

Elemental concentrations in aerosol samples determined by Artificial Neural Networks from PIXE spectrum.

M.I. Dinator^(a), R. Correa^(b), I. Requena^(c) and J.R. Morales^(d)

^(a) *Universidad de Chile, Facultad de Ciencias, Departamento de Física
Las Palmeras 3425, Ñuñoa, Santiago, Chile.
mdinator@uchile.cl*

^(b) *Universidad Tecnológica Metropolitana, Departamento de Física
Av. José Pedro Alessandri 1242, Ñuñoa, Santiago Chile
rcorrea@utem.cl*

^(c) *Universidad de Granada, Departamento de Ciencias de la Computación e Inteligencia Artificial, España
Daniel Saucedo Aranda s/n, 18071 Granada, España
requena@decsai.ugr.es*

^(d) *Universidad de Chile, Facultad de Ciencias, Departamento de Física
Las Palmeras 3425, Ñuñoa, Santiago, Chile.
rmorales@uchile.cl*

Abstract. An application of artificial neural networks, ANN, has been performed to determine elemental concentrations in atmospheric aerosol samples. A feed-forward ANN was used in the learning phase. The network was trained with a back propagation algorithm in order to obtain the elemental concentrations in these samples. The inputs to the ANN analysis were experimental data obtained from the PIXE irradiations plus one from each element in the spectrum obtained directly in real time. The ANN output was the elemental concentration. A set of thirty seven PIXE spectra from aerosols collected in Santiago, Chile, provided the raw data which were used in the ANN application. Following a random selection, thirty spectra were used in the training phase, while the rest was used to test the ANN capability in the determination of unknown concentrations. Several networks of similar topology were independently trained for each one of the five elements considered in this study, Al, Si, S, K, and Fe. These networks operated in parallel, thus allowing a simultaneous determination of the elemental concentrations. The results were in good agreement with those obtained by standard PIXE analysis. This ANN application to the study of atmospheric aerosols has proved to be reliable and easy to extend to a large number of samples of similar characteristics.

Keywords: Artificial neural networks; PIXE, aerosols

INTRODUCTION

Application of PIXE (Proton Induced X-ray Emission) analysis to determine elemental concentration has proved to provide valuable data for environmental studies. These determinations rely heavily in the analysis of characteristic X-ray spectra, which is usually performed by the use of computational codes. In this work we present an application of artificial neural network (ANN) to obtain elemental concentrations from the spectra and experimental parameters.

The major motivation of this work is to get benefits from the ANN capability to give similar answers in shorter times than those obtained by common PIXE analysis. In fact, the experience of the authors has shown that the analysis of one PIXE spectrum with a code like AXIL [1], takes about 5 minutes to an analyst. Instead a trained ANN can perform the same analysis in milliseconds using the same computer hardware. Other ANN advantage is the possibility of manage a large number of samples.

The fundamentals of the ANN technique have been described elsewhere [2,3] and several commercial and free codes are accessible nowadays.

The uses of new information technologies around artificial intelligence, such as ANN, applied to atomic and nuclear physics has been developed and incremented during the last two decades [4]. Some applications of ANN to analyze alpha, gamma and X-ray spectra have been published in the last fifteen years [5,6,7].

PIXE is a nuclear accelerator based technique whose X-ray spectral analysis demands expert knowledge and significant calculation time to obtain the elemental concentrations of the samples [8, 9]. Although PIXE analysis has reached certain degree of automation, ANN technique could simplify and accelerate the process to get final results.

In this work ANN, has been applied to determine elemental concentrations in atmospheric aerosol samples. A set of thirty seven PIXE spectra from aerosols collected in Santiago, Chile, provided the raw data used in the ANN application. Following a random selection, thirty spectra were used in the training phase, while the rest was used to test the ANN capability to determine unknown concentrations.

ANALYSIS

The complete analysis of a sample implies two steps: PIXE and ANN.

PIXE Analysis

PIXE analyses were performed at the Center for Experimental Physics, Faculty of Science, University of Chile. Proton beams of 2 MeV were provided by a Van der Graaff accelerator. Details of the PIXE line setup has been given elsewhere [10].

Aerosols were obtained in downtown Santiago in spring and winter periods in year 2004. Thirty seven Teflon filters containing atmospheric aerosols were irradiated for 15 minutes with beam currents of 5 nano amperes.

Spectra were obtained with a Canberra Si(Li) detector having a resolution of 200 eV approximately and standard pulse electronic units and MCA. Thirteen elements were detected in most of the filters (Al, Si, S, K, Fe, P, Cl, Ca, Ti, Cr, Mn, Cu, and Zn). Figure 1 illustrates a typical spectrum. These X-ray spectra were analyzed using AXIL code provided by IAEA, which determines the number of counts in each peak.

The relative elemental concentration, C_Z , expressed by the ratio of the elemental surface mass density and the total mass per unit area of the sample, was obtained by the equation:

$$C_Z = \frac{I_Z(E_{K\alpha})}{M \cdot Q \cdot J(E_{K\alpha})}, \quad (1)$$

where $I_Z(E_{K\alpha})$ is the number of counts in the $K\alpha$ peak from the Z-element, M [g/cm^2] is the total mass surface density of the target, Q [C] is the integrated charge in the Faraday cup, and $J(E_{K\alpha})$ [$\text{g}^{-1} \text{cm}^2 \text{C}^{-1}$] is the value of the sensitivity function at the peak energy obtained from experimental data [11].

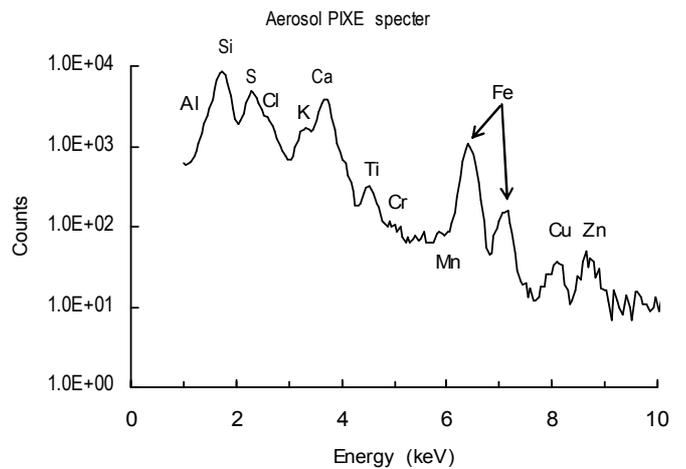


Figure 2 illustrates a flux diagram that shows the PIXE route and the experimental parameters used as inputs for the ANN analysis. Concentrations determined by PIXE analysis are only used in the ANN training phase.

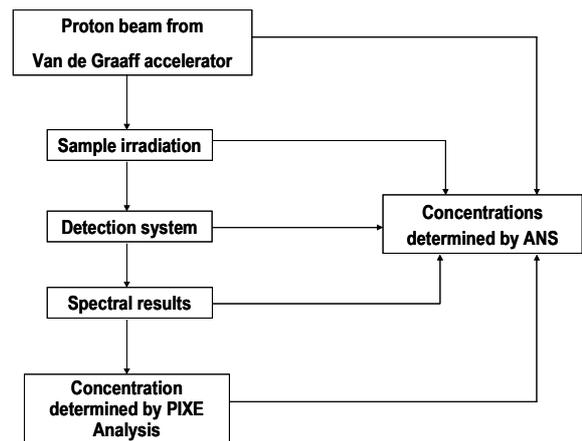


FIGURE 2. Flux diagram showing the relationship between PIXE experimental steps and ANN inputs

ANN Analysis

For each analyzed element in the sample, an ANS is designed. Each Artificial Neural System (ANS) has been constructed to determine the elemental concentrations in the analyzed aerosol samples. ANS is composed with five feed-forward ANN of similar topology (Figure 3), which operate in parallel inside the ANS. Each ANN was trained with well know back propagation algorithm in an independent process in order to learn first, and reproduce after, the process to obtain the concentrations of elements present in the samples. The input and output parameters in the training phase are the same for all ANN. The output value given by the ANS is obtained by combining the ANN outputs.

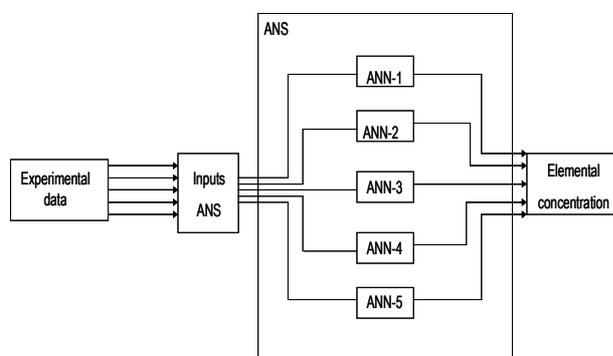


FIGURE 3. ANS spectral analysis schema for aerosol samples.

An ANN with three layers was used to generate the mapping of 4-input parameter values to produce 1-output value. The output is the concentration of the considered element present in the sample, while the inputs are: proton beam intensity impinging the sample (Q), total mass of the sample (m), area of the sample (S), and the element peak information obtained from the spectra (n). The mapping is carried out using the weights of the neuron connections, computed in the supervised training process. The structure of the employed ANS and the topology of the ANN are shown in Figure 4.

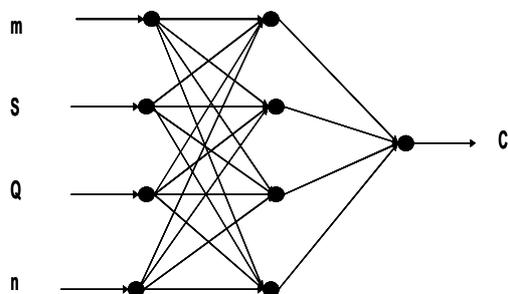


FIGURE 4. The topology of the ANN.

In the training phase, the least square error method was used in the back propagation algorithm.

The experience in ANN advises that the number of training cases must be much greater than the number of connections in order to guarantee the ANN learning [2]. In the architecture of this ANN, 20 connections were defined and their values were determined in the training process using 30 samples. After the training phase has been accomplished, a test procedure was performed verifying that the neural net effectively has learned

RESULTS AND CONCLUSIONS

Elemental concentrations were calculated by standard PIXE methodology in the way described above.

In this ANN application only the major elements, Al, Si, S, K and Fe observed in the spectra were considered. Even though in some samples trace elements were observed, the number of cases was so few that did not sustained the training of the network to obtain reliable concentrations. In order to determine trace element concentrations, more cases are required.

In ANN applications, detection limits depend on the range of experimental PIXE concentrations used in the training phase. Detection limits determined by PIXE can be included in ANN calculations in order to avoid eventual meaningless concentration values. Concerning uncertainties associated to ANN applications, in general these are determined by an overall comparison between experimental and ANN values used during the training and test processes. Here, results from PIXE, named as “expected concentrations”, C_{exp} and from ANN, denoted by C_{nn} , both from the training phase, were compared obtaining a linear correlation factor $r^2 = 0.96$ as is illustrated in Figure 5. Similarly, for the test phase a linear correlation factor $r^2 = 0.88$ was obtained and shown in figure 6.

In both cases, it is observed that the best results are obtained for high concentrations while the dispersion is rather large for low concentrations, as would be expected from statistical uncertainties in the corresponding peaks. C_{nn} and C_{exp} results for the test phase are given in table I.

According to these results ANN has been successfully applied to the analysis of major elements in aerosols samples. This ANN application has proved to be reliable and easily could be extended to a large number of samples of similar characteristics

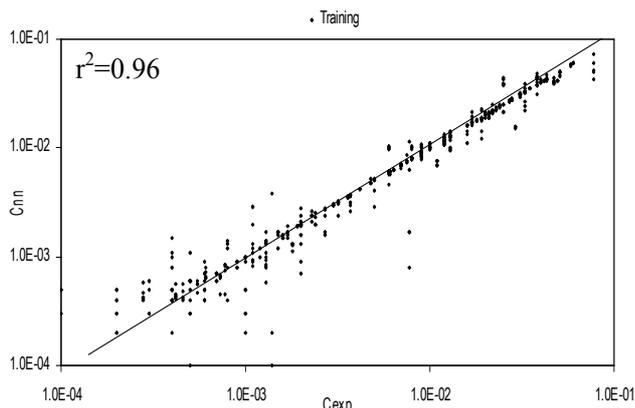


FIGURE 5. Predicted concentrations from ANS (C_{nn}) and expected concentration (C_{exp}) obtained from the training set

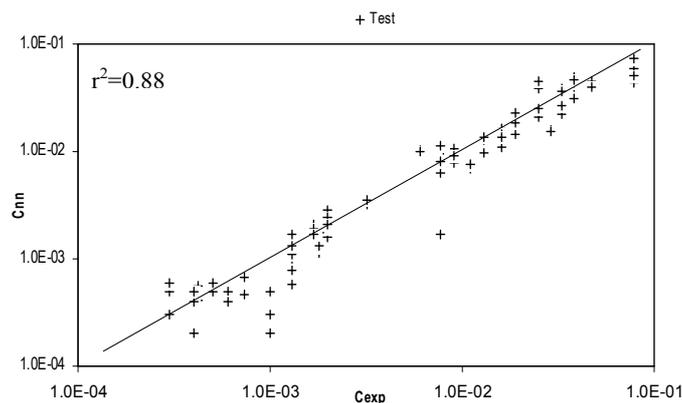


FIGURE 6. The predicted concentrations from ANS (C_{nn}) and expected concentrations (C_{exp}) in the test set.

TABLE I. Predicted concentrations (C_{nn}) from ANS analysis and expected concentrations (C_{exp}) in the test set

Aluminum - Al		Silicon - Si		Sulfur- S		Potassium- K		Iron - Fe	
C_{nn}	C_{exp}	C_{nn}	C_{exp}	C_{nn}	C_{exp}	C_{nn}	C_{exp}	C_{nn}	C_{exp}
%	%	%	%	%	%	%	%	%	%
4.2E-02	4.9E-02	2.8E-01	2.5E-01	3.0E-02	3.3E-02	1.0E-02	8.0E-03	4.6E-02	3.8E-02
4.3E-02	3.8E-02	2.9E-01	1.7E-01	1.9E-02	1.9E-02	1.0E-02	6.0E-03	2.9E-02	2.5E-02
3.9E-02	2.5E-02	1.3E-01	1.4E-01	1.3E-02	1.6E-02	9.5E-03	9.0E-03	4.1E-02	4.7E-02
1.6E-02	2.9E-02	1.1E-01	1.7E-01	1.1E-02	1.3E-02	7.4E-03	1.1E-02	5.1E-02	7.8E-02
1.3E-03	1.8E-03	7.6E-02	6.9E-02	7.0E-04	1.0E-03	6.0E-04	5.0E-04	2.2E-03	1.7E-03
6.0E-04	7.0E-04	1.5E-03	1.3E-03	5.0E-04	3.0E-04	5.0E-04	6.0E-04	1.4E-03	1.3E-03
8.0E-04	1.2E-03	1.5E-03	1.2E-03	4.0E-04	4.0E-04	2.1E-03	2.0E-03	4.5E-03	7.7E-03

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