DESIGN OF AEROSOL SAMPLER TO REMOVE RADON AND THORON PROGENY INTERFERENCE FROM AEROSOL SAMPLES FOR NUCLEAR EXPLOSION MONITORING

S. R. Biegalski and O. A. Ezekoye

The University of Texas at Austin

Sponsored by Army Space and Missile Defense Command

Contract No. W9113M-05-1-0016

ABSTRACT

An aerosol sampler is being designed to physically separate aerosols containing radioactivity from natural origin from aerosols containing radioactivity produced in a nuclear weapons explosion. Studies show that aerosols with natural activity have an aerodynamic diameter in the range of 0.1 to 1 μm. In contrast, atmospheric nuclear explosions produce radioactive aerosols with aerodynamic diameters less than 0.1 μm. Surface nuclear explosions produce a bimodal distribution of radioactive aerosol particles. The first group of particles in a surface explosion is produced by spontaneous nucleation and is very similar in distribution to that of the atmospheric explosion. These particles do not combine with material from the ground entrained within the explosion and have aerodynamic diameters less than 0.1 μm. The second group of particles from a surface explosion contains admixed particles entrained in the explosion and has particles with aerodynamic diameters greater than 1 μm. These differences in aerosol sizes are quite fortuitous because they allow aerosol aerodynamic diameter to be utilized as a physical property to separate aerosols of natural origin from those produced in a nuclear explosion.

The benefits of this aerosol sampler are clear for nuclear explosion monitoring. Since aerosols with natural radioactivity are the primary contributors to background in gamma-ray spectroscopy of the aerosol filters, separation of the natural radioactivity from the aerosol samples will result in detection limit improvements. Not only will the background be reduced, but the air filters will not require significant decay times between collection and the start of the gamma-ray spectrum acquisition. For locations where radon and thoron levels are high, these detection limit improvements will be significant. In addition, the aerosol size provides information that may be utilized to distinguish between a surface explosion and an atmospheric explosion. This information is not available from the aerosol samplers currently in use for nuclear explosion monitoring. The capabilities of the U.S. nuclear explosion monitoring system will be significantly enhanced with this aerosol sampler.

OBJECTIVE(S)

Background

The United States has maintained an interest in nuclear explosion monitoring since the Trinity test shot at Alamagordo, New Mexico on July 16, 1945. In August 1948, the U.S. Air Force created the Office of Atomic Energy-1 (AFOAT-1), giving it responsibility for managing the Atomic Energy Detection System (AEDS) discovering foreign atomic tests and other nuclear-weapons related activities (Ziegler and Jacobson, 1995). The AFOAT-1 would later be renamed the Air Force Technical Applications Center (AFTAC). The initial monitoring was based off of atmospheric aerosol collection from airplanes during transoceanic flights. The first USSR nuclear test conducted at the Semipalatinsk nuclear test site on August 29, 1949, and was subsequently observed by the United States through this atmospheric monitoring (Rhodes, 1995; Ziegler and Jacobson, 1995).

These first nuclear explosion tests were easily observed by aerosol collection. The aerosols were measured on the on the opposite side of the world for up to a month after the explosion (Perkins et al., 1995). Nuclear explosion monitoring became more difficult after the Limited Test Ban Treaty (LTBT) in 1963. The LTBT prohibited atmospheric testing by the signatories and nuclear weapons testing consequently moved underground. While the underground testing did contain significant portions of the fission products, many tests vented and released substantial radioactive material. Nuclear explosion monitoring focused on new technologies to monitor the underground tests. The aerosol collection stations were upgraded to enhance detection limits. Noble gases (e.g., Xe) have a higher probability for release from underground explosions, so noble gas monitoring was adopted into the nuclear explosion monitoring efforts. Additional focus was also given to seismic monitoring that could precisely determine the time of the event, accurately determine the location of the explosion, and estimate the yield.

The United States currently maintains a program to look for nuclear explosions (Welch, 1997). Atmospheric radionuclide monitoring is one technology among the many that are used. The atmospheric monitoring efforts consist of aerosol monitoring and noble gas monitoring. This proposal will focus on a way to improve the detection capability of the aerosol monitoring stations.

Aerosol Collection and Measurement

Current aerosol monitoring stations are stationary and work by filtering a large volume of air at flow rates between 500 and 1000 m^3 hr^{-1}. A nominal collection methodology is to collect the air filter for 24 hours, allow the radionuclides on the filter paper to decay for 24 hours, and then acquire a gamma-ray spectrum of the air filter is for 24 hours. The total turnaround time for a sample collection, decay, and gamma-ray spectrum acquisition is 72 hours.

Natural background radiation is one of the main factors that inhibit the detection of nuclear weapons debris through aerosol monitoring. Figure 1 shows an example of an aerosol filter gamma-ray spectrum exhibiting signal from just natural radionuclides. The presence of these natural radionuclides in the gamma-ray spectrum raises the Compton continuum and decreases the detection capability for radionuclides resulting from nuclear weapons tests.

The main contributors to the natural radiation spectrum are thoron progeny, radon progeny, ^{40}K (a primordial radionuclide), and ^{7}Be (produced through cosmic-ray interactions in the atmosphere). Specifically, ^{208}Tl, ^{212}Bi, and ^{214}Bi cause the majority of the Compton continuum in the rage most fission products are expected to be measured (300 to 1300 keV). Thus, the concentrations of these radionuclides in an aerosol sample directly affect the detection limits for fission products. In the thoron decay series, ^{212}Pb is the longest lived progeny with a half-life of 10.6 hours. In the radon decay series, ^{214}Pb is the progeny to be noted with a half-life of 26.8 minutes (Willeke and Baron, 1993). In order to decrease the Compton continuum contributions from thoron and radon progeny, air filters are normally allowed to decay for up to 24 hours. While ^{210}Pb has a half-life of 22 years, it does not contribute significant Compton continuum interference due to its low energy gamma-ray.

The relationship between detection limit and the concentration of thoron progeny is shown in Figure 2. The figure shows the relationship between ^{140}Ba minimum detectable concentration (MDC) as calculated by the Currie method (Currie, 1968) and ^{212}Pb concentration. The concentrations of ^{212}Pb, ^{212}Bi, ^{212}Po, and ^{208}Tl reach a state of equilibrium during the decay period after sample collection. Therefore, the relationship shown in Figure 2 represents

the combined effect of thoron progeny on ^{140}Ba MDC. Likewise, the MDC for ^{140}Ba (main gamma-ray energy at 537 keV) should be considered illustrative of other radionuclides that have gamma-rays in the same energy region.

Figure 2 shows ^{212}Pb concentrations ranging up to 0.02 Bq m^{-3}. These concentrations are actually quite moderate in comparison to regions of the world that have high thoron concentrations. For example, ^{212}Pb atmospheric aerosol concentrations in Europe range up to 0.1 Bq m^{-3} (Gäggeler, 1995). This is a factor of five higher than what was observed in Charlottesville, Virginia during the measurement period from November 18, 2000 to January 21, 2001. For the range shown in Figure 2, the relationship between ^{140}Ba MDC and thoron concentration appears linear. However, the relationship is actually a function of the square root of the Compton continuum at the point of the ^{140}Ba primary gamma-ray line. Therefore, extrapolation of this relationship to higher ^{212}Pb concentrations is difficult. The conclusion should be that the adverse effect of local thoron concentrations can be much worse than what is shown with the data from Charlottesville, VA.

Detection limit improvements may also be seen if the sample decay time is shortened. The 24 hour decay time is currently needed in the sampling methodology to allow for decay of the short-lived radon and thoron progeny. This consequently reduces the magnitude of the Compton continuum. While this is advantageous for detection of radionuclides with half-lives longer than a few days, the detection of shorter-lived radionuclides is depreciated by the 24 hour decay time. Many radionuclides of interest to nuclear explosion monitoring fall into the category of having a half-live less than 24 hours (e.g., ^{91}Sr, ^{93}Y, ^{97}Zr, ^{99m}Tc, ^{133}I, ^{135}I, ^{156}Sm and ^{157}Eu). By removing the natural radioactivity from the aerosol sample, the gamma-ray spectrum acquisition may start with no need for decay time after the end of the sample collection. In fact, gamma-ray spectrum acquisition could be conducted during aerosol collection without significant interference from natural radioactivity. A reduction in decay time would result in a detection limit improvement for nearly all radionuclides of interest for nuclear explosion monitoring.

The MDC for a given radionuclide may generally be calculated as shown in equation 1 (Biegalski and Biegalski, 2001). This formula assumes that the radionuclide is not a daughter involved in a complex decay chain.

$$MDC = \frac{L_D \lambda^2}{\varepsilon I_\gamma F \left(1 - e^{-\lambda t_c}\right)\left(e^{-\lambda t_d}\right)\left(1 - e^{-\lambda t_a}\right)} \qquad (1)$$

where

L_D is the detection limit
ε is the detector efficiency
I_γ is the radionuclide gamma-ray yield
λ is the radionuclide decay constant
t_c is the aerosol collection time
t_d is the sample decay time
t_a is the gamma-ray spectrum acquisition time

This formula may be utilized to calculate the MDC achieved by shortening the decay time. Table 1 shows the factor improvements expected from a reduction in decay time from 24 hours to 0, 1, and 4 hours. These improvements are directly related to the radionuclide half-life. The shortest-lived radionuclide in this example, ^{99m}Tc, would receive more than a factor of 15 improvement in MDC with a reduction in decay time from 24 hours to 0 hours. These factors assume that all other factors in equation 1 remain constant for the comparison. Due to the time-dependent dynamics of the Compton continuum structure these values should be referred to as only rough estimates.

The combination of reduced Compton continuum and reduced decay times will produce significant MDC improvements for the detection of aerosols containing nuclear explosion debris. Reduction in MDC by a factor of 3 may be expected for high radon and thoron days at locations like Charlottesville, VA. Far greater improvements may be expected for sampling locations with much higher radon and thoron concentrations. Reduction in decay time will also result in improvements for radionuclides with half-lives on the order of one day or less. Depending on the radionuclide and the decay time, this improvement will reduce the MDC by a factor ranging between 2 and 15. The combination of these effects could easily result in a factor of six improvement in MDCs for various radionuclides of interest. Factors above ten would be realistic for station locations that have high radon and thoron levels.

Aerosol Size of Natural Radioactive Aerosols and Radioactive Aerosols from Nuclear Explosions

In order to improve the detection capability for ground-based aerosol collection systems, it is advantageous to physically separate the natural radioactive aerosols from the radioactive aerosols produced in a nuclear explosion. Figure 3 is an illustration of the size range that these aerosol particles may be categorized in. Separation of the natural radioactive aerosol particles by size will allow for significant improvements to be made in ground based aerosol detection system MDCs for nuclear explosion debris.

Work published by Grundela and Porstend (2004) show that the majority of aerosol particles containing thoron progeny lie in the size range between 0.1 μm and 1.0 μm. The measurements were made with an on-line alpha cascade impactor for many natural radionuclides. The results for ^{212}Po and ^{218}Po are very similar and their consistency illustrates that all thoron progeny likely fall within this aerosol size range. While these data are specific to the aerosols collected in Göttingen, Germany, the results are similar to those found in other studies for thoron progeny (Bondietti et al., 1987).

Storebø (1974) published an extensive study on the aerosol size distributions of debris from nuclear explosions. For an atmospheric explosion, nearly all the aerosol particles fall below the 0.1 μm mark for cases where the vapor mixing ratio is 10^{-4} or less. For surface explosions, the particles are generated in a bimodal distribution. The first group of particles in a surface explosion is produced by spontaneous nucleation and is very similar in distribution to that of the atmospheric explosion. These particles do not combine with material from the ground entrained within the explosion and have aerodynamic diameters less than 0.1 μm. The second group of particles from a surface explosion contains admixed particles entrained in the explosion and has particles with aerodynamic diameters greater than 1 μm. An underground nuclear explosion may be categorized as a surface explosion with a high initial surrounding concentration. The data provided in the Storebø (1974) publication is consistent with aerosols collected in Sweden from the September 26, 1976 Chinese nuclear explosion (De Geer et al., 1978), an above-ground test performed at Lop Nor in the range of 20-200 kt.

Water surface and underwater bursts were not included in the study by Storebø (1974). Glasstone and Dolon (1977) state that the particles entering the atmosphere from a sea burst consist mainly of salts and water drops. When dry, these particles are very small and light. As a result, it may be assumed that a water surface or underwater burst will produce aerosol particles with size similar to those produced in an atmospheric explosion. However, during transport the hygroscopic nature of the seal salt particles may cause the particles to grow if they enter a region with high humidity.

RESEARCH ACCOMPLISHED

Research has been initiated to design an aerosol sampler that will selectively remove aerosols in the 0.1 to 1 μm range. Many different designs exist for aerosol samples to separate a particle by size. These designs are largely based off the inertia of the particles as they are pulled through the sampler. In a curved flow field inertia makes the particle trajectories deviate from the flow streamlines (Willeke and Baron, 1993). The particles that deviate from the flow streamlines are then removed from the air flow and consequently collected. The two main designs used for aerosol separation by size are the cyclone design and the impactor design. In a cyclone, particles are removed from the air flow by the centrifugal force. In general, a cyclone does not yield as sharp a cutoff in particle size as an impactor (Willeke and Baron, 1993). For this reason, the impactor design if preferred for this project.

By combining several single-stage impactors in a series, a multistage impactor may be constructed. Each stage within the impactor would collect particles with a specific inertial range. The first stage would collect the particles with the most inertia (largest particles) and subsequent stages would collect smaller particles. This is achieved by decreasing the nozzle diameter in each successive stage.

Figure 4 shows how the aerosol sampler will collect aerosol particles in three separate stages. The first stage will contain the largest aerosol particles with an aerodynamic diameter greater than 1 μm (second group of particles from surface nuclear explosions). The second stage will collect aerosol particles with an aerodynamic diameter between 0.1 μm and 1 μm (aerosols of natural origin). The third stage will collect aerosol particles with an aerodynamic diameter less than 0.1 μm (aerosols from atmospheric nuclear explosions and the first group of particles produced in

a surface explosion). To increase the volumetric flow through the sampler, a grid of cascade impactors as shown in Figure 4 will likely be utilized in the prototype sampler.

This sampler will result in the majority of the natural radioactivity being deposited on the Stage 2 filter. The Stage 1 and Stage 3 filters will contain the aerosols in the size region produced during nuclear explosions. A number of scenarios are possible for gamma-ray spectrum acquisition. One scenario would be to acquire a gamma-ray spectrum for the Stage 1 and Stage 3 filters with no decay time after collection. A second gamma-ray spectrum of the Stage 1, 2, and 3 filters could be collected after 24 hours of decay. Experiments will have to be conducted to determine the optimum methodology, but it should be clear that all aerosol material is collected with this sampler and will be available for subsequent analysis. This sampler will in no situation reduce the detection capabilities for aerosols from nuclear explosions.

The flow field for these computations will be found from using a commercial computational fluid dynamics (CFD) package (Fluent 6.1). The CFD solver has been benchmarked to solve transport equations in complex geometries. Our most recent study used this software tool to investigate aerosol transport in a smoke detector geometry. The user defined function (UDFs) for the smoke detector analysis will be modified to analyze the impactor design in the proposed project. The CFD simulations will be used to determine both the streamline behavior through the impactor plates as well as to determine how close to the theoretical value of $\sqrt{St_{50}} \approx 0.49$ our system can be designed.

CONCLUSIONS AND RECOMMENDATIONS

Within the next year, The University of Texas at Austin will complete the design and construction of this prototype aerosol sampler. Testing will be conducted to verify the aerosol cut-off diameters achieved by the sampler. Environmental sampling with confirm the size range of the natural aerosols. Once the prototype sampler has been fully tested, an effort will be placed to design a modification to allow size separation in the aerosol samplers currently deployed by the U.S. Government for nuclear weapons test monitoring.

REFERENCESS

K. M. F. Biegalski and S. Biegalski (2001), Determining detection limits and minimum detectable concentrations for noble gas detectors utilizing beta-gamma coincidence systems, *J. Radioanal. Nucl. Chem.,* 248 (3), 673.

E. A. Bondietti, C. Papastefanou, and C. Rangarajan (1987), Aerodynamic size associations of natural radioactivity with ambient aerosols, in *Radon and is Decay Products-Occurrence, Properties and Health Effects*, ed. P. K. Hopke. ASC-Symposium Series 3131, 397.

L A. Currie (1968), Limits for Qualitative and Quantitative Detection, *Anal. Chem.* 40: 586.

Lars-Erik De Geer, Rune Arntsing, Ingemar Vintersved, Jan Sisefsky, Siv Jakobsson, Jan-Ake Engstrom (1978). Particulate Radioactivity, Mainly from nuclear explosions, in air and precipitation in Sweden mid-year 1975 to mid-year 1977, FOA Report, FOA-C—40089

H. W. Gäggeler (1995). Radioactivity in the Atmosphere, *Radiochemica Acta*, 70/71: 345.

S. Glasstone and P. J. Dolan (1977), *TheEffects of Nuclear Weapons, 3rd Edition*, United States Department of Defense and the United States Department of Energy.

M. Grundela and J. Porstend (2004), Differences between the activity size distributions of the different natural radionuclide aerosols in outdoor air, *Atmos. Env.* 38: 3723.

Perkins, Miley, Hensley, Abel (1995). Airborne Radionuclides of Concern and Their Measurement in Monitoring a Comprehesive Test Ban Treaty, PNL-SA-25481.

R. Rhodes (1995), *The Making of the Atomic Bomb*, Simon & Schuster, New York.

P. B. Storebø (1974), Formation of Radioactivity Size Distributions in Nuclear Bomb Debris, *Aerosol Sci.* 5: 557.

M. Welch (1997), AFTAC Celebrates 50 Years of Long Range Detection, *AFTAC Monitor*.

K. Willeke and P. A. Baron (1993), *Aerosol Measurement: Principles Techniques and Applications*, Van Nostrand Reinhold, New York.

C.A. Ziegler and D. Jacobson (1995), *Spying Without Spies: Origin of America's Secret Nuclear Intelligence Surveillance System*, Praeger.

Table 1. Estimated Improvement in MDC Resulting from Decay Time Reduction (assuming all other factors equal).

Nuclide	Half-life (hr)	Factor Improvement in MDC Resulting from Decay Time Reduction From 24 Hours		
		0 hr Decay	1 hr Decay	4 hr Decay
^{91}Sr	9.63	5.6	5.2	4.2
^{93}Y	10.18	5.1	4.8	3.9
^{97}Zr	16.91	2.7	2.6	2.3
^{99m}Tc	6.01	15.9	14.2	10.0
^{133}I	20.8	2.2	2.2	1.9
^{135}I	6.57	12.6	11.3	8.2
^{156}Sm	9.4	5.9	5.5	4.4
^{157}Eu	15.18	3.0	2.9	2.5

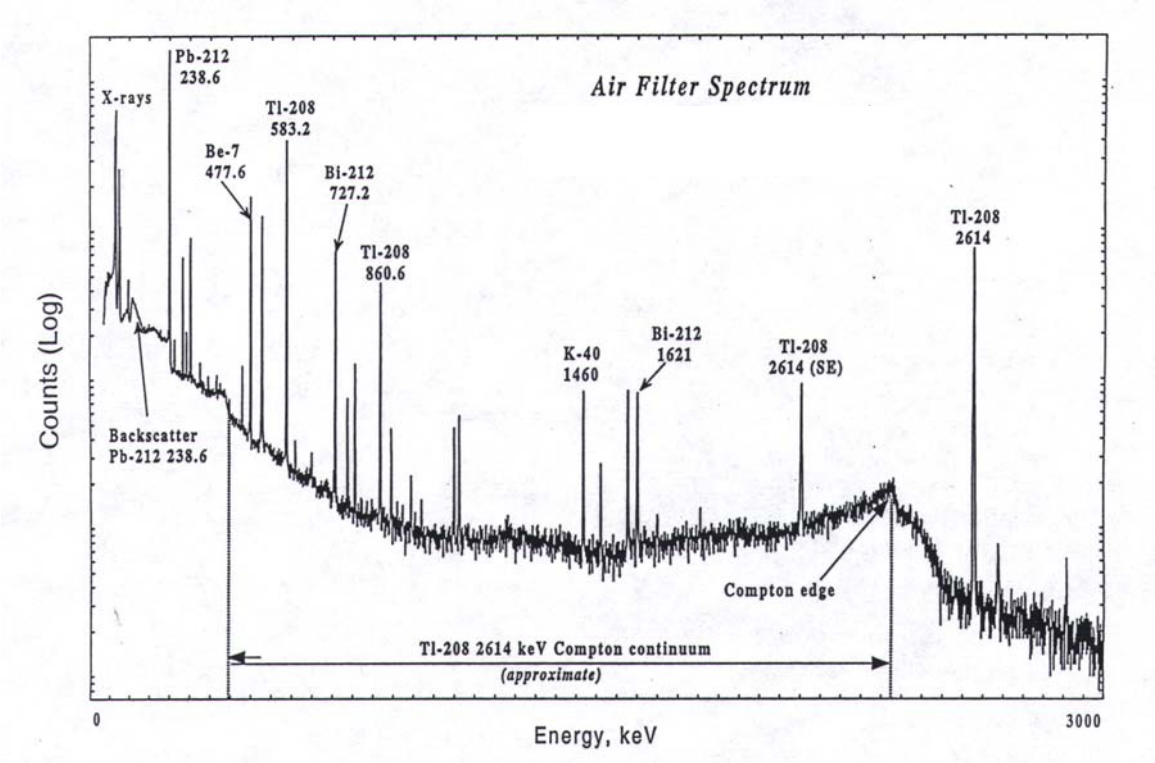

Figure 1. Gamma-ray spectrum of air filter acquired on a HPGe detector (24 hour collection, 24 hour decay, and 24 hour spectrum acquisition).

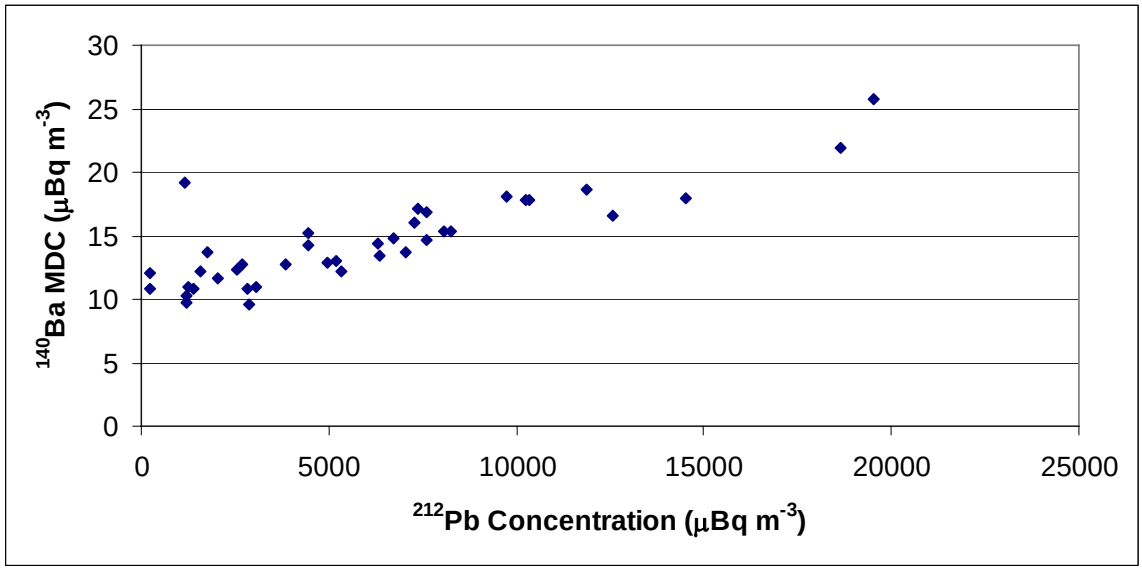

Figure 2. Relationship between ^{140}Ba minimum detection limit and ^{212}Pb concentration for aerosol sampling station in Charlottesville, VA, USA. The collection, decay, and gamma-ray spectrum acquisition periods were 24 hours each. These samples were collected November 18, 2000 to January 21, 2001.

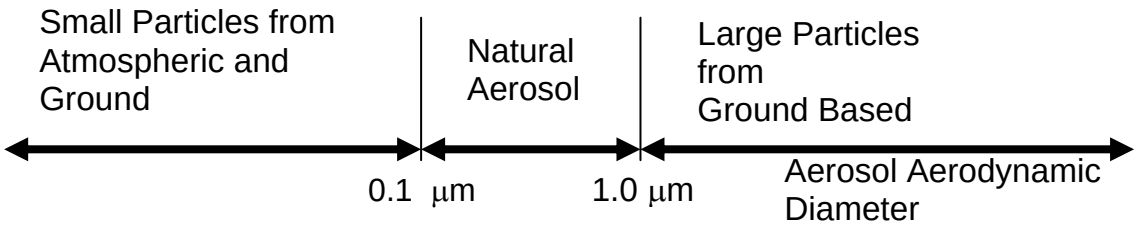

Figure 3. Size range of aerosol particles from natural sources and nuclear explosions.

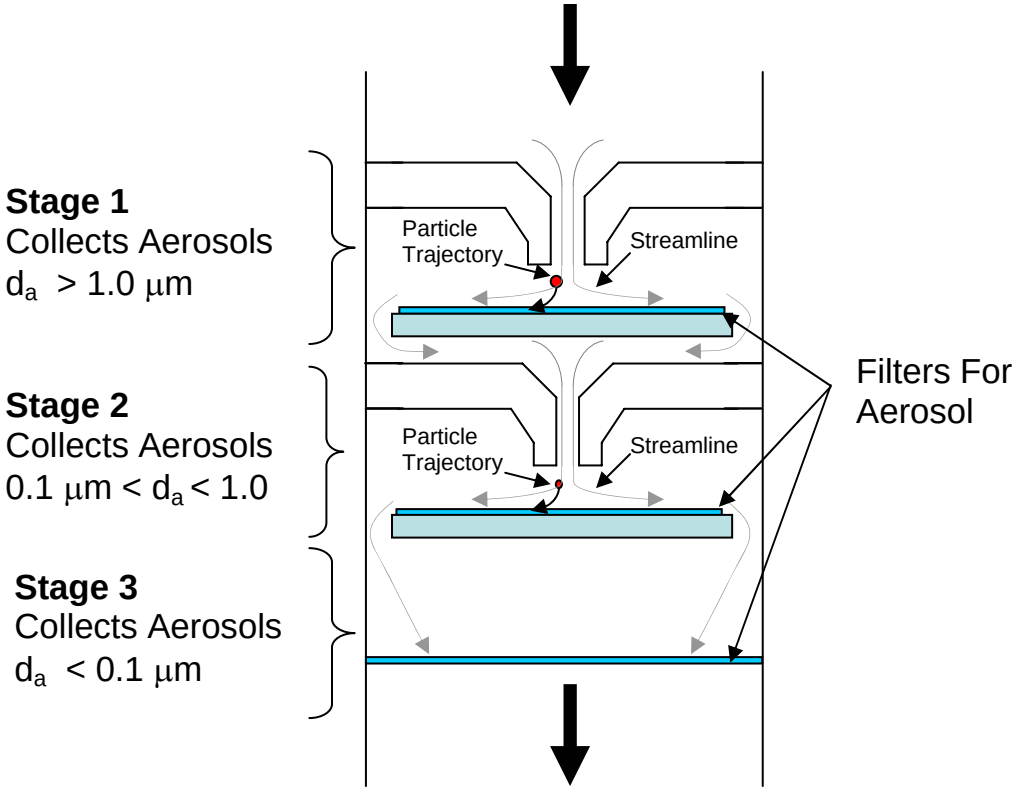

Figure 4. Three stage impactor.