Properties of Smoke from Overheated Materials in Low-Gravity

David L. Urban[1], Gary A. Ruff[2] and William Sheredy[3],
NASA Glenn Research Center, Cleveland, Ohio 44135

Thomas Cleary[4] and Jiann Yang[5]
National Institute of Standards and Technology, Gaithersburg, Maryland 20899

George Mulholland[6]
The University of Maryland, College Park, Maryland 20742

and

Zeng-guang Yuan[7]
National Center for Space Exploration Research, Cleveland, Ohio 44135

Smoke particle size measurements were obtained under low-gravity conditions by overheating several materials typical of those found in spacecraft. The measurements included integral measurements of the smoke particles and physical sample of the particles for Transmission Electron Microscope analysis. The integral moments were combined to obtain geometric mean particle sizes and geometric standard deviations. These results are presented with the details of the instrument calibrations. The experimental results show that, for the materials tested, a substantial portion of the smoke particles are below 500 nm in diameter.

I. Introduction

Appropriate design of fire detection systems requires knowledge of both the expected signature of the events to be detected and the background levels of the measured parameters. Terrestrial fire detection systems have been developed based on extensive study of terrestrial fires.[1, 2] Unfortunately there is no corresponding data set for spacecraft fires and consequently the fire detectors in current spacecraft were developed based upon terrestrial designs. There are a number of factors that can be expected to affect the particle size distribution of the smoke from spacecraft fires. The absence of buoyant flow in low-gravity increases the residence time in microgravity fires and increases the transit time from the reaction zone to the detector.[3] Microgravity fires have been found to have radically different structure from their 1-g counterparts. The limited options available to respond to a spacecraft fire increase the importance of early detection. Finally the materials used in spacecraft are different from typical terrestrial applications where smoke properties were previously evaluated. All of these effects can be expected to change the smoke particle size distribution. The objective of this work was to make sufficient measurements of smoke from spacecraft fires to enable improved design of future detectors.

Smoke Background

---

[1] Branch Chief, Combustion and Reacting Systems Branch, 21000 Brookpark Road, MS 77-5, Senior Member.
[2] Advanced Capabilities Project Office, 21000 Brookpark Road, MS 77-7, Associate Fellow.
[3] ISS Projects Office, 21000 Brookpark Road, MS 77-5.
[4] Engineered Fire Safety Group, 100 Bureau Drive, Stop 8664.
[5] Fire Measurements Group Leader, 100 Bureau Drive, Stop 8662.
[6] Visiting Professor of Mechanical Engineering, Glenn L. Martin Hall.
[7] Staff Scientist, 21000 Brookpark Road, MS 77-5.

Smoke is a general term that encompasses aerosol materials produced by a number of processes. In particular it can include unburned, recondensed, original polymer or pyrolysis products that can be either liquid or solid, hydrocarbon soot, condensed water vapor, and ash particles. Soot particles dominate the smoke particulate in established flaming fires while unburned pyrolysis products and recondensed polymer fragments are produced by smoldering and pyrolysis in the early stage of fire growth. Given the constrained space on any spacecraft, the target for the fire detection system is necessarily the early phase and not established flaming fires; consequently, the primary target for detection is the pyrolysis products and not the soot.

Prior spacecraft systems are summarized in more detail in papers by Friedman and Urban.[4, 5] In the Mercury, Gemini and Apollo missions, the crew quarters were limited and mission durations were short, consequently it was considered reasonable that the astronauts would rapidly detect any fire. The Skylab module, however, included approximately 30 UV-sensing fire detector.[4] These devices were limited to line-of-sight and were reported to have difficulties with false alarms. The Space Shuttle Detectors were based upon ionization fire detector technology, the most advanced technology available at the time and used an inertial separator designed to eliminate particles larger than 1-2 micrometers. The International Space Station (ISS) smoke detectors use near-IR forward scattering, rendering them most sensitive to particles larger than a micrometer, outside of the range of sensitivity of the shuttle detector. As described by Friedman[4] there have been six overheat and failed component failures in the NASA Orbiter fleet in addition to several similar incidents that have occurred on the ISS. None of these events spread into a real fire but as mission durations increase, the likelihood of failures increases. The experience on Mir in 1997 has shown that failure of oxygen generation systems can have significant consequences. As a result, improved understanding of spacecraft fire detection is critically needed.[6]

Previous work on smoke particles from low-gravity sources by Urban et al.[5] found that the particulate produced by low-gravity flames (soot or unburned fuel particles) tends to have larger size particles than in normal gravity. Results from the CSD (Comparative Soot Diagnostics) Experiment[5] which studied smoke properties in low-gravity from several spacecraft materials suggested that liquid smoke particles could achieve sizes larger than 1 µm while solid particulate remained in the sub-micrometer range. However, the CSD experiment did not produce sufficient data concerning the size of the liquid smoke particles to guide detector design. The combined impact of these limited results and theoretical predictions is that, as opposed to extrapolation from 1-g data, direct knowledge of low-g combustion particulate is needed for more confident design of smoke detectors for spacecraft.

II. SAME Experiment

To address the limited data from the CSD experiment concerning the likely size of spacecraft smoke particulate, another experiment the Smoke Aerosol Measurement Experiment (SAME) was developed. The SAME experiment sought to avoid the problems experienced by the CSD experiment by obtaining the particulate size statistics on-orbit with a reduced dependence upon sample return to Earth. This is a challenging endeavor because existing aerosol instrumentation is typically large, incompatible with spacecraft experiment constraints, and may require substantial sample return to Earth. As will be described below, an alternative approach was employed that used three discrete instruments to measure separate moments of the size distribution. When combined, these moments provide useful aggregate statistics of the size distribution. The measurements were made using smoke generated by overheated spacecraft materials in much the same manner as the CSD experiment however the sample temperature, flow field, and particle aging time were more rigorously controlled.

A. Moment Method

The approach used by the SAME experiment is termed the 'moment method' for convenience.[7] As will be described below, the approach consists of measuring three moments of the size distribution (zeroth, first and third) and using the properties of the log-normal distribution to estimate the geometric mean diameter and the standard deviation.

The average particle size and an estimate of the width of the size distribution will be estimated from various moments of the size distribution. The number distribution, $f_N(D)$, is defined as

$$f_N(D) = \frac{dN}{dD} \tag{1}$$

where dN is the number of particles per cm^3 with diameter between D and D + dD. The moments of interest consist of the number concentration, M_0, the first moment M_1, and the volume or mass concentration moment, M_3 and are defined as

$$M_i = \int D^i f_N(D)dD \quad i = 0,1,3 \tag{2}$$

When i=0, the zeroth moment of the distribution, M_0, equation (2) is simply the number of particles per unit volume. In the SAME experiment, this was measured using a condensation nuclei counter. The first moment, i=1, can also be thought of as the "diameter concentration" or integrated diameter per unit volume and is approximately proportional to the ionization detector moment (signal). For particles in the Mie scattering regime, particles sizes from 0.3λ to about 3λ (~0.2 µm to 2.0 µm for a red laser), the light scattering signal is approximately proportional to the third moment, i=3. From these moments, and a measurement of M_0 using a condensation particle counter, two mean diameters can be computed: the count (arithmetic) mean diameter $D_{0.5}$ or $\overline{d}$, which is equal to M_1/M_0 and the diameter of average mass $D_{1.5}$ or $d_{\overline{m}}$, which is equal to $(M_3/M_0)^{1/3}$. The basis for the subscript naming convention for $D_{0.5}$ and $D_{1.5}$ will be discussed later). The log-normal size distribution is widely used for describing the size distribution of aerosols including non-flaming smoke because for most aerosols; the bulk of the number concentration is associated with smaller particles.[8, 9] The number distribution fN(D) for the lognormal distribution is expressed as follows:

$$f_N(D) = \frac{N_t}{(2\pi)^{1/2} D \ln \sigma_g} \exp\left(- \frac{(\ln D - \ln D_g)^2}{2 \ln^2 \sigma_g} \right) \tag{3}$$

where N_t is the total number concentration of the aerosol (=M_0), and D_g and σ_g are the geometric mean diameter and geometric standard deviation defined by

$$\ln D_g = \int_0^\infty \ln D f_N(D)dD \Big/ \int_0^\infty f_N dD \tag{4}$$

$$\ln \sigma_g = \left[\int_0^\infty (\ln D - \ln D_g)^2 f_N(D)dD \Big/ \int_0^\infty f_N(D)dD \right]^{1/2} \tag{5}$$

For the log-normal distribution, one finds that the various diameter definitions given above are related to the geometric mean number diameter, D_g, via the equation:[8 9]

$$D_p = D_g \exp(p \ln^2 \sigma_g) \tag{6}$$

For the count mean diameter, $D_{0.5}$, and the diameter of average mass, $D_{1.5}$, the corresponding values of p are 0.5 and 1.5. Fig. 1 shows a typical log-normal distribution for a D_g=1.0 and σ_g= 1.6. For this distribution, the corresponding values of $D_{0.5}$ and $D_{1.5}$ are1.17 µm and 1.39 µm, respectively. Using equation (6), one can relate σ_g to the ratio of $D_{1.5}$ and $D_{0.5}$ via the equation:

$$\sigma_g = \exp\left(\ln(D_{1.5} / D_{0.5})\right)^{1/2} \tag{7}$$

By combining these three moments it is possible to compute three mean diameters of the size distribution and the geometric standard deviation. Validation of this approach is discussed in Cleary, Weinert and Mulholland.[7] These statistics provide a strong basis for design of spacecraft smoke detectors.

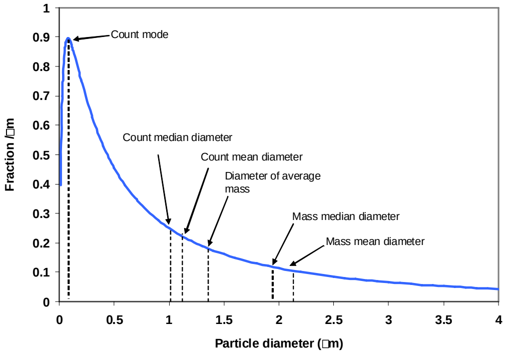

Figure 1: Diameters for a log-normal distribution with D_g=1.0 and σ_g= 1.6.

B. Instruments

These measurements were made using an assembly of three separate instruments. Two are industrial hygiene instruments manufactured by TSI and one is a modified residential smoke detector.

The zeroth-moment instrument is a condensation nuclei counter P-Trak™ (TSI Inc.). This device operates by passing the aerosol-laden particle stream through a region saturated with isopropanol vapor and then into a cooler region where the vapor condenses onto the particles increasing their diameter such that they can be readily counted by a light scattering device. This instrument is very robust and operates over a range of 0 to 10^5 particles/cm^3 and 20 nm to 1 µm diameter. Some dilution is required, since the smoke concentration ranges from about 0.5 × 10^6 to 5 × 10^6 particles / cm^3 There was also a concern that the isopropanol condensate would not return to the wick in low-gravity.[5] To mitigate this issue, the condensing section of the device was modified with very small grooves to improve conductance of the condensate back to the wick. These changes were tested in a separate space experiment with good results indicating the modified device could be used successfully in low gravity.[5]

The first-moment instrument is the ionization chamber from a residential smoke detector. This device uses an alpha-particle emitter to generate ions in a region within a DC electric field. The drift of the ions in the electric field results in a current. The presence of particulate reduces the current as a result of the attachment of the ions to the particulate. The mobility of the charged aerosol is too small for it to be collected on the ionization chamber electrode. The required particle concentrations are on the order of 10^5 particles/cm^3 and no sample dilution was required.

The third-moment instrument is a light scattering device DustTrak™ (TSI Inc.). The device uses a 90 degree light scattering signal to quantify the aerosol mass density. For terrestrial dust particulate this signal correlates well with the mass concentration, however additional compensation will be needed to account for the range of particle sizes that will be seen in the SAME experiment. The device's operating range is from 0.001 mg/m^3 to 100 mg /m^3. These devices are equipped with an aerodynamic impactor at the inlet which captures particles larger than the selected size. The SAME experiment included 2 DustTraks™ one with a 1 µm impactor and one with a 10 µm impactor. The difference in the signal from these two devices provided a measure of the fraction of the particulate that was larger than 1 µm. In some cases dilution was required owing to the high smoke concentration levels.

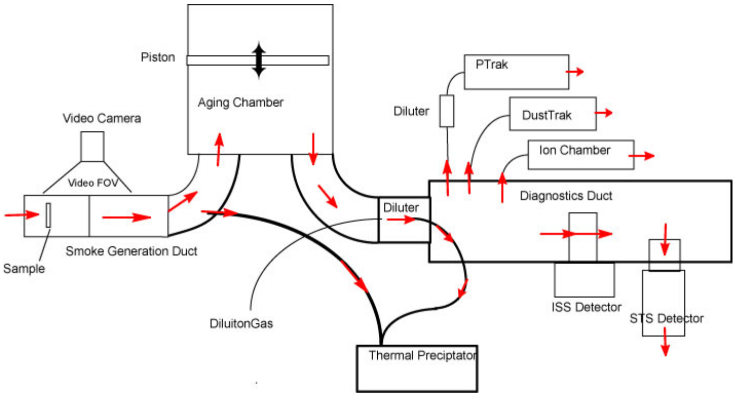

Figure 2. Schematic of the SAME hardware**.**

A schematic of the assembled hardware appears in Fig. 2. The system was installed in the Microgravity Science Glovebox, an ISS facility that provides many resources including: containment, power, data, video and uplink commanding. Smoke was generated by overheating a small sample of material in the smoke generation duct for approximately 60 seconds. During this interval, controlled flow was induced by a moving piston in the aging chamber which drew the smoke into the chamber. The smoke was held in the chamber for a predetermined time, allowing the particles to coagulate. After a specified aging time, the smoke was then pushed by the piston into the diagnostics duct where the moment instruments made their measurements. Also installed in the diagnostics duct were space shuttle and ISS smoke detectors. As the smoke was monitored by the moment instruments, a sample of the smoke particles was deposited on Transmission Electron Microscope (TEM) grid via a thermal precipitator which uses thermophoresis to deposit the particles on the grids.

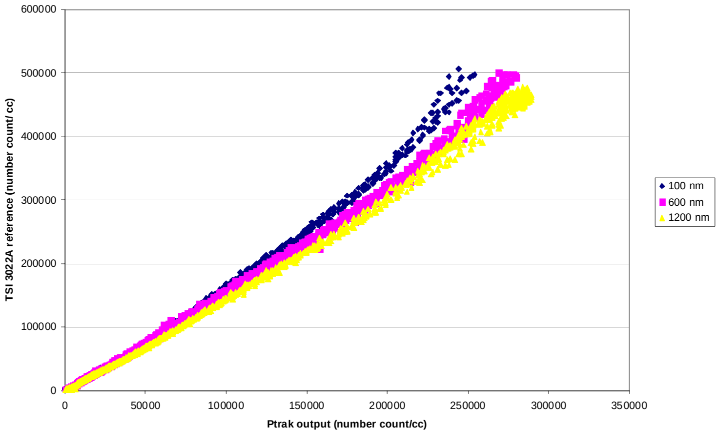

Figure 3: Ptrak Calibration results with Mono disperse DOP droplets.

Calibration of these instruments was essential to properly interpret the flight data and was performed on the ground before the flight. Calibration was accomplished using two different aerosol generators one using mono-disperse particle generation using dioctyl phthalate (DOP) according to the approach by Mulholland and Liu[10] and the other using polystyrene spheres. The monodisperse droplet generator functioned by producing a spray of DOP diluted with isopropanol which is then evaporated and recondensed producing monodisperse droplets. The droplet size is controlled by the DOP dilution level. The generator will operate stably for tens of minutes. The aerosol from the generator was sampled simultaneously by the SAME instrument under test and a reference standard. For the number count, the reference instrument was a TSI 3022 particle counter, for the Mass Concentration, a Tapered Element Oscillating Microbalance and for the first moment a TSI Electrical Aerosol Detector was used. The results for the P-Trak are shown in Fig. 3. As the number concentration increased, the effect of the particle diameter became more evident. Separate correlations were developed for each particle size and the closest correlation was used to analyze the flight data based on the initial estimates of the average particle size.

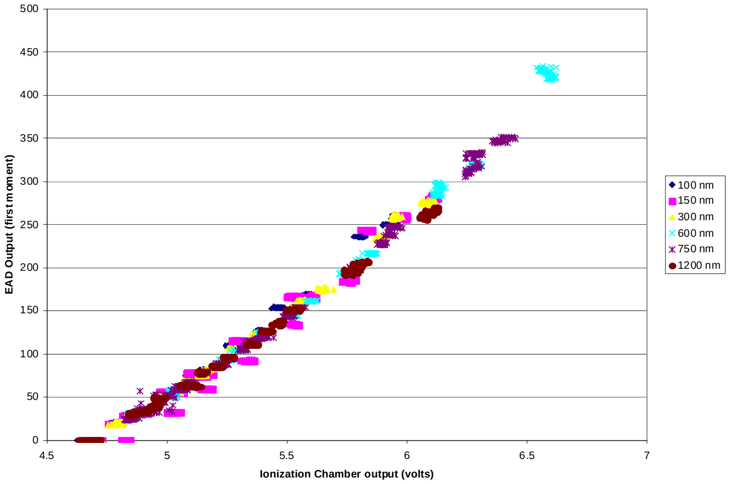

Figure 4: Ion Chamber Calibration results with Mono disperse DOP droplets.

The first moment device, the ion chamber showed little effect of particle size as seen in Fig 4. Consequently a single correlation was used for all particle sizes.

The third moment device (TSI-Dust Trak) is theoretically predicted to show non-monotonic behavior as particle size is increased. The calibration results for the DustTrak shown in Fig. 5 was obtained using monodisperse DOP particles and indicates a strong dependence on particle size. This dependence becomes even more complicated when the particle size distribution is polydisperse.

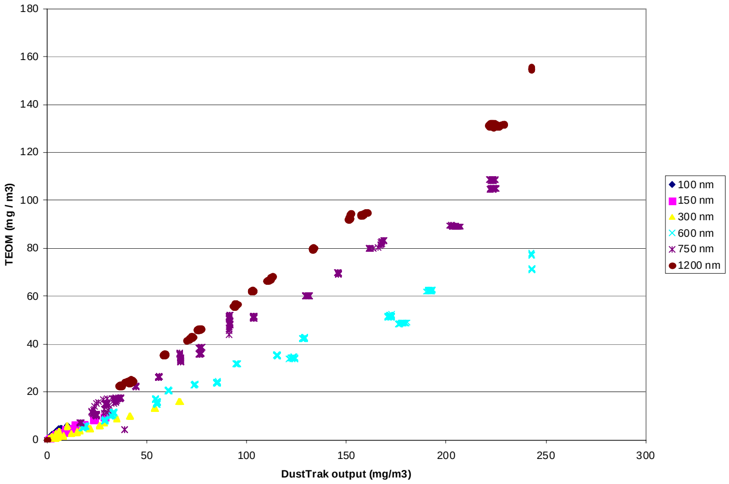

Figure 5: Dust Trak results with Mono disperse DOP droplets.

To address this calibration while minimizing the assumptions required about the particle size distribution, a model of the operation of the DustTrak instrument was developed using Mie scattering theory. Using the zeroth and first moment data along with the refractive index of the particulate, the third moment was predicted by assuming a log-normal distribution and making initial guesses for the geometric mean diameter and the geometric standard deviation. The output of the DustTrak was then predicted by the scattering model and the model proceeded in an iterative manner until the predicted DustTrak output matched the actual output. The geometric mean diameter and geometric standard deviation were those that produced convergence of the predicted and measured DustTrak output. In some cases, convergence was not obtained and for these cases, the geometric mean diameter can not be determined. This non-convergence is currently under investigation but may be due to the presence of a bimodal particle size distribution.

C. Experimental Results

Overall 30 sample materials were tested. These were comprised of six samples each of 5 materials: Teflon™, Kapton™, silicone rubber, cellulose (lamp wick), and dibutyl-phthalate deposited on a porous wick. The TEM girds recovered from the thermal precipitators were unfortunately contaminated with extraneous particles whose origin is currently under investigation. Despite the contamination, the lamp wick and Teflon samples produced distinct particles consistent with ground based experience. Typical particles are shown in Fig. 6 Geometric Mean, Arithmetic Mean, and Average Mass diameter results from the moment instruments for baseline runs are presented in Table 1.

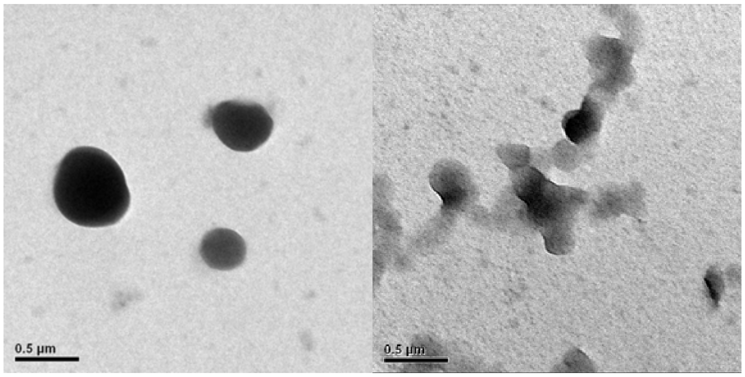

Figure 6: Test point 32 Lamp wick Test point 26, Teflon

	Geometric Mean Diameter (micro meters)	Arithmetic Mean Diameter (micro meters)	Diameter of Average Mass (micro meters)
Kapton	0.080	0.105	0.172
Lamp wick	0.186	0.227	0.420
Silicone	0.189	N/A	N/A
Teflon	0.159	0.205	0.219

Table 1:Diameter results from baseline runs. The Silicone results did not converge for these conditions

Overall, the Teflon and Kapton particles were very small with limited increase in the size from the diameter of average mass compared to the arithmetic mean diameter. The Lamp wick results exhibited substantially larger diameter of average mass. As reported previously,[11] the Dust Trak results for Lamp wick and Silicone demonstrated substantial portions of the particle distribution possessed aerodynamic diameters larger than 1 micrometer. The small diameters for the Kapton and Teflon will make detection of this smoke challenging for light scattering devices, on the other hand the large sizes seen with the Lamp wick and Silicone would generate very large signals on a light scattering system, suggesting that detection of these particles against the background environment will require a detection system capable of measuring more than one moment of the particle size distribution.

III. Acknowledgments

The support of the SAME experiment team and the crews of ISS increments 10, 13, and 15 are gratefully acknowledged. The SAME project was conducted through the ISS Exploration Research Project of the Exploration Technology Development Program. The support of this program is also acknowledged.

Certain commercial entities, equipment, or materials may be identified in this document in order to describe an experimental procedure or concept adequately. Such identification is not intended to imply recommendation or endorsement by the National Aeronautics and Space Administration or the National Institute of Standards and Technology, nor is it intended to imply that the entities, materials, or equipment are necessarily the best available for the purpose.

IV. References

[1] Bukowski, R.W. and G.W. Mulholland, “Smoke Detector Design and Smoke Properties,” NBS Technical Note 973, 1978.

[2] Bukowski, R. W, R. D. Peacock, J. D. Averill, T. G. Cleary, N. P. Bryner, W. D. Walton, P. A. Reneke, and E. D. Kuligowski, “Performance of Home Smoke Alarms, Analysis of the Response of Several Available Technologies in Residential Fire Settings,” NIST Technical Note 1455, December 2003.

[3] Brooker, J. E., Urban, D.L., Ruff, G.A., “ISS Destiny Laboratory Smoke Detection Model”, 07ICES-113, *2007 International Conference on Environmental Systems*, Chicago, Ill. July 2007.

[4] Friedman, R., “Fire Safety Practices and Needs in Human-Crew Spacecraft,” *Journal of Applied Fire Science*, Vol. 2, 1992, pp. 243-259.

[5] Urban, D.L., Griffin D., Ruff, G.A., Cleary, T., Yang, J., Mulholland, G., Yuan, Z.G., “Detection of Smoke from Microgravity Fires”, *2005 International Conference on Environmental Systems,* Rome Italy, Paper # 2005-01-2930 July 2005, SAE Transactions, pp 375-384.

[6] Ruff, G.A., D.L. Urban, and M.K. King, “A Research Plan for Fire Prevention Detection and Suppression,” AIAA-2005-0341, *43rd Aerospace Sciences Meeting and Exhibit*, Reno, NV, January 9-12, 2005.

[7] Cleary, T.G., D W. Weinert, and G.W. Mulholland, ”Moment Method for Obtaining Particle Size Measures of Test Smokes", Natl. Inst. Stand. Technol., NISTIR 7050, 2003.

[8] Raabe, O.G., *J. Aerosol Sci.*, Vol. 2, 1971, p. 289.

[9] Reist, P.C., *Introduction to Aerosol Science*, Macmillan Pub. Co., NY, NY, 1984.

[10] Mulholland, G.W. and Liu, B.Y.H., 1980. Response of Smoke Detectors to Monodisperse Aerosols, *J. Research of the National Bureau of Standards*, 85:223-238.

[11] Urban, D.L. Ruff, G.A., Mulholland, G.W., Cleary, T.G., Yang, J.C., Yuan, Z.G., “Measurement of Smoke Particle Size under Low-Gravity Conditions,” *2008 International Conference on Environmental Systems,* Chicago, IL, Paper 2008-01-2089.