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Experimentally Investigating Water Aerosol Formation Via Alpha Radiation in a Humid Nitrogen Atmosphere

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The University of Southern Mississippi

Experimentally Investigating Water Aerosol Formation Via Alpha Radiation in a Humid Nitrogen Atmosphere

by

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Abstract

This project aims to verify the formation of water aerosols induced by an ionizing radiation source in a Nitrogen atmosphere with various levels of humidity. This work is part of an effort to characterize the overall signature of ionizing radiation in Earth's typical atmosphere. By signature, it is meant all the chemical reactions and physical processes that take place between Earth's atmosphere and the ionizing radiation. This signature can potentially be used to identify a radiation source out in the field. The identification of a radiation source could potentially be used in a variety of defense applications.

To characterize this signature, Cavity Ring Down Spectroscopy is used. This is a method in spectroscopy which can be used to quantify the optical absorption or scattering caused by various species present in a larger gas sample. This is done by measuring the time it takes for different samples to absorb or scatter a light beam with a known wavelength. By identifying the formation of water aerosols via an ionizing source in an environment that resembles our own, the concentration and formation rate of these water aerosols can be later determined. This information will then be added to the overall working model for the signature of ionizing radiation left in the Earth's atmosphere.

Key Words: Water Aerosols, Alpha Radiation, Humidity, Nitrogen, Physics, Experimental Physics

Dedication

Karen Payne and Burney Payne:

Thank you for granting me life’s opportunities.

Acknowledgements

Thank you to my advisor, Dr. Winstead, and Tyler Reese for mentoring me through this project. I could not have done it without either of you. I would also like to thank my research group, Laken Camp, Alex Roberts, and Andrew Giovengo, for always participating in group collaboration and encouragement.

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List of Abbreviations

CRDS	Cavity Ring Down Spectroscopy
N_2	Molecular Nitrogen
H_20	Dihydrogen Oxide, Water
Nd:YAG	Neodymium-Doped Yttrium Aluminum Garnet

Chapter I: Introduction

Long-range radiation detection devices are needed for a variety of applications in fields such as defense and atmospheric science. Detection of ionizing radiation emitted from materials used or generated as by-products in production of nuclear weapons could potentially help stop the production of a bomb before it undergoes further development. Characterization of radiation effects in the atmosphere could also provide more information about our atmosphere's relationship with solar radiation. This project is specifically geared towards defense applications and is funded by the Defense Threat Reduction Agency. Currently, the most common instruments used to detect radiation are modern variations of the traditional Geiger counter. Radiation interacts with and loses energy to the atmosphere. Because of this, a major restriction of these devices is their reliance on direct contact with radiation, which is dependent on the range of travel of the radiation in the atmosphere. There are three main kinds of ionizing nuclear radiation: Gamma, Beta, and Alpha. These can travel up to kilometers, meters, and centimeters respectively in air under typical ambient conditions on the Earth's surface. The atmospheric effects due to alpha radiation were studied in this project. For a device that must interact with the radiation to get readings from a source emitting alpha radiation, it must be within centimeters of the source, because that is how far the radiation can travel in the atmosphere. A method of detecting the chemical effects that the radiation induces into the atmosphere, rather than the radiation itself, has been proposed. By detecting these chemical effects in the atmosphere from a much greater distance, a prediction of the presence of radiation can be made. The chemical effects left in the atmosphere all come together to form what is called the radiation’s signature. For a long-range radiation detection device based on atmospheric effects to be optimized, it is essential to first build a model that explains how radiation interacts with our atmosphere, or to understand its signature. A variety of molecules can be produced from the various reactions that happen between ionizing radiation and the naturally occurring chemical species in the atmosphere.

This project will study just one of the many resultant effects of radiation induced reactions, namely the formation of water aerosols. Eventually, the data obtained for aerosols will be added to the overall model for describing reactions induced by radiation. The end goal of the entire project is to build a complete model for our atmosphere that accurately describes all important reactions that can take place and the concentration of each reactant and product species, given a certain sample of radioactive material in a known environment. In other words, the larger project’s goal is to determine the signature left in the air from an ionizing source. With this model, one would potentially be able to identify the presence of a radioactive source if the tested atmosphere contained certain concentrations of certain molecules. The end goal of this portion of the project is to provide the missing water aerosol data to the pool of working data for other species formed from radiation induced reactions. More specifically, this project aims to identify the formation of water aerosols in a nitrogen environment with some level of humidity and exposure to an ionizing source. Nitrogen is used for these initial experiments because approximately 80 percent of the Earth’s atmosphere is composed of nitrogen, and pure nitrogen does not interact with the wavelength used here when exposed to ionizing radiation. First, it must be verified that water aerosols are being produced in this environment, and if so, whether the current experimental system can effectively detect them.

To do this, an important laser method is used called cavity ring down spectroscopy (CRDS). CRDS is a very sensitive optical absorption spectroscopy method that can be used to determine the identity and concentration of various molecular species in gas samples. CRDS works by detecting how well a sample absorbs or scatters a certain wavelength of light that has been trapped in an optical cavity formed from two highly reflective mirrors. From the absorption/scattering properties a prediction of the product present can be made. This is because each molecule or atom has a unique set of wavelengths that are absorbed/scattered at varying efficiency. For example, water's peak absorption wavelength is in the infrared range.

Ionizing radiation often creates species that do not normally exist in the atmosphere, including ionized atoms and molecules. There are two types of ions that can be formed, ones that carry a positive charge when an electron is ripped off and ones that carry a negative charge when a neutral atom or molecule captures that free electron. Water molecules are polar molecules, meaning they have one positively charged side and one negatively charged side. Because opposite charges attract, in humid atmospheres, one side of the water molecule is attracted to the charged ion, and this starts the formation of a water aerosol. A visual of a negatively charged ion attracting polar water molecules is given in Illustration 1.

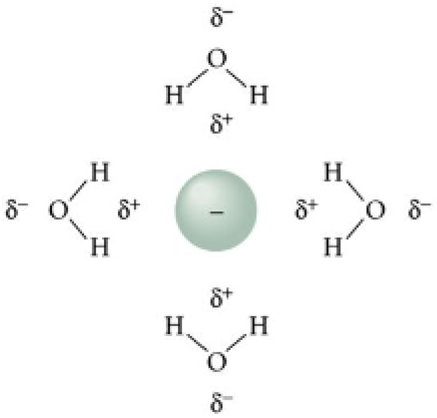


Illustration 1

Structure of a Water Aerosol[1]

CRDS uses the time it takes for the light to be absorbed/scattered, and from this, the identity and concentration of molecules or scattering particulates, such as aerosols in the sample, can be determined. Even slightly altering the wavelength of the laser can change the effective absorption rate. With this information, we can measure how much and what kind of molecules are being produced by the radiation to provide laboratory validation for models that will later be developed. Eventually, these models can be used to determine whether nuclear radiation is present based on the presence and concentration of the molecules found in the atmosphere.

Just as each molecule has a different absorption spectrum, every aerosol has its own scattering properties. These properties depend on the size and composition of the aerosol. CRDS can also be used to study the scattering properties of aerosol particles. The data will help determine how quickly the water aerosols are formed and to what size. This information can be used to validate the larger model and to predict the presence of radiation if these aerosol characteristics are reproduced.

This project is contributing to a much larger project, with the end goal being to provide models to support development of long-range radiation detection devices. For now, a working model capable of predicting the exact concentrations of each product made from reactions between the radiation and known atmospheric conditions is needed. Before this can be done, the production of water aerosols formed in this environment needs to be validated. Once this model is validated, the performance of various long-range radiation detection devices can be optimized.

Chapter II: Review of the Literature

O'Keefe and Deacon published the first paper on cavity ring down spectroscopy in 1988 and introduced a new way of detecting the absorption rate of light through a sample.[2] Traditional absorption spectroscopy methods had been used for decades prior to the invention of CRDS, but these methods were limited in their sensitivity. CRDS can increase the sensitivity of optical absorption measurements by factors of 10^3 or even 10^4 in some cases and can be used to measure species concentrations to the parts per trillion level in some cases.[2] This is an extreme case and only holds true when the species being measured absorbs the light very well.

To study the absorption of light, it is essential to have a sound understanding of Beer's Law. August Beer formed the equation in 1852, which describes how the intensity of light decays while traveling through a material.[3]

Equation 1

$$I(x,\lambda) = I_0 e^{-\alpha(\lambda)x}$$

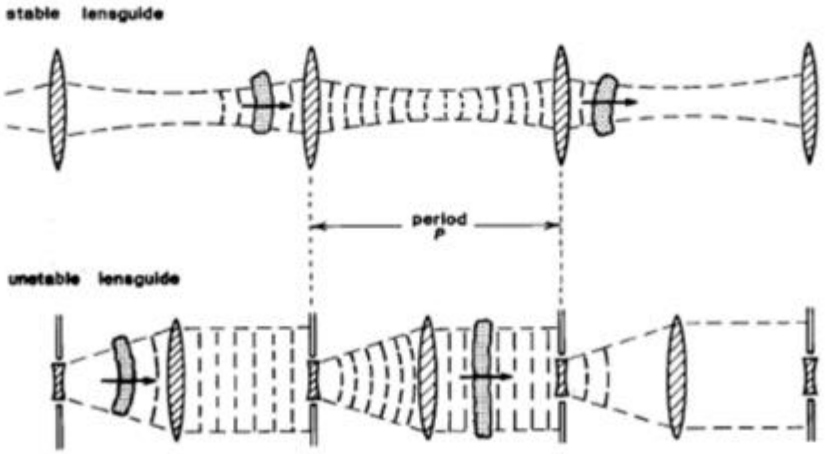
Beer's Law describes how the intensity of light behaves while moving a certain distance x, through a medium.[3]

This equation demonstrates how the intensity of the light from the source is affected by traveling through a medium.[3] I is intensity and is a function of the wavelength of light sent through the sample and the distance the light travels. I_0 is the initial intensity of the light source. Alpha is the absorption co-efficient of the sample and is also dependent upon the wavelength of light. Finally, x is the distance the light travels through the material. The absorption coefficient is dependent on the effective area, known as the absorption cross-section, of the particle that is absorbing or scattering light as well as the number density of the particles. Effectively, the larger the cross-section of a particle, the more efficiently it will absorb or scatter a light beam. Importantly, for optical absorption, the cross-section does not equate to the physical size of a molecule but is determined by the quantum mechanics governing light absorption by a specific molecule. For scattering, the cross-section is determined by the relative size of the particle to the wavelength of light and by the composition of the particle. By studying the rate at which the intensity of various wavelengths of light changes, an analysis of the sample in the cavity can be made. This is because every molecule has a specific "fingerprint" for the electromagnetic radiation it will absorb. An experimental problem indicated by this law is that alpha might be a very small value (on the order of 10^{-9}/cm). That means x, the distance the light travels, must effectively be very large for there to be a noticeable change in intensity of light.

CRDS offers a solution to the short distance problem. Because light travels so quickly, approximately $3\text{x}10^8$m/s in a vacuum, light trapped inside a relatively small optical cavity formed from highly reflective mirrors can effectively travel several kilometers by making the trip between the mirrors multiple times. For a half meter cavity and 99.99% reflective mirrors, a ring down time of 16.66 microseconds is predicted. In that amount of time light travels 5000 meters, or 5000 round trips in the cavity. This effectively makes our x value larger, and therefore there is a noticeable change in the initial intensity of the light and the final intensity of the light, which allows for the absorption coefficient, alpha, to be measured.

Optical cavities are typically constructed from curved mirrors. For a given radius of curvature, there is a maximum distance at which the mirrors can be placed from one another for the cavity to be stable, i.e. for the light to remain trapped in the cavity.[4] If the cavity is unstable then the light will be lost outside of the cavity. Below is a diagram comparing a stable and unstable cavity, where the multiple reflections from the curved mirrors are modeled as a series of lenses.[4]

Illustration 2



The top image is a stable cavity and the bottom is an unstable cavity, which means light escapes from the cavity.[4]

Both mirrors have a g parameter that is given in the following equation: $g= 1-L/R$, where L is length of the cavity, and R is the radius of curvature of the mirror.[4] A cavity is considered stable if and only if it satisfies the condition $0<g_1g_2<1$.[4] In this equation g_1 is for mirror 1 and g_2 is for mirror 2.

Equation 2

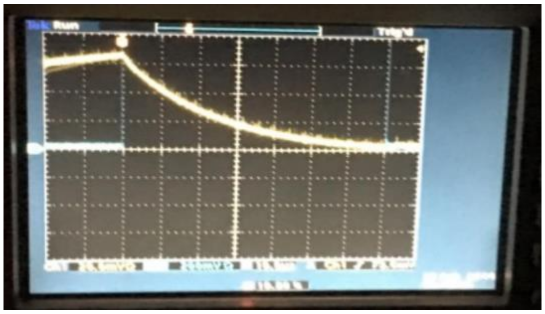
$$I(t) = I_0 e^{-\frac{t}{\tau}}$$

Alternate form of Beer's Law in Cavity Ring down Spectroscopy

Another formula that applies to light intensity inside the cavity is given in equation 2. This equation relates the intensity of the initial light as a function of time. Where I(t) is intensity of light as a function of time, I_0 is initial intensity of light, t is the specific time at which one is measuring the intensity, and τ is the ring down time constant. The ring down time constant is dependent on the mirror reflectivity and the absorption and scattering properties of the sample being tested.

To determine the concentration of a species in a sample in the cavity it is important to know the reflectivity of the mirrors. Some of the light's energy will be either transmitted through or absorbed by the mirrors. To measure the effective reflectivity of the cavity mirrors, a baseline decay time must be found without the species of interest present in the cavity, this is called τ_0. By comparing how long it takes for the light to ring down under baseline conditions to how long it takes for the light to ring down when exposed to a sample, the species and concentration of the sample can be found. Below is an intensity vs. time graph obtained using an oscilloscope to measure the output from a photodetector placed behind a cavity that had been excited by a diode laser. The decay starts when the diode laser is turned off. This measurement can be done because the light exiting the cavity strikes the detector and induces a current which, after amplification, is read as a voltage by the oscilloscope.

Illustration 3



Intensity of light vs. time graph

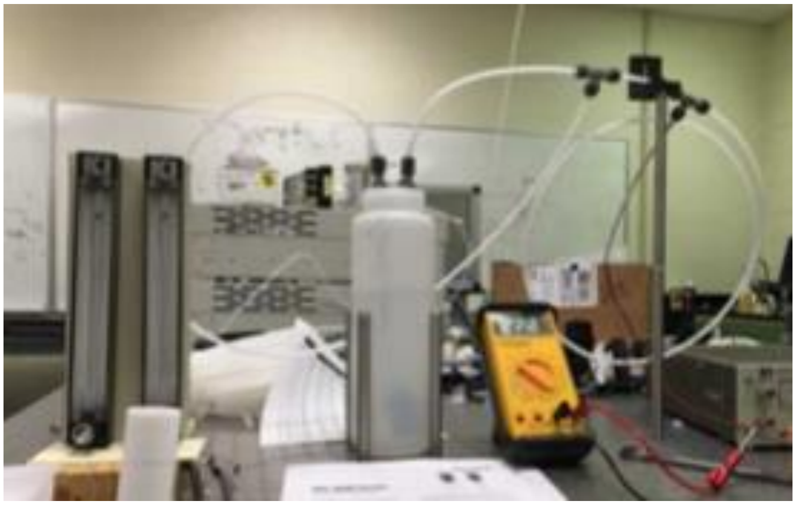
This graph indicates that, as time increases, the intensity of the light in the cavity dies down until it is zero. Although this run was done without being exposed to a specific sample, it was not done in a vacuum, so the loss in intensity is due to a combination of particles that were present in the lab air at this time along with the loss in intensity due to the mirrors. These additional baseline losses are combined with the actual mirror reflectivity into one effective mirror reflectivity value that is slightly lower than the actual value. Once this baseline ring down time has been established, it can be compared to the ring down time when a sample is exposed to a radiation source. By comparing the time it takes for the light to be absorbed in a medium before and after exposure to a radiation source, along with knowing the absorption coefficient of the product being characterized, the concentration of that product can be found.

The production of water aerosols in a nitrogen atmosphere with varied humidity levels can be verified using this method. It is known that ions can enhance the formation of aerosols. In 1895, Wilson used an expansion chamber to show for the first time that ionizing radiation in the form of x- rays has an impact on the rate of nucleation of particles in the gas phase.[5] Nucleation is defined as the first step in the formation of a new structure. The experiment was repeated with the sample exposed to uranium, which emits alpha radiation. The experiment yielded the same results as the first experiment and Wilson concluded that droplets formed on radiation-induced ions to produce aerosol particles.[5] In 1997, it was reported that cosmic rays played a role in cloud formation by ionizing atmospheric molecules which in turn served as nucleation centers for water molecules.[6] Udelhofen and Cess (2001) found that cloud cover in the United States for the last 100 years correlated with solar activity.[6] This is of importance because it shows that radiation in the atmosphere positively corresponds to water aerosol production (to produce the clouds). The discovery that radiation could increase the nucleation rate of water aerosols in the atmosphere leads scientists to believe that by studying aerosol formation (along with other molecule formation), they may be able to develop methods for identifying when radiation is present.

Chapter III: Methodology

To begin collecting data, i.e. the cavity ring down time in both humid and dry atmospheres with and without radiation, the correct equipment is needed. A laser with a known wavelength that matches the desired absorption/scattering behavior, two highly reflective mirrors, a non-reactive chamber for containing an atmospheric sample, a system to regulate humidity levels, and a photo detector are all needed. The laser being used is a pulsed neodymium doped yttrium aluminum garnet laser, which is abbreviated Nd:YAG. The fourth harmonic of this laser has a wavelength of 266nm. Illustration 4 shows the humidity introduction system used in this project.

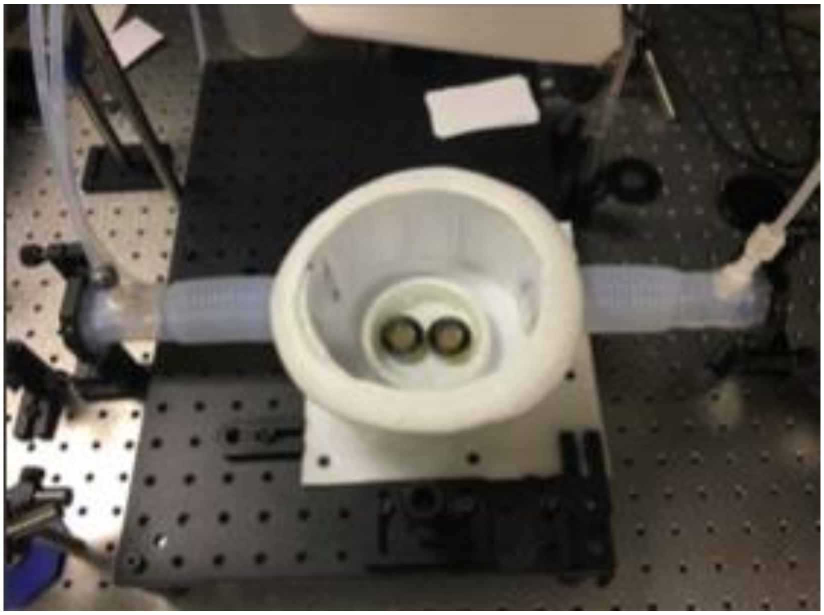
Illustration 4



Humidity Regulation System

In this image the dry nitrogen is bubbled through the bottle of water. The amount of nitrogen being sent through the bottle is regulated using the flow tubes show. One flow tube adjusts the flowrate of humidified nitrogen (~100% relative humidity) into the system, and the other flow tube regulates the flowrate of dry nitrogen that enters the system. Using these two flow tubes, the desired amount of relative humidity can be achieved. This gas is then flowed into a non-reactive chamber containing two alpha sources. This non-reactive chamber is shown in Illustration 5.

Illustration 5



Non-Reactive Chamber

The humidified nitrogen flows in through the tube on the left and flows out through the tube on the right. In this photograph the two cylindrical disks in the cavity are the alpha sources, Polonium-210. They are placed in the center so that the incoming air is directly in the pathway above the alpha sources. The two mirrors are situated at the ends of the cavity. The chamber itself is non-reactant with any of the products being formed and has a lid on it whenever data is being taken. The chamber and tubing are all Teflon or Teflon lined which does not react with the products made in the experiment. To set up the cavity, the furthest mirror from the laser source must be aligned. It will need to be facing the light source. Once the beam is reflected into the laser, indicating that the mirror and the laser are perpendicular to one another, the second mirror is placed facing the first mirror to create a cavity. A jig (a piece of paper with a pin sized hole in it) is placed in between the mirrors to help align the beam. Once both mirrors are aligned so that reflections from their surface go through the jig, light intensity will increase in the cavity and in some cases the beam will start to become visible. This cavity excitation can be referred to as ringing up. This is true for optical cavities with continuous wave lasers. Once the cavity has rung up, the initial light source can be turned off and the intensity of light in the cavity will start to decay. This is called ringing down. To collect data for measuring this decay time, a photo detector is placed behind the furthest mirror to read intensity as a function of time. As previously mentioned, to study the ring down time in the presence of a radiation source, the ring down time with no radiation present must be measured as a baseline. Equations 3 and 4 are for τ_0, the time it takes for the cavity to ring down with no absorbing/ scattering sample present, and τ, the time it takes for the cavity to ring down with an absorbing/ scattering sample present, respectively.

Equation 3

$$\tau_0 = \frac{d}{-clnR}$$

The time it takes the light intensity to die down without radiation present

Equation 4

$$\tau = \frac{d}{c[-lnR+\sigma Nl]}$$

The time it takes light to ring down with radiation

Here d is spacing between the mirrors, l is the length of the sample, σ is the absorption cross-section, N is the number density and R is mirror reflectivity. As seen from the equation the larger the value for R, the reflectivity of the mirrors, the longer time it takes for the light beam to ring down inside the cavity. Longer ring down times mean a longer effective pathlength and thus an increased sensitivity. This value for R is determined not only from the reflectivity of the mirrors but also from how well species or particles already in the environment absorb or scatter the light source. The second equation can be rewritten to solve for the absorption cross-section times the number density of molecules as seen in Equation 5.

Equation 5 $$\sigma N = \frac{d}{lc}\left(\frac{1}{\tau} - \frac{1}{\tau_0}\right)$$

absorption/scattering coefficient

By dividing by the appropriate cross-section for scattering, if known, the number density of the water aerosols present can be found. However, even if the specific cross-section is not known, this quantity is proportional to the number density of aerosols present. This experiment will be done first with pure dry nitrogen. In this experiment Nitrogen and the alpha radiation should not be forming any new chemical products that would change the ring down time. Then the experiment is repeated, but this time water vapor is introduced into the system in the form of humidified nitrogen gas. Once humidity has been introduced, new products are formed. The ring down time is now decreased as the new products absorb/scatter the light from the laser. From these two pieces of data it can be determined that water aerosols are being formed. By studying the concentration of the water aerosols formed under different circumstances, a model can be made to predict the concentration of water aerosols under any given circumstance.

Chapter IV: Results

Illustration 6 shows the ring down time of the cavity with respect to time in a pure nitrogen atmosphere. This data was taken at a flow rate of 50 milliliters per minute to ensure a slightly positive pressure inside the chamber. The black line represents the running average of 50 samples in Illustration 6 and 104 samples in Illustration 7. To obtain this data, dry nitrogen was flowed into the non-reactive chamber containing the alpha sources. The alpha sources went through intervals of being covered and uncovered while the ring down time was measured. This was accomplished by raising and lowering a cover inside the chamber using a Teflon wrapped wire passed through the top of the chamber. In pure nitrogen the alpha radiation ionizes and dissociates a small fraction of N_2 molecules into N_2^+, N, and N^+. However, these species do not absorb the laser wavelength, so without humidity there are no water molecules to nucleate into aerosol particles that would change the ring down time.

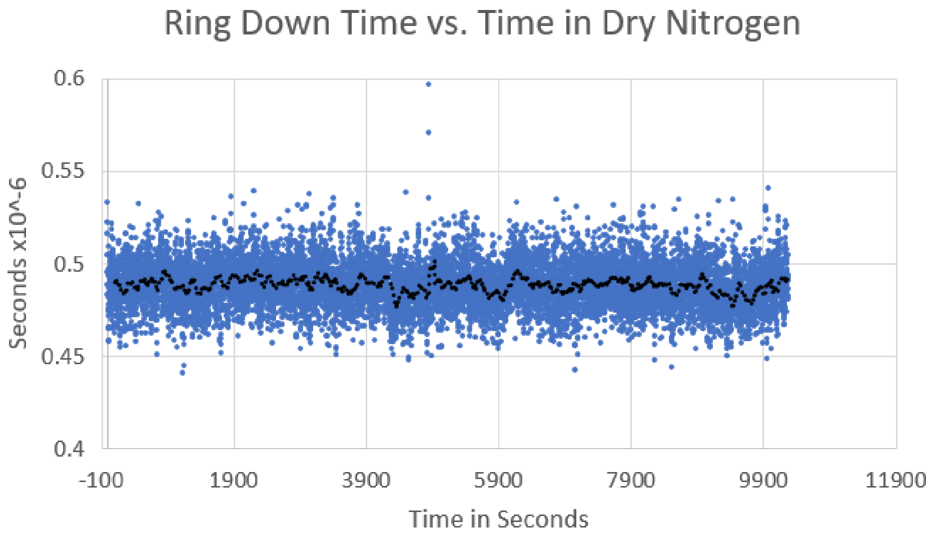


Illustration 6

Illustration 7 shows the ring down time of the cavity with respect to time in an environment that was Nitrogen at approximately 28% relative humidity. The same steps from above were repeated. In this illustration, the shorter ring down times evident in the data occurred when the atmosphere was exposed to the alpha sources. The longer ring down times in the data represent when the atmosphere was not exposed to the alpha sources, i.e. the cover was in place over the alpha sources.

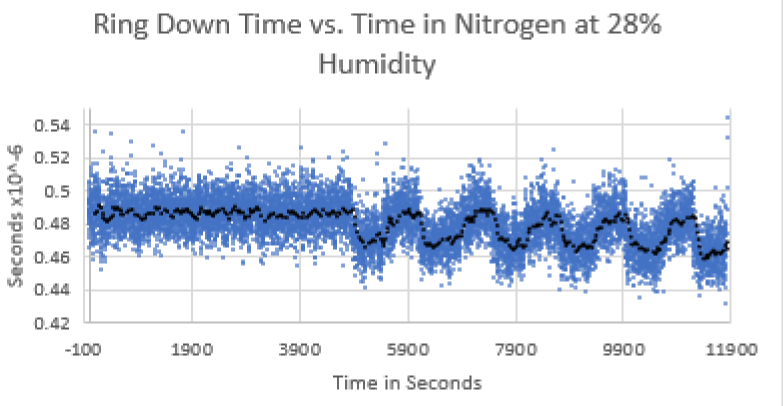


Illustration 7

Chapter VI: Discussion

An initial interpretation based on aerosol formation will be made from analyzing the graphs of ring down time vs. time. In illustration 7, ring down time vs. time in Nitrogen at 28 percent relative humidity, the graph displays a distinct response to radiation exposure. This pattern can be used to predict the formation of water aerosols. The regions of the data with shorter ring down times represent when the environment was exposed to a radiation source. The ring down time is longer when there is no radiation present because there are fewer products in the environment to absorb/scatter the light from the laser, whereas when radiation is present, the ring down time drops. When the environment comprised of water and Nitrogen is exposed to a radiation source, it is predicted that the product, namely water aerosols, is being produced. We hypothesize that this can be observed in the data because when the environment was exposed to a radiation source in the presence of humidity, the ring down time dropped drastically. Yet when the pure nitrogen sample was exposed to radiation, little to no change in ring down time was observed with the presence or absence of radiation. This implies that a product is being formed that requires both the presence of radiation and humidity that is absorbing or scattering the light at a higher rate than when the cavity contained dry gas. In illustration 6, the graph does not display the same distinct response to radiation as it does in the runs with humidity. It is interpreted that this is because alpha radiation in pure nitrogen does not generate other chemical species that absorb this wavelength of laser light, and without humidity, water aerosols to scatter the light cannot form. Therefore, the ring down time is not affected by the exposure to radiation. From these two graphs it can be hypothesized that the ring down time is decreased because of the formation of water aerosols in the cavity. Once the value for sigma from Equation 4 and 5 is determined for water aerosols, an accurate prediction of water aerosol concentration can be made. Work will continue to further develop and interpret aerosol models. In the future, this project aims to develop a program that can use certain input characteristics of an environment and produce output information about the formation of aerosols. Dr. Fangqun Yu has done this with water aerosol production via solar radiation with some level of sulfuric acid present.[7] Dr. Yu has provided a website for calculating aerosol characteristics based on his work.[7] The algorithm below provides an example from this website that is comparable to what this research aims to do computationally, but instead, with an alpha radiation as our ionizing source as well as consider gas mixtures that do not include sulfuric acid.

```
INPUTTED VALUES:
--> [H2SO4] = 1.00E+07 /cm^3
--> Temperature = 280.0 K
--> Relative Humidity = 50.0 %
--> Ionization Rate = 5.0 ion-pairs/cm^3s
--> Surface Area = 40.0 um^2/cm^3
OUTPUT based on ion-mediated nucleation model (Yu, 2006; Yu and Turco, 2008)[8]:
--> Ion-mediated nucleation rate = 2.8E-01 /cm^3s
--> Number of H2SO4 molecules in critical cluster = 16.5
--> Number of H2O molecules in critical cluster = 58.2
--> Diameter of critical cluster = 1.8 nm
```

This code takes input values about the environment, such as temperature, humidity, and size and concentration of reactants, and the output values describe the ratio of components of the aerosol product, size of the product, and the rate of production.[8] Once the concentration of water aerosols formed in this nitrogen environment due to an ionizing source can be determined computationally, this missing piece of information will be added to the overall signature left in the atmosphere from an ionizing source. This signature can be used to identify when an ionizing source is present in the environment. Finding this signature out in the field could be an indicator of a radioactive material.

One of the challenges the project faced was building a stable cavity and non-reactive chamber with no leakage of the room air into the cavity. Initially, when the ring down time vs. time graph for pure nitrogen was measured, changes in ring down time were unexpectedly observed. It was later discovered that room air was diffusing into the system and was reacting with the alpha radiation, mostly likely forming ozone, which absorbs strongly at this wavelength. This was causing the ring down time to drastically drop as the new products absorbed the light. However, by maintaining a small positive pressure on the chamber during measurements to prevent inflow of room air, this effect was removed.

Chapter VII Conclusion

In a time when the threat of a nuclear war is possible, it is important that new methods of detecting radiation emitted from materials potentially used in radiological weapons are being developed and implemented. A method of detecting the chemical or physical signature left in the atmosphere vs. detecting the radiation itself is a way to extend the range at which radiation can be detected. One of the physical components of ionizing radiation's signature is the development of water aerosols. This project successfully verified that water aerosols were being formed in a nitrogen environment with an average relative humidity level of 28%. This was done using CRDS to measure the time it takes for a light beam to be scattered when the sample was exposed to radiation and when the sample was not exposed to radiation. The ring down times for these two types of runs were compared, and it was noted that the ring down time drops when a humidified sample is exposed to an ionizing radiation source. It was concluded that this was due to the formation of water aerosols. Later this project will be taken over by an upcoming undergraduate and the concentration and size distribution of these water aerosols will also be determined.

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