

Background considerations in the analysis of PIXE spectra by Artificial Neural Systems.

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Abstract. In order to study the importance of background in PIXE spectra to determine elemental concentrations in atmospheric aerosols using artificial neural systems ANS, two independently trained ANS were constructed, one which considered as input the net number of counts in the peak, and another which included the background. In the training and validation phases thirty eight spectra of aerosols collected in Santiago, Chile, were used. In both cases the elemental concentration values were similar. This fact was due to the intrinsic characteristic of ANS operating with normalized values of the net and total number of counts under the peaks, something that was verified in the analysis of 172 spectra obtained from aerosols collected in Mexico city. Therefore, networks operating under the mode which include background can reduce time and cost when dealing with large number of samples.

1. Introduction.

Since implementation of the PIXE method by Johansson et al [1] it became one of the most used accelerator-based analytical method. Its capability to detect many elements simultaneously, roughly from Na to Pb, in concentration in the order of tens of parts per million, ppm, without significant damage to the sample, make it specially adequate to analyze low mass samples. One successful application has been the analyses of atmospheric aerosols. In environmental studies it is important to identify the most abundant elements in the sample and their concentrations. This knowledge permits the identification of natural and anthropogenic sources of aerosols, thus contributing to evaluate the air quality of the site where the samples were collected. Some countries have implemented systematic collection campaigns in cities and non urban sites. In these studies, a rather large amount of samples is collected for later irradiation by a proton beam in a PIXE laboratory. At least one energy spectrum is obtained for each sample requiring a careful analysis to identify the emitting atom and to determine its concentration. Usually, an analytical function fits the spectrum which is composed by peaks associated to each element detected in the sample, and by a continuous background covering the energy range accepted by the X-ray detector. The net number of counts in each peak, which is related to the elemental concentration, is calculated by integration around the peak and subtraction of background. In general, elemental concentrations are obtained by comparison with spectra given by targets of reference material with known concentrations. Some computational codes like AXIL, GUPIX, and others are used in the analysis of the spectra.

In previous works [2,3] we tested the application of artificial neural networks (ANN), to the



analysis of this type of spectrum from materials like atmospheric aerosols and organic tissues. Also, it has been shown that a configuration based on a system of networks presents some advantages with respect to the use of single networks. In this work an Artificial Neural System (ANS) was constructed with several feed-forward ANN of the similar topology and operating in parallel inside the ANS. Following the training phase, elemental concentrations are calculated in very short time, in the order of microseconds.

The determination of the net number of counts in each peak, require the subtraction of the counts in the background, implying an additional mathematical operation. Here, we show that when applied to the analysis of PIXE spectra, trained ANNs working with the total number of counts including background, or with the net number of counts in each peak, give statistically similar results of the elemental concentration.

Our results are based on the analyses of a large number of fine atmospheric aerosols samples, thirty eight collected in Santiago, Chile, and one hundred seventy two collected in the metropolitan area of Mexico city [4].

2.- Structure of the ANS

The construction of the ANS, followed the same pattern used in a previous application [3] with several ANS operating in parallel, one for each chemical element detected in the sample. In Figure 1 the neural system schema is shown. Several feed-forward ANN, of similar topology, operate in parallel inside each ANS trained with the well known back propagation algorithm in an independent process. The only difference is in the training process, so that the initial weights are different in each ANN, i.e., several ANN with the same topology and training set, are running at the same time. The ANS obtain the capacity to learn and apply this knowledge to unknown cases not included in the training phase.

The input and output parameters in the training phase are the same for all ANN in the ANS. Each ANS output value is obtained by combining the ANN outputs in the decision system. The advantages in the use of a system of networks rather than single networks have been already shown [3].

For each one of the selected element a map with four input parameters was generated in an ANN, with three layers in the ANS, in order to provide a single output parameter, i.e. the elemental concentration $C(Z)$. The four input parameters were: i) m , the deposited mass in the filter; ii) Q , the total electric charge transported by the ions while crossing through the filter during irradiation; iii) S , the size of the filter surface irradiated, and iv) $N(Z)$, the total number of counts in peak.

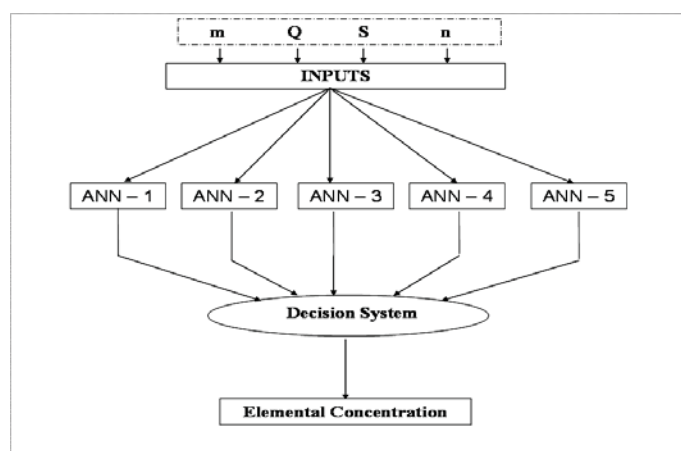


Figure 1. Schematic for ANS spectrum analysis for aerosol samples.

In the training process the weights of the connections between each neuron in the input layer with the hidden layer and between this one and the output layer are adjusted through an iterative process that minimize the error by a function considering the approximation between the network output and the expected answer. Now, if one decides to take as an input the net number of counts instead of the total number of counts, in the training process the network will learn to work under that condition, and the unknown concentration will be calculated accordingly.

Less computer work is required by taking the total number of counts rather than the net number of counts in the peak.

The architecture of the ANN has the topology shown in Figure 2, where a feed forward neural network has I neurons in the input layer, H neurons in the hidden layer and one neuron in the output layer. The aggregation function Σ_j in each neuron h_j in the hidden layer, $j = 1, \dots, H$, represents the scalar product between the input vector $X = (X_1, \dots, X_I)$ and the associated weight vector $W_j = (W_{j1}, \dots, W_{jI})$ where W_{ji} is the value of the connection between the neuron X_i in the input layer and the neuron H_j in the hidden layer. The activation function $f_j = f(\Sigma_j)$ in each hidden neuron is a sigmoid function of type

$$f_j = (1 + \exp(-\Sigma_j X W_j))^{-1} \quad (1)$$

representing the output value of each neuron in the hidden layer. In the output layer Σ_o represents the aggregation function of the outputs from the hidden layer and corresponds to the scalar product of the output vector in the hidden layer $f = (f_1, \dots, f_H)$ and the weight vector $W_o = (W_1, \dots, W_H)$ associated to the value of the connections between each neuron H_j of the hidden layer with the neuron O in the output layer.

The output value O is determined by a linear activation function depending on Σ_o .

$$O = \Sigma_o = \sum f W_o \quad (2)$$

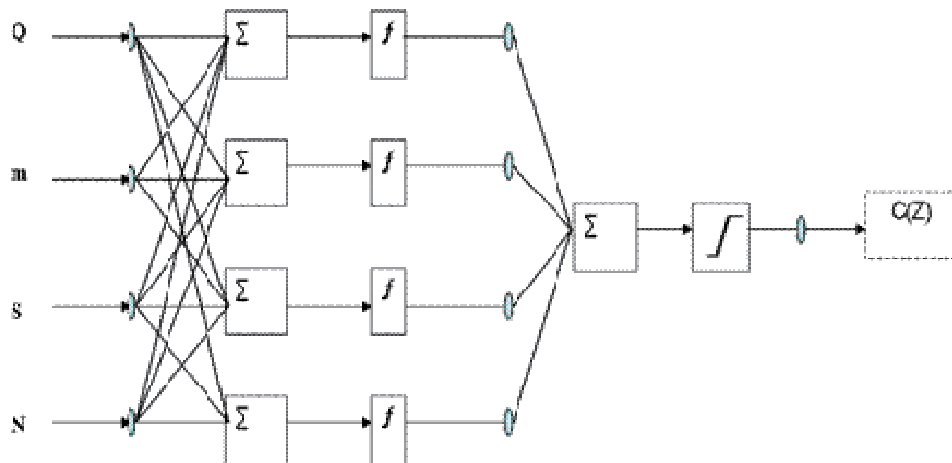


Figure 2. Architecture of ANN used inside ANS.

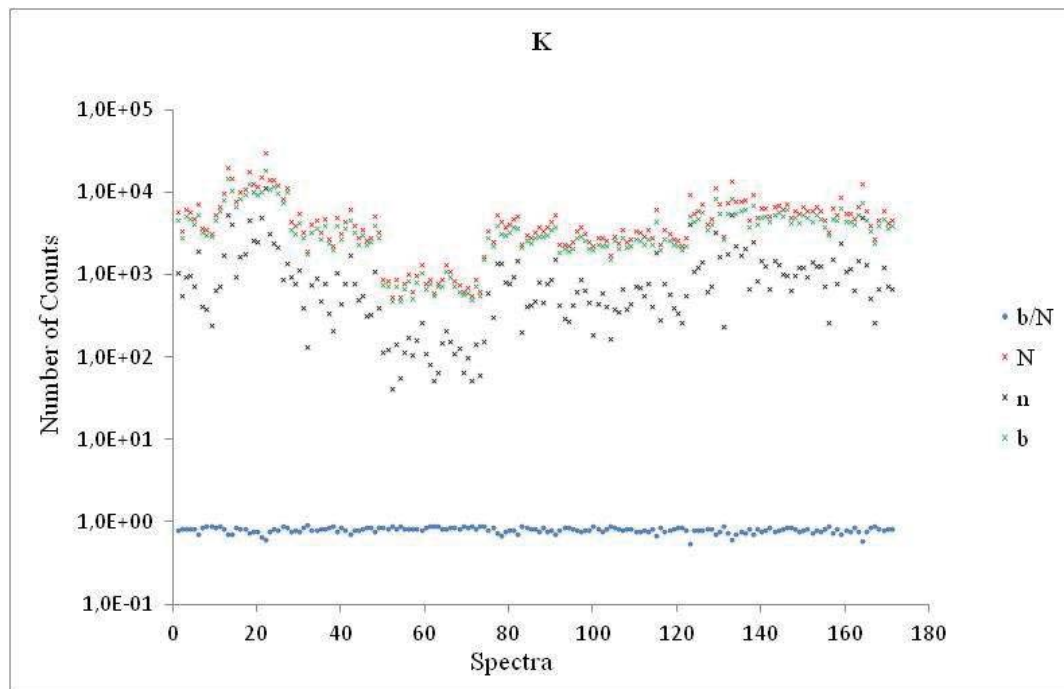
The neural networks used architecture with three layers having four input neurons, four hidden neurons and one output neuron. The input parameters form the vector (m, S, Q, N) and the network output is the concentration $C(Z)$ of the element detected in the sample using the

proper values of the inputs. Input parameters were normalized to the electric charge passing across the filter and collected at the Faraday cup during irradiation.

3. Background considerations.

In a typical PIXE spectrum the peaks lie on a continuum radiation background generated by the interactions of the incoming ions with electrons belonging to the atoms in the target, beam collimators, Faraday cup and eventually in the chamber walls. Effects like secondary electron bremsstrahlung, quasi free electron bremsstrahlung, atomic bremsstrahlung and ion bremsstrahlung produce a continuous radiation from zero to energies higher than those from K, L, or M transitions. Several authors have studied and calculated the probabilities associated to these effects. [5,6,7,8,9]. Prediction of peak to background ratio in gamma ray spectra have been performed using an artificial neural network [10].

In general, there are two contributions to the background under the peak, one is due to the interaction of the ions in the beam with collimators, Faraday cup, target holder, and the substrate to which the material under analysis is fixed, and secondly that produced by the interaction with the material itself. With an adequate experimental setup, the first contribution can be reduced to a minimum while the second would fluctuate with the mass deposited on the filter. This fact has been observed in the spectra considered in this work and is illustrated in Figure 3 for silicon and potassium, where the ratio between the number of counts in the background, b , to the total number of counts in the region of interest around the peak, N , for each spectrum, remains almost constant within a small statistical deviation. Defining this ratio as $B(Z) = b(Z)/N(Z)$, the mean values of the ratios for the major elements detected in the set and their corresponding standard deviation, σ , are presented in Table 1.



(a)

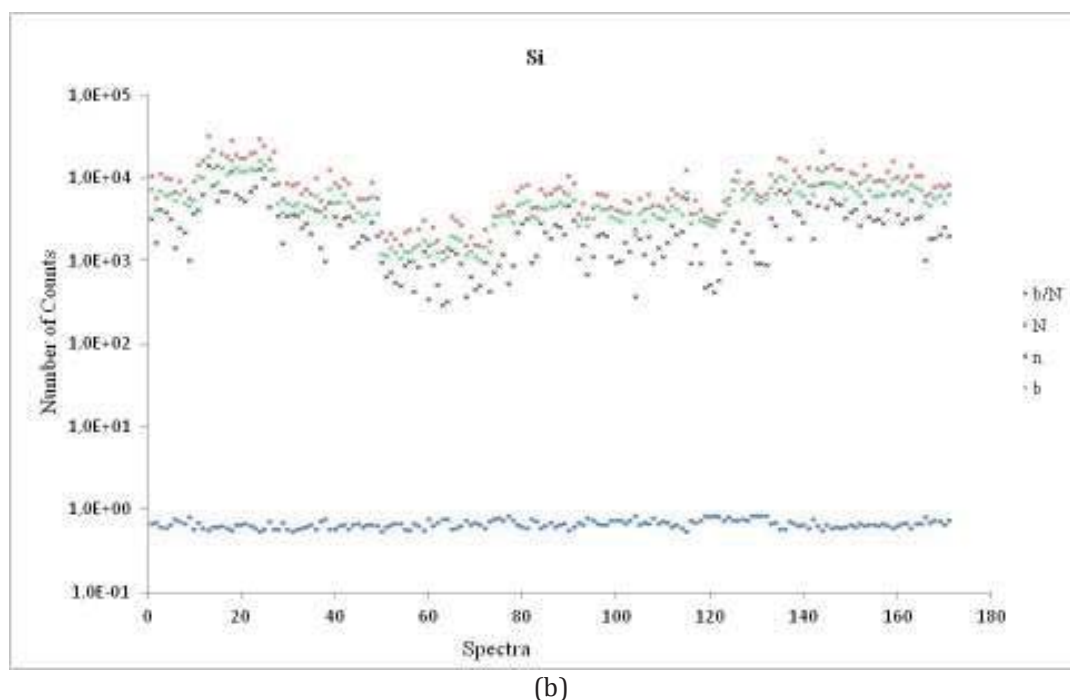


Figure 3 Illustrates ratio between the number of counts in the background, b , to the total number of counts in the region of interest around the peak, N , for all 172 spectra collected in the metropolitan area of Mexico city analyzed for (a) K and (b) Si

Table 1: shows the mean values $B(Z)_{\text{mean}}$ of the ratios for the major elements detected in the set and their corresponding standard deviation,

	$B(Z)_{\text{mean}}$	σ
Si	0.686	5.8×10^{-3}
S	0.692	6.0×10^{-3}
K	0.822	4.8×10^{-3}
Ca	0.574	6.0×10^{-3}
Fe	0.310	4.7×10^{-3}

In general, the total number of counts in the region of interest around a characteristic peak of each detected element, of atomic number Z , can be expressed as

$$N_i(Z) = n_i(Z) + b_i(Z) \quad (3)$$

where the subscript i denotes one of the spectrum in the set, $N_i(Z)$ corresponds to the total counts, $n_i(Z)$ represents the net number of counts and $b_i(Z)$ is the number of counts due to the background. In the evaluation of the elemental concentration, $C_i(Z)$, by standard methods, it is assumed that this value is proportional to the net number of counts in the peak.

Since ANS operates with values normalized, i.e. between 0 and 1, all inputs are normalized to the maximum value of the set. Then, we define the normalized values $N_i'(Z)$ and $n_i'(Z)$ as

$$N_i'(Z) = N_i(Z) / N(Z)_{\max} \quad (4)$$

and

$$n_i'(Z) = n_i(Z) / n(Z)_{\max} \quad (5)$$

Since it has been observed that for each element, $B_i(Z)$ fluctuates in a narrow range around the mean value $B(Z)_{\text{mean}}$, it can be expressed approximately as

$$B_i(Z) = B(Z)_{\text{mean}} \pm \sigma(Z) \quad (6)$$

From equations (6), $b_i(Z)$ can be expressed as

$$b_i(Z) \approx N_i(Z)(B(Z)_{\text{mean}} \pm \sigma(Z)) \quad (7)$$

and from equation (3)

$$n_i(Z) \approx N_i(Z)(1 - B(Z)_{\text{mean}} \pm \sigma(Z)) \quad (8)$$

where

$$k(Z) = 1 - B(Z)_{\text{mean}} \quad (9)$$

is a constant and then

$$n_i(Z) \approx N_i(Z)(k(Z) \pm \sigma(Z)) \quad (10)$$

So, the values $n_{\max}$ and $N_{\max}$ become related by a constant

$$n_{\max}(Z) \approx N_{\max}(Z)(k \pm \sigma(Z)) \quad (11)$$

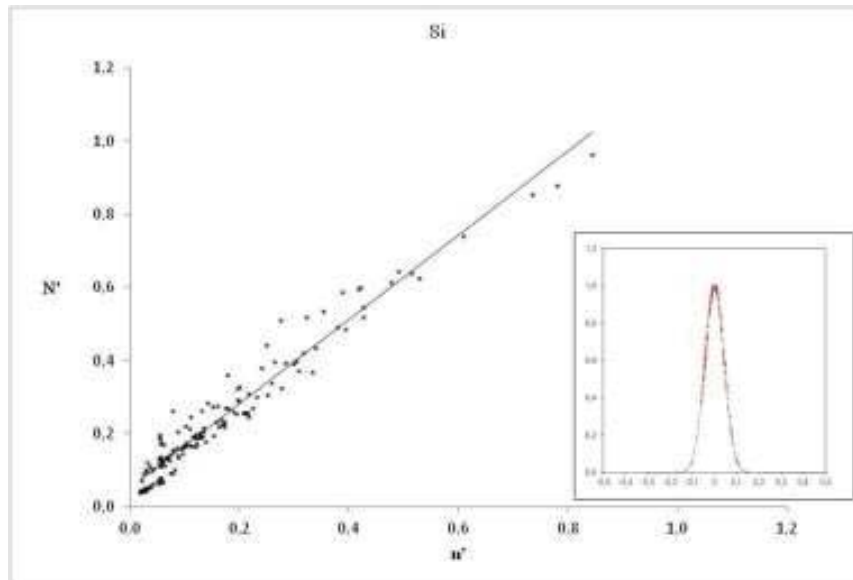
where

$$(k \pm \sigma(Z)) \approx n_{\max}(Z) / N_{\max}(Z) \quad (12)$$

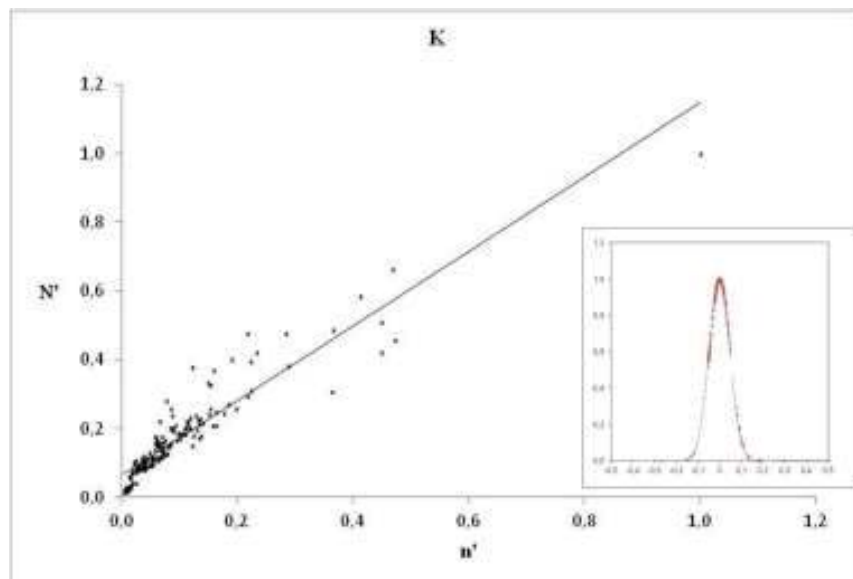
Combining with (10) it is found that the normalized values $n_i'(Z)$ and $N_i'(Z)$ are alike

$$n_i'(Z) \approx n_i(Z) / n(Z)_{\max} \approx N_i(Z) / N(Z)_{\max} \approx N_i'(Z) \quad (13)$$

The similarity between $n_i'(Z)$ and $N_i'(Z)$ was verified for the most abundant elements in 172 spectra of aerosols collected in Mexico city. The correlations between $n_i'(Z)$ and $N_i'(Z)$ for Si and K are shown in Figure 4, and for other elements are given in Table 2.



(a)



(b)

Figure 4 Correlation between the normalized total number of counts N' and the normalized net number of counts n' for Si and K are presented in (a) and (b) respectively. The residual distributions are shown in the inserted squares. A total of 172 spectra collected in the metropolitan area of Mexico city were considered.

In Table 2 the linear correlation factors r^2 and their standard errors σ for Si, S, K, Ca and Fe are given. This result supports the assumption that the neural network system provides similar elemental concentration values operating with either one of the normalized values. Values of

r^2 and σ show strong statistics dependence and relation linear between N' and n' for each element analyzed.

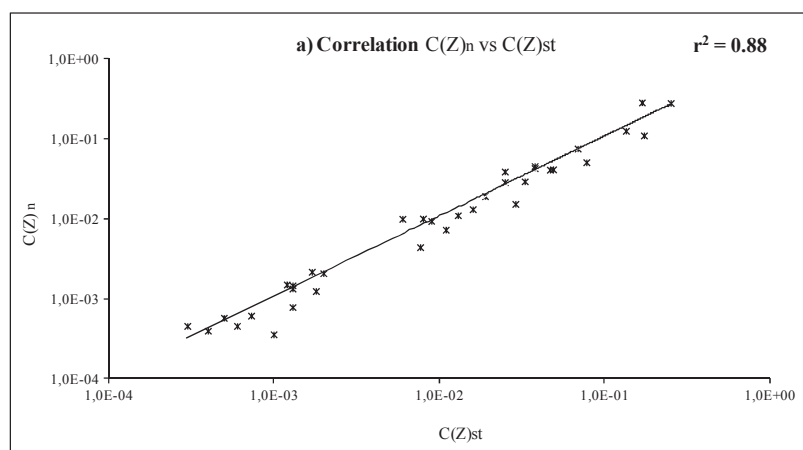
Table 2: Presents the linear correlation factors r^2 and their standard errors σ for Si, S, K, Ca, Fe

	r^2	σ
Si	0.95	4.07×10^{-2}
S	0.93	5.48×10^{-2}
K	0.87	4.84×10^{-2}
Ca	0.97	3.58×10^{-2}
Fe	0.99	3.19×10^{-2}

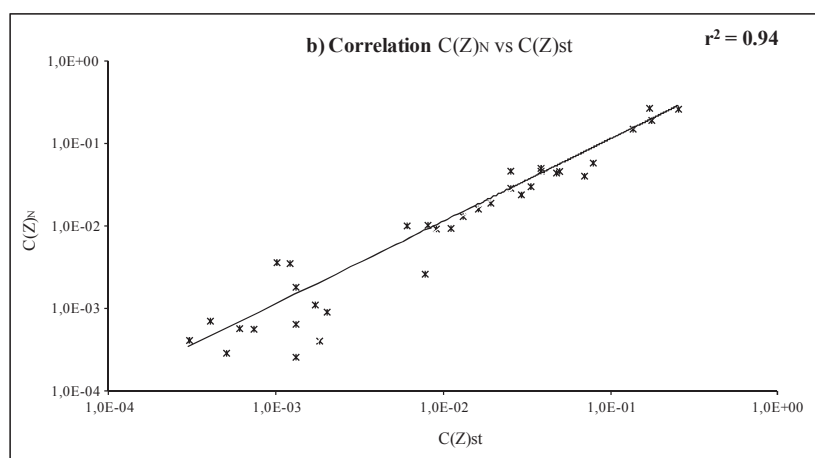
3. Results.-

In order to verify the above mentioned assumption, a different set of 38 aerosol PIXE spectra with known elemental concentrations, obtained by a standard method using the computer code AXIL [11] were taken as expected concentrations for ANS $C(Z)_{st}$ were reanalyzed by an ANS working either with the net number of counts or with the total number of counts. The comparison between $C(Z)_{st}$ and concentrations obtained by ANS using $n'(Z)$ or using $N'(Z)$, gave linear correlation factors of 0.88 and 0.94 respectively (See Figure 5). The correlation between normalized values $n'(Z)$ and $N'(Z)$, was verified with a set of 172 PIXE spectra of aerosols from Mexico city.

It is worth to mention that both sets of filters, those from Mexico city and those from Santiago, were irradiated in the same laboratory and under similar experimental conditions.



(a)



(b)

Figure 5 Concentrations of five elements, Si, S, K, Ca and Fe obtained by ANS using a) the net number of counts in the peak, b) and the total number of counts in the peak.

4. Conclusions.

Peaks associated to detected elements in aerosol PIXE spectra have been analyzed by artificial neuronal systems trained to operate with either the net number of counts or, with the integral number of counts. The elemental concentrations obtained in both cases turned out to agree quite well with concentration obtained by standard methods. This result is consistent with the fact that a constant added to the value of an input parameter in an artificial neuronal system do not affect the final results.

Networks operating under this mode can reduce time and cost when dealing with large number of samples.

Acknowledgments.

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