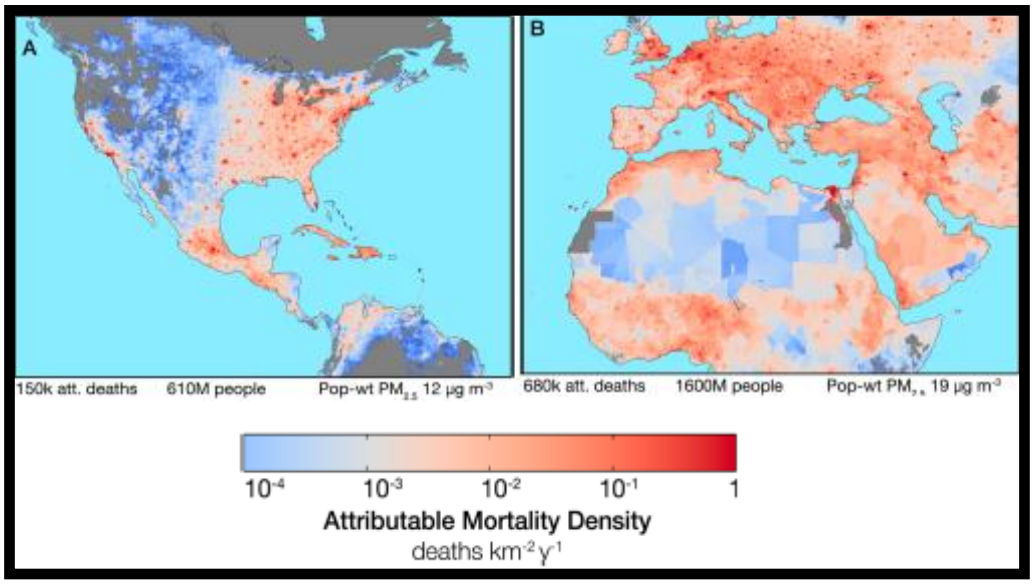
Vol. 3, No. 2 May 2015



IASTA E-Bulletin

Published by Indian Aerosol Science and Technology Association

Editor

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Cover Photo: Attributable premature mortality from ambient $PM_{2.5}$ exposure for (A) the northern America and (B) Europe and Africa. Dark gray regions indicate areas without attributable mortality due to ambient $PM_{2.5}$ below theoretical minimum-risk concentration (Courtesy: Apte et al., Environ. Sci. Tech., 2015)

CONTENTS

Generation of high concentration nanoparticles using Glowing Wire Technique

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1. Introduction

Nanoparticles (size < 100 nm) exhibit strikingly different properties than their bulk counterparts due to higher surface area and quantum effects. Because of their properties, these find application in various fields like medical, industrial and research. The Glowing wire generator (GWG) is one such technique for generation of Nanoparticles, where material is evaporated by the heating of high resistance wire and subsequent quenching by gas stream (Schmidt-Ott et al, 1980). Precise control over contamination and generation of particles in nano size range is the main advantage of this method (Peineke et al. 2009). Another advantage of generating nanoparticles using this method is significantly higher number emission rates which make them suitable to be used for specific purposes such as validation of coagulation based aerosol evolution models etc.

The generation and measurement of nanoparticle aerosols from electrically heated wire (glowing wire) is challenging, considering the control of parameters and characteristics of generated aerosols. The chemical composition of wire, electronic stability, variation in its electrical resistance with increasing temperature and carrier gas flow effects etc. affects the generation and stability of aerosols.

In the present work, we focused on developing a prototype hot wire generator for continuous generation of nanoparticle aerosols. This generator is operated at low power (~30-50 W) and can be used for continuous production of nanoparticle aerosols of concentrations up to the order of 10^7 cm^{-3} with mean size of particle agglomerates in the range of ~10-15 nm.

2. Materials & Methods

The prototype aerosol generator was parametrically studied for its response and an optimized set of operating parameters were selected and implemented in the experiments intended for generator performance and usage of generated particles. For parametric studies, GRIMM 5.403 CPC (measuring aerosols having diameter larger than 4.5 nm) was used to measure the integral aerosol number concentration, while GRIMM SMPS+C (11 nm to 1000 nm in 44 size channels) was employed for number size distribution measurements.

2.1 Hot Wire Generator description

The prototype generator (Fig. 1) consists of a metallic coil (1mm Diameter, 20 mm Long, 20 turns) enveloped in a quartz glass providing an interaction volume of 450 cm^3 for the generated aerosols. The upper part of the generator was specially fabricated for electrical connection of the metal wire (handling currents in Ampere range) in the generator volume. A variac was used for supplying AC voltage in the range of 5-10 V and circuit current produced was measured to be in the range of 5-10 A. The horizontal sampling ports have been provided to guide the generated aerosols to the aerosol measuring instruments. An excess port has been provided for other parallel measurements and for maintaining the pressure/flow rates within working ranges of measuring instruments. Wire of a specific chemical composition (single or composite) can be used for the generation of nanoparticle aerosols of the material of interest. In present work, commercially available electrical heater wire and nichrome wire were used

as aerosol sources. Particle free air was supplied to transport the test aerosols from the generation volume.

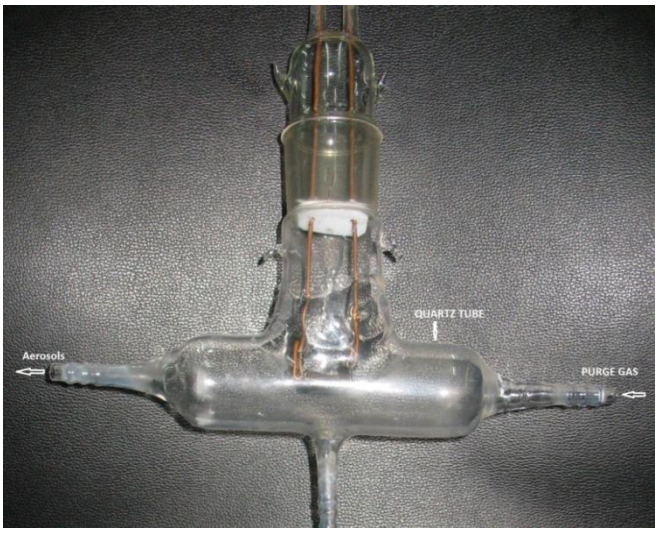


Fig. 1: Hot wire generator

2.2 Chemical characterization of wire material & its evaporation rate

As evaporation rate of heated wire forms the basis of vapor source term which subsequently nucleates, the chemical composition of wire is required to be known precisely. The saturation vapor pressure of constituents of wire is one important input for calculating its evaporation rate. Elemental composition of the tested wires has been obtained using XRF analysis. Following table shows the elemental composition of two wire materials used for this study. Conventional electrical heater wire was used as 1st wire (wire A) while commercial grade nichrome wire was the 2nd wire (wire B) for all experiments.

Table 1: Elemental Composition of wire material used in experiments

Components	Weight % of components (Wire A)	Weight % of components (Wire B)
Cr	18.2±0.1%	17.6±0.2 %
Fe	78.2±0.3 %	0.4±0.1 %
Ni	ND	79.8±0.3 %
Mn	3.2±0.1 %	2.1±0.1 %

Evaporation rate (r) for these wires can be calculated using Hertz–Knutsen equation under some assumptions

$$r = \lambda \sqrt{\frac{1}{2\pi M N_a k_b T}} \left(p_s(T) - p_0 \right)$$

Where λ = accommodation coefficient, M = molar mass, T = temperature, $p_s\ (T)$ = saturation vapour pressure, p_0 = vapour pressure, N_a= Avogadro number and k_b = Boltzmann constant

In above equation, accommodation coefficient accounts for vapour scattering back to the wire surface (for that correction, λ should be lesser than 1), however it can safely be assumed to be equal to 1 for the generation conditions. Since a carrier flow was used in the system, p_0 can also be assumed to be zero.

Saturation vapor pressure for these elements can be taken from standard vapor pressure look up tables. In absence of the measurement of the surface temperature of the hot wire, melting

point of the constituent was taken as T in the calculations. Table 2 shows the calculated p_s values and the corresponding evaporation rate for elements which were obtained by XRF analysis of these wires.

Table 2: Thermo-physical properties of wire elements

Element	Melting Temperature, T_m (K)	$P_s(T_m)$, Pa	Evaporation rate, r (mole $m^{-2}s^{-1}$)
Cr	2180	7.39×10^{2}	9.6
Fe	1811	3.39	4.66×10^{-2}
Ni	1728	1.04×10^{-1}	1.42×10^{-3}
Mn	1519	1.42×10^{2}	2.15

From the above table, it is evident that chromium contributes maximum towards vapor generation for present wires/study cases (percentage of Mn was very low for both wires and vapor pressure of Fe is significantly lower compared to Cr). The value shown in the table are upper bound of the evaporation rate as they are estimated based on individual metal melting point instead of alloy melting point (1673 K). Considering Cr as the vaporized constituent (100%), calculated evaporation rate (mole $m^{-2}s^{-1}$) can be converted to mole $m^{-2}s^{-1}$ which subsequently can be used to calculate source emission rate (SE) using the following equation

$$SE = \frac{(r*S)}{V}$$

Where, S is total surface area of the evaporating material and V is generator volume. In present study, evaporation rate of wire material was estimated ~5.78×10^{24} mole $m^{-2}s^{-1}$ while source emission rate (for generator volume of 450 cm^3) was ≈ 2.0×10^{25} mole $m^{-3}s^{-1}$.

3. Results and discussions

3.1 Optimization of Hot Wire Generator's parameters

A series of experiments have been carried out for obtaining optimum operating conditions for generation of stable high aerosol number concentrations. Results indicated that carrier gas flow rate and applied voltage are two main factors affecting the generator output.

Carrier gas flow rate plays an important role for generation and transport of the aerosols. During the generation process hot vapors carried by a cooler carrier gas become supersaturated, and nucleate and to form critical size nuclei. In presence of flow, residence time of vapor in hot regions changes, resulting in stable generator output.

3.1.1 Flow rate optimization

In order to know the differences with respect to changes in carrier gas flow rate, the wire/generator (wire A) was set-up at a fixed voltage of 7 V. As indicated in Fig. 2(a), the optimum carrier flow rate was observed to be between 1-1.5 L min^{-1}, resulting in maximum and stable number concentration. As the flow rate increased above 4 L min^{-1}, aerosol concentration was reduced appreciably. At higher carrier gas flow rates, wire cools more efficiently, reducing its temperature and, hence, lowering the evaporation rate. Also these vapors no longer remain supersaturated due to lesser temperature gradient between vapor and carrier gas affecting the number concentration.

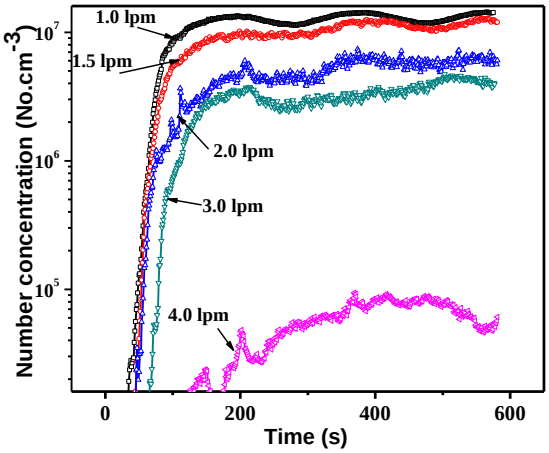
3.1.2 Voltage optimization

Although applied voltage in the range of 6-7 V looked optimum for generation of high concentration of nano-aerosols, another experiment was performed for studying the effect of the applied voltage on the maximum number concentration. This experiment was performed at a fixed flow rate of 1 L min^{-1}. The result has been shown in Fig. 2 (b). As observed, the maximum aerosol concentration reached to a saturation level after about 6 V (for wire A).

Based on the above experiments, optimized operating parameters (for wire A & B and of similar composition) were fixed as follows:

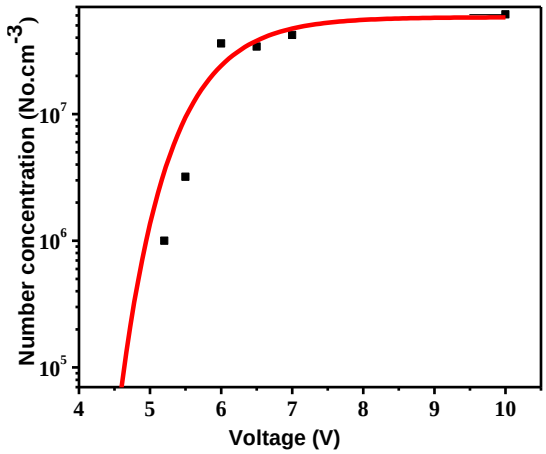
1) Applied voltage : 6-9 V (Power: 30-50 W)

2) Carrier gas flow rate (Air /Argon): 1-1.5 L min^{-1}
3) Maximum aerosol concentration achieved: ~ > 10^7 cm^{-3}.



(a)

Fig. 2(a): Effect of carrier gas flow rate on number



(b)

Fig. 2 (b): Effect of applied voltage concentration

3.2 *Generator output characterization*

In the second step, experiments were carried out to characterize the output of generator in terms of aerosol properties. Also, long time stability of generator was tested in some of the experiments. Size characteristics were interpreted using SMPS.

3.2.1 Number concentration emission

The electrical heating by the applied voltage requires some time to overcome the vaporization threshold after which the vapor emission from the wire starts. These vapors then self-nucleate to form aerosols in the nano size range and are guided to the measurement point using carrier gas. Fig. 3 shows behavior of aerosol concentration as measured by CPC for wire A in different set of experiments. As observed, wire A took about 50 seconds to warm up for vapor production and another 50-100 seconds for achieving saturation aerosol concentration. It can also be seen, saturation number concentration was of the order of 10^7 cm^{-3} for this wire which was more or less similar for the other wire as well.

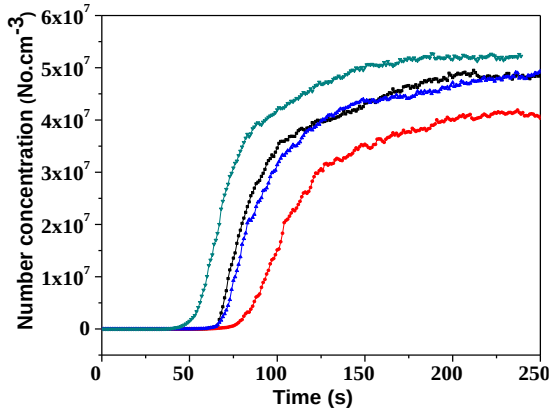


Fig. 3: Aerosol number concentration at generator 1.0 L min^{-1} carrier flow and applied voltage of 7 V

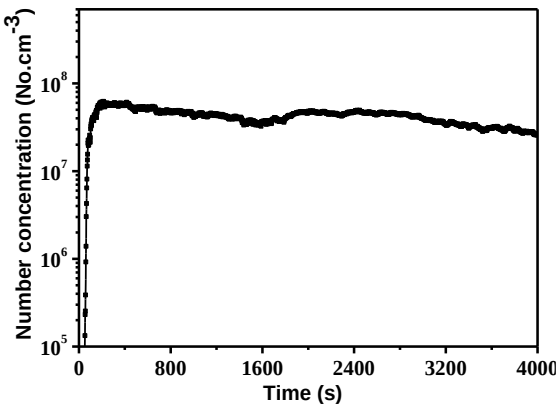


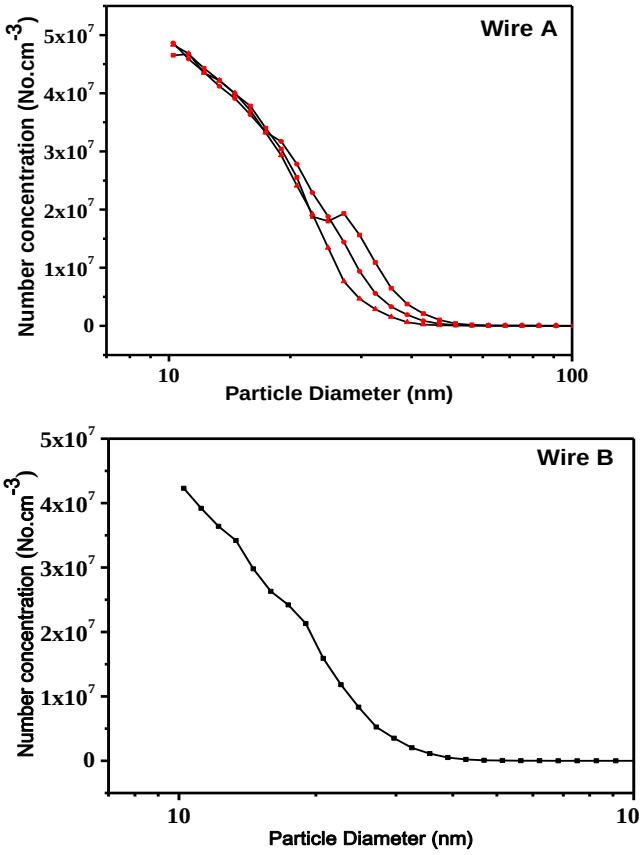
Fig. 4: Long Term response of output

Fig. 4 shows the long term behavioral response of wire A with the optimized parameters. It can be seen that, the number concentration remained in order of 10^7 cm^{-3} or more when

output was observed at the generator exit ensuring the steady nano-aerosol output form the generator.

3.2.2 Particle size distribution

The developed generator is capable to produce nanoparticle aerosols of high number concentration and appreciably less dispersity. SMPS was used in a separate set of experiments intended to measure size distribution of the generated aerosols. The results have been shown in Fig. 5 (a & b) for wire A & B, respectively. As seen from figures below, for wire A, the particle geometric mean diameter obtained was ~15-16 nm with geometric standard deviation (GSD) of ~1.4 and for wire B; it was ~14.8 nm with GSD of ~1.4.



(a) **(b)**

Fig. 5: Aerosol size distribution from generator for (a) Wire A, (b) Wire B

4. Conclusions

A prototype glowing wire nanoparticle aerosol generator has been developed. The generator is optimized for the carrier gas flow rate of 1-1.5 L min^{-1} and operating voltage of 6-9 V (electrical power: 30-50 W) to give steady and continuous number concentration of the order of 10^7 cm^{-3}. Two chemically different metal wires were used in experiments focused on evaluating the performance of generator and specifying the characteristics of the generated aerosols. Measurements carried out using SMPS (> 10 nm) at generator exit showed that the geometric mean size of aerosols coming out from generator is ~ 15 nm with a geometric standard deviation of 1.4. The high concentration nanoparticles generated through this instrument provides controlled and tunable source term for performing some of the aerosol behavioral experiments aimed at understanding processes such as coagulation effects. The generator can further be used for synthesis of coated or uncoated metal nanoparticles for specific use.

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(Full version of this work can be found in J Nanopart Res (2014) 16:2776, DOI 10.1007/s11051-014-2776-5)

Charge size distribution of common lab generated aerosols

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Introduction

Particle size, shape, concentration and charge are some of the important parameters governing aerosol evolution, transport and deposition. Among these, charge was not an easily measurable parameter because of the tedious indirect measurements and hence ignored until recent times. In the past, charge measurement studies have been conducted for nebulizers and combustion sources (Bruce Forsyth et.al, 1998; Chuen-Jinn Tsai et. al, 2005). Earlier studies used Electrical analyzer or Dual mobility particle sizer to measure the signal in terms of number concentration which was then converted to charge fractions using charge equilibrium theories (Rodrigues et.al, 2006; Sahu et.al, 2012). With the advent of Electrical Low Pressure Impactor (ELPI), charge measurement has become more direct and easy.

Precise knowledge of charge status of particles is key information for sampling, transport, deposition on surfaces or in the human respiratory tract system, and applications like micro-contamination control. Electrical effects of particles are also studied extensively for applications in drug delivery, electrostatic precipitators used in industries, effect on residence time of aerosols in atmosphere etc.

Aerosol particles can be electrically charged by different techniques and the number of charges on a particle depends on the charging mechanisms, which in turn, are sensitive to material properties, temperature, humidity etc. There are several routes for generation of aerosol particles. They can be synthesized from the liquid phase by atomizing a solution of specific composition to form droplets that crystallize to solid particles upon subsequent evaporation of the solvent. They can also be formed from gas-to-particle conversion via nucleation and growth by condensation and coagulation. Gas-to-particle synthesis can be achieved by using furnace, flame, plasma, or laser reactors, glowing wires and spark discharges. The magnitude and/or polarity of the charge are generally very difficult to predict even if the initial conditions during the generation process are known.

Most commonly utilized laboratory generator for aerosol studies is the nebulizer, which produces aerosols by mechanical dispersal of a solution. Combustion sources, on the other hand, are used to generate aerosols with very high number concentration (at source) of ultrafine size. Electrical heating of metal wires (usage for heating) is another source of aerosols which distinguishably generates high number concentrations in nano-size regime. Although characteristics such as concentration, size, chemical nature etc. have been measured

and studied for these generators; very few studies are available related to their charge measurement.

The aim of the present study was to measure the charge size distribution of aerosols generated via three different mechanisms viz., Nebulizer (NaCl), Combustion (Incense stick) and Hot wire generator (Nichrome wire). Electrical low pressure impactor (ELPI) has been employed for charge measurements in this work.

Instrumentation and methods:

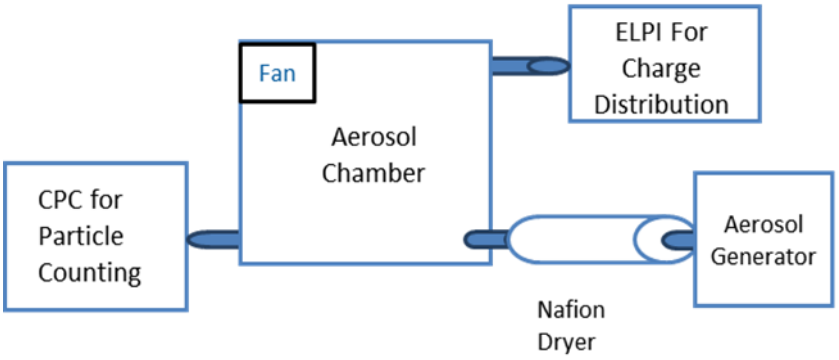

Fig.1. Experimental Set up

Experiments were carried out in an aerosol chamber equipped with fan for homogenization and particle measurement systems such as ELPI and Condensation Particle Counter (CPC) (fig. 1). Condensation Particle Counter (CPC) was used for integral number concentration measurement and to validate the steadiness of the aerosol concentration in the chamber whereas, ELPI is a particle spectrometer for measuring airborne particle size distribution (number and charge) in real-time. The operating principle of ELPI is based on particle charging using corona charger, size classification in cascade impactor and electrical charge detection with sensitive electrometers.

The generated aerosols were conditioned using Nafion dryer and passed to the aerosol chamber. Only under steady conditions, charge measurements were carried out. The measurement of size wise charge distribution (net charge per particle) was obtained by keeping ELPI with its internal particle charger ON alternatively for 20 seconds. The particles are charged to a known charge level in a corona charger of ELPI, hence for charge measurement, the charger is put off. Under charger ON condition, ELPI measures number concentration using calibrated penetration function while it measures current over all stages for charger OFF condition.

Results and discussions:

Fig. 2, 3, 4 and 5 show number of charges/particle plotted as a function of aerodynamic diameter of each stage of ELPI for aerosols generated from nebulization of NaCl, electrical heating based aerosol generator using nichrome wire (Hot wire generator) and combustion of incense stick.

Different concentrations of NaCl viz. 0.1%, 1%, 10% were used to generate aerosols. For each concentration, charge/particle increases with increase in particle size.

Charge/particle at mode viz., 120 nm was found to be 0.66. Study of charge obtained at mode is important because this charge is carried by maximum number of particles.

1. Atomization of NaCl particles

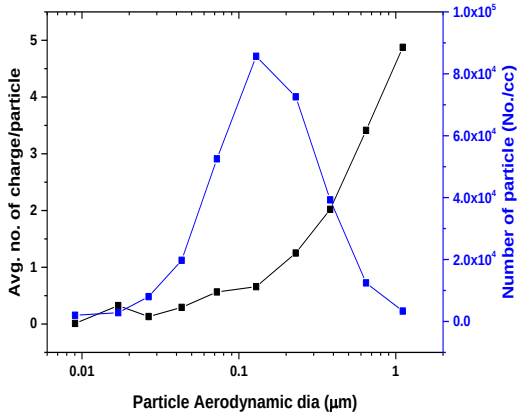

Fig.2 Particle charge and size distribution for particles generated from Nebulization of 10% NaCl solution

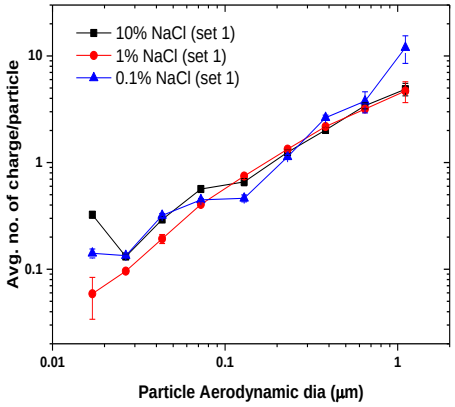

Fig.3 Particle charge distribution for particles generated from Nebulization of 10%, 1% and 0.1% NaCl solution

2. Hot wire generator

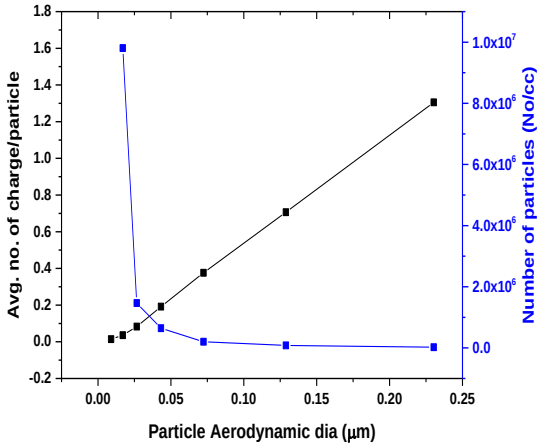

Fig.4. Particle charge and size distribution for particles generated from HWG

3. Combustion of incense stick

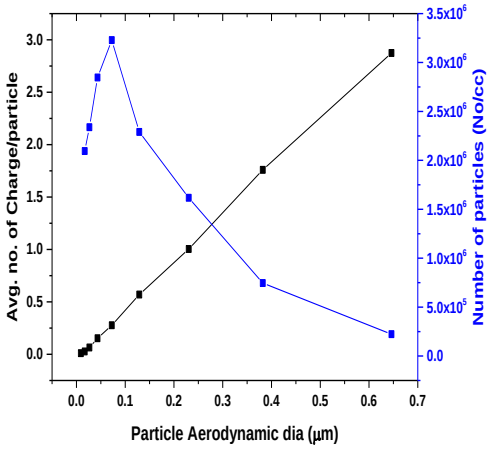

Fig.5. Particle charge and size distribution for particles generated from Combustion of Incense stick

Particles generated in case of combustion of incense stick and hot wire generator were in the lower size ranges. For them as well, charge was considered at mode concentration. The

charge/particle at modal size for hot wire generator was 0.036 at 17 nm and for combustion source 0.28 at 72 nm. For both sources, charge/particle increases with size as expected.

Conclusion:

Charge size distribution of aerosols generated from nebulization, combustion and hot wire generator were measured. Charge/particle was seen to be increasing with particle size for all the three sources. Charge at modal size was considered important because maximum particles would carry that charge. Charge/particle at modal size for nebulization, hot wire generator and combustion was measured to be 0.66 at 120 nm, 0.036 at 17 nm and 0.28 at 72 nm respectively.

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RESEARCH NEWS

1. Global mortality from ambient $PM_{2.5}$ exposure

Apte, J. S., J. D. Marshall, A. J. Cohen and M. Brauer (2015): *Addressing global mortality from ambient $PM_{2.5}$*, Environmental Science and Technology, DOI:10.1021/acs.est.Sb0123.

Evidence of premature mortality attributable to ambient $PM_{2.5}$ exposure continues to grow. In a recent study, *Apte et al.* (2015) have estimated global premature mortality from ambient $PM_{2.5}$ exposure using high resolution satellite-derived $PM_{2.5}$ and integrated concentration-response function to cover widely varying demography. Main focus of the study is to understand the mitigation measure implication. It was found that a modest improvement in relatively clean areas (e.g. North America and Europe) would result in large avoided premature mortality, while a substantial effort is required in the polluted regions (e.g. China and India). Average $PM_{2.5}$ would need to decrease by 20-30% over the next 15 years just to maintain present day premature mortality rate. The study also highlights the complexity involved in mitigation measures. For example, population exposure benefit per unit emission control is very high in densely populated countries like India. There is a need to carry out region-specific study to comprehend the problem and reduce the uncertainty in burden of disease.

2. Evidence of invigoration of aerosol-limited warm clouds

Koren, I., G. Dagan and O. Altaratz (2014): *From aerosol-limited to invigoration of warm convective clouds*, Science, 344: 1143-1146.

Invigoration effect is the most elusive and debated among all aerosol-cloud interactions. Earlier studies have found evidence of invigoration in deep convective clouds with cold top and warm base. This study demonstrated the effect to be evident on warm clouds in the pristine (aerosol optical depth, AOD is less than 0.2) regions of the world. Significant increase in rain rate and cloud fraction and a decrease in cloud top pressure was observed with an increase in AOD from as low as 0.06 (equivalent to 100 CCN/cm^3) to 0.1 (equivalent to 300 CCN/cm^3). Cloud microphysics is different than what is expected in polluted regions. Cloud droplet growth is fast, leading to quicker, but weak rain. The results suggest that the clouds forming in aerosol limited (i.e. pristine) environment are more sensitive to change in AOD (proxy for CCN). The study hypothesized that invigoration of warm clouds to freezing level is favourable over the pristine oceans.

3. A new size-composition resolved aerosol model (SCRAM)

Reference: Zhu, S., K. N. Sartelet, and C. Seigneur (2015): *A size-compsotion resolved aerosol model for simulating the dynamics of externally mixed particles: SCRAM (v 1.0),* Geoscientific Model Development, 8: 1595-1612.

A new size-composition resolved aerosol model (SCRAM) was presented in this study. The model is capable of simulating the dynamics of externally mixed aerosols. The new model categorizes aerosols by size-resolved composition and simulates dynamic evolution of aerosols due to coagulation, condensation/evaporation, and nucleation. The composition is represented either by mass fraction of individual species or group of species (e.g. inorganic, hydrophobic etc.). Composition and size of the particles are defined by the user. The model offers the possibility to change the mixing state of aerosols. The source code of the model is available at URL *http://cerea.enpc.fr/polyphemus/src/scram-1.0.tar.gz.* It can be modified under the terms of the GNU General Public License.

4. Quasi-biennial oscillation of stratospheric aerosol detected

Hommel, R., C. Timmreck, M. A. Giorgetta, and H. F. Graf (2015): *Quasi-biennial oscillation of the tropical stratospheric aerosol layer,* Atmospheric Chemistry and Physics, 15: 5557-5584.

Stratospheric aerosol is a key component in the concept of geo-engineering because of their ability to create a global dimming. It is therefore important to understand the natural variability of stratospheric aerosol load in order to quantify radiative impact of stratospheric aerosols on the Earth's climate. This study quantifies the influence of quasi-biennial oscillation (QBO) on stratospheric aerosols, when the stratosphere is unperturbed by volcanic eruptions. QBO is a dominant mode of stratospheric variability. Stratospheric aerosol layer extent in the tropics is modulated by QBO by up to 6 km (16-33 km). Strongest signal is

detected above 10 hPa pressure level (~31 km), while the signal is moderate below 10 hPa level where aerosol mixing ratio is largest. The study shows that the life cycle of sulfate droplets in the tropical lower stratosphere is influenced by processes which are coupled in a nonlinear manner to QBO.

5. Europe Aerosols Campaign

http://www.globe.gov/web/europe-aerosols-campaign/overview.

Europe Aerosols Campaign is a nice initiative involving student (school and college) community and educating them about aerosol science. The campaign is launched under GLOBE program. Two intensive observation periods are planned for the year 2015 (March 2 to May 8 and Sep-Oct). People are invited to take measurements with sunphotometer (following a common data protocol), upload the data to a common server and even analyze them with the help of aerosol scientists. Low cost sunphotometers can directly be purchased from the authorized vendors. This is a novel concept for spreading awareness and interest about aerosols through direct involvement in data collection and enlarging the measurement network in a systematic framework.

FORTHCOMING EVENTS

1. 4th Workplace and Indoor Aerosols Conference 2016 -

The 4th Workplace and Indoor Aerosols Conference will be held in Barcelona, Spain during April 21-22, 2016. The conference website is: **http://www.aerosols2016.eu/**.

2. Upcoming IGAC Events -

Several events related to atmospheric chemistry and aerosols are planned under International Global Atmospheric Chemistry (IGAC) annual program. Detailed information can be found in: **http://www.igacproject.org/igac-events**.