

## **Relations between chemical variables in Acid Mine Drainage process: An Application of Fuzzy Clustering Algorithms to the Characterization**

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### **ABSTRACT**

In the present work, Acid Mine Drainage processes in the Chorrito Stream, which flows into the Cobica River (Iberian Pyrite Belt, Southwest Spain) are characterized by means of clustering techniques based on fuzzy logic. Also, pH behavior in contrast to precipitation is clearly explained, proving that the influence of rainfall inputs on the acidity and, as a result, on the metal load of a riverbed undergoing Acid Mine Drainage processes highly depends on the moment when it occurs. In general, the riverbed dynamic behavior is the response to the sum of instant stimuli produced by isolated rainfall, the seasonal memory depending on the moment of the target hydrological year and, finally, the own inertia of the river basin, as a result of an accumulation process caused by age-long mining activity.

### **INTRODUCTION**

Since their inception in the mid-1960s by Professor Lofti A. Zadeh from the University of California, Berkeley fuzzy sets have triggered mixed feelings among the scientific community. On the one hand, there has been a growing number of devotees who have early recognized the potential of fuzzy sets to model and solve many real world problems. On the other hand, however, there has been quite a considerable number of opponents – quite often prominent scholars and researchers – who have fiercely fought against these emerging new tools. One of their arguments has been its lack of applications.

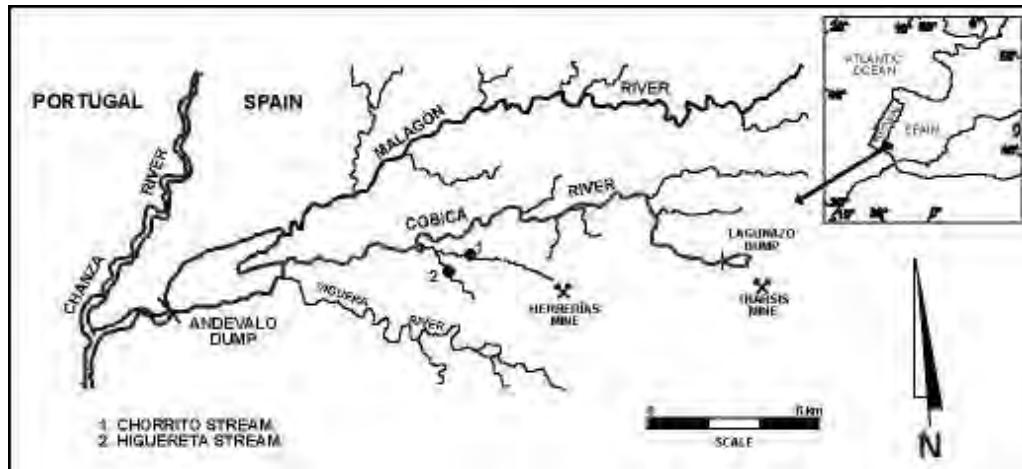
The situation has changed since the mid 1980s when the so-called “fuzzy boom” occurred, primarily in Japan, but then also in Korea and Europe, much less so in the USA. Basically, the turning point was the launching on the market of fuzzy logic control based appliances and other equipment exemplified by subway trains, cranes, elevators, etc. They had primarily been successful applications of fuzzy logic control originated by Mamdani 1974, Takagi (Takagi and Sugeno 1985), Sugeno (Sugeno and Yasukawa 1993) and other people.

### **AMD processes**

AMD processes are one of the most hazardous types of water pollution due to their nature, extent and difficult solution (Azcue 1999), as well as to their remediation economic costs (Commonwealth of Pennsylvania 1994). Rivers affected by this type of contamination are characterized by their acidity, as well as by the high sulfate and heavy metal content in their waters, and by the metal content of their sediments. The caused damage ranges from sub-lethal alterations for some individuals of the affected ecosystems in the cases of very low pollution, with associated problems of bioaccumulation and biomagnification (Nebel and Wrigth 1999), to the disappearance of the river fauna, and the loss of water resources as water becomes useless for human, agricultural or industrial use (Sáinz *et al.* 2002 ).

### **GENERAL SETTINGS**

This work was developed starting from the analysis of water samples taken at the Chorrito Stream. This stream flows into the Cobica River and is located in the SW of Spain, very close to the border with Portugal. The Chorrito Stream is a typically mining channel that receives discharges from a abandoned mine operation: Herrerías Mine. (Figure 1).



**Figure 1: Location map**

The channel regime in the environment of the Chorrito stream is directly associated to precipitation given the impermeable nature of the outcropping rocks in the basin. This fact gives the channel its practically torrential characteristics with large floods in winter and almost null flow in low water periods (Grande *et al.* 2003). Mining liquid pollution arriving to the Cobica watercourse coming from the Chorrito stream is produced by mine underground water, waste dump leachate and general washout of all pyrite waste dispersed through the numerous mining operations. AMD process undergone by the drainage network in the regional environment has been widely described for the Tinto and Odiel rivers by different authors (Borrego 1992 ; Borrego *et al.* 2002 ; Braungardt *et al.* 1998, Davis *et al.* 2000; Elbaz-Poulichet *et al.* 1999-2000-2001; Leblanc *et al.* 2000; Sáinz 1999; Sáinz *et al.* 2002-2003-2004; Grande *et al.* 2000-2003).

Geologically, the described sector is located in the Iberian Pyrite Belt, in the Spanish South-West, with numerous huge and over-huge Paleozoic massive sulfide deposits, the biggest in the world according to Sáez *et al* 1999. The massive sulfide bodies contain pyrite to which sphalerite, galena and chalcopyrite and other minor stages are associated (Sáez *et al.* 1999). These deposits have been exploited for over 2000 years.

## OBJECTIVES AND METHODS

The present work aims at characterizing AMD processes in the Chorrito stream, which flows into the Cobica River. For this purpose, fuzzy logic-based clustering techniques will be used (Aroba 2003).

### Sampling and analytical methodology

The sampling campaign in the Chorrito Stream was carried out daily from December 2000 to March 2001 during the rainy period, when water runs along both streams, and was finished when watercourses stopped running in the target sector. At the sampling time, pH and conductivity were measured *in situ* using a CRISON pH meter 507 equipment, and a CRISON conductivity meter 524 equipment, respectively. After pH and conductivity field measurements, a sample was taken to determine sulfate and another one to determine heavy metals.

### Analytical procedures

Metal concentrations in sample waters were determined by Atomic absorption spectrometry using flame FAAS (Fe, Cu, Mn and Zn).

Sulfates were determined by Ion Chromatography with Chemical Suppression of Eluant Conductivity (Standard Methods, 4110). Water samples were filtered with nylon 0.22-0.45 µm (Millipore, Millex Ref. SLCR 013) filters disc.

### Fuzzy logic and data mining

Fuzzy logic (Zadeh 1965) operates with reasoning rules which are very close to the human approximate and intuitive way of thinking. The main characteristic of fuzzy logic is that it allows us to define values without specifying a precise value, something which is not possible with classical logic, upon which computer development has been based so far. In classical logic, the membership to one class or set is binary, i.e., one is either member or not, so that only two precise values are worked with (1 and 0, yes or no). Thus, if "very low pH" is defined for some samples, it is evident that a sample with pH = 2 belongs to the cluster and another one with pH = 6 does not, but how do we classify a sample with pH = 4.99? It is precisely in the answer to this kind of questions where classical logic shows its limitations.

Fuzzy logic allows us to associate each sample with a certain degree of fulfillment of the "very low pH" prototype. This grade is called "membership grade"  $\mu_{VLpH}(x)$  of the element  $x \in X$  to the set "very low pH". The set  $X$  is called universe of discourse –range of values– of the variable  $x$ . The range of  $\mu_{VLpH}$  ranges from 0 to 1, each value representing the absolute non-membership or membership to the set, respectively.

The membership grade may be represented by a function (von Altrock 1995). Fuzzy sets are a generalization of traditional sets. The  $\mu_{VLPH}(x) = 0$  and  $\mu_{VLPH}(x) = 1$  cases which would correspond to conventional sets, are just special cases of fuzzy sets. The use of fuzzy sets defined by means of membership functions in logic expressions is called *fuzzy logic*. In these expressions, the membership grade of a set is the degree of certainty of the sentence. The geometric form of membership functions is totally arbitrary, but in general, simple geometry and known equation functions, such as trapeziums, triangles or sigmoids, are used. Once all variables involved in the problem are coded to the qualitative domain by means of membership functions, it is possible to write a set of rules representing the relation between input and output variables. These rules present the format *if-then*, and are made up of an antecedent and a consequent.

The process of extracting knowledge from a database is called KDD (*Knowledge Discovery in Databases*). This process is made up of several stages ranging from data preparation to achievement of results (Fallad and Uthurusamy 1996, Zaïane 1999). One of these stages is called *data mining* and can be defined as the non-trivial process of extracting implicit, *a priori* unknown useful information from the stored data (Holsheimer and Siebes 1994).

#### **The computer tool PreFuRGe: Predictive Fuzzy Rules Generator (Aroba 2003)**

Classical clustering algorithms generate a partition of the population in a way that each case is assigned to a cluster  $c$ . These algorithms use the so called “rigid partition” derived from the classical sets theory: the elements of the partition matrix (with  $n$  elements) obtained from the data matrix can only contain values 0 or 1; with zero indicating null membership and one indicating whole membership to each one of the  $c$  partitions. That is, the elements must fulfill:

- (a)  $\mu_{ik} \in \{0,1\}, 1 \leq i \leq c, 1 \leq k \leq n$
- (b)  $\sum_{i=1}^c \mu_{ik} = 1, 1 \leq k \leq n$  (3)
- (c)  $0 < \sum_{i=1}^c \mu_{ik} < n, 1 \leq i \leq c$

Fuzzy partition is a generalization of the previous one, so that it holds the same conditions and restraints for its elements, except that in this case real values between zero and one are allowed (partial membership grade).

Therefore, samples may belong to more than one group, so that the selecting and clustering capacity of the samples increases. From this we can deduce that the elements of a fuzzy partition fulfill the conditions given in (3), except that now condition (a) will be written as:

$$\mu_{ik} \in [0,1], 1 \leq i \leq c, 1 \leq k \leq n \quad (4)$$

The best known general-purpose fuzzy clustering algorithm is the so called *Fuzzy C-Means* (FCM) (Bezdeck 1981). It is based on the minimization of distances between two points (data) and the prototypes of cluster centres (*c-means*). The rule used for measuring distances is the Euclidean rule.

The described algorithm was used (Sugeno and Yasukama 1993) to build a fuzzy model based on rules of the form:

$$R' : \text{IF } \mathbf{x} \in \mathbf{A}' \text{ THEN } y \in B' \quad (5)$$

Where  $\mathbf{x} = (x_1, x_2, \dots, x_n) \in \mathfrak{R}^n$  are input variables,  $\mathbf{A} = (A_1, A_2, \dots, A_n)$  are  $n$  fuzzy sets,  $y \in \mathfrak{R}$  is the output variable and  $B$  is the fuzzy set for this variable.

The developed computer tool, *PreFuRGe* (Aroba 2003), is based on the previously described methodology (Sugeno and Yasukama 1993) and represented by (5).

#### **RESULTS**

The results obtained by means of data analysis with the PreFuRGe tool are shown in figures 2, 3, 4, 5 and 6. In figures 2, 3, 4, and 5, the universe of discourse of the rainfall variable is normalized and divided into 6 partitions.

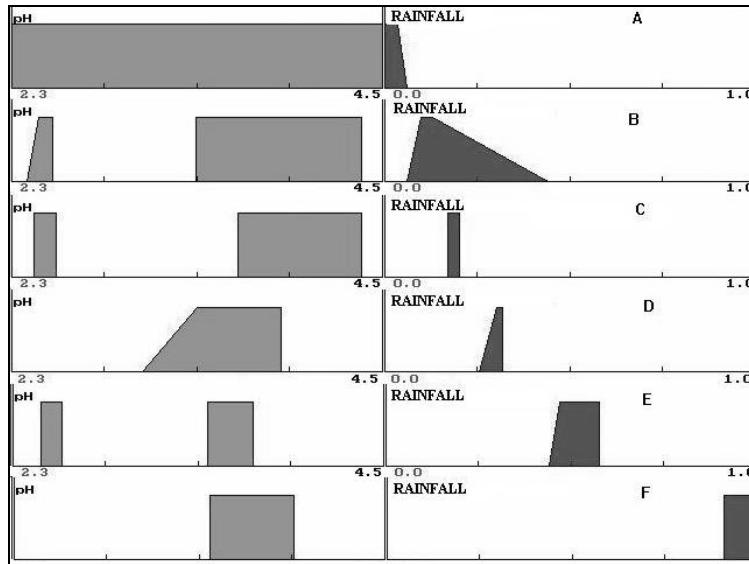


Figure 2

In Figure 2, the response of pH mean values with respect to the partitioned rainfall values is shown. The existence of non linear interdependency reasons is evident, with a higher definition as the target rainfall sector becomes higher, i.e., for extremely high rainfall values (Figure 2f), pH takes specific values which, contrary to what would be expected in these environments, are not the highest in the studied group of values. At the same time, the extremely low sector (Figure 2a) of rainfall may correspond with any pH value in water, therefore showing no apparent relationship between both variables. Precipitation central values (Figures 2b, 2c, 2e) do not provide decisive information, but it is worth noting that in almost all of them a similar response in pH is obtained, which becomes specific in the coexistence of two distant sectors for a specific rainfall interval. Thus, for rainfall mean values we can find either very low or average-high pH values, but no intermediate values. For interpreting the described phenomenon, in this work we fall back on the clustering of rainfall, together with the date it occurred, which will be analyzed in Figure 6.

Figure 3 shows the response of conductivity mean values with respect to partitioned precipitation values. Conductivity values are extremely low in 0.6 ( $\mu\text{S}/\text{cm}$ ) and extremely high in 5.9 ( $\mu\text{S}/\text{cm}$ ). In Figure 3a, precipitation extremely low values match a wide range of conductivity values, from the average-low limit to very high. Note that precipitation values within this sector comprise days with null rainfall and days with minimum –but different from null, rainfall.

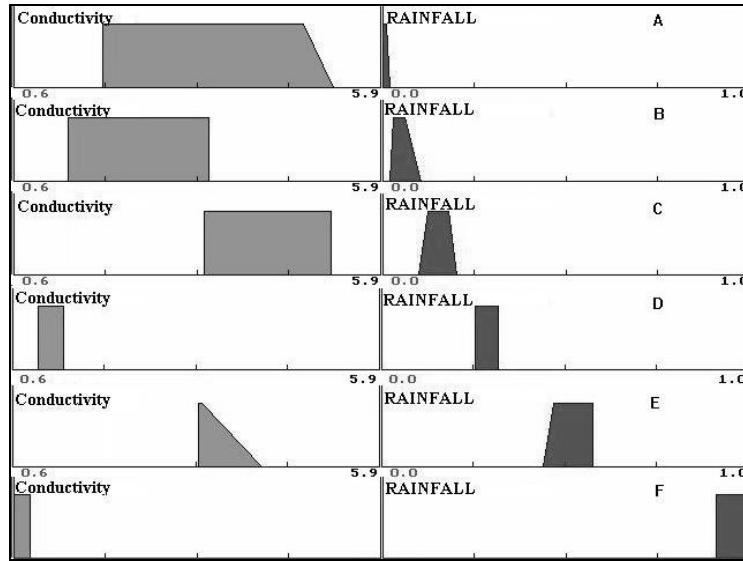


Figure 3

In Figure 3b we can observe that the immediate higher sector of precipitation match conductivity values which occupy a narrower and lower interval than the previous. But when we observe Figure 3c, with rainfall close to the low to average-low limit, the behavior is quite different: now conductivity takes clearly higher values (and can reach the highest values in the whole sampling space).

Figure 3f proves that the highest precipitations match the lowest conductivities, but not the other way round.

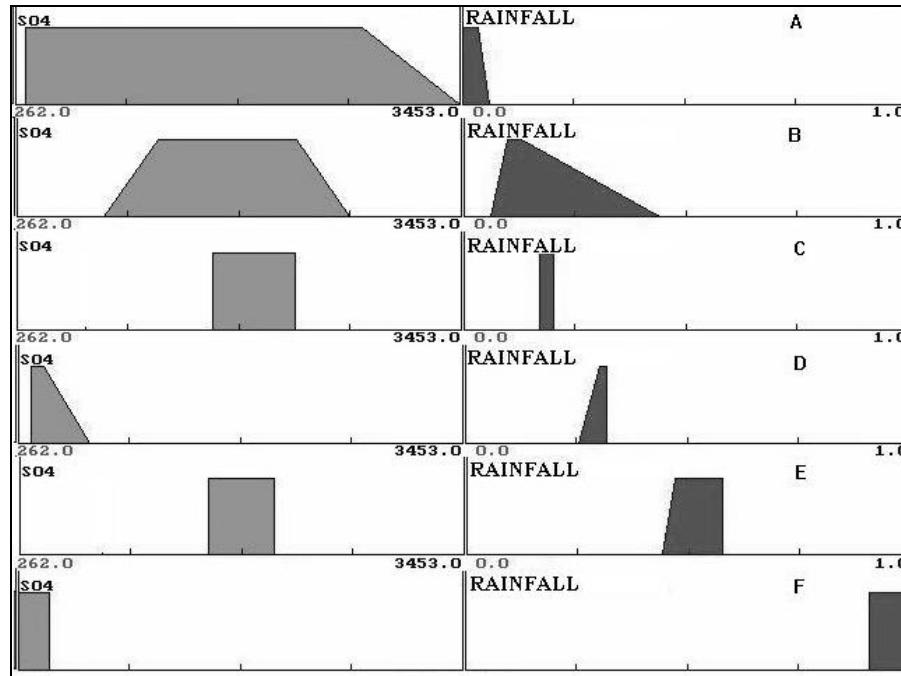


Figure 4

Figure 4 shows the response of  $\text{SO}_4$  mean values with respect to the partitioned rainfall values.  $\text{SO}_4$  values are extremely low in 262 ( $\text{mg l}^{-1}$ ) and extremely high in 3453 ( $\text{mg l}^{-1}$ ). In Figure 4f, rainfall extremely high values match extremely low values for sulfate concentration with a very precise adjustment. Conversely, when considering the extremely low value for rainfall (Figure 4a), sulfates can take any value. Therefore, we are again in a situation similar to that occurred for pH and conductivity. It must be emphasized again that precipitation values for the target sector comprise both null rainfall days and minimum rainfall days regardless of the moment they occurred. Figure 5 shows the joint response of all sampled variables: pH, Zn, Cu, Fe,  $\text{SO}_4$ , Mn and conductivity, with respect to the partitioned precipitation values.

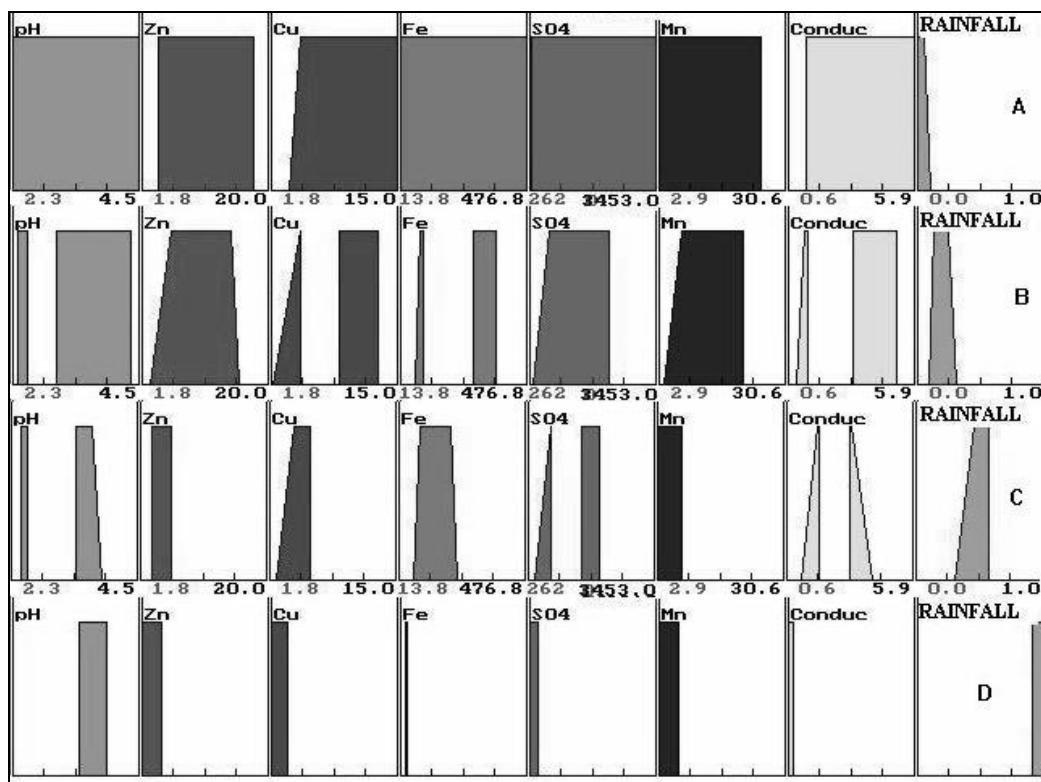
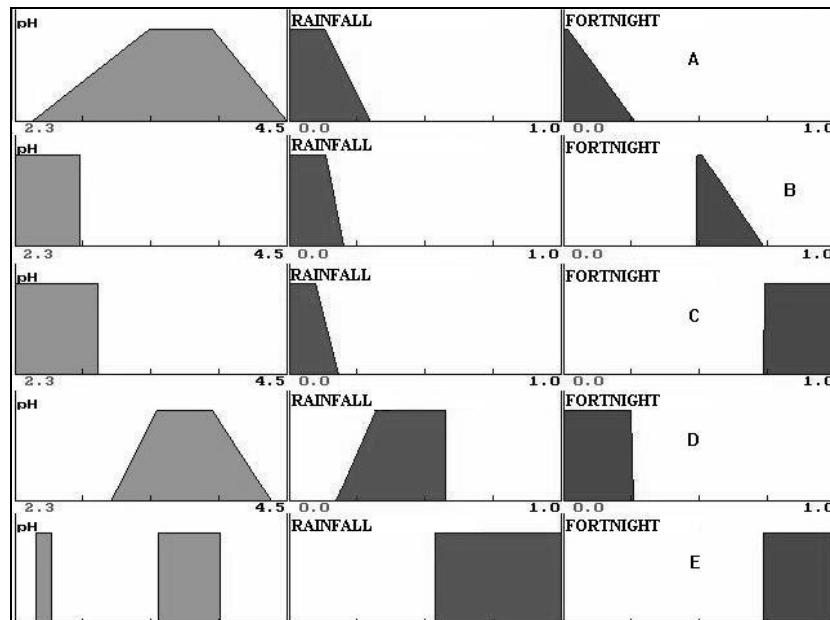


Figure 5

For extremely low or null precipitation (Figure 5a), we observe an almost total dispersion of the values taken by the studied variables, which may present any value along the universe of discourse.

The rest of rules shown in Figure 5 offer an overall view of the response of the analyzed variables in contrast to the "rainfall" stimulus, considered as a whole. Thus, as we considered an interval of higher precipitation, a higher definition of this response around increasingly more concentrated values becomes evident. Note that for extremely high rainfall values all metals show minimum and highly defined concentrations, whereas pH is over its mean value but not at its maximum value.



**Figure 6**

Figure 6 shows clusterized precipitation data together with the dates when they occurred. Each tuple has a consequent (the target rainfall fortnight) and two antecedents (precipitation and pH). Fortnight periods correspond to the following dates and figures: 1<sup>st</sup> fortnight, 23/12/00 to 06/01/01, figure 6a; 2<sup>nd</sup> fortnight, 07/01/01 to 21/01/01, figure 6b; 3<sup>rd</sup> fortnight, 22/01/01 to 05/02/01, figure 6c; 4<sup>th</sup> fortnight, 06/02/01 to 20/02/01, figure 6d; and 5<sup>th</sup> fortnight, 21/02/01 to 06/03/01, figure 6e.

It can be observed that low precipitation periods occur at very different dates which consequently generate very different pH values. Thus, in Figure 6a, which corresponds to the cluster of the first sampling fortnight (December/January), coinciding with the first rains of the hydrological year, pH takes values grouped within the average-high sector with few points towards lower and higher values. However, Figures 6b and 6c, corresponding to the penultimate and last sampling fortnights, when the last precipitations of the hydrological year occurred, coinciding with the end of rainfall and the coming of the dry period and, as a result, the stoppage of water flow into the river, show pH values concentrated at the low end.

Figure 6d shows that average-low rainfall values in the first and second fortnights generate pH values concentrated at the average-high end, with few points towards higher and lower values.

Chart 6e proves that the high precipitation occurred at the end of the sampling period generate highly specific pH values centered at very low values, but not at the far end, and average-high values.

## DISCUSSION

The review of antecedents leads to the general proposal of how AMD river basins operate in accordance with the following outline:

Rainfall extremely low values provide closely grouped pH values in the extremely low sector. However, the response achieved in Figure 2a is clearly contrary to what would be expected, where any pH value for the target sector of rainfall values is obtained. Justification of this phenomenon could lay on the fact that, in this figure, rainfall data are not ordered temporally, so that consideration of null rainfall may correspond to either a period where it has not rained for a long time or to another where rain has just stopped falling after a period of continuous raining.

To this must be added that at the same precipitation low end, null rainfall days are grouped with minimum rainfall values. Part of these values match the first rains after drought and intense meteorization of dumps, with abundant highly meteorized surfaces and the subsequent existence of easily leachable and first annual rainfall-sensitive oxide crusts and salts. It seems therefore evident that days with very different hydrochemical meanings are included in the interval of very low rainfall. As a result, these precipitations may provoke any pH value, as shown by Figure 2a.

In order to explain the apparently anomalous behavior of pH/rainfall relationships shown in Figure 2a, where rainfall extremely low values match any pH value, chart 6 shows clusterized precipitation together with the time period when it occurred. Here we try to establish cause-effect relationships according to the moment when rainfall occurred. Thus, minimum rainfall values at the end of the sampling period match pH values ranging from low to extremely low, as would be expected with scarce rainfall (Figures 6b and 6c). However, non trivial data contained in this figure is supported above all by Figure 6a, where it can be observed that scarce precipitation occurred at the beginning of the sampling period provokes any pH value, although most values are found in the average-high zone of the range. All this allows us to explain the apparent incongruence of Figure 2a, because if we superpose low precipitation values in Figures 6a, 6b and 6c, we obtain the pH values in Chart 2a.

In essence, the first annual rains provoke the leaching of meteorization surficial crusts, which in turn provoke an instant pH decrease, after which a dilution of the river water occurs, caused by the massive water discharge to the riverbed. This translates into an increase of pH to values close to 4.5. From this moment on, a normal hydrological year for this type of climate provokes a progressive decrease of pH from this value up to values close to 2.3 (Sáinz *et al.* 2003) where it remains stabilized along the whole dry period and until the next hydrological year, when the cycle is again repeated.

Chart 2b allows us to explain the process more precisely: the largest density of points, centroid of the fuzzy set (trapeze ceiling) correspond to scarce though not null rainfall days, which provoke a pH that can be minimum (with very concrete values) or from average to maximum. The tool recognizes a multiple projection of pH values in contrast to rainfall values corresponding to a single partition. This is interpreted as a result of the fact that the target rainfall amounts correspond to different rainy seasons which, consequently, generate different pH. The same reasoning is suggested from the interpretation of figures 2c and 2e.

Figure 5 shows the response of the studied variables as a whole in contrast with the stimulus "rainfall", and allows us to establish the following overall operational model of the river basin: null or minimum precipitation (Figure 5a) provokes the coexistence of values scattered along the whole universe of discourse for the rest of the studied variables, which is interpreted as the response of the environment to similar rainfall amounts occurred at different moments, in which the available ionic load in the environment is radically different and dependent on: a) total concentration of already leached solutes incorporated to the riverbed that, in turn, along with the flow, define the inertia of the receptor system; b) the dilution factor provoked by the rain that occurred at each moment.

## CONCLUSIONS

In the described context, the PreFuRGe computer tool used in this work gains a dimension of remarkable efficiency for the qualitative overall diagnosis of the situation, and can also be applied for establishing cause-effect relationships that, in contrast with classical statistical treatments, improve work considerably and make the understanding of the involved processes easier.

The application of fuzzy logic and data mining for characterizing AMD processes in the same sector and from the same mass of data confirm and enrich operational models previously proposed for this sector by means of multivariate analysis.

Multivariate (rainfall + time) fuzzy clustering used in this work allows us to clearly explain the behavior of pH in contrast to rainfall, which has not been possible up to date without the application of fuzzy clustering on one single parameter (rainfall), proving that the influence of rainfall inputs on the acidity of a riverbed undergoing AMD processes and, as a result, on the metal load, depends largely on the moment when it occurred.

In general, the dynamic behavior of the riverbed responds to the sum of instant stimuli produced by isolated rainfall, the seasonal memory depending on the moment of the target hydrological year and, finally, the own inertia of the river basin, as a result of an accumulation process caused by age-long mining activity.

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