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# Trapping charged nanoparticles in the nano aerosol mass spectrometer (NAMS)

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#### 1. Introduction

Particles between about 1 and 50 nm in diameter (referred to as nanoparticles herein) are an important subset of ambient aerosol that have significant climate [1,2] and human health [3–5] impacts. Chemical composition measurements of ambient nanoparticles can assist research in these areas by providing insight into the sources and transformations of these particles in the atmosphere [6,7]. From a chemical analysis perspective, this is a difficult task because of the small amount of material involved. For example, a 25 nm unit density particle has a mass of about 8 attograms (8 × 10<sup>-18</sup> g).

We have developed a nano aerosol mass spectrometer (NAMS) [8,9] to sample and analyze individual, singly charged 10–30 nm dia. particles from atmospheric pressure [10–12]. Chemical analysis is performed by irradiating individual particles with a tightly focused, high energy laser pulse. The laser radiation produces a plasma that reaches the complete ionization limit, where the particle is completely disintegrated into positively charged atomic ions [13]. The method of sampling nanoparticles into this instrument must meet two objectives. First, particles must be efficiently transmitted from atmospheric pressure into the vacuum. Second, particles must be spatially constrained within the focal region of the laser beam, preferably in a manner that can be used for size-selection. To accomplish the first objective, particles are transmitted from atmospheric pressure into the vacuum through a combination of aerodynamic and electrodynamic focusing

## ABSTRACT

Particle trapping in the nano aerosol mass spectrometer (NAMS) is investigated through a combination of modeling and experimental measurements to understand and optimize the trapping process. In the NAMS, charged nanoparticles pass through an aerodynamic lens, digital ion guide (DIG) and field adjusting lens (FAL) prior to entering a digital ion trap (DIT) where they are captured and subsequently analyzed. The DIT is operated with a square wave potential applied to the ring electrode. SIMION modeling was used to study particle motion from the exit aperture of the DIG into the DIT. Several parameters were characterized including particle starting position, kinetic energy, trajectory and FAL design. A new FAL assembly was designed to increase the number of trapped particles. The new design was found to increase the rate that ambient particles are analyzed by over an order of magnitude. With the new design, an ambient aerosol concentration  $(dN/d\log d_m)$  of  $1 \times 10^3$  cm<sup>-3</sup> yields approximately 2–3 particles analyzed

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elements. To accomplish the second objective, an electrostatic lens assembly is used to inject particles into an ion trap in a manner that maximizes trapping [8]. The complete instrument employs (in order of particle transit and analysis) an aerodynamic lens, a digital ion guide (DIG), a field adjusting lens (FAL), a digital ion trap (DIT), and a Nd:YAG laser along with time-of-flight mass analyzer for creating and analyzing atomic ions.

In order to maximize the rate at which particles are trapped and analyzed by NAMS, a fundamental understanding is needed of the trapping process and how the instrument design affects it, especially as a function of particle size. In this work, SIMION<sup>®</sup> is employed to model nanoparticle trapping. SIMION has been used in many studies to model ion trapping in conventional mass spectrometers [14–17] and to model low m/z ions in a digital ion trap (DIT) [18]. We apply the knowledge gained to the design, implementation and demonstration of an improved configuration for NAMS for the trapping and subsequent mass spectrometric analysis of nanoparticles.

### 2. Experimental

#### 2.1. Experimental measurements with NAMS

A schematic of the NAMS is shown in Fig. 1. From right to left (direction of charged particle and atomic ion motion as indicated in the figure), the instrument consists of an aerodynamic lens inlet, a digital ion guide (DIG), a digital ion trap (DIT), and a reflectron time-of-flight (TOF) mass analyzer. A full description of the NAMS is given elsewhere [9]. Below, we describe only those aspects of the instrument that relate directly to this study.

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Nano Aerosol Mass Spectrometer (NAMS)



Fig. 1. A schematic of the nano aerosol mass spectrometer (NAMS) with the "old" FAL design.

Test aerosols used in this study were generated with an Electrospray Aerosol Generator (model 3480, TSI Inc., Minnesota). This unit electrosprays analyte solutions (typically 0.2%, w/w, in 0.02 M ammonium acetate buffer) in the presence of a radioactive source to neutralize the primary droplets and minimize Raleigh breakup as the solvent evaporates. This results in an aerosol whose size distribution can be adjusted within the 10-30 nm size range (size distribution depends primarily on the analyte concentration of the electrosprayed solution), and each particle has either zero or one charge. The aerosol is sent through a unipolar charger [19] which increases the fraction of particles having a single positive charge [20]. The particle size distribution is monitored and specific particle sizes are selected for experiments requiring a monodisperse distribution with a Scanning Mobility Particle Sizer (SMPS; Electrostatic Classifier, model 3080, Condensation Particle Counter, model 3025a, TSI, Inc., St. Paul, Minnesota), which also helps pull the aerosol through the unipolar charger to the NAMS inlet. With the SMPS, particle size is measured and/or selected on the basis of mobility diameter  $(d_m)$ .

Instrument performance was also assessed through ambient aerosol measurements. Data are presented for two measurement campaigns: Pasadena, California in May–June 2010 using the "old" NAMS design (128,761 ambient particles analyzed); Hyytiälä, Finland in March–April 2011 using the "new" NAMS design (93,951 ambient particles analyzed).

Aerosol passing across the NAMS inlet is sampled through a 0.1mm-dia. flow limiting orifice into the aerodynamic lens similar to the design of Wang et al. [8], which is maintained at a subambient pressure of 1.5 torr. Particles exiting the lens traverse a  $\sim$ 6-mm field-free, differentially pumped region and pass through a 1-mm-dia. entrance aperture to the DIG. The DIG consists of four cylindrical rods with square wave potentials applied to them. The rods are configured similar to a quadrupole ion guide, that is the two opposing rods have a waveform that is 180° phase-shifted from the other two. The frequency of the square wave potential is fixed at 50 kHz with amplitudes of -500/+500 V. The dynamic electric field produced by these rods is sufficient to focus singly charged particles in the targeted size range of 10-30 nm dia. toward the centerline of the DIG. Particles exit the DIG through a 1-mm-dia. aperture that permits differential pumping. Argon gas is leaked into the DIG to collisionally cool the particles as they traverse this region, both to assist the focusing effect of the DIG and to increase the probability of trapping further downstream in the DIT. As discussed later, particles exiting the aerodynamic lens are much too energetic to be efficiently trapped, and must be translationally cooled prior to entering the DIT. As particles exit the DIG, they pass through the so-called field adjusting lens (FAL) [18]. This lens is slightly different in form and function than that described by Ding et al. [18] for a conventional DIT mass spectrometer. For the NAMS, this lens serves to manipulate the particle trajectory into the DIT.

The DIT consists of two end caps and a ring electrode in a configuration similar to that of a conventional quadrupole ion trap ( $r_0 = 10 \text{ mm}, z_0 = 7.1 \text{ mm}$ ). A square wave potential is continuously applied to the ring electrode, which operates at an amplitude of -507 V/+504 V with a variable frequency based on the particle size being analyzed. In the work presented here, a frequency of 10 kHz is used. As discussed previously, the DIT traps particles over a size range based on the mass normalized diameter ( $d_{mn}$ ), which is defined as the diameter of a spherical particle with unit density that has the same mass as the particle being trapped. Eq. (1) shows the relationship between the mobility diameter ( $d_m$ ) and mass normalized diameter ( $d_{mn}$ ) for spherical particles:

$$d_{\rm mn} = d_{\rm m} \left(\frac{\rho}{\rho_0}\right)^{1/3} \tag{1}$$

where  $d_{\rm m}$  is the mobility diameter,  $d_{\rm mn}$  is the mass normalized diameter,  $\rho$  is the particle density and the reference density is assumed to be 1 g/cm<sup>3</sup>.

A pulsed valve attached directly to the DIT supplies argon gas to this region in a manner that maximizes the DIT pressure during the trapping period while maintaining an average pressure of  $\sim$ 1–3  $\times$  10<sup>-4</sup> mbar in the differentially pumped chamber surrounding the DIT. Argon collisionally cools the particles to increase the probability of trapping and to focus them to the center of the DIT. Particle trapping occurs over a 250 ms time period. At the end of this period, a Nd:YAG laser (model CFR400, Big Sky Lasers, Billings, Montana) operating at ~200 mJ/pulse, is fired and the laser radiation is tightly focused to the center of the DIT. The laser radiation interacts with the trapped particle to form a plasma that disintegrates the particle into positively charged atomic ions. Immediately before the laser fires, the end caps are pulsed to +1300 V and -500 V (front to back in the direction of particle and ion motion inside the DIT) to extract the atomic ions into the TOF drift tube (R. M. Jordan Co., Inc., Grass Valley, California). The ion extraction period is timed to coincide with the +504V portion of the square wave potential applied to the ring electrode.

#### 2.2. Modeling particle motion through the NAMS

In this study, particle motion from the exit aperture of the DIG through the FAL and into the DIT is explicitly modeled. Particle motion through the aerodynamic lens and DIG is not modeled. Instead, the net effect of these components is incorporated into the model through the selection of starting conditions for particles leaving the exit aperture of the DIG. The range of starting

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