

Chemical characterization of dry deposition of pollutants in one industrial site in Guanajuato, Mexico

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Abstract— Dry deposition samples were collected in a weekly basis during 2010 in a site located at Central Mexico in the State of Guanajuato. Samples were collected with an automatic wet/dry deposition sampler using nylon filters as surrogate surfaces. Samples

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were extracted with deionized water and analyzed for pH, conductivity, Na^+ , K^+ , Ca^{2+} , Mg^{2+} , NH_4^+ , NO_3^- , SO_4^{2-} and Cl^- . The most abundant ions were K^+ , Ca^{2+} , SO_4^{2-} and NO_3^- , suggesting that the influence of local sources located at SW from the sampling site contributed with significant amounts of alkaline particles (from the cement plants) and acid gases precursors (from the power plant and oil refinery). From meteorological analysis, it could be observed that the sampling site was all time under the influence of the oil fired power plant. However, in spite of the high levels of NO_3^- and SO_4^{2-} found in the samples collected, pH values ranged between 5.1 and 6.3, with an average value of 5.66. From the quantification of the neutralization effect of the main alkaline components, the prevalence role of K^+ and Ca^{2+} was completely evident, suggesting the influence of the cement plants located upwind the sampling site. Dry deposition fluxes were estimated for all ions measured. Nitrogen dry deposition at Salamanca City is already in the upper extreme of the threshold value reported for sensible ecosystems. On the other hand, sulphur deposition exceeded slightly the critical load value reported for some ecosystems in Europe. Considering that results reported in this study not include the wet deposition fluxes, even at this moment, nitrogen and sulphur deposition is not a problem, it is necessary to take steps to avoid that total deposition of these elements exceeds critical loads.

Keywords— Combustoleo, Dry deposition, Dry fluxes, Mexico, Oil-fired power plants.

I. INTRODUCTION

ATMOSPHERIC pollutants are transferred to the ground by two pathways: dry and wet deposition. However, wet deposition can also occur by the contribution of other hydrometeors known as atmospheric deposits which are products of condensation of atmospheric water vapour [1]. On the other hand, dry deposition consists of gaseous particles and aerosols absorbed or deposited directly from atmosphere onto vegetation, soils, or another kinds of surfaces without any hydrometeors as a medium; this process is surface dependent [2]. Atmospheric deposition (wet deposition plus dry deposition) constitutes the last stage in the pathway of the acidifying pollutants from the source to the receptor. It has been demonstrated that the atmosphere is a significant contributor to the total pollutant load to many terrestrial and aquatic ecosystems.

The significance of the acid rain problem has been well established since several years ago, however, it is necessary to note that in the total load of acids to the surface (soils, vegetation and water bodies), precipitation is one component of this problem but not the unique, in such a way that dry deposition should be also considered.

The phenomenon of acid deposition has been largely studied around the world over last twenty years [3],[4], and it has been found that episodes of acid deposition are directly related to industrial emissions, being the coal and fuel burning power plants the main sources [5].

Fossil-fired power plants represent the larger contributor to acid rain of any industrial activity, since they are the larger source of sulphur and nitrogen oxides [6]. Fossil-fired power plants emit six common criteria air pollutants. These pollutants are by-products of electricity generation and include volatile organic compounds (VOCs), sulphur dioxide (SO₂), nitrogen oxides (NO_x), carbon monoxide (CO), lead and particulate matter less than 10 microns in diameter (PM₁₀) [7].

In Mexico, 72 % of generated energy comes from burning of fossil fuels, being “combustoleo” (a blend of residua and gas oil cutter stock) the fuel more used in the main productive sectors. Combustion process of “combustoleo” results in large amounts of gases such as SO₂ and NO_x that contribute to the acid deposition in the surroundings of the plants [8].

Studies about critical loads have been carried out in specific ecosystems using different sensitive species as indicators of atmospheric pollution [9],[10],[11],[12],[13],[14], the most of them have been carried out in the European countries and in the United States. However, in the case of Mexico, only few studies including wet deposition has been carried out, whereas some studies have been limited to understand the role of atmospheric deposition in the biogeochemical cycles [15].

In the other hand, it is necessary to take in account that the deposition patterns and the ecosystems responses may be different from those reported for mid-latitudes. Furthermore, eco-tourism activities constitute an important source of economic resources in Mexico, therefore, it is essential to provide an outline that let to determine if critical loads are exceeded, to assess annual trends and for demonstrating the efficiency of new environmental policies about atmospheric pollution. This information will let to the environmental authorities in Mexico to establish target loads considering the degree of ecosystem protection desired and the time needed to achieve this protection.

The present study aims to investigate the composition of dry deposition in one site located in the surroundings of a power plant burning “combustoleo”.

II. MATERIALS AND METHODS

A. Study Area

Samples were collected during autumn and winter seasons of 2010 at Salamanca City in the State of Guanajuato, located at central part of Mexico (Fig.1). The main characteristics of the sampling site are shown in Table I.

Table I. Main characteristics of the study area.

Altitude m a.s.l.	1720
Latitude North	20° 59'
Longitude East	101° 17'
Average population	789,548
Effective capacity installed of the power plant	866MW



Fig.1. Location of the study area.

This region is a complex site where total atmospheric inputs to the ecosystems come from mixed sources. Intensive agricultural activities are developed at the south of the city, whereas, in the north region, there is an important industrial zone that includes a fuel-fired power plant, an oil refinery and two cement plants. Considering that the industrial activities developed in Salamanca City result in great amounts of atmospheric pollutants, this city has been classified as a critical zone by the Mexican

environmental regulations, for this reason, authorities carry out intensive and continuous watching programmes, and monitoring of air quality; however, dry and wet deposition are not included as a part of this monitoring network.

Climate in Salamanca City is sub-humid warm with rains occurring along the summer season; however, rains are scarce with an annual average precipitation of 700 mm. For this reason, this study only reports data for dry deposition.

A portable meteorological station (Davies Inc) was operating during the whole study period. From these meteorological data, wind roses were constructed by using WRPLOT VIEW 6.5.2 (Lakes Environmental Inc). To trace the origin of the air masses, backward trajectories were calculated by using HYSPLIT model from US-NOAA. From wind roses and backward trajectories, it was observed that prevailing winds came from SW, just from the power plant and the oil refinery.

B. Sampling Procedure

When The sampling was performed by automated wet-dry deposition collector (Tisch). The sampler was equipped with two polyethylene buckets and a lid controlled by a rain sensor, which moves depending on the beginning and the end of the rain event. In this work only dry deposition was sampled because the rain events were scarce during 2010 in Salamanca City.

Samples in the dry bucket were collected in a weekly basis by using a surrogate surface according to the method used by Alonso *et al* [16]. Nylon filters (Nylasorb, Pall Corporation; 47 mm diameter and 1 μm pore-size) were used as surrogate surfaces, and they were exposed in horizontal position (using a filter support) inside dry bucket. Each week, samples were sent to the laboratory, where filters were ultrasonically extracted with 80 ml of deionized water and filtered through 0.45 μm cellulose membrane filters. The extracts were stored at 4°C until analysis, which was performed within three weeks after collection.

C. Chemical Analysis

pH and conductivity were determined immediately by using precision pHmeter Termo Orion 290 and a conductometer CL 135. Cl^- , NO_3^- , SO_4^{2-} were analyzed by Ion Chromatography (Agilent 1100) coupled to a conductivity detector (Alltech).

Na^+ , K^+ , Ca^{2+} and Mg^{2+} were determined by Atomic Absorption Spectroscopy with the Flame Technique (Thermo-scientific ICE 3300). NH_4^+ was determined colorimetrically using a spectrophotometer UV (Hach DR 2800).

The detection limits were estimated as three times the standard deviation of six blank samples. Repeatability was determined by analysis of samples from at least 2 replicate measurements. The quality of the analytical data was checked out by a cation-anion balance (Fig. 2) and by comparing measured conductivity with the conductivity calculated (from the concentration of all measured ions and their specific conductivities).

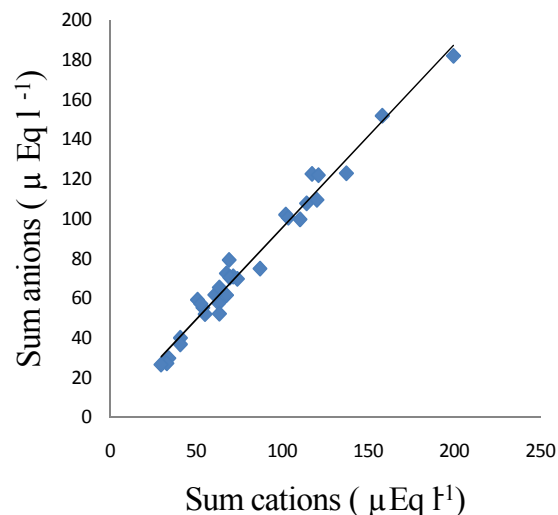


Fig. 2. Quality control (ionic balance) of chemical analysis for dry deposition at the study site.

From Fig. 2, it can be observed that practically the majority of the ions were analyzed. In addition, the measured conductivities were in agreement with the calculated conductivities.

III. RESULTS

The results obtained from analysis of dry deposition at Salamanca site are presented in Table II. It can be observed that pH values ranged from 5.1 to 6.3 with an average value of 5.66. The relative ionic abundance was: $\text{K}^+ > \text{SO}_4^{2-} > \text{NO}_3^- > \text{Ca}^{2+} > \text{NH}_4^+ > \text{Mg}^{2+} > \text{Na}^+ > \text{Cl}^-$.

Fig. 3 shows the frequency distribution for pH, it can be observed that the most of the collected samples showed pH values almost neutral.

Table II. Summary statistics for dry deposition in Salamanca site.

Ions	Concentration ($\mu\text{Eq l}^{-1}$)			Standard deviation
	Minimum	Maximum	Mean	

Na ⁺	0.01	2.17	0.62	0.63
K ⁺	25.83	174.17	69.66	35.5
Ca ²⁺	0	20.45	5.45	5.43
Mg ²⁺	1.65	6.58	2.69	1.18
NH ₄ ⁺	0	18.29	2.72	4.49
Cl ⁻	0.03	1.89	0.59	0.51
SO ₄ ²⁻	16.44	105.93	49.71	22.76
NO ₃ ⁻	9.84	75.32	27.82	16.1
pH	5.1	6.3	5.7	0.28

The contribution percentage of cationic components (Na⁺, K⁺, Ca²⁺, Mg²⁺ and NH₄⁺) towards the total ion mass balance was 43%, and acidifying components (SO₄²⁻, Cl⁻, and NO₃⁻) accounted 57% of total ionic mass.

Fig. 4 shows the individual contribution of different ions towards the total ionic mass. K⁺ was the dominant cation contributing with 38% to the total ionic mass. Among anions, SO₄²⁻ was the predominant anion followed by NO₃⁻, contributing with 32.5% and 24% of the total ionic mass, respectively.

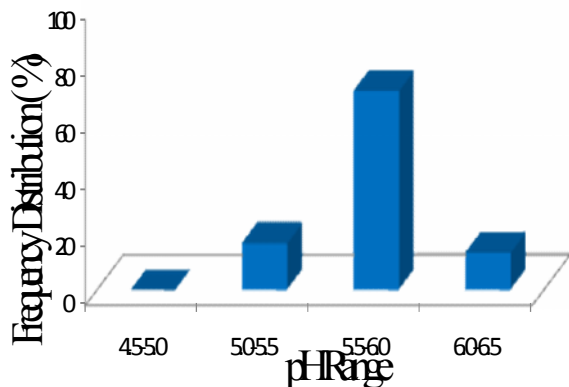


Fig. 3. Frequency distribution for pH at the study site (%).

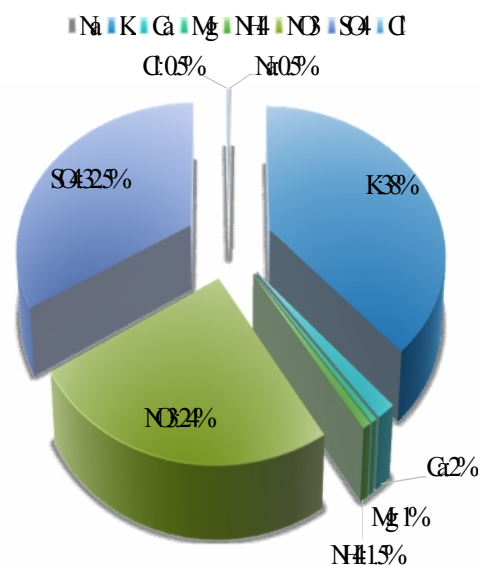


Fig. 4. Contribution of different ionic species toward the total ionic mass.

From Table II, it can be observed that sulphate levels (49.71 $\mu\text{Eq l}^{-1}$) exceeded almost 5 times the hemispheric values (10 $\mu\text{Eq l}^{-1}$) reported for remote sites by Galloway *et al* [17]; it suggests that there were a significant influence of local and regional sources that contributed with great amounts of this ion.

In addition, nitrate levels (27.82 $\mu\text{Eq l}^{-1}$) exceeded almost ten times the hemispheric values (2.8 $\mu\text{Eq l}^{-1}$), it is agree with the residence times of SO₂ and NO₂ in the atmosphere. SO₂ has a regional contribution due to it is commonly subjected to long-range transport, whereas NO₂ is removed in the surroundings of the source and it is well known as a local pollutant. It suggests that there was an important local source contributing to these nitrate levels. K⁺ was the most abundant ion, and it was evident that this ion played an important role in the neutralization process, resulting in an average pH value almost neutral.

Considering that Salamanca is not a coastal site, it can be assumed that the presence of SO₄²⁻ and NO₃⁻ was associated to the emissions from the power plant and the oil refinery, it is agree with the prevail winds. From Fig. 5 and Fig. 6, it can be observed that prevailing winds came from SW during the study period. In addition, to trace the origin of the air masses during the study period, air-mass backward trajectories were obtained daily 48 h before using NOAA HYSPLIT Model. It was observed that during the study period, air masses came from SW, just from the power plant located near the sampling site.

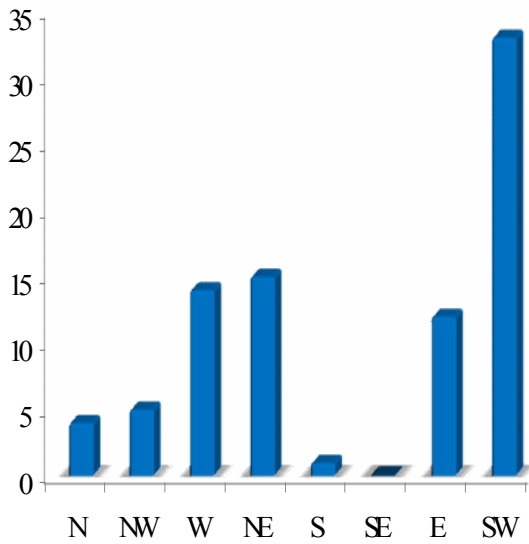


Figure 5. Wind direction frequency distribution for Salamanca during the study period.

The origin of the elements in atmospheric deposition samples can be inferred from correlation between ion pairs, identifying common sources, similar removal process or mechanisms, and interactions acid-base.

Table III. Correlation matrix of ions in dry deposition at the study site.

	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	NH ₄ ⁺	NO ₃ ⁻	SO ₄ ²⁻	Cl ⁻
Na ⁺	1							
K ⁺	-0.1	1						
Ca ²⁺	0.62	0.55	1					
Mg ²⁺	0.04	0.68	0.47	1				
NH ₄ ⁺	-0.2	0.04	-0.2	0.05	1			
NO ₃ ⁻	0.04	0.93	0.62	0.74	0.05	1		
SO ₄ ²⁻	-0.1	0.95	0.50	0.61	0.17	0.84	1	
Cl ⁻	0.97	-0.1	0.68	0.09	-0.2	0.08	-0.1	1

From Table III, it can be observed a good correlation between K⁺-NO₃⁻ and K⁺-SO₄²⁻; it suggests an important relationship acid-base between these ion pairs, and that these compounds were deposited as KNO₃ and K₂SO₄ as a result of neutralization process between alkaline particles and strong acids. In spite of alkaline particles could come from crustal (soil particles), it is important consider that the two cement plants located upwind the sampling site could contributed with significant amounts of Ca²⁺ and K⁺.

On the other hand, from Table III, it also can be observed an evident relationship between NO₃⁻ and SO₄²⁻, demonstrating that both ions had a common source, probably combustion processes. It is supported by the analysis of time trends shown in Fig.7.

In spite of, nitrate and sulphate levels were high and even higher than hemispheric values, the contribution of alkaline particles was significant and played an important role: K⁺ > Ca²⁺ > NH₄⁺ > Mg²⁺ > Na⁺.

The interaction between cations and anions can be described by the neutralization factor (NF). (1):

$$NF_x = \left[\frac{X}{(NO_3^-) + (SO_4^{2-})} \right] \quad (1)$$

Where X is the component of interest, (NO₃⁻) and (SO₄²⁻) are the concentrations for nitrate and sulphate ions, respectively.

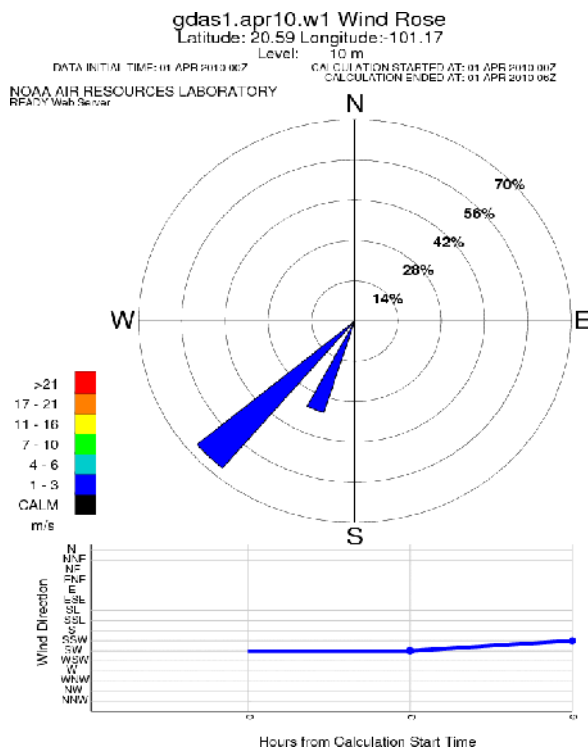


Fig. 6. Typical wind rose for Salamanca during the study period.

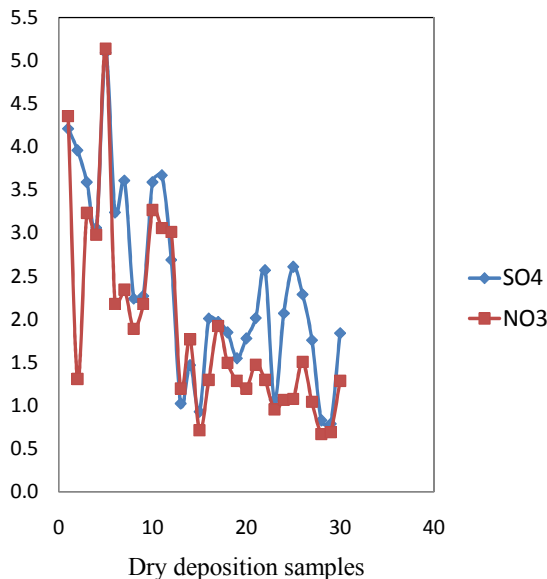


Fig. 7. Time-trends (concentrations in $\mu\text{Eq l}^{-1}$), of NO_3^- and SO_4^{2-} in dry deposition at the study site.

To verify which cation (Ca^{2+} , K^+ and NH_4^+) more frequently neutralized the acidic components in dry deposition, neutralization factors (NF) were estimated and a triangular diagram was drawn (Fig. 8). The triangle relating the neutralization factor of Ca^{2+} , K^+ and NH_4^+ revealed that Ca^{2+} was the predominant neutralizer. It is well known that the high concentrations of Ca^{2+} are related to the surface soil and mining activities.

Neutralization factors demonstrated that calcium played an important role in the neutralization process, suggesting probably the presence of $\text{Ca}(\text{NO}_3)_2$ and CaSO_4 in the atmosphere, originating from alkaline particles derived from the earth crust and the cement plants, and from SO_2 and NO_x emitted in combustion processes in power plants.

Factor analysis is a useful statistical tool and it can be applied condensing the information contained in a number of variables into a smaller set of new composite dimensions. Principal component analysis (PCA), a linear method for multivariate ordination was used to visualize the relationship among trace elements, focused to identify the possible sources of the major ions. ANOVA test by permutation were performed in order to test the significance of the factors to explain the found variations.

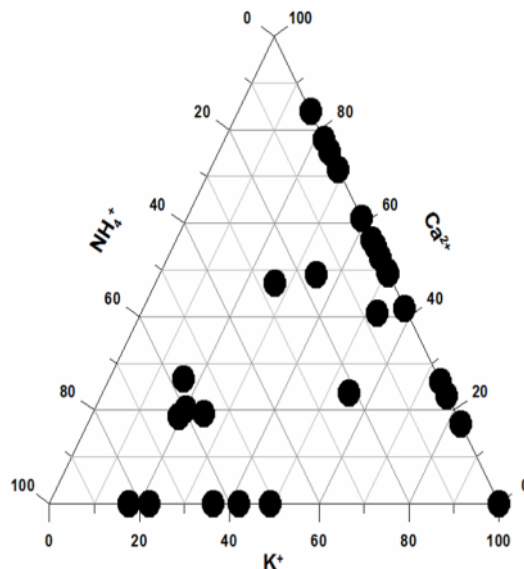


Fig. 8. Triangular diagram relating neutralization factor (NF) of Ca^{2+} , K^+ and NH_4^+ .

As can be observed from Fig. 9, there were a significant relationship between Na^+ and Cl^- , in addition, it can be observed the evident influence of alkaline ions like K^+ and Mg^{2+} on the neutralization process of strong acids.

It is necessary to know the ionic concentration levels of deposition for estimating its dynamic effect on natural ecosystems. However, the deposition fluxes and patterns of the species of interest constitute the most important parameters to be considered to assess the real pollution effect in a specific area.

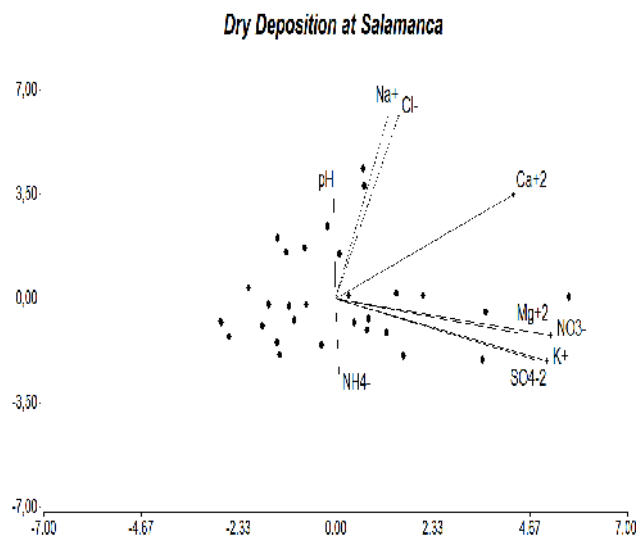


Fig. 9. PCA results for factor identification for analytical data of Salamanca City.

From Fig. 10, it can be observed that dry deposition fluxes showed a great variability. However, K^+ , SO_4^{2-} ,

NO_3^- and Ca^{2+} exhibited the highest rates of dry deposition. It is well known that the dry deposition is generally more connected with local emissions, contrary to rain, which better reflects long range transport [8]. It suggests that the sampling site probably was under the influence of local sources located at SW, in this case, power plant, oil refinery and cement plants.

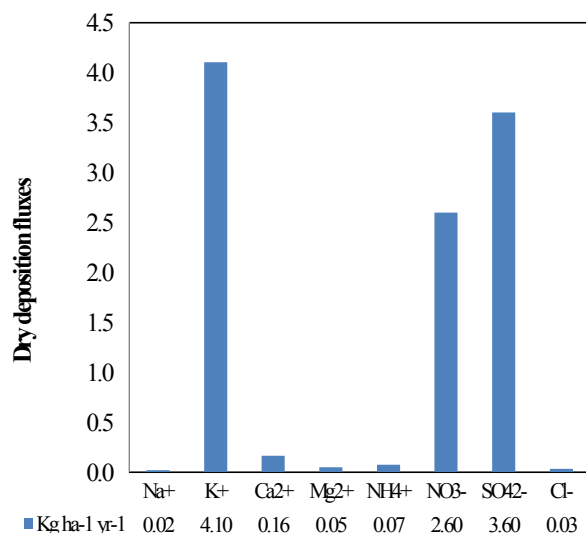


Fig. 10. Dry deposition fluxes for Salamanca City.

Neutralization factors demonstrated that calcium played an important role in the neutralization process, originating from the earth crust, and the emissions from cement plants and combustion processes.

Critical loads have been defined as the amount of one chemical compound that one ecosystem can tolerate before show damages [14]. Several studies about critical loads for N and S using sensitive species have been reported, however, in Mexico, only a few studies in pine wood and oak forests in Mexico Valley have been carried out, and data about critical loads are not available.

From Fig. 10, it can be observed that nitrogen deposition in Salamanca City is lower than $5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, the critical load value reported for sensitive ecosystems in Europe [11]. However, considering that the hemispheric values reported for tropical remote sites [17] range between 1 to $3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, dry deposition at Salamanca City is already in the upper extreme of this threshold value. Even at this moment, nitrogen deposition is not a problem, it is necessary to take steps to avoid that nitrogen deposition exceeds these critical loads. In addition, it is necessary to mention that this study only reported dry deposition data since rain events were scarce during the study period.

Critical loads used as reference values in this discussion correspond to total deposit (wet + dry deposition), therefore, fluxes reported in Figure 10 are

sub-estimated since the contribution of wet deposition was not considered.

On the other hand, sulphur deposition at Salamanca exceeded slightly the critical load value reported by Grennfelt and Nilsson [14] for sensitive ecosystems ($3 \text{ kg S ha}^{-1} \text{ yr}^{-1}$). In addition, the hemispheric value reported by Galloway et al [17] for remote sites ($0.5 \text{ kg S ha}^{-1} \text{ yr}^{-1}$) was exceeded almost 7 times, it suggests that sulphur deposition is already an environmental problem in this site, considering that the contribution by long range transport of wet deposition was not included.

IV. CONCLUSIONS

The study of dry deposition in Salamanca, showed a high content of K^+ and Ca^{2+} as the dominant cations and a high content of SO_4^{2-} followed by NO_3^- , as the most important anions.

Even though, there are two important industrial facilities (the power plant and the oil refinery) within this city and it could be expected that the dry deposition would be acidic, the contribution of alkaline components was significant resulting in dry deposition with pH values relatively neutral. It means that in Salamanca City, there is a complex environment with mixed sources driving the atmospheric chemistry.

However, it is necessary to appoint that pH itself, it is not a conservative tracer of the potential ecological effects as a result of the deposition process of atmospheric pollutants. In spite of, pH values obtained in dry deposition samples were not in the acid range, the deposition fluxes of S and N represent a threat to the ecosystems in this site.

It is necessary to carry out more studies focused on critical loads and measurements of wet and dry deposition during at least three years, it will let to set a baseline to understand the atmospheric chemistry in this site. In addition, it is necessary to achieve monitoring programs to assess trends and to set goals that let to carry out an adequate environmental management of this site.

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