Contract No:

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eDPS Aerosol Collection

The eDPS Aerosol Collection project studies the fundamental physics of electrostatic aerosol collection for national security applications. The interpretation of aerosol data requires understanding and correcting for biases introduced from particle genesis through collection and analysis. The research and development undertaken in this project provides the basis for both the statistical correction of existing equipment and techniques; as well as, the development of new collectors and analytical techniques designed to minimize unwanted biases while improving the efficiency of locating and measuring individual particles of interest.

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publically published in its current form.

SRNL Legal Signature

Signature	**Date**

eDPS Aerosol Collection

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Thrust Area: SI4-3

Project Type: Strategic

Project Start Date: October 1, 2013
Project End Date: September 30, 2015

The eDPS Aerosol Collection project is focused on improving current electrostatic particle collection science and technology. By leveraging selected characteristic features, particles of interest can be manipulated during the collection process into a physical configuration conducive to subsequent microscopy and micro-scale laboratory analyses. In order to achieve this goal, a fundamental research and development effort is required to build an accurate model/understanding of how the desired particles behave in electric fields at atmospheric pressure and what physical and chemical characteristics dominate, or could be made to dominate, their trajectories. Aspects of this work are related to ion mobility mass spectrometry and traditional aerosol science, but critical knowledge is lacking and must be developed as part of this effort.

FY2015 Objectives

- Procure computational fluid dynamic (CFD) software.
- Simulate existing collector design.
- Identify physics and chemistry effecting particle collection.
- Evaluate several collector technologies
- Define bias and uncertainty metrics for collectors

Introduction

SRNL has successfully leveraged electrostatic precipitator (ESP) based aerosol collection technology for national security applications and made it available to selected U.S. agencies and departments. The current state of ESP technology is fundamentally an indiscriminate design, i.e. used to collect a broad range of sizes and types of particles. To date, this feature has been a benefit for SRNL's current customers. However, in order to expand into new markets, SRNL needs to use the empirical knowledge gained to date through the application of the Lab's current technology to guide fundamental research into aerosol particle collection physics and the manipulation of particles during the collection process.

The ability to collect samples in such a way that the particles of interest can be easily and quickly found for individual analysis increases the efficiency of numerous techniques, such as scanning electron microscopy, secondary ion mass spectrometry, x-ray diffraction, Raman microscopy, etc. These and other similar per-particle analytical techniques are being used to obtain ever more information from the source of the collected particles, and finding the informative particles amongst background material is challenging for all of them. The ultimate goal of this work is the ability to deploy aerosol collectors that provide a level of "sample preparation" during the collection process itself.

This project will not only allow SRNL to make a significant contribution to collection technology within the DOE mission space, it will also reinforce and grow SRNL's reputation as a DOE center of expertise in aerosol sampling. In addition to maintaining our current customer relationships, the basic research being performed in this LDRD is enabling SRNL to expand its reputation into the larger open aerosol science community and will provide a vehicle to transition existing particle knowledge and expertise into the broader community as appropriate. The potential applications for advanced particle collection for particulate analysis are broad and the ability to perform this type of collection is of interest, not only to the nonproliferation community, but the biology, industrial hygiene and pharmaceutical fields as well.

Approach

The eDPS Aerosol Collection project focuses on building the knowledge and infrastructure to perform advanced aerosol collection research and development. This overarching objective is being executed via three main areas. The primary research area is developing the capability to simulate the complex interaction of forces that influence a particle's trajectory in an ESP type collector. Commercially available modeling software is being used in to simulate the effects of air flow, static and dynamic electric fields, and magnetic fields on particles. Once developed and validated against existing collector designs these models will be incorporated into the R&D and engineering processes currently serving our customers.

The secondary key area is the upgrading of an existing aerosol chamber to an automated collector test bed. Computer controlled power supplies, particle generators, data acquisition, and modular test components make it possible to quickly build and test new designs. These areas focus on basic research and long term returns. The eDPS project also incorporated a third area to explore possibilities based on commercially available equipment adapted for this purpose.

Results/Discussion

A primary concern for aerosol and particulate scientists is understanding and accounting for biases introduced throughout the generation, transport, sampling, and analysis processes. This year the eDPS team has evaluated several aerosol collectors. Both commercially available and SRNL developed collectors were studied. Figure 1 shows the probability distribution functions for the equivalent circular diameter of sea spray aerosol particles collected by four different aerosol collectors. The SKC (Sioutas Personal Cascade Impactor, SKC Inc., Eighty Four, PA) and the NGI (model 170 Next Generation Impactor, MSP Corporation, Shoreview, MN) both function based on aerosol mobility physics while the ACE (SRNL) and ImpACE (SRNL) are electrostatic precipitators. The four collectors were set up to simultaneously collect sea spray for three hours at a beach front location near Charleston, SC. Probability density functions for the feature areas for each of the detectors. A smooth kernel distribution is calculated from the raw data with bandwidth selection following Silverman's rule. The plot gives the

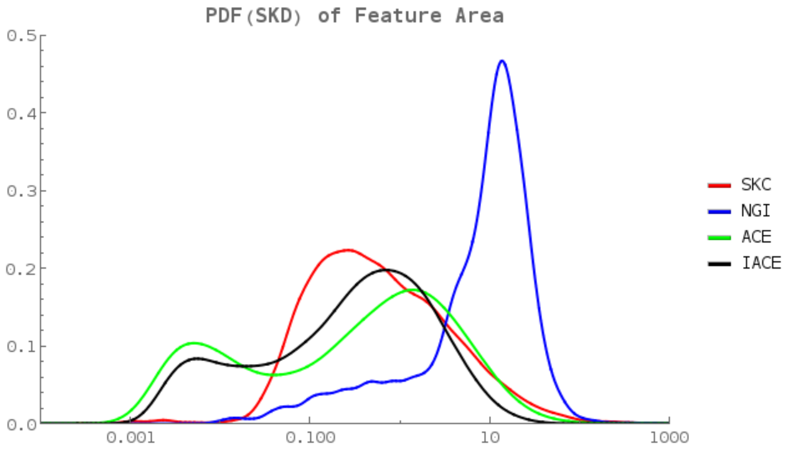

Figure 1: Particle size distributions for four simultaneous sea spray aerosol collections use four different collectors.

probability that an observed value will fall between x and x+dx. This has the effect of normalizing the four data sets (i.e. the area under each curve is 1). Table 1 gives the particle (feature) count for each collector, giving an indication of overall efficiency. Figure 1 clearly highlights the potential for miss-interpretation of the data if the effects of a given collector are not taken into account.

Table 1: Feature Count

	Net Features
SKC	9994
NGI	10539
ACE	2021
IACE	4301
Total	**26855**

In order to understand the underlying principles that give rise to the collection bias seen above, it is necessary to develop a model that captures the physics that define the fate of a particle in the collector. Figure 2 illustrates the electric potentials (a) and air velocity (b) in a simulation of the SRNL ACE collector. Due to the device being symetrical in the vertical and horizontal axes, it is only necessary to model one quarter of the device when the appropriate mirroring functions are included. The model was developed in COMSOL Multiphysics (COMSOL, Inc., Burlington, MA).

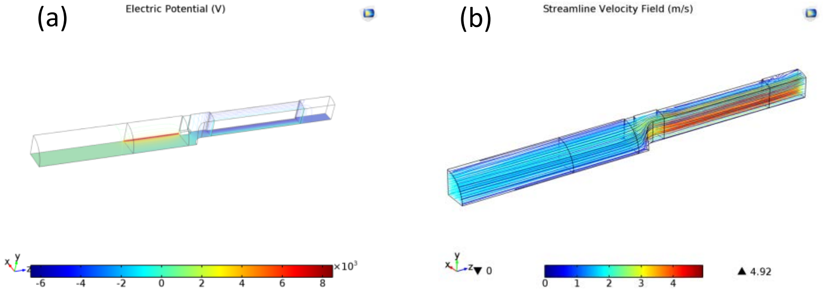

Figure 2: Electric potential (a) and air flow velocity (b) in the SRNL ACE electrostatic precipitator.

Once the device has been simulated and the static forces calculated, it is possible to calculate the trajectories of individual particles. Figure 3 shows the trajectories of a population of particles consisting of four sizes (0.01 µm, 0.1 µm, 1 µm, and 10 µm) emitted from each of 100 random locations. Each particle picks up electric charges in the corona region according to Cochet's model[1,2].

$$\left\{(1+2\,\lambda/d_p)^2+\left(\frac{2}{1+2\lambda/d_p}\right)\cdot\left(\frac{\epsilon_r-1}{\epsilon_r+2}\right)\right\}\cdot\pi\epsilon_0\,d_p^2\,E\cdot\left(\frac{t}{t+\tau_Q}\right)$$

$$\tau_Q=\frac{4\,\epsilon_0}{j_{\mathrm{NE}}/E}$$

λ – Mean Free Path
ϵ – Electrical Permittivity
E – Electric Field
t – Time in Electric Field
j_{NE} – Current Density
d_p – Particle Diameter

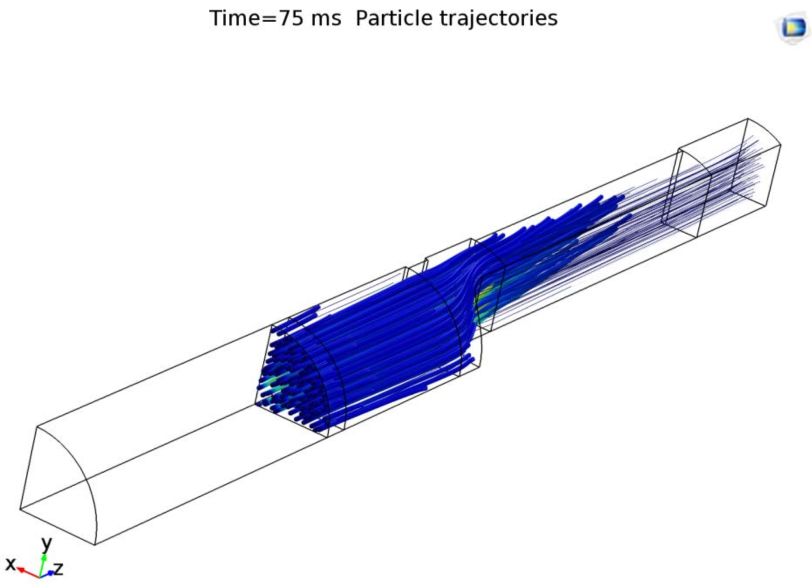

Figure 3: Particle trajectories fin the ACE simulation. Probability of collection is a function of particle size (charge and drag) and initial proximity to corona wire (charge).

These simulations provide insight into the balance of forces needed to collect any given particle and the potential for introducing collection bias. The probability that a particle is collected in an ACE device fundamentally depends on the balance between the horizontal drag force or air resistance and the number of charges on the particle creating an electric force in the collector's electric field. Figure 4 shows the fate of the 400 particles based on their initial location in the collector inlet.

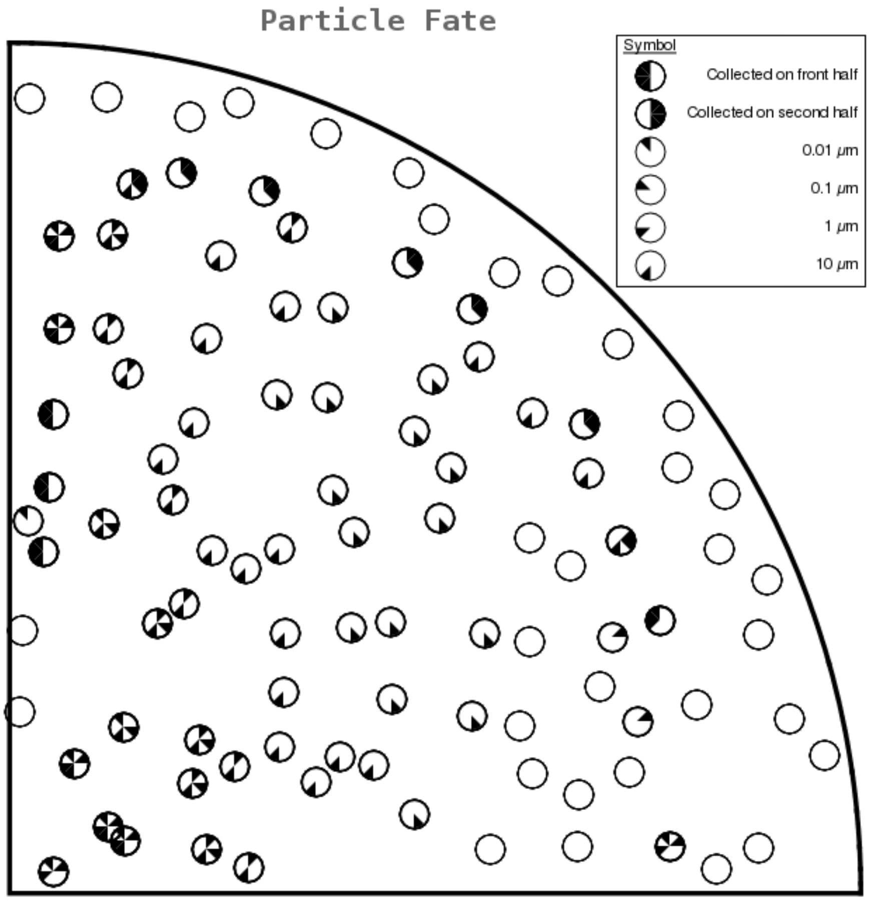

Figure 4: Particle fate as a function of initial position prior to entering charging region. One quarter of the end view is shown, and the corona wire is located at the 0,0 origin.

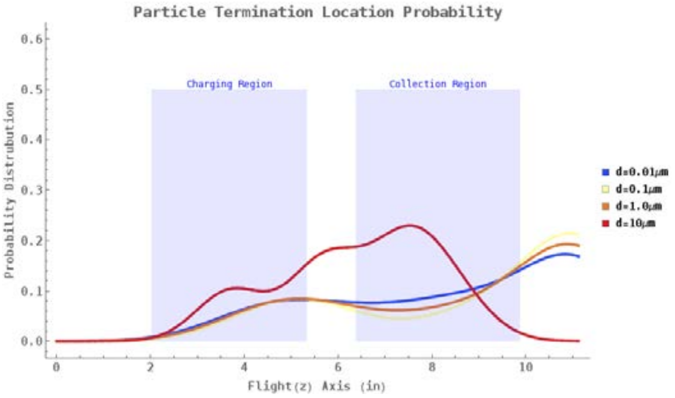

Figure 5: Probability of collection locations for the 400 particles.

Figures 4 and 5 show how the particle population and specific collector geometry can affect collection efficiency and bias. Figure 5 also illustrates how the selection of the collector plates, selected for SEM analysis, can introduce bias and could potentially lead to miss-interpretation of the particle population being analyzed. Collection efficiencies were calculated to be approximately 70% by depletion, i.e.

determined by measuring the number of particles that remain in the exiting air flow. The efficiency looking at the collector plate was approximately 30%. This discrepancy is consistent with published work out of PNNL[3].

FY2015 Accomplishments

- Developed, built and tested an impactor-electrostatic precipitator (Imp-ACE) based collector designed to prevent the collection of unwanted macroscopic particles.
- Performed two sea spray aerosol (SSA) collections for inter-collector characterization.
- Developed cluster analysis tools used to improve the automated classification of particles by the scanning electron microscopes.
- Established computational capabilities integrated with R&D, design and production within the National Security directorate.
- Upgraded aerosol science laboratory capabilities for aerosol physics experiments and characterization of new designs.
- Improved fundamental understanding of collector dynamics using complex real-world samples.
- Developed computational simulations that have provided physics based explanations for empirical observations.

Future Directions

Information and capabilities developed as part of this LDRD project have already been incorporated into several proposals to R&D funding agencies including:

- Defense Threat Reduction Agency J9 NTFC/A
- Department of Homeland Security / National Technical Nuclear Forensics Center
- National Nuclear Security Administration Office of Defense Nuclear Nonproliferation Research and Development

This project has also enhanced SRNL's ability to characterize bulk powder samples and the mathematical and statistical techniques needed to extrapolate meaningful information about the bulk material from SEM and other per-particle analytical measurements. Spin-off projects already begun in fiscal year 2016 are exploring techniques for identifying probative signatures based on the clustering in multiple orthogonal data sets.

FY 2015 Publications/Presentations

1. J. Venzie; Quality Control and Verification/Validation in Technical Nuclear Forensics R&D; University of Washington; July 2015.
2. J. Venzie, H. Ajo, N. Bridges, T. Shehee, R. Rogers; Analysis of SRS HB-Line Alternative Feedstock Two Plutonium Oxide; DHS NTNFC Academic Collaboration and Program Review, August, 2015.
3. J. Venzie; Finding a Needle in the Haystack and What Does It Mean?; University of Missouri, March 2015.

References

1. Parker, K. R. (1997). Applied Electrostatic Precipitation. London, UK, Blackie Academic & Professional.
2. Hinds, W. C. (1999). Aerosol Technology : properties, behavior, and measurement of airborne particles. New York, NY, John Wiley & Sons, Inc.
3. Laskin, A. and J. P. Cowin (2002). "On deposition efficiency of point-to-plate electrostatic precipitator." Journal of Aerosol Science 33(3): 405-409.

Acronyms

ACE	Aerosol Contaminant Extractor – an electrostatic precipitator developed at SRNL.
CFD	Computational Fluid Dynamics – modeling of fluid (or air) flow.
DOE	U.S. Department of Energy
eDPS	Title of this project.
ESP	Electrostatic Precipitator – particle collector technology using electric fields.
ImpACE	Impactor-Aerosol Contaminant Extractor
LDRD	Laboratory Directed Research and Development
NTS	Nonproliferation Technology Section – SRNL organization executing this LDRD.
PDF	Probability Distribution Function – probability that a variable will be less than or equal to a given value.
SEM	Scanning Electron Microscope
SRNL	Savannah River National Laboratory

Intellectual Property

The research team is working with the SRNL intellectual property group to assess the products of this project.

Total Number of Post-Doctoral Researchers

Dr. Robert Rogers joined SRNL in July of this year. His background is in aerosol classification and he has performed the sea spray aerosol analysis and data clustering tasks. Dr. Rogers is a post-doctoral fellow supported through the National Nuclear Forensics Expertise Development Program, part of the National Technical Nuclear Forensics Center within the U.S. Department of Homeland Security Domestic Nuclear Detection Office.