Proton-Induced X-Ray Emission Analysis of Atmospheric Aerosols Collected at Piseco Lake in the Adirondack Mountains

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Introduction

We are developing a research program in ion-beam analysis (IBA) of atmospheric aerosols at the Union College Ion-Beam Analysis Laboratory to study the transport, transformation, and effects of airborne pollution in Upstate New York. The simultaneous applications of the IBA techniques of particle-induced X-ray emission (PIXE), Rutherford back-scattering spectrometry, particle-induced gamma-ray emission, and proton elastic scattering analysis is a powerful tool for the study of airborne pollution because they are non-destructive and provide quantitative information on nearly all elements of the periodic table. Here we present some preliminary results of a PIXE analysis of aerosol samples collected at Piseco Lake in the Adirondack Mountains.

PIXE Analysis

PIXE is the main IBA technique used for elemental analysis of atmospheric aerosols in the Union College Ion-Beam Analysis Laboratory because it is a very powerful tool for the study of environmental pollution. Some of the properties that make it well suited for this kind of work are its high sensitivity and low detection limits for elements from Na to U, it is non-destructive, requires little sample preparation, and has short analysis times [1].

A schematic of a basic PIXE experimental setup is shown in Figure 1. The sample of interest is bombarded with a beam of protons, occasionally knocking an inner-shell electron out of an atom in the sample, creating a vacancy (Figure 2). This allows an outer-shell electron to fill the hole, emitting an X-ray that can be detected (Figure 3). Each element emits characteristic X-rays which allow us to determine the elements present in the sample. The concentration C_Z of an element Z present in the sample is given by

$$C_z = \frac{Y_z}{Y_t \cdot H \cdot Q \cdot \varepsilon \cdot T}$$

where Y_Z is the intensity of the principle X-ray line for element Z, Y_t is the theoretical intensity per micro-Coulomb of charge, H is an experimental constant determined by taking data on a set of standards, Q is the measured beam charge incident on the sample, ε is the intrinsic efficiency of the detector, and T is the coefficient for transmission through any filters or absorbers between the target and the detector.

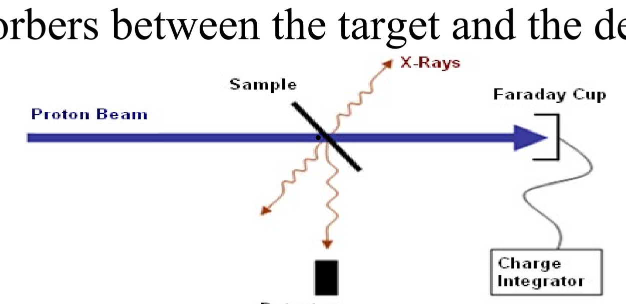

Figure 1: A schematic of a basic PIXE experimental setup for thin targets

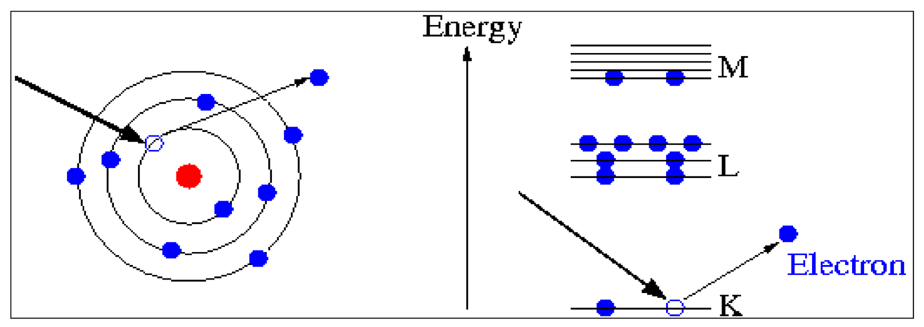

Figure 2: The ejection of an inner shell electron by a proton.

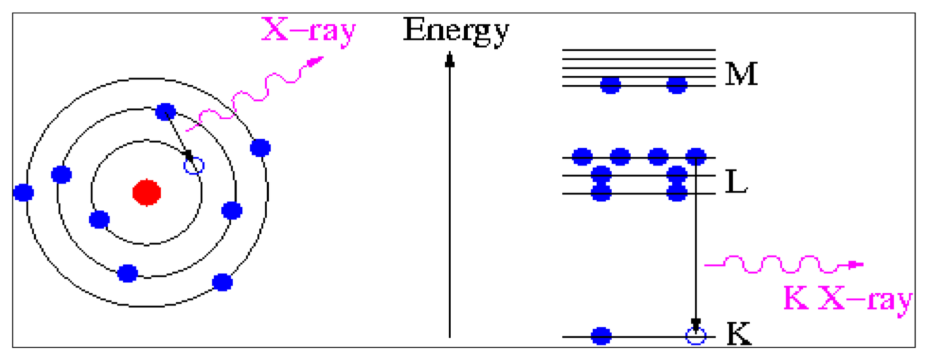

Figure 3: An X-ray is emitted when the void is filled by an outer shell electron.

Sample Collection

A nine-stage cascade impactor was used to collect the aerosols and separate them based on their particle size [2]. Shown in Figure 4 is a schematic and photograph of the impactor. The impactor was attached to a vacuum pump which drew air through the impactor at a rate of 1 L/min for approximately 48 hours. This corresponds to a total of approximately 2.7 m^3 of air that flowed through the impactor. Particles of different aerodynamic diameter ranges were impacted on Kapton foils in each stage. The thin Kapton foils, shown in Figure 5, were removed and used as targets in the PIXE experiments with the accelerator.

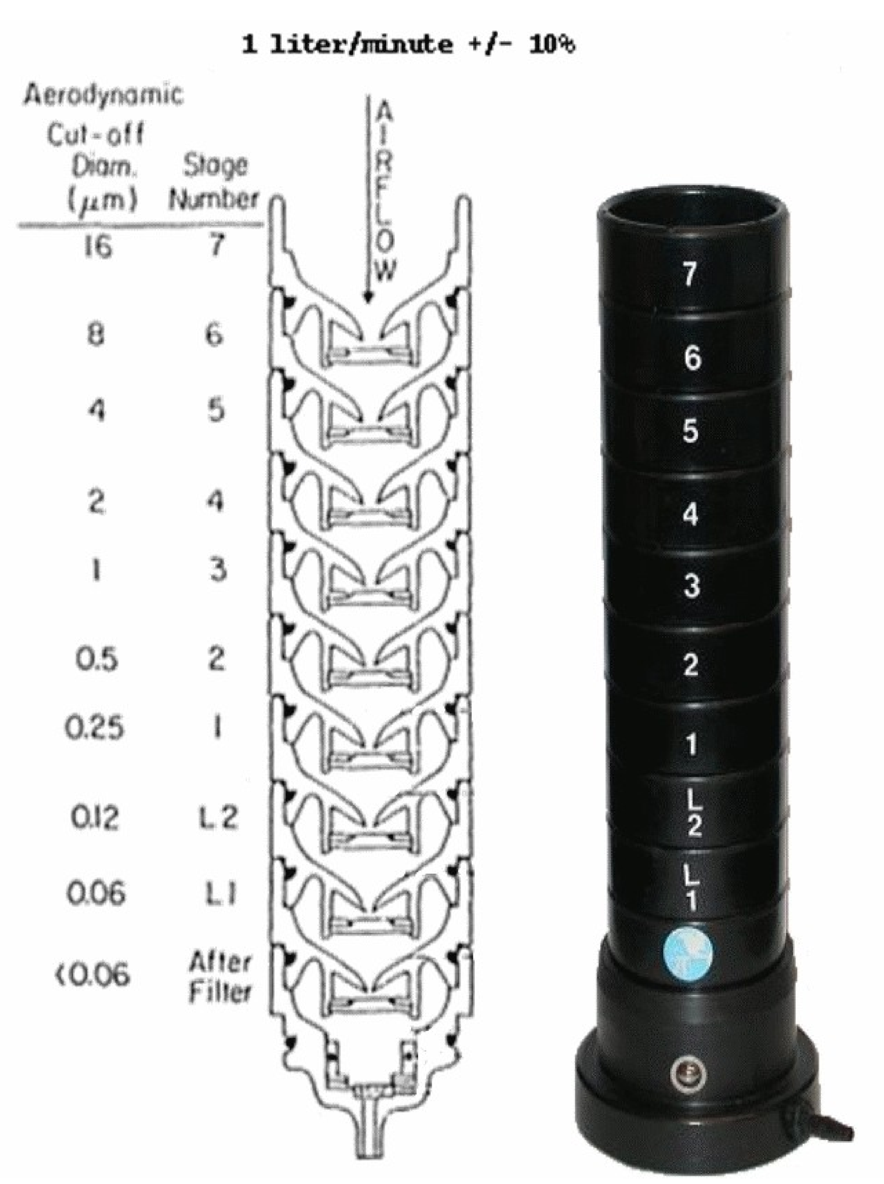

Figure 4: A schematic and photograph of the nine stage cascade impactor [2].

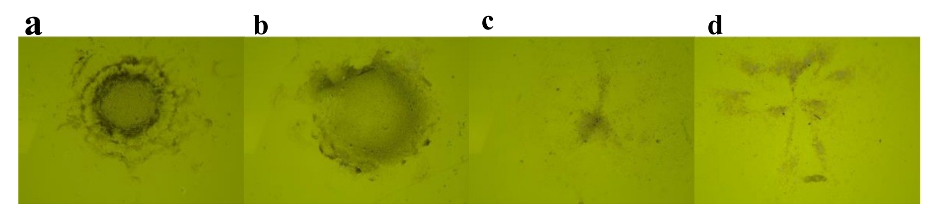

Figure 5: A photograph of microscope images of some of the impacted Kapton foils.

Experiment

The PIXE experiments were performed using the Union College Pelletron Accelerator shown in Figure 6. Proton beams with an energy of 2.2 MeV and a diameter of ~1 mm were incident on the samples positioned normal to the beam in a scattering chamber. Beam currents of 2 to 5 nA were measured in a Faraday cup behind the scattering chamber. The X-rays were detected with an Amptek silicon drift detector (SDD) at 135°, after passing through a 76-μm thick Be absorber on the detector. The SDD detector was calibrated using an ^{241}Am source. Energy spectra of the emitted X-rays were collected for all the aerosol samples and a set of Micromatter standards [3]. A picture of the inside of the scattering chamber is shown in Figure 7.

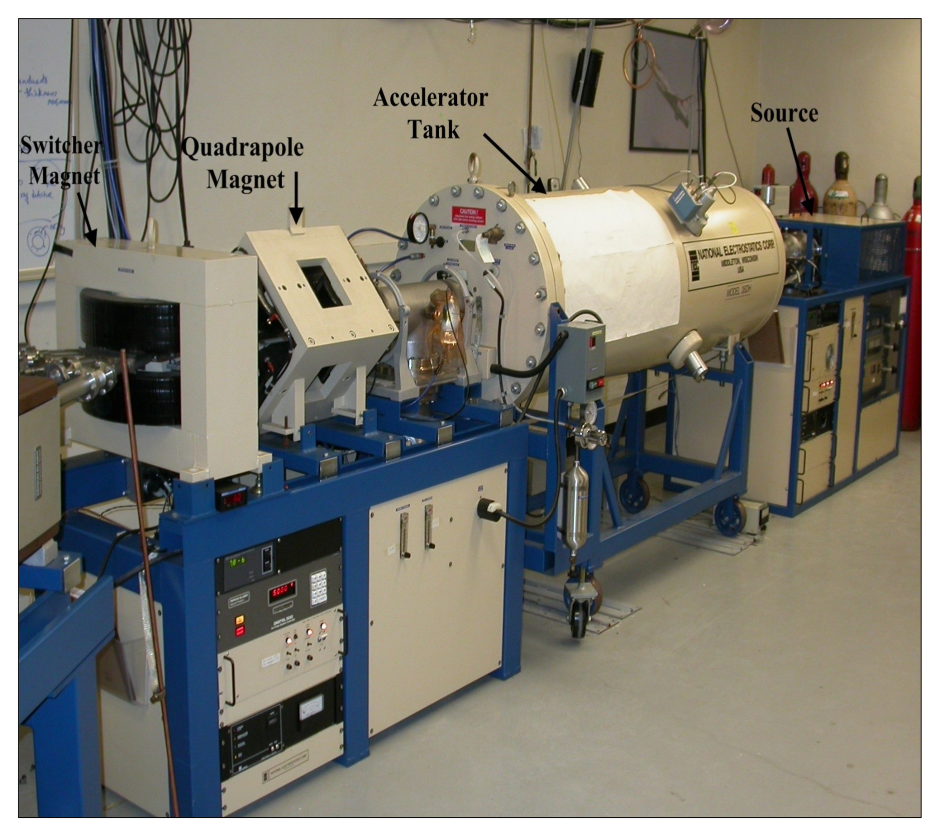

Figure 6: A photograph of the Union College Pelletron Accelerator.

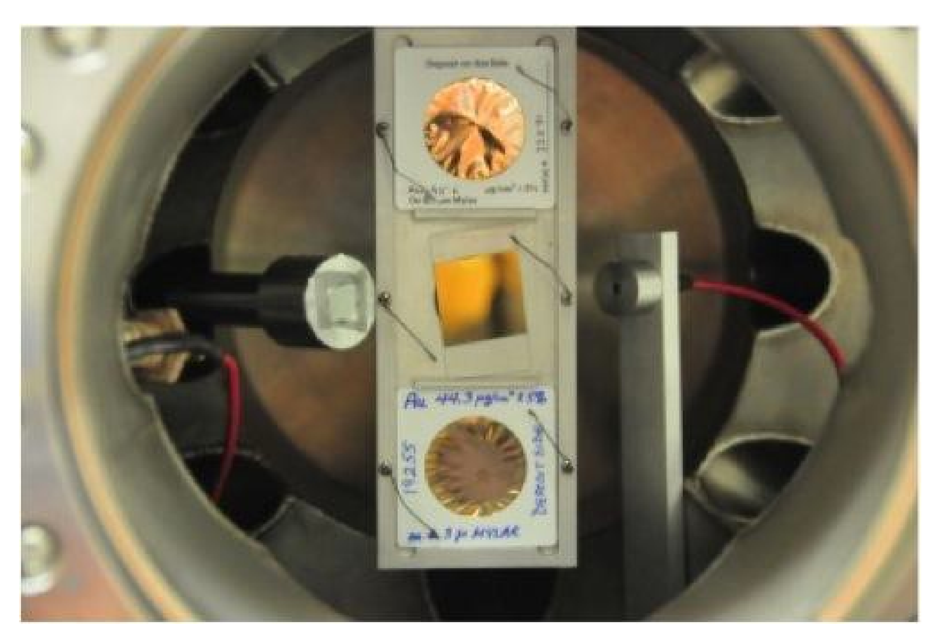

Figure 7: A photograph of the inside of the scattering chamber showing the target ladder (canter), the X-ray detector (left), and a charged particle detector (right).

Preliminary Results

Preliminary results are presented in Figures 8-12. Figures 9 and 10 show comparisons of PIXE spectra taken on a blank Kapton foil and foils impacted with particulate matter 0.5-1 and 1-2 μm in diameter, respectively. All the spectra were fitted using GUPIX [4] and GeoPIXE [5] software to determine the concentration of each element in ng/cm^2 on the foils. Figures 10 and 11 show bar graphs of concentration for each element for particulate matter 0.5-1 and 1-2 μm in size, respectively. A bar graph of concentration in ng/m^3 of sampled air versus element for all foils is shown in Figure 12. The significant concentrations of phosphorous, sulfur, and potassium in many of the particulate matter size ranges are suggestive of coal combustion and industry [6]. Sulfur, along with nitrogen, are responsible for acid rain which has been a well documented problem in the Adirondack Mountains.

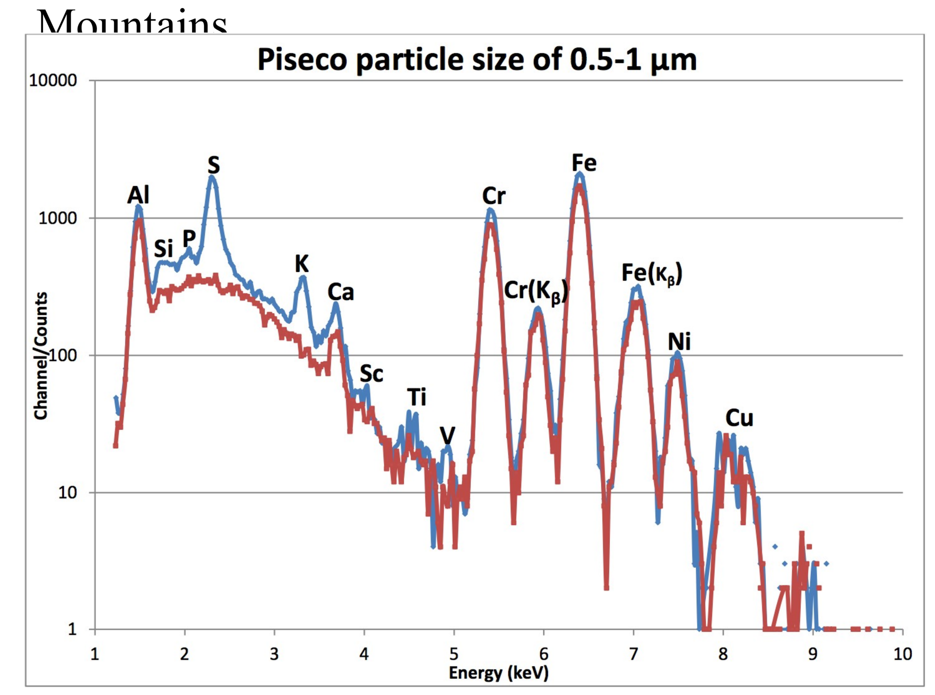

Figure 8: Comparison of PIXE spectra taken on impacted Kapton foils (blue) for particle size of 0.5-1μm and a blank Kapton foil (red).

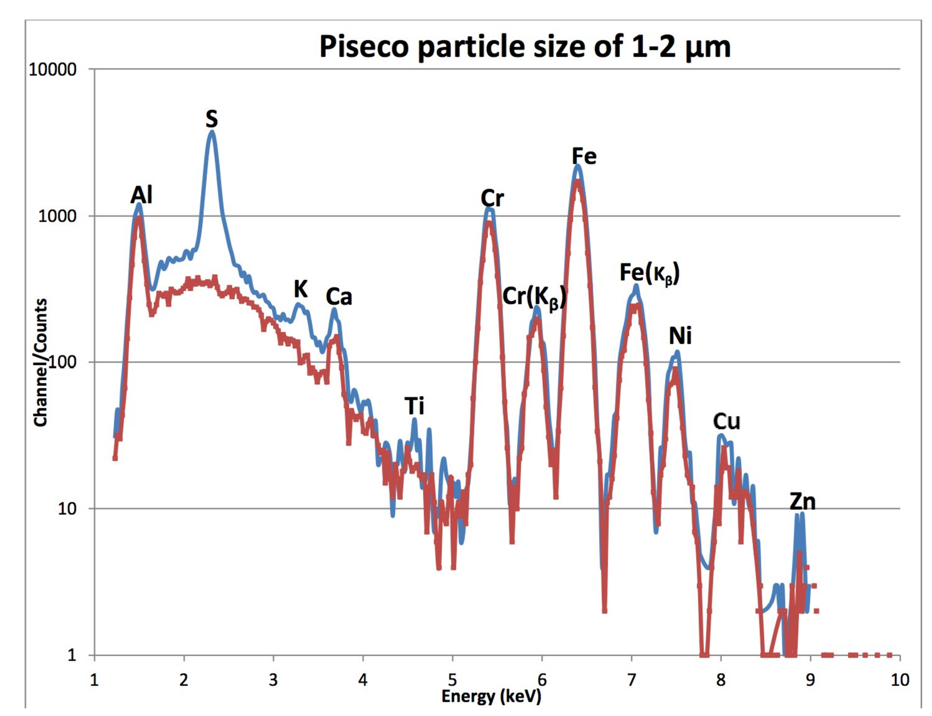

Figure 9: Comparison of PIXE spectra taken on an impacted Kapton foil with particulate matter between 1-2 μm (blue) and a blank Kapton foil (red).

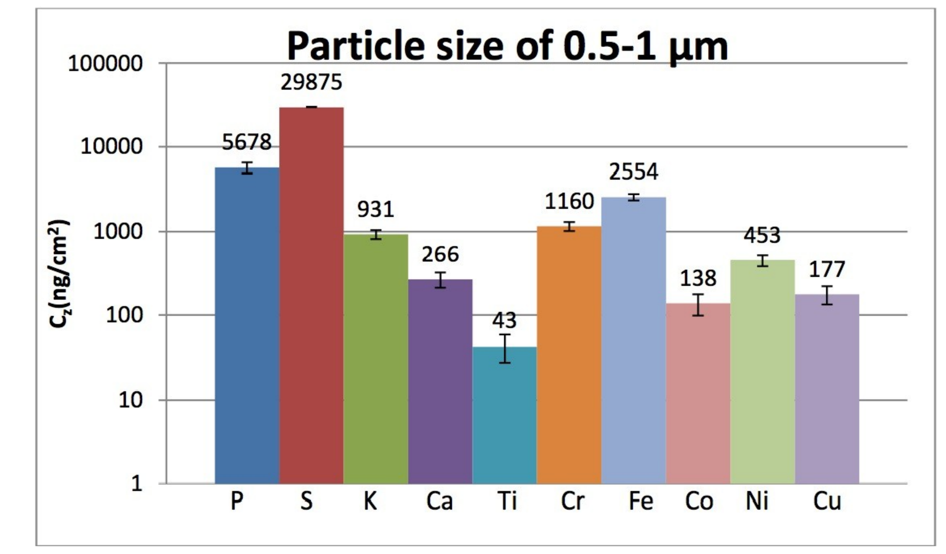

Figure 10: A bar graph of concentration in ng/cm^2 vs. element for particulate matter of 0.5-1 μm in size.

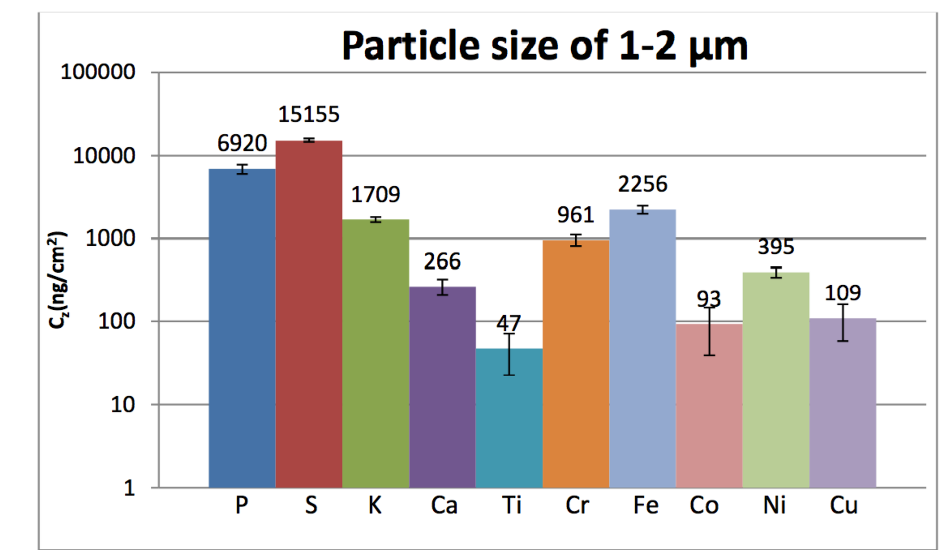

Figure 11: A bar graph of concentration in ng/cm^2 vs. element for particulate matter of 1-2 μm in size.

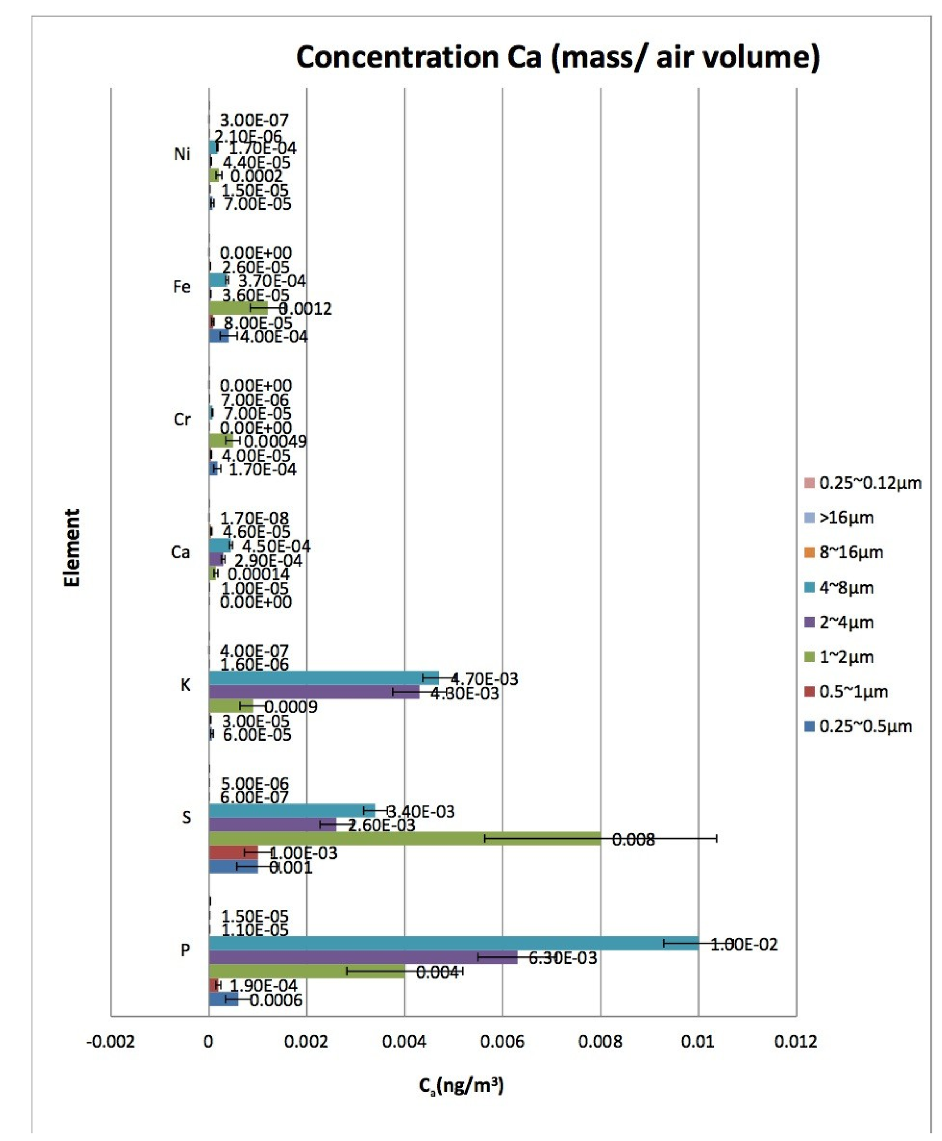

Figure 12: A bar graph of concentration in ng/m^3 of sampled air vs. element for all foils.

References

[1] Johansson, Sven, John Campbell, and Klas Malmqvist. *Particle Induced X-Ray Emission Spectrometry (PIXE)*. New York, NY: John Wiley & Sons, 1995.
[2] PIXE International Corporation, P.O. Box 2744, Tallahassee, FL 32316 U.S.A.
[3] MicroMatter Co., 18218 18th Ave. NW, Arlington, WA 98223, U.S.A.
[4] GUPIX, the versatile PIXE spectrum fitting software, University of Guelph.
[5] GeoPIXE, CSIRO Earth Science and Resource Engineering, Australia.
[6] D.D.Cohenetal. Nimb 109,218 (1996).