

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

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ABSTRACT

Studies show that aerosols with natural activity have an aerodynamics diameter in the range of 0.1 to 1.0 μm . In contrast, atmospheric nuclear explosions produce radioactive aerosols with aerodynamic diameters of less than 0.1 μm . Surface nuclear explosions produce a bimodal distribution of radioactive aerosol particles with aerodynamic particles greater than 1.0 μm and less than 0.1 μm . A high-volume ($70 \text{ m}^3\text{h}^{-1}$), low-pressure aerosol impactor has been developed that separates the particles into the three size distributions: aerosols with aerodynamic diameters greater than 1.0 μm , between 0.1 and 1.0 μm , and smaller than 0.1 μm .

This sampler has been tested and compared to the performance of a micro-orifice uniform deposit impactor (MOUDI). Testing was completed by measuring natural aerosol samples as well as testing with a submicrometer monodisperse aerosol generation system. Additional pressure taps have been included in the system to improve system characterization. As a result of the testing and measurements, design modifications are being made for system optimization.

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OBJECTIVES

This work seeks to develop an aerosol sampler that physically separates aerosols containing radioactivity from natural origin from aerosols containing radioactivity produced in a nuclear weapons explosion. Aerosols with natural radioactivity have been shown to have a size distribution predominantly between 0.1 and 1.0 μm . Grundela and Porstend (2004) showed this was the case for data collected in Göttingen, Germany for thoron and radon progenies. Similar studies, including Bondiotti, et al. (1987) also found this size range for Thoron progeny. Aerosols with radioactivity from nuclear explosions were found to have a bimodal size distribution depending on the type of detonation. Aerosols were collected after a 20 kt nuclear explosion. For air explosions, the aerosol particles had a diameter of less than 0.1 μm and for surface explosions, the particle size was larger than 1.0 μm (Storebø, 1974). These data were consistent with aerosols collected in Sweden from a September 26, 1976, Chinese 20–200 kt nuclear explosion conducted above-ground (De Geer et al., 1978). It is therefore advantageous to separate aerosol particles into at least three size groups by particle diameter: greater than 1.0 μm , between 0.1 and 1.0 μm , and less than 0.1 μm .

There are many designs of aerosol samplers that separate the particles by size. Inertial impactors, especially cascade impactors, are ideal for this task. Particle inertia in a curved flow makes the particles deviate from the streamline, impacting a collection surface (Willeke and Baron, 1993). By controlling the fluid flow through the geometry of the sampler, particular sizes can be separated. When multiple single-stage impactors are placed in series, particle sizes of decreasing diameter may be collected. One difficulty with the current study is the small particle size of radioactive aerosols from aboveground nuclear explosions (particle size is less than 0.1 μm). Conventional impactors normally only have a particle diameter limit of 0.3–0.4 μm (Vanderpool et al., 1990). Two methods that have been used previously to obtain lower particle diameters are using a MOUDI or using a low-pressure impactor. The nozzles in a MOUDI are difficult to manufacture, especially for the case of a high volume flow since thousands of nozzles are needed for the lower stages. Lowering the pressure in a stage changes the mean free path between fluid particles and allows for smaller diameter particles to be collected. A critical stage is needed to create a pressure drop and a vacuum pump is needed in order to pull the desired pressures.

Once the aerosols containing radioactivity from natural sources are separated from those with radioactivity from nuclear explosions, the samples are analyzed. With the reduced background, gamma-ray spectroscopy of the aerosol filters will result in detection limit improvements. The filters will not need significant decay times between collection and gamma-ray spectrum acquisition. These improvements will be most notable where radon and thoron levels are high. The size distribution will also enable identification of a surface or atmospheric explosion.

The current focus of the study is the design and validation of the previously mentioned aerosol sampler. A generic impactor was modeled using a computational fluid dynamics program, FLUENT 6.1. Both circular and rectangular nozzles were modeled and benchmarked with previous studies. This provided insight into the physics of the fluid flow and particle behavior. By calculating the necessary geometries and pressures needed to obtain the desired aerosol particle sizes, the cascade impactor was manufactured and assembled. In order to verify the size distribution for each stage, fluorescent test aerosol particles of known size were produced and collected by the sampler onto the filters of each stage. The filters were then analyzed for size distribution for design validation.

RESEARCH ACCOMPLISHED

In the previous work, computer fluid modeling of round and rectangular nozzles was benchmarked against previous literature and the designs for critical orifice stages were developed to modify a commercial six-stage impactor (TE-230 high-volume cascade impactor with TE-5000 high-volume air sampling system made by Environmental Tisch). The TE-230 impactor has a variable motor that can vary the volumetric flow rate. At 20 cfm, the cutoff diameters are 10.2, 4.2, 2.1, 1.4, 0.73, 0.41, and 0 μm . At 40 cfm, the cutoff diameters are 7.2, 3.0, 1.5, 0.95, 0.49, and 0 μm (the 6th stage is not used because the pump cannot handle the pressure drop). New plates were machined as mentioned above, and a new pump was obtained that would accommodate the high flow rate and large pressure drop. In order to attain the relatively high flow rate of 66 m^3/h (1100 L/min) with the low pressure of approximately 0.2 atm (absolute) after the last stage, a Rietschle Thomas Zephyr DLR60 dry compression claw compressor with a 5-HP motor was acquired.

To test the effect of the introduction of the critical stages into the Tisch impactor, a submicrometer monodisperse aerosol generator was procured and assembled to produce aerosols containing test particles with various diameters at the following values: 0.051, 0.10, 0.20, 0.92, 1.0, and 2.1 μm . A TSI Model 2076 constant output atomizer, seen in Figure 1, creates submicrometer aerosols from solutions or suspension. Compressed air expands through an orifice, forming a high-velocity jet, as shown in Figure 2a (TSI, 2005). As the liquid is drawn through the atomizing section, the large droplets impact on the wall opposite the jet and drain back. A fine spray leaves the atomizer and is dried by silica gel in an annular tube (TSI Model 3063 diffusion dryer), seen in Figure 2b.

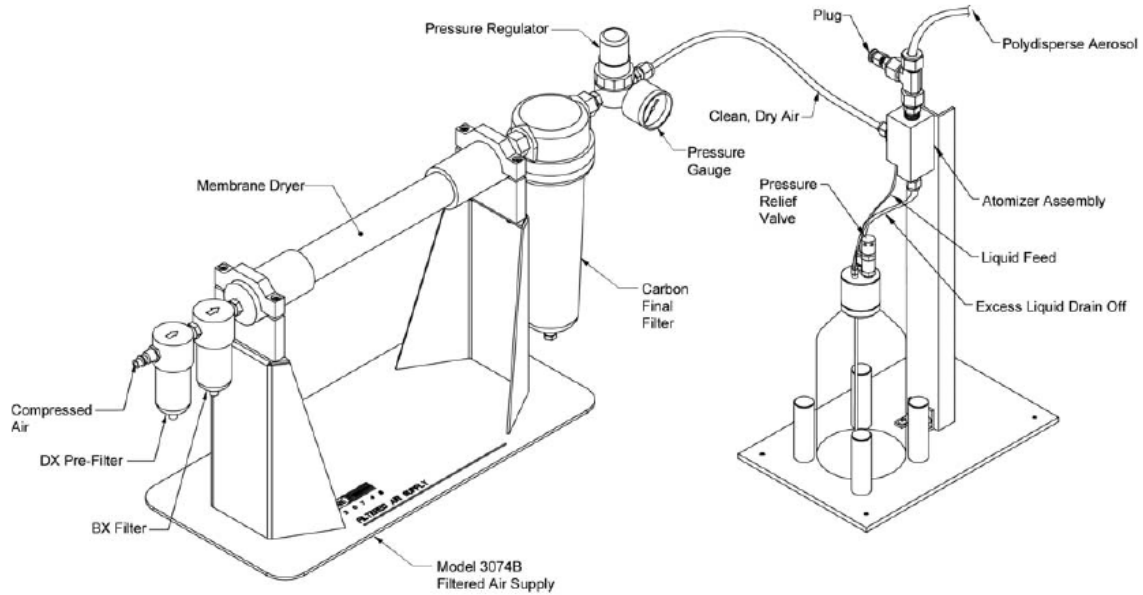


Figure 1. TSI Model 2076 Constant Output Atomizer.

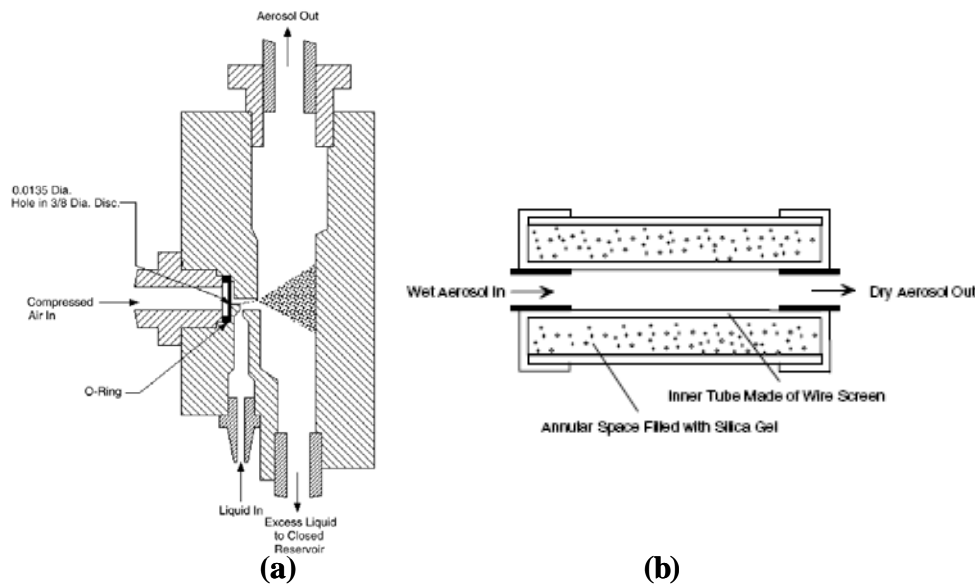


Figure 2. (a) TSI Model 2076 Constant Output Atomizer and (b) TSI Model 3063 Diffusion Dryer.

After the aerosols were conditioned by the diffusion dryer, they were run through a TSI Model 3012A Krypton-85 Neutralizer so that any charge on the particles can be removed. The actual aerosol generation system can be seen fully assembled in Figure 3. To verify that the system was correctly producing the desired aerosols, the assembly was calibrated using the MOUDI, which has twelve stages with cutoff diameters of 18, 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.1, 0.056, and 0 μm . Achieving quantitative results for the collection efficiency was not necessary as the particle collection could be verified visually as being almost completely collected in the appropriate stage, with only minimal collection on the adjacent stages. The time needed for sufficient test particle collection is on the order of 1 to 1.5 hours. The impaction on the adjacent stages is the result of the collection efficiency for each stage being less than 100%, allowing some particles to be collected too early as well as too late. To visually inspect the collection on each stage, the filters were exposed to ultraviolet light which allowed the fluorescent test particles to be easily seen. Each test particle size was verified and all proved to be generated as expected.



Figure 3. Assembled aerosol generation apparatus.

Further tests were conducted with the MOUDI to confirm that the size distribution of naturally occurring radionuclides lies within the diameter range of 0.1 to 1.0 μm (Grundela and Porstend, 2004). For these tests, the MOUDI was set up outside of the UT Nuclear Engineering Teaching Laboratory to collect for approximately four days to achieve a saturation activity for the radionuclides within the Radon and Thoron progeny that are to be investigated. The buildup of the activity on the filters due to one particular isotope will reach saturation at about 5 to 7 half-lives. For example, ^{212}Pb has a half-life of 10.64 hours and would reach saturation after about 3 days. Other isotopes that were analyzed were ^{214}Pb , ^{214}Bi , ^{212}Pb , and ^{208}Tl .

Once the MOUDI collection was stopped, the twelve stages were grouped into three stages that were representative of the three size regimes of interest. Stages that collected particles of size 1.0 μm and higher were referred to as the course region, size 0.1 to 1.0 μm were identified as the fine region and anything below 0.1 μm were referred to as the superfine region. To reduce the decay time for the samples, each of the groups were quickly prepared and their gamma spectrums were simultaneously counted on three Hyper-Pure Germanium detectors for a total of 12 hours each. At the end of the three counts, the spectra were analyzed to determine the initial activity on the filters at the time the MOUDI collection ended.

The various spectra were back-corrected to account for the decay in the time between ending sample collection and starting the gamma-ray spectrum acquisition, t_d , the decay during the spectrum collection, t_a , and losses due to the efficiency, ϵ , of the various detectors at a particular energy, as well as the probability, γ , for a particular gamma ray

to be emitted. This correction can be seen in the following equations that relate the number of counts a detector sees at a particular energy to the initial activity, where λ is the decay constant for the particular isotope:

$$\begin{aligned}
 \text{\# of counts} &= \text{\# of decays} * \varepsilon * \gamma \\
 &= \int_{t_d}^{t_a+t_d} (\text{Activity}) dt * \varepsilon * \gamma \\
 &= A_o \varepsilon \gamma \left[\frac{e^{-\lambda t}}{-\lambda} \right]_{t_d}^{t_a+t_d} \\
 \Rightarrow A_o &= \frac{\text{counts} * \lambda}{\varepsilon \gamma (e^{-\lambda t_d}) (1 - e^{-\lambda t_a})}
 \end{aligned} \tag{1}$$

Once the data had been back-corrected to reflect the changes mentioned above, the initial activity, A_o , of each set of filters was compared to one another for the activity of a particular isotope. The activities were then represented in an activity per unit of volume using Equation 2 to see the size distribution within one cubic meter of air, where Q is the concentration of radionuclides in the air in atoms per m^{-3} and F is the volumetric flowrate in $m^3 s^{-1}$.

$$\begin{aligned}
 Q &= \frac{\text{counts} * \lambda}{F \varepsilon \gamma (e^{-\lambda t_d}) (1 - e^{-\lambda t_a}) (1 - e^{-\lambda t_c})} \\
 \text{where, } A \left(\frac{Bq}{m^3} \right) &= Q \lambda
 \end{aligned} \tag{2}$$

In Table 1, the activity of each group for various isotopes is shown and the coarse group (0.1 to 1.0 μm) can be seen to have the most activity in all isotopes studied in this test. Figure 4 also shows the relative activity of ^{212}Pb found on each set of filters. These results provide further evidence that naturally occurring radionuclides in the radon and thoron progenies are found to be of sizes that can be filtered out to reduce the background activity in an aerosol sample.

Table 1. MOUDI results for naturally occurring activity from 4 day collection.

Isotope ($t_{1/2}$)	Superfine Activity	(+/-)	Fine Activity	(+/-)	Coarse Activity	(+/-)
^{212}Pb (10.64 hrs)	0.095	0.033	0.299	0.028	0.009	0.019
^{214}Pb (26.8 min)	-	-	0.593	1.403	-	-
^{214}Bi (19.9 min)	1.571	2.613	5.312	2.629	1.912	1.647
^{208}Tl (3.053 min)	22.569	43.943	299.777	119.666	-	-

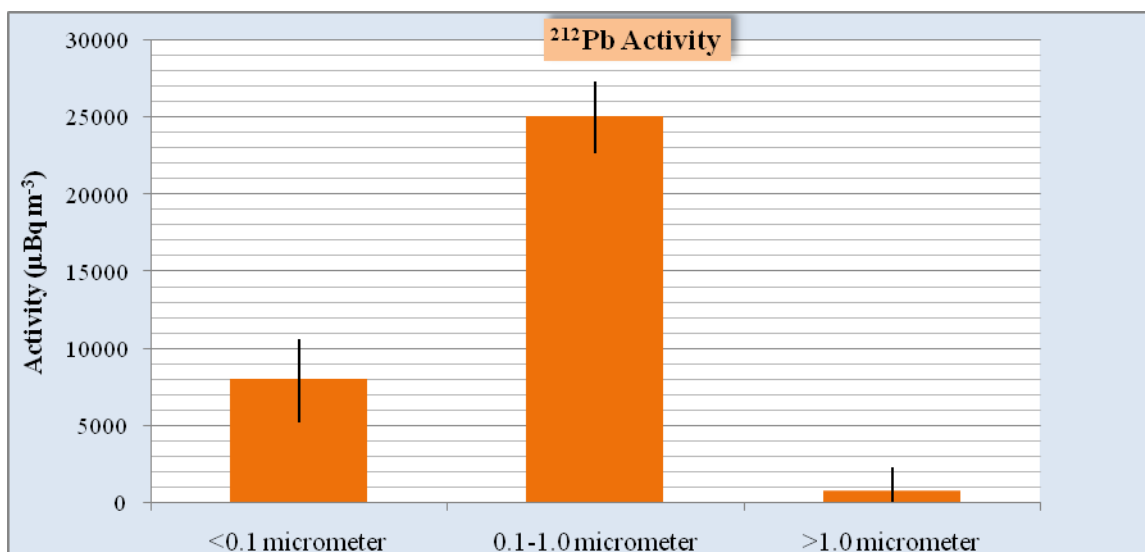


Figure 4. Activity due to Pb-212 after 4 day MOUDI collection.

The MOUDI results were also used to model the detection limits if a radionuclide aerosol sampler analyzer (RASA) configuration were used. The data used to determine the ^{140}Ba concentration from the ^{212}Pb was the USP075-Charlottesville, VA RASA data collected from November 19, 2000 to January 21, 2001. From Figure 5, the detection limits in the regions < 0.1 μm and > 1.0 μm have been greatly reduced using the aerosol separation method.

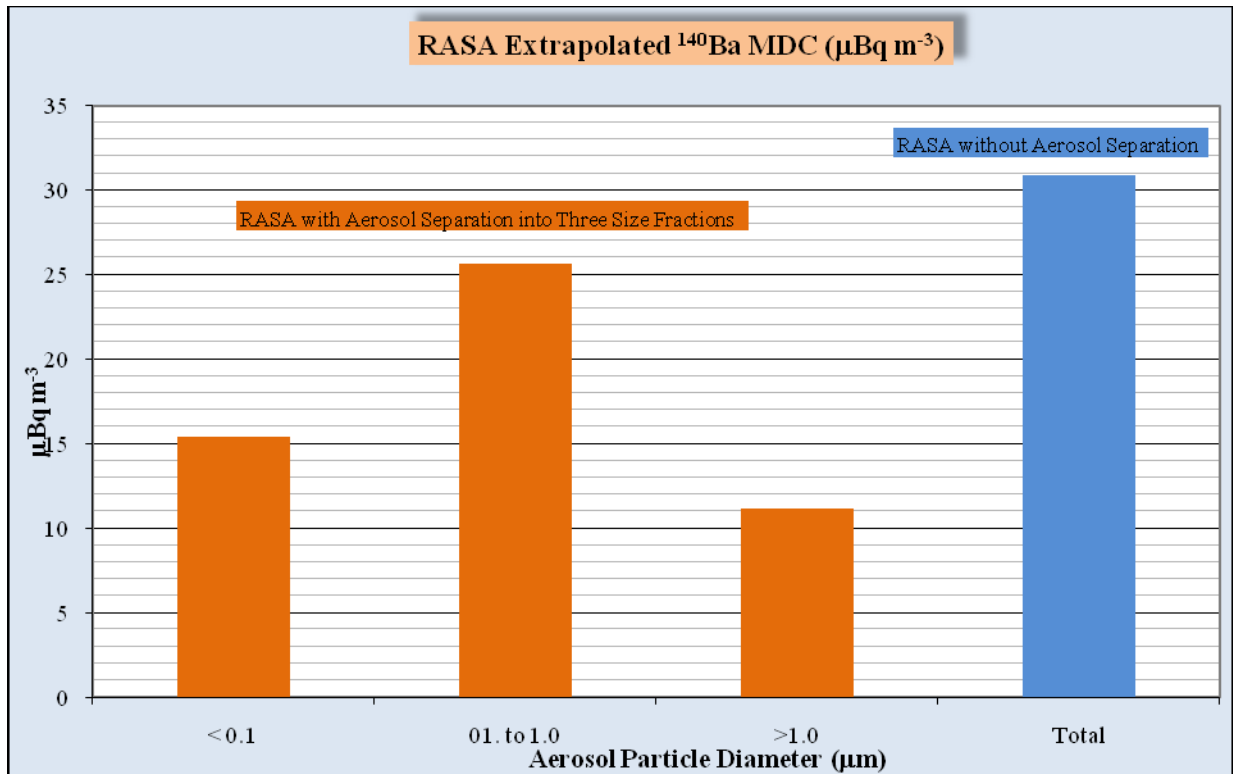


Figure 5. MOUDI data extrapolated to represent the detection limits with RASA using particle separation

To determine the abundance of longer-lived isotopes within the aerosol samples taken by the MOUDI, Neutron Activation Analysis (NAA) was performed on the sets of filters that resulted from a two-day collection outside of the University of Texas Nuclear Engineering Teaching Laboratory. To irradiate the samples and perform the analysis, the Mark II TRIGA Research Reactor was used to expose the samples to a neutron flux and the gamma-ray spectra were collected using the same labs as for the above analysis. The samples were irradiated for 8 hours and allowed to decay for approximately 4 weeks before starting the spectra acquisition. The long decay time was necessary as all of the elements investigated are characterized as relatively long-lived and the decay period reduces the background as well as lowers our detection limits. The results found for the masses of various isotopes are listed and shown in Figure 6.

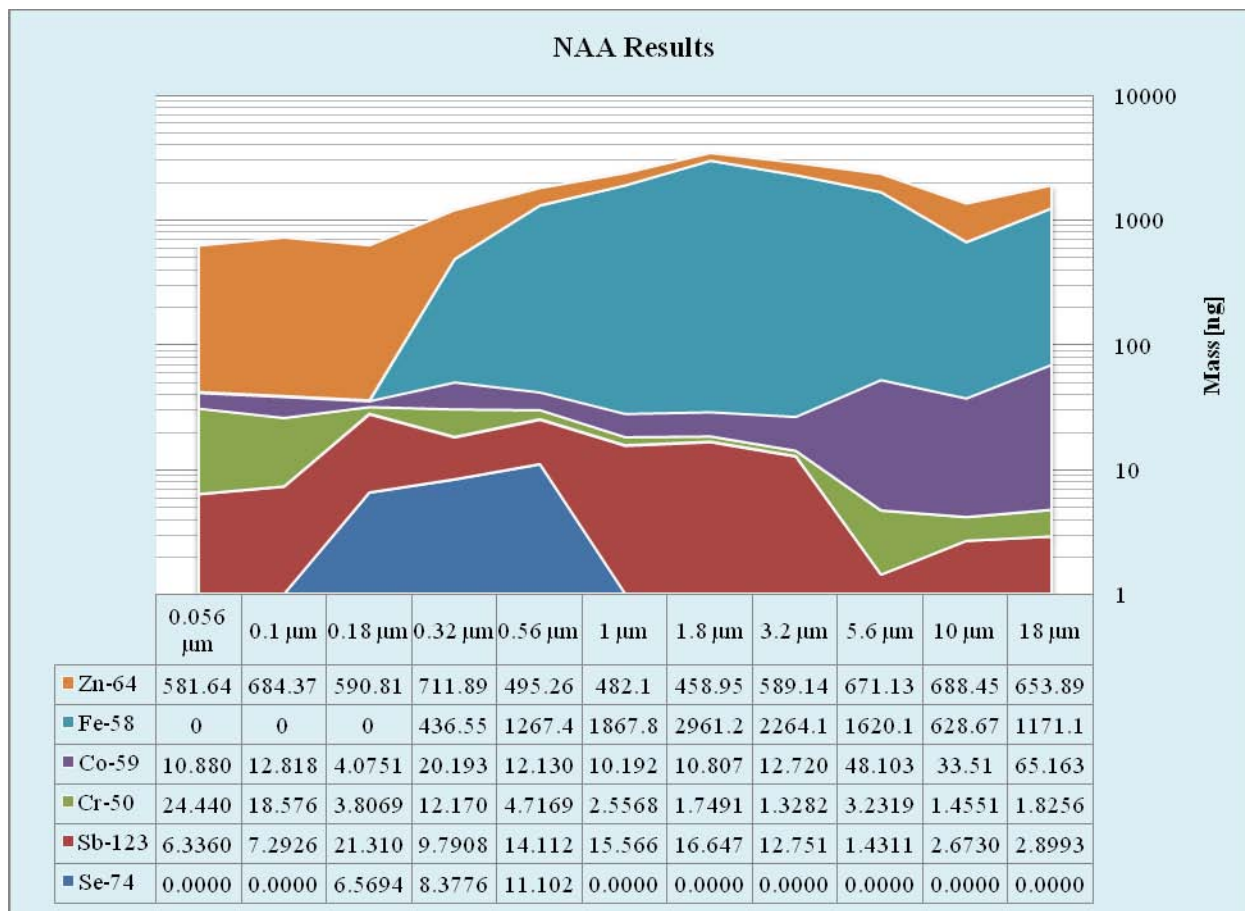


Figure 6. The masses of various isotopes found in the MOUDI sample filters using Neutron Activation Analysis.

Using the Nuclear Explosion Monitoring Inertial Impactor (NEMII) system, initial tests were performed to determine the effectiveness of the modifications designed in the previous phase of the project. The system was tested using both the aerosol generation system mentioned earlier as well as taking prolonged (24+ hours) aerosol collections and using a Geiger-Müller Counter to compare the relative activity of each filter. The results from both of these tests showed that the system was not operating as expected concerning the 0.1 µm impaction stage. The test aerosols were not being collected at this stage and were being collected on the following filter which is the last filter stage of the system where only 0 to 0.1 µm sized particles should be collected. The GM counter results also show that most of the radioactivity from the aerosol collection was located on the last filter, again suggesting that the particles 0.1 to 1.0 µm were passing the appropriate filter. To better characterize the pressure levels within the critical stages, low-pressure gauges, which can be seen in Figure 6, were installed to give a real-time measurement of the behavior of the system in these important regions of the impactor. According to the calculations using the nozzle design tools developed in the previous phase of the project, the pressures after the first and second critical orifice stages should be 0.55 atm and 0.30 atm, respectively, corresponding to pressure drops of 0.45 and 0.70 from atmospheric pressure. The measurements taken showed that the actual pressures in these stages were 0.917 atm after the first stage and 0.883 after the second stage.

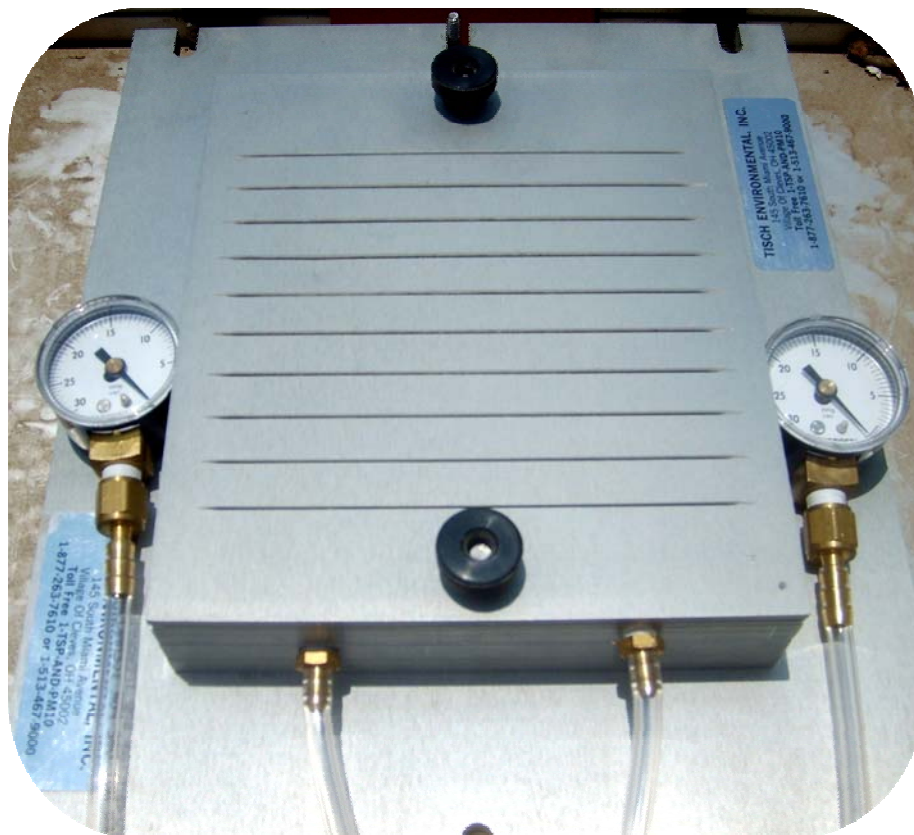


Figure 6. Installation of Low-Pressure Gauges at the critical orifice stages.

The pressures in these stages were lowered by restricting the nozzle flow using rubber gasket material in an attempt to achieve the desired pressure drops, thus allowing for correct particle separation. The pressures expected were eventually maintained, yet this change still had no effect on the particle separation. At this point, further investigation into the calculation of these stages was made and is currently ongoing in an effort to more accurately model the flow within the impactor stages.

Recent tests were done using the methods of impactor design that do not rely on achieving low-pressures within the impactor (Demokritou, 2002), and these have shown improved results for the $0.1 \mu\text{m}$ collection stage. The impactor design in previous work has shown that for high flow rates of 900 liters/minute or higher (NEMII: 1100 L/min) the impactor does not need to achieve low pressure to acquire a cutoff diameter size of $0.1 \mu\text{m}$. The major difference between our current setup and this alternate method is that the Stokes number will not remain constant throughout the impactor but will be dependent on the fluid velocity as the Demokritou group has shown. Further characterization of the flow within the impactor will be calculated and new nozzle plates will be manufactured using this alternate model.

CONCLUSIONS AND RECOMMENDATIONS

Over the past year, tests have been conducted which verify the size distributions of naturally occurring radionuclides, especially those found within the Radon and Thoron decay chain. The tests were conducted using a MOUDI, which is a commercially available impactor with particle size separation as fine as $0.056 \mu\text{m}$. The results agree with past experiments that found that there is a preferred size regime for these particles starting at $0.1 \mu\text{m}$ and extending to $1.0 \mu\text{m}$. The MOUDI was also used to calibrate an aerosol generation system from TSI, Inc. which is able to disperse fluorescent polystyrene particles for collection. The aerosol generation system has been assembled and accurate particle size dispersion has been verified. Neutron Activation Analysis has been conducted on MOUDI collection filters from a 48-hour aerosol sampling and long-lived isotopes have been identified and masses have been found for each.

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The NEMII system has been tested using both the TSI aerosol generation system and by measuring radioactivity amounts within aerosol samples taken at the UT Nuclear Engineering Teaching Laboratory. Each set of measurements has shown incorrect particle collection on the 0.1 μm cutoff stage. Low-pressure gauges were installed on the two critical orifice stages to monitor the pressures at these points. The pressures achieved did not agree with those calculated in the design of the stages. After a series of modifications to reach the target pressures were made, the collection at the 0.1 μm stage did not improve.

Further tests and calculations are currently being done to improve the particle separation based upon results from past experiments (Demokritou, 2002) that do not rely on critical orifices, but instead exploit the benefit of using a high-volume sampler and allow the Stokes number to vary accordingly. These tests have seen improved particle separation. Additional work will include Neutron Activation Analysis on the filters from the NEMII system in comparison to the NAA work done on the MOUDI samples once reasonable particle separations have been accomplished.

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