows up to 5 l min^{-1}. A modified version of this neutralizer has been produced at the Institute for Tropospheric Research, Leipzig, Germany (designated IfT) with an inner diameter of 4 cm, a volume of 60 cm^3 and a conical inlet with a 30° taper. The ^{85}Kr sources were less than two years old at the time of testing.

Russell et al. (1996), in collaboration with Aerosol Dynamics (Berkeley, CA), presented an alternative design employing ^{210}Po as a source of alpha particles. This design (designated CIT) provides a larger internal volume and, thus, a long residence time in the unit, ca. 1 s at a flow of 6 l min^{-1}. The body of the neutralizer is 4.7 cm inner diameter and 18.0 cm in length and houses four removable 0.5 mCi ^{210}Po sources (model 2U500, NRD Inc., Grand Island, NY). Since ^{210}Po has a half-life of only 138 days, the instrument specifications are for a source 120 days old, so that four 0.5 mCi strips are estimated to provide a minimum source strength of 1.0 mCi. This half-life requires replacement of the radioactive sources about every four months. At the time of the tests described here, the sources were between 60 and 90 days old. The inlet of this unit has an entrance cone with a 7° divergent angle. This angle minimizes development of turbulence and a more nearly laminar flow is maintained, thus minimizing particle losses and providing a more uniform exposure time for particles in the active region of the neutralizer than with an inlet jet that may be generated with the TSI and IfT neutralizers.

The neutralizing efficiency depends on the residence time of particles in the neutralizer. The flow profile within the neutralizer is strongly affected by the geometry of the entrance region of the housing. Eddies of jets can cause a distribution of residence times in the neutralizer, such that some particles exit the neutralizer before sufficient time for recharging. Although a detailed investigation of the flow profiles of the existing designs is beyond the scope of

this study, it is instructive to note that the neutralizer with the highest charging efficiency was specifically designed to maintain laminar flow.

DIFFUSION LOSSES

This experiment employed a monodisperse aerosol source, two ultrafine condensation particle counters (UCPC, model 3025, TSI Inc., St. Paul, MN), and a test section for the charge neutralizers as shown schematically in Fig. 1a. All connecting tubing was stainless steel (4 mm i.d.). The source of particles was a tube furnace operating between 600°C and 620°C to volatilize sodium chloride (NaCl) which nucleated to form a narrow distribution of particles size centered at or below the desired test diameter. A monodisperse increment between 3 and 15 nm diameter was selected from this distribution with a differential mobility analyzer (DMA). The resulting negatively charged aerosol was neutralized (TSI model 3077) to minimize losses in the tubing downstream.

Tests of particle transmission efficiency were performed by sequentially measuring the particle concentration downstream of either the neutralizer or a section of bypass tubing (equivalent in length and connections to those on the neutralizer) by a manually operated, three-way valve. The desired flow through the neutralizer as well as that demanded by the UCPCs was adjusted by adding or removing air flow at the points in the system indicated in Fig. 1a. The reference UCPC had two normalizing functions: first, to account for any drift in particle concentration from the aerosol source with time, and second, to account for changes in dilution needed to provide several flows through the neutralizers while maintaining source and UCPC sample flows at their nominal values.

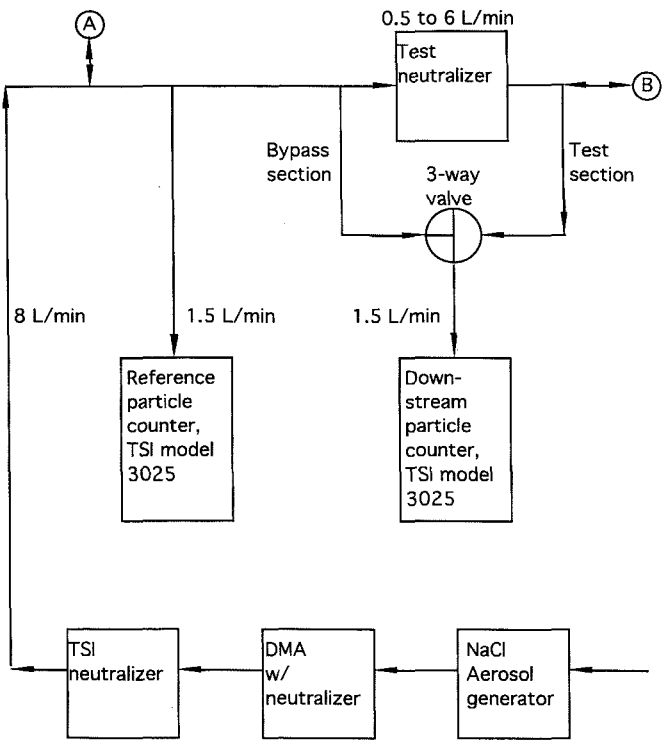

FIGURE 1. Schematic of the experimental apparatus for particle loss tests. Points labeled A, B, and C indicate sources and/or sinks of air used to balance flows through the various components of the system.

For each measurement in a test (for a given size, flow rate, and neutralizer unit) the UCPC particle counts were taken over three or more fixed time intervals with total counts of 20,000–100,000 particles in each interval. Measurements in the bypass and neutralizer flow configurations were alternated to yield three or more measurements per test. The transmission efficiency was calculated as

$$\text{transmission efficiency} = E = \frac{N_{\text{neut},i}}{N_{\text{ref},i}} \Big/ \frac{N_{\text{bypass},j}}{N_{\text{ref},j}},$$

where i and j are indices referring to specific, corresponding UCPC counting periods, $N_{\text{neut},i}$ is the count downstream of the neutralizer during measurement i, $N_{\text{bypass},j}$ is the reference count during measurement j, $N_{\text{ref,i}}$ is the count downstream of the bybass during measurement i. $N_{\text{ref},j}$ is the reference count during measurement j. Table 1 shows the results for the three neutralizers as a function of flow rate and particle size.

The primary source of uncertainty in this experimental design is the precision of flow rate measurements, in particular changes during a series of measurements. All pressure and vacuum sources were regulated, and flows were controlled by needle valves. The UCPCs were calibrated with bubble flow meters before the experiment to have an aerosol sample flow of 0.5 $cm^3\ s^{-1}$ ± 2%. The UCPCs were compared on a common aerosol source and shown to be stable and to agree within 3%. Sample flows through the neutralizers were measured and adjusted before each test and checked again after each test; they were found to be stable to within ±3%.

Figure 2a–c provides a graphical presentation of the diffusion loss results. Assuming laminar flow, the transmission efficiency expected for each neutralizer can be calculated theoretically as a function of particle size using the formula of Gormley and Kennedy (1949). Actual losses in the neutralizers are likely to be larger due to nonlaminar flow and irregular internal surfaces. For the CIT neutralizer, which was

TABLE 1. *Particle Transmission Efficiency for the Three Neutralizers as a Function of Flow Rate and Particle Size:* **Mean Values and Standard Deviation of the Measurements are Given**

	Diameter nm	Flow	Flow	Flow
CIT		6 l min^{-1}	1.5 l min^{-1}	
	15			
	10	0.97 ± 0.01	0.92 ± 0.02	
	7			
	5	0.91 ± 0.01	0.82 ± 0.03	
	3	0.84 ± 0.02	0.79 ± 0.07	
IfT		2.5 l min^{-1}	1.5 l min^{-1}	
	15	0.96 ± 0.02		
	10	0.95 ± 0.01	0.93 ± 0.02	
	7	0.91 ± 0.01		
	5	0.89 ± 0.02	0.83 ± 0.04	
	3	0.85 ± 0.02	0.77 ± 0.02	
TSI 3077		2.5 l min^{-1}	1.5 l min^{-1}	0.5 l min^{-1}
	15	0.96 ± 0.01		0.96 ± 0.01
	10	0.93 ± 0.01	0.94 ± 0.02	0.94 ± 0.01
	7	0.92 ± 0.01		0.89 ± 0.01
	5	0.89 ± 0.01	0.84 ± 0.01	0.83 ± 0.00
	3	0.84 ± 0.01	0.75 ± 0.03	0.76 ± 0.00

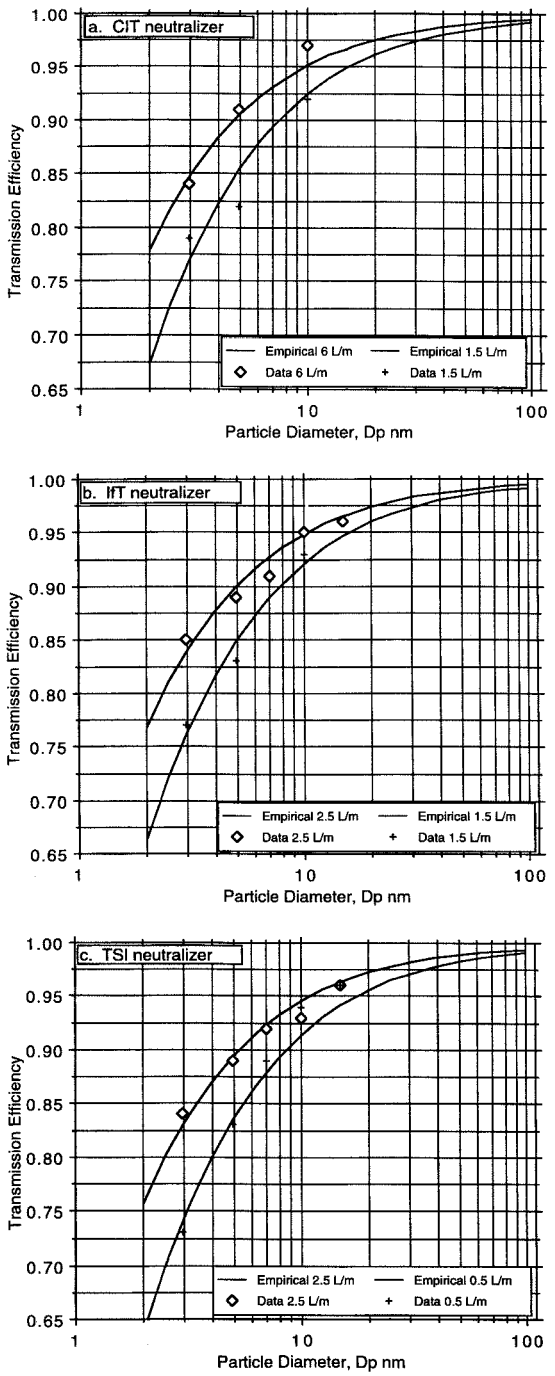

FIGURE 2. Transmission efficiency for three neutralizer designs. Data points are experimentally determined values and lines are the calculated values from a least square fits to data using an exponential equation of the form of Gormley and Kennedy (1949).

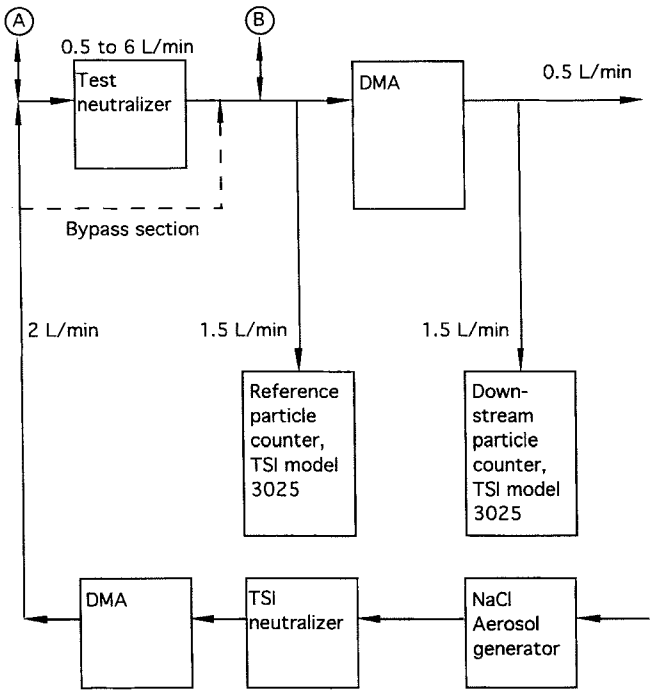

FIGURE 3. Schematic of experimental apparatus for recharging efficiency tests. Points labeled A, B, and C indicate sources and/or sinks of air used to balance flows through the various components of the system.

designed to be the most laminar of the three types tested, the transmission efficiency for 5 nm particles at 6 l min^{-1} is calculated to be about 0.96; the experimentally determined transmission efficiency at that flow and size was 0.91. The IfT neutralizer has the lowest volume and hence the shortest residence time. Its transmission efficiency at 1.5 l min^{-1} was 0.83 compared to a theoretical value of 0.93. An empirical fit to the experimental data (shown in Fig. 2) was made using an exponential equation similar to that of Gormley and Kennedy (1949) but with only one term.

PARTICLE CHARGING EFFICIENCY

To investigate the charging efficiency of each neutralizer the apparatus in Fig. 3 was used. NaCl particles of a given size with a single negative charge from the DMA were passed through one of the test neutralizers to be recharged, ideally to an equilibrium distribution. The resulting aerosol was separated according to charge in a second DMA. Particles of 10, 20, and 50 nm diameter at concentrations of 10^3–10^4 cm^{-3} were used. A flow of 6 l min^{-1} was used for the CIT neutralizer while the IfT and TSI

TABLE 2. Fraction of Singly, Negatively Charged Particles after Neutralizer

Particle Diameter (nm)	CIT 6 l min^{-1}	IfT 2 l min^{-1}	TSI 2 l min^{-1}	Wiedensohler Empirical
50	0.204	0.255	0.342	0.223
20	0.104	0.176	0.381	0.110
10	0.048	0.150	0.417	0.052

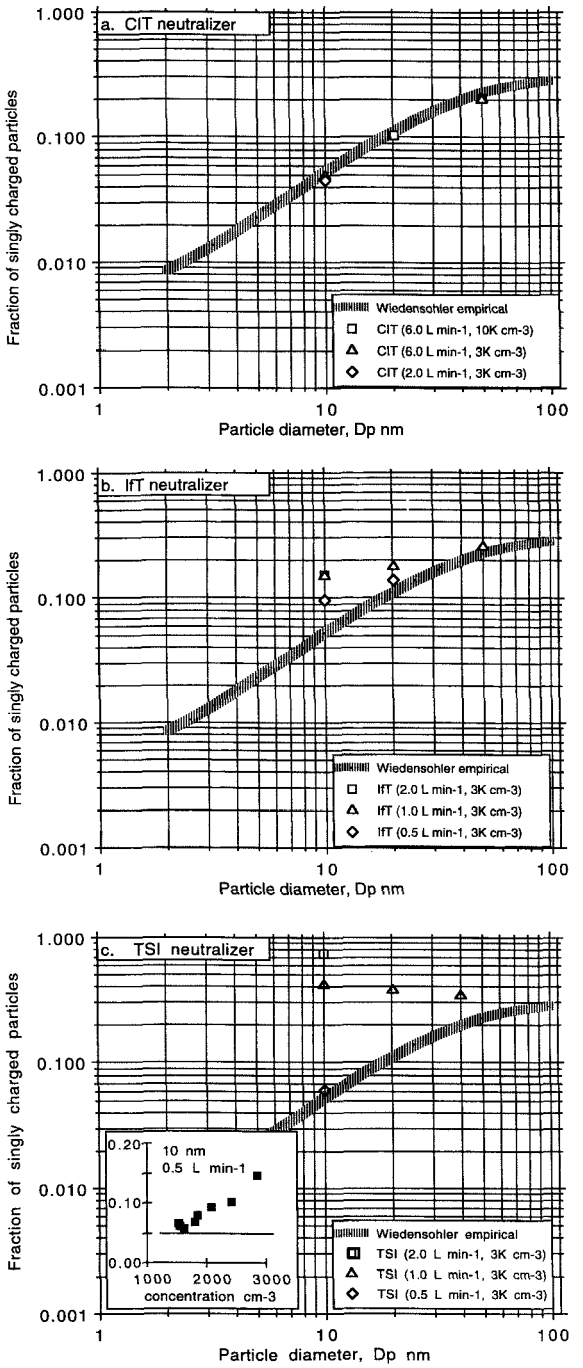

FIGURE 4. Particle charging efficiencies for three neutralizer designs. Data points are the experimentally determined values and lines are the theoretically predicted fraction of charged particles according to Wiedensohler (1988). The inset in the graph of the TSI data (c) shows the measured concentration dependence of recharging.

units were tested at 2 and 0.5 l min^{-1}. A summary of the results is presented in Table 2 and Fig. 4.

After passing through the CIT neutralizer at 6 l min^{-1}, the fraction of singly charged, 50 nm particles was 0.204, averaged over all concentrations tested. This is about 10% less than the theoretically predicted fraction of 0.223, but is still within the combined uncertainty of the measurement and theory. Charge fractions at the other sizes tested were low by a similar factor. The IfT and TSI neutralizers recharged the particles much less efficiently but approached the theoretical values at the lowest concentration and flow rate tested, $\approx 10^3$ cm^{-3} and 0.5 l min^{-1}, respectively. When two of these neutralizers were used in series at a flow rate of 2 l min^{-1} and concentration of 3×10^3 cm^{-1}, recharging was nearly at the theoretical value as well. It is not clear why their recharging efficiency was so low. Considering the strength of the radiation sources and the ion pair production rate for beta particles from ^{85}Kr in air (Cember, 1983) there should have been an adequate ion concentration for recharging to within a percent of the equilibrium value given the residence time in the chambers (Reist, 1993). There are two possible explanations for the discrepancy. The first is that in spite of theoretical considerations insufficient ions were present to bring the high concentration of aerosol to equilibrium in the specified residence time. This could be due to low beta source strength from the stainless steel foil cylinder housing the ^{85}Kr or rapid loss of ions from the interior volume to the walls of the neutralizer. The second possible explanation is that the flow profile is sufficiently nonideal that most of the flow passes directly from the inlet to the outlet inside the neutralizer such that the actual residence time of a large fraction of the particles is very much shorter than the calculated residence time for uniformly mixed flow.

IMPLICATIONS FOR AEROSOL MEASUREMENTS

All neutralizer designs tested showed significant but well-defined diffusion losses for particle sizes less than 10 nm and typical flows. These measurements can be used to correct for these losses in aerosol sampling measurements to retrieve the actual atmospheric aerosol concentration.

Within the combined measurement and theoretical uncertainties, the CIT ^{210}Po neutralizer was effective at 6 l min^{-1} and produced the predicted equilibrium, singly charged fraction even when the incoming aerosol was 100% singly charged. Therefore, we expect that it would be appropriate both for bringing atmospheric aerosol to an equilibrium distribution as well as for recharging more highly charged aerosol from a generator.

Both ^{85}Kr neutralizers failed to bring a singly charged aerosol to equilibrium charge for the range of concentrations and flows reported here. However, at the lowest of the tested flows and concentrations the ^{85}Kr neutralizer nearly achieved the predicted equilibrium singly charged fraction. In most atmospheric or clean-room aerosol measurement applications the particle population is largely uncharged or near equilibrium. Under such conditions, these neutralizers may well provide sufficient charging to establish an equilibrium distribution. Certainly, at the low particle concentrations, generally much less than 1000 cm^{-3}, that prevail in the marine boundary layer atmosphere of the southern hemisphere where ACE1 was conducted, the ^{85}Kr neutralizers should provide adequate recharging capability to produce an equilibrium charge distribution.

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Stolzenburg (Aerosol Dynamics, Inc., Berkeley, CA; aerosol@dnai.com), who may be contacted directly for further information.

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