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Chemometrical Analysis of 18 Metallic and Nonmetallic Elements Found in Honeys Sold in France

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The elemental analysis of 86 honeys sold in France was performed with an inductively coupled plasma atomic emission spectrometer in order to measure significant concentrations of Ag, Ca, Cr, Co, Cu, Fe, Li, Mg, Mn, Mo, P, S, Zn, Al, Cd, Hg, Ni, and Pb. Principal component analysis, correspondence factor analysis, and hierarchical cluster analysis were used to rationalize and interpret the analytical data. Crude relationships were found between the elemental profiles of the honeys and their botanical origin. Some honeys were highly polluted by heavy metals and/or other xenobiotics. Explanations for these contaminations are proposed.

KEYWORDS: Honey; heavy metals; elemental analysis; ICP-AES; multivariate analysis; health risk

INTRODUCTION

In France, between 80 000 and 100 000 individuals own a total of approximately 1 500 000 beehives (*1*). Professional and semiprofessional beekeepers represent 1500−2000 and 5000−6000 people, respectively. They hold approximately 900 000−1 000 000 hives. The rest are held by amateur beekeepers. The minimum number of hives required for a beekeeper to be considered a professional is 400 (figure legally fixed). These professionals are farmers possessing an apiary in addition to another agricultural activity or specialist bee-farmers practicing the rearing of honeybees. Many are newcomers attracted to the "natural way of living" who try to produce their honey as traditionally as possible. It is also worth mentioning that a retired professional may continue with up to 80 hives (figure also legally fixed) (*1*). The semiprofessionals are largely workers or employees who use beekeeping as a complement in revenue, often not negligible. The amateurs are found in all the sociological categories of French society (*1*).

Only honey produced by professionals and semiprofessionals enters into the commercial circuit, either by direct farm sales and markets or via hypermarkets, supermarkets, or other types of stores (*1*). The honey produced by amateurs is consumed by the household or given away. Sometimes, but rarely, it is sold to neighbors or colleagues at work (*1*).

Between 40% and 50% of the total honey consumed is placed in pots by the producers. The rest enters the commercial circuit, sold in barrels containing 300 kg to wholesalers specializing in the potting of honey and selling to supermarkets and the like (*1*). The number of French conditioners specialized in the conditioning of honey is very low and decreases from year to year due to the general crisis in apiculture in France. The largest French conditioner treats near 10 000 tons of honey per year. For comparison purposes, the leader in Germany treats twice that amount per year (*1*).

The total consumption (table honey and bakers' honey) is in the order of 500−700 g per inhabitant per year, which means that the annual French production of honey is about 30 000−40 000 tons. From 6000 to 12 000 tons are imported, and 2000−5000 tons are exported (*1*). Obviously, the production of honey is subject to considerable variations in quantity resulting from meteorological conditions as well as many other factors. For comparison purposes, the world production of honey is about 1 200 000 tons per year (*2*).

One of the responsibilities of government authorities is to ensure that beekeepers, processors, distributors, and retailers provide French consumers with safe and wholesome honey. Consequently, analyses are regularly performed to detect adulterations and pollution by pesticides, antibiotics, and to a lesser extent heavy metals (*3*). Indeed, surprisingly, while heavy metals can have hard adverse effects on living species, including humans, their potential presence in honey is less investigated than those of pesticides and antibiotics. In addition, the analyses are often only focused on specific elements such as Pb, Cd, and Hg (*3*). To fill this gap, recently, elemental analysis of 150

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Table 1. Botanical and Geographical Origins of the 86 Honeys Sold in France

no.	type	country	no.	type	country	no.	type	country
1	multiflora	Spain	30	chestnut	France	59	multiflora	imported
2	lavender	France	31	multiflora	France	60	acacia	Hungary
3	chestnut	France	32	acacia	France	61	acacia	France
4	multiflora	imported[a]	33	forest	France	62	lavender	France
5	acacia	China	34	lavender	France	63	heather	France
6	clover	Canada	35	mountain	France	64	thyme	France
7	multiflora	France	36	multiflora	imported	65	acacia	France
8	"garrigue"	France	37	clover	Canada	66	"garrigue"	France
9	orange tree	USA	38	multiflora	France	67	heather	France
10	lavender	France	39	orange tree	USA	68	chestnut	France
11	eucalyptus	Spain	40	multiflora	France	69	acacia	France
12	multiflora	France	41	multiflora	Poland	70	flowers from Provence	France
13	fir	France	42	multiflora	Argentina	71	lavender	France
14	acacia	Hungary	43	thyme + conifer	Crete	72	acacia	France
15	acacia	Hungary	44	multiflora	France	73	raspberry bush	France
16	acacia	imported	45	multiflora	France	74	"garrigue"	France
17	rosemary	France	46	multiflora	France	75	lavender	France
18	multiflora	France	47	multiflora	France	76	acacia	France
19	multiflora	imported	48	multiflora	France	77	"garrigue"	France
20	lime tree	France	49	orange tree	France	78	lavender	France
21	mountain	France	50	acacia	China	79	acacia	France
22	heather	France	51	multiflora	Caribbean	80	multiflora	France
23	buckwheat	France	52	multiflora	France	81	lavender	France
24	mountain	France	53	mountain	France	82	multiflora	France
25	forest	France	54	mountain	Switzerland	83	multiflora	France
26	"garrigue"	France	55	multiflora	Italy	84	multiflora	France
27	multiflora	France	56	multiflora	France	85	acacia	Hungary
28	lavender	France	57	multiflora	France	86	multiflora	France
29	acacia	France	58	multiflora	France			

[a] The honey was not produced in France, but the geographical origin was not clearly given on the label.

French acacia honeys collected directly by beekeepers from hives located in polluted and nonpolluted environments was performed to measure the detectable concentrations of 18 elements (*4*). The results highlighted a lack of significant contamination, except in some samples originating near industrial areas in which Ag, Cu, Al, Zn, and S could be found in fairly high concentrations. Conversely, Cd, Hg, Ni, and Pb were not detected in all the analyzed samples (*4*). The aim of the present study was to perform a similar investigation on the various types of honeys which are sold in France.

MATERIALS AND METHODS

Sample Collection. Because the goal of our study was to detect the concentrations of metallic and nonmetallic elements in the honeys sold in France, the samples were collected in all places where honey might be sold. Thus, 86 different honeys were purchased directly from bee farms, dietetic stores, markets, hypermarkets, or supermarkets. Online stores were discarded from the collection process because this commercial option is not widely used for honey in France. Samples were purchased in all the French geographical regions, excluding Corsica and the overseas departments, except for one sample from Guadeloupe. For each sample, a questionnaire was filled out indicating the nature of the container, commercial origin (e.g., name of the conditioner with the identification numbers), type of honey, geographical origin, and so on. Most of the honeys were packaged in glass jars and, to a fairly lesser extent, in plastic buckets. **Table 1** shows the type and geographical origin of the 86 honey samples. This table reveals that samples of all the main types of honeys were collected. It is important to note that no palynological analysis was performed to determine more precisely the botanical origin of the honeys studied. About 73% of the honeys were produced in France, and 27% originated from other European or non-European countries.

Elemental Analysis. Prior to the preparation and chemical analysis of the honeys, the samples were coded and randomized to avoid their identification by the chemists. The mineralization of the honey samples was performed in polypropylene-stoppered vials of 10 mL volume (Plastiques Gosselin, ref. TR 95 PPN 10TT (vials) and ref. B135 (stoppers)) by dissolution in 69.5% HNO_3 (63.01 g/mol; $d = 1.409$) (Carlo Erba, ref. 408071). The nitric acid was diluted in a 2/3 ratio with water previously purified according to the guidelines of the French Pharmacopoeia (11th edition). For each honey sample, amounts of 1 and 2 g, exactly weighed, were digested in 5 mL of the above acidic solution. Stoppered vials were safely placed in a bain-marie and were warmed to a mineralization temperature of 60 °C. After 3−4 h under these experimental conditions, the volume of each vial was adjusted to exactly 10 mL with HNO_3 (2/3), and mineralization at 60 °C was continued as described above. The time required to obtain complete mineralization of a sample ranged from 6 to 7 h, and the product was analyzed after being kept for 15 h at room temperature. A 5 mL sample of the solution was injected into an inductively coupled plasma atomic emission spectrometer (ICP-AES, Panorama, Jobin & Yvon) previously calibrated for the 18 studied metallic and nonmetallic elements. The zero point was obtained from the acidic solution used to mineralize the honey and corresponded with a blank. The wavelengths (nm) of the emission peaks of the 18 studied elements were the following: aluminum, 396.152; cadmium, 226.502; calcium, 317.933; chromium, 267.716; cobalt, 228.616; copper, 324.754; iron, 259.940; lead, 220.353; lithium, 670.776; magnesium, 279.553; manganese, 257.610; mercury, 184.887; molybdenum, 202.032; nickel, 231.604; phosphorus, 178.225; silver, 328.068; sulfur, 180.672; and zinc, 213.856. All samples were automatically analyzed in triplicate by the spectrometer. In addition, for each sample, both quantities (i.e., 1 and 2 g) were analyzed. The standard deviations were always less than 5%. Last, it is noteworthy that the limit of detection for S, Al, Ni, Ca, Mg, P, and Pb in the honey samples was 1 ng/g. For Hg the limit was 0.5 ng/g, while Ag, Cr, Fe, Li, and Mn were not detected at concentrations less than 0.2 ng/g. Last, the limit of detection of Co, Cu, Mo, Cd, and Zn was 0.1 ng/g.

Data Analysis. To correctly analyze the analytical data, multivariate statistics were needed (*5*, *6*). Because it is always interesting to confront the results obtained from different approaches (*7*, *8*), three different unsupervised methods were used to reduce the dimensionality of the obtained data matrix in a visualizable two-dimensional space. Due to the nature of the analytical results, a classical principal component analysis (PCA) on covariance matrix was first used (*9*). Briefly, PCA replaces the original variables of a data set with a smaller number of

Table 2. Number of Positive Responses (Nb/86) for the 18 Studied Elements with Their Corresponding Mean, Lowest, and Highest Concentrations (in ppm)

element	Nb/86	mean	range
Ag	6	0.127	0.09–0.16
Ca	86	54.06	8.90–130.90
Cr	9	0.203	0.08–0.36
Co	28	0.149	0.10–0.23
Cu	82	0.305	0.06–1.71
Fe	26	11.03	0.56–86.76
Mg	86	19.16	3.62–68.78
Mn	86	3.685	0.11–42.81
Mo	12	0.264	0.15–0.33
P	86	129.3	84.39–354.45
S	63	41.88	9.61–118.10
Zn	64	1.343	0.17–6.42
Al	35	2.329	0.18–9.72
Cd	19	0.152	0.08–0.25
Ni	5	0.198	0.09–0.34
Pb	3	0.793	0.28–1.08
Li	2	0.04	0.03–0.05
Hg	0	na[a]	na

[a] na = not applicable.

Table 3. Number of Positive Responses (Nb/150) for 18 Elements Measured in French Acacia Honeys (*4*) with Their Corresponding Mean, Lowest, and Highest Concentrations (in ppm)

element	Nb/150	mean	range
Ag	10	0.596	0.08–2.16
Ca	150	22.86	2.98–108.50
Cr	33	0.187	0.05–0.52
Co	46	0.091	0.03–0.25
Cu	72	0.163	0.03–2.30
Fe	107	1.167	0.13–10
Mg	150	8.708	1.43–109.50
Mn	141	0.777	0.06–10.34
Mo	86	0.441	0.07–0.81
P	150	73.45	32.12–397.5
S	84	15.39	1.60–67.66
Zn	67	0.746	0.04–5.96
Al	99	0.374	0.05–1.44
Li	5	0.07	0.02–0.24
Ni	0	na[a]	na
Hg	0	na	na
Cd	0	na	na
Pb	0	na	na

[a] na = not applicable.

uncorrelated variables called the principal components (PCs). The method is linear in that the new variables are a linear combination of the original ones (*9*).

Correspondence factor analysis (CFA) (*10*) was also tried because it had been successfully used on similar data matrixes (*4*, *11*−*13*). CFA is primarily a technique for displaying the rows and the columns of a two-way contingency table as points in corresponding low-dimensional vector spaces. These spaces may be easily superimposed to obtain a joint display. Its χ^2 metrics allows us to work on data profiles. CFA has been extended to display other matrixes of non-negative data. Consequently, it appeared particularly suited for the analysis of the data matrix (in ppm) produced by the chemical analysis.

Hierarchical cluster analysis (HCA) also deals with rectangular tables of variables and objects. The aim of HCA is to uncover some latent structure of the objects (and variables) in terms of groups of similar elements and, possibly, in terms of a hierarchy of embedded groups. Briefly, two main steps are repeated alternately. The first one is to search the distance matrix for the two closest objects (or variables). The second step is to consider this pair of objects as a single individual and to recompute the distances between this new element and the rest of the objects. The first step is repeated on the reduced distance matrix, and so on. Thus, at each cycle of two steps, a new supergroup is formed by the aggregation of the two nearest groups, or single objects, of the previous cycle. The aggregation is represented in the dendrogram by the junction of the corresponding branches, which is called a node of the tree. The procedure is stopped when all groups have been aggregated (*14*). The two difficulties with HCA deal with the selection of a distance formula and an aggregation strategy. In our study, the χ^2 distance and the average linkage procedure were selected because they are commonly used on this type of data (*11*, *14*).

All the statistical analyses were performed with ADE-4 (*15*), a powerful statistical software package offering numerous graphing tools for optimal data display.

RESULTS AND DISCUSSION

Analytical Results. The elemental analyses obtained from 1 or 2 g of honey yielded similar results and hence were averaged. The number of positive responses (i.e., concentrations superior to the different limits of detection) for each metallic or nonmetallic element in the 86 analyzed honeys and their corresponding average, lowest, and highest concentrations (in milligrams per kilogram to raw (wet) weight) are given in **Table 2**.

Table 2 shows that calcium, magnesium, and phosphorus were detected in all the samples analyzed. These results are not surprising due to the nature, role, and ubiquity of these fundamental elements. Manganese was also present in all the honey samples but in concentrations generally lower than those recorded for Ca, Mg, and P. Copper was found in most of the honey samples, and sulfur and zinc were detected in more that 70% of the honeys. Undoubtedly, pollution by humans can be suspected to be the source of these elements. About 41% of the analyzed samples included measurable concentrations of aluminum and, to a lesser extent, cobalt or iron, or both. Surprisingly, cadmium was found in about 22% of the honey samples. This is a particular concern due to the high toxicity of this heavy metal. In 1994, the Centre National d'Etudes Vétérinaires et Alimentaires (CNEVA, the National Center for Veterinary and Alimentary Studies), in the frame of their annual control of the quality of honeys, analyzed 122 French honeys and 28 foreign honeys for their concentrations in Pb and Cd (*3*). While Cd was not detected in the foreign honeys, 3% of the French honeys were contaminated by detectable amounts of Cd, with a mean concentration of 0.07 ppm. The percentage of Cd found in our study is higher (**Table 2**). Conversely, Fléché et al. (*3*) showed that lead was not detected in the French honeys, while 43% of the foreign honeys were contaminated by detectable concentrations of this element, with a mean concentration of 3.8 ppm. In our study, Pb has been detected in three samples (numbers 22, 69, and 73), with concentrations of 0.28, 1.08, and 1.02 mg/kg, respectively. It is interesting to note that all of these honeys originated from France (**Table 1**). Fortunately, mercury was not detected in the 86 samples studied, but silver, chromium, and nickel were found in some honeys. Last, it is worth noting that lithium was found in two samples (numbers 18 and 25 in **Table 1**), with concentrations of 0.03 and 0.05 ppm, respectively.

For comparison purposes, the results of the elemental analysis recently performed (*4*) on 150 French acacia honeys are given in **Table 3**. While Ni, Cd, and/or Pb have been found in the present study, sometimes in fairly high concentrations, these elements were not detected in the acacia honeys, even when a large part of the samples were collected directly from hives located near sources of industrial pollutions (e.g., highways, petroleum industries). Conversely, Al was found with a high frequency in both studies. This confirms the wide contamination level of this element. In addition, while in the previous study

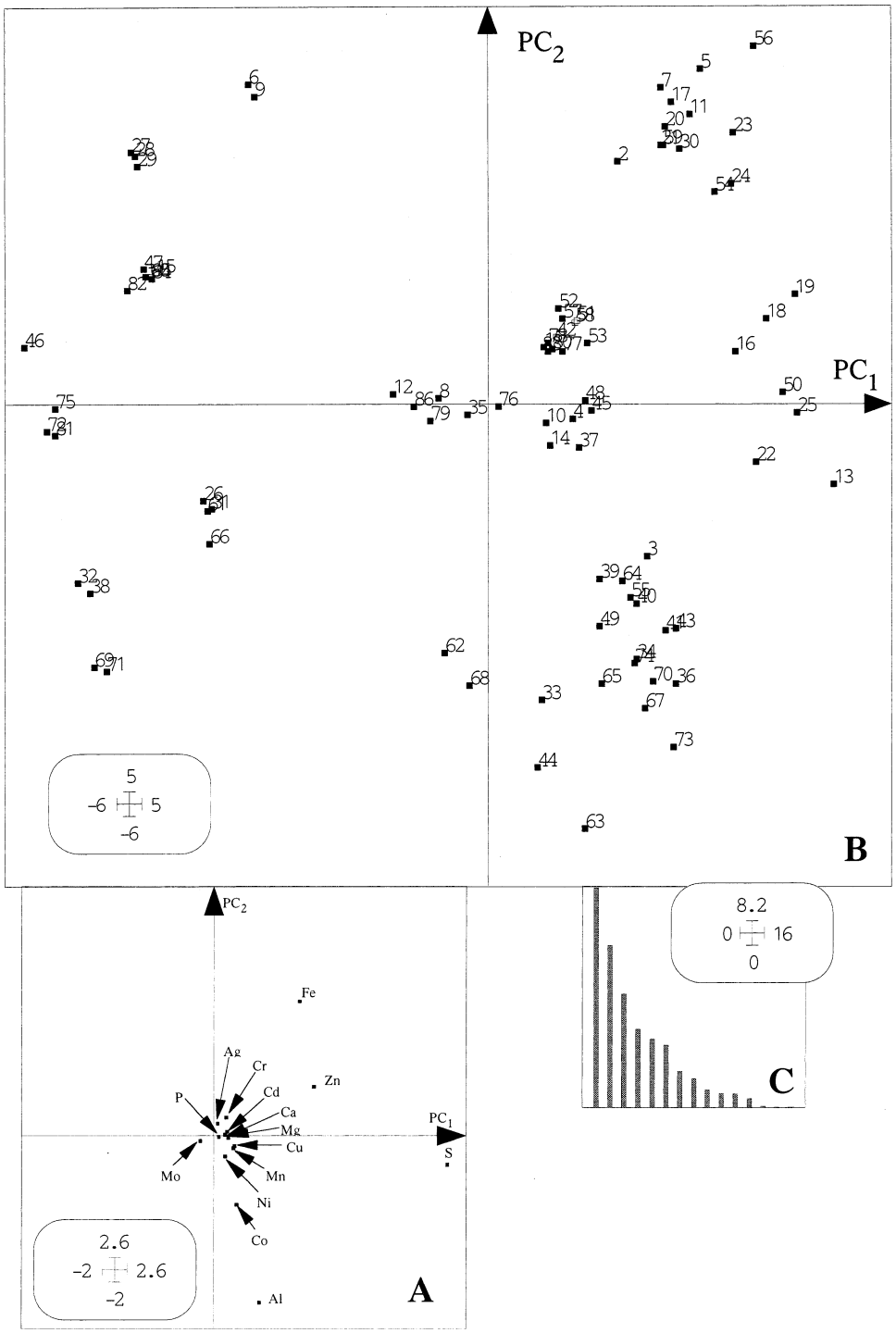

Figure 1. PC_1PC_2 factorial maps for the 15 elements (A) and 86 honey samples (B). Graph of the eigenvalues (C).

the highest concentration found was 1.44 ppm (**Table 3**), our study reveals that one sample included 9.72 ppm of Al (**Table 2**). This is a particular concern because evidence exists that Al may play a role in the etiology of Alzheimer's disease (*16*).

Table 3 shows that Fe and Mo were frequently detected in acacia honeys. Conversely, their presence has been detected with a fairly low frequency in the honeys analyzed in the present study (**Table 2**), since only about 30% and 14% of the honeys present detectable concentrations of Fe and Mo, respectively.

While elemental analyses have been performed on honeys found in other countries (*17*, *18*), it is difficult to compare our results with those published in the literature due to differences in the methodologies, analytical methods, and so on. Kump et al. (*19*), comparing the performances of radioisotope X-ray fluorescence spectrometry, total reflection X-ray fluorescence spectrometry, atomic absorption spectrometry, and inductively coupled plasma atomic emission spectrometry methods for detecting metallic and nonmetallic elements in honeys, pollens, and bees, have clearly addressed this problem.

To perform a rational analysis of the honey samples, linear multivariate analyses were used. Only elements with a frequency of occurrence greater than 5% were considered for statistical analysis (*9*). This yielded the design of a 15 × 86 (elements/samples) data matrix.

Multivariate Analysis of the Honey Samples. Due to the nature of the analytical results, a classical PCA on covariance

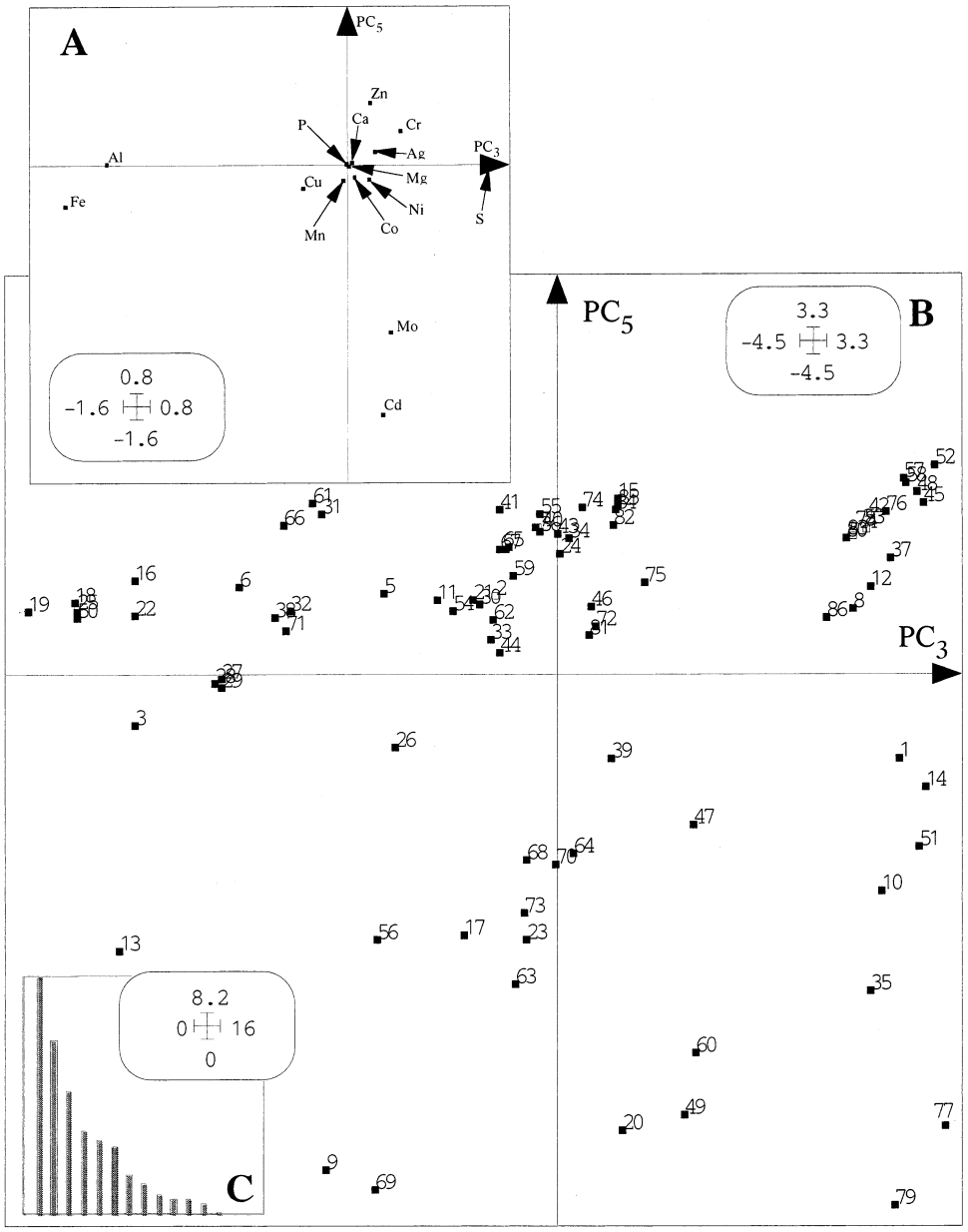

Figure 2. PC_3PC_5 factorial maps for the 15 elements (A) and 86 honey samples (B). Graph of the eigenvalues (C).

matrix (*5*, *20*) was used to reduce the dimensionality of the 15 × 86 data matrix. A $\log_{10}$ data transformation was first employed to reduce the size effects, which could hide information on the factorial maps.

The first factorial map, PC_1PC_2 (**Figure 1**), accounts for 46.24% of the total inertia of the system (i.e., 26.64% + 19.60%). Variables (i.e., elements) are represented in **Figure 1A**, and samples (i.e., objects) are displayed in **Figure 1B**. Inspection of **Figure 1B** reveals a first gradient of distribution of the samples along PC_1 with, for example, honey numbers 13, 25, or 50 in the right part of PC_1 and honey numbers 46, 75, or 81 in the left part of this axis. This distribution is mainly dependent on the concentration of sulfur found in the honeys (**Figure 1A**). For example, honey number 13 includes the highest concentration of S (i.e., 118.10 mg/kg), while conversely, in honeys numbers 46, 75, or 81, the concentrations of sulfur are below the limit of detection of the ICP-AES for this element. Obviously, the other metallic and nonmetallic elements influence the distribution of the samples on PC_1 and PC_2. Samples located in the bottom part of **Figure 1B** are contaminated by aluminum. Thus, honeys numbers 63 and 73 present the highest concentrations of Al. Al contaminates most of the honey samples located below the PC_1 axis. Conversely, iron is present in the honeys located at the top of **Figure 1B**, especially in the right part. For example, samples 19 and 56 include 86.76 and 85.25 ppm of Fe, respectively. The location of nickel in **Figure 1A** tends to indicate that samples including this pollutant are located in the bottom right part of **Figure 1B**. This is absolutely true, since the five honeys with Ni are numbers 14, 37, 43, 63, and 73. The central location of cadmium in **Figure 1A** reveals that this element is not correctly represented on PC_1PC_2. Consequently, honeys contaminated by this heavy metal, such as numbers 9, 10, 13, and 26, do not form a cluster in **Figure 1B**. The points located at the origins of PC_1PC_2 (**Figure 1A**,**B**) and the graph of the eigenvalues (**Figure 1C**) clearly indicate the necessity to consider other principal components for the complete analysis of the data. The PC_3PC_5 factorial map (**Figure 2**), which accounts for 21.98% (13.74% + 8.24%) of the total inertia of the system, provides additional information about the affinities of the honeys based on their analytical profiles. Thus, honeys without Fe and Al form a strong cluster and are located on the right part of the PC_3 axis. Conversely, samples with Al and/or

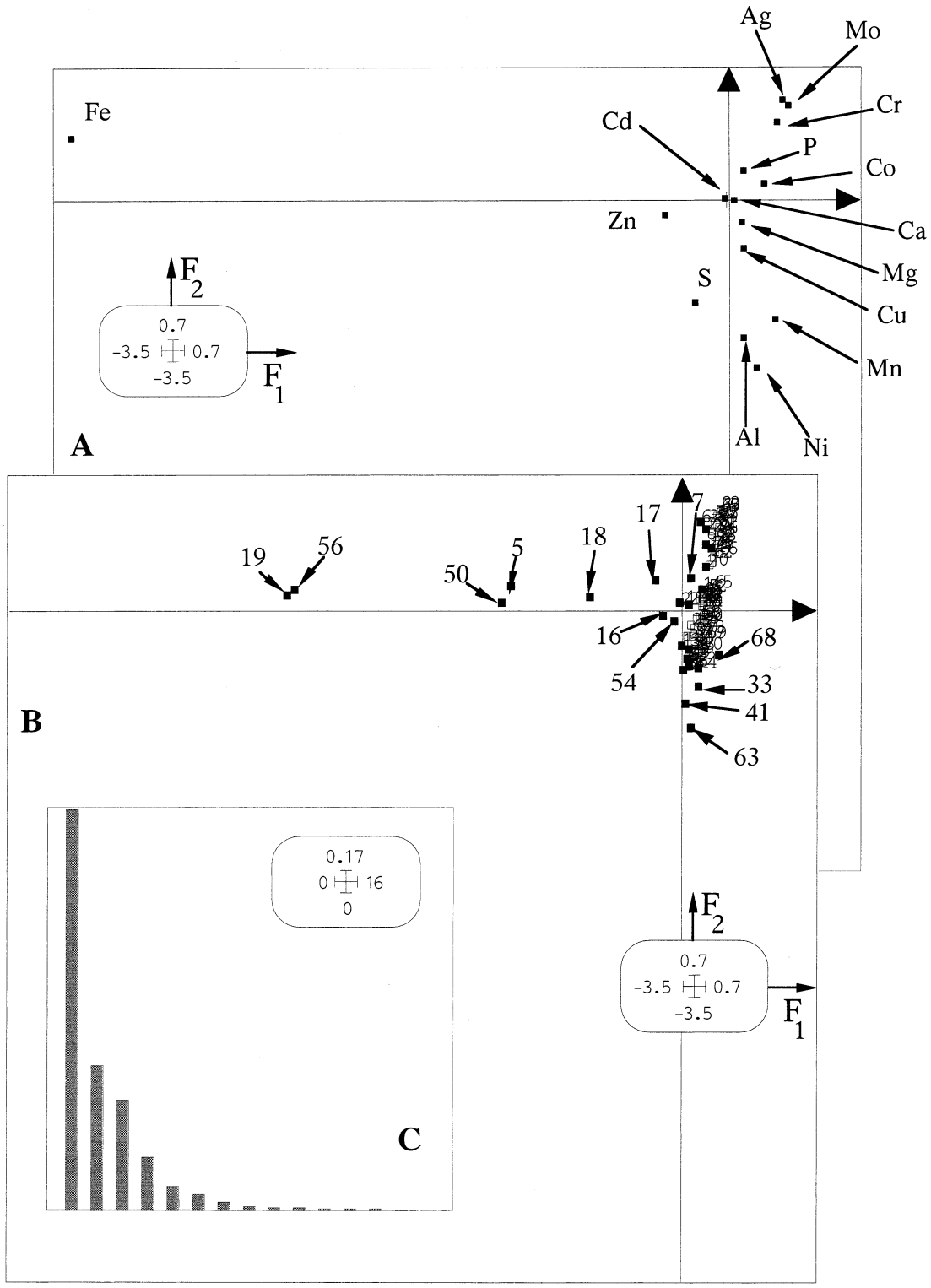

Figure 3. F_1F_2 factorial maps for the 15 elements (A) and 86 honey samples (B). Graph of the eigenvalues (C).

Fe are located on the opposite side. In the same way, a strong opposition exists between all the honeys located below the PC_3 axis which include Cd and/or Mo, except for sample 3 and also the strong cluster above this point constituted of honeys numbers 27−29. Samples 1, 14, 26, and 39 have only Mo. Samples 9, 20, 49, 60, 69, 77, and 79 include Cd and Mo. Those samples located between these two groups of honeys are contaminated by Cd.

The analytical data being comprised of only positive values with the same units (i.e., ppm), a CFA was performed on this matrix by means of ADE-4 (*15*).

CFA allows us to significantly reduce the dimensionality of the 15 × 86 data matrix since the five first axes (i.e., F_1-F_5) account for 95.35% of the total inertia of the system. The factorial map F_1F_2 (52.21% + 18.85% of the variance) clearly reveals that samples 19 and 56 are outliers due to their high concentrations in Fe (**Figure 3A**,**B**). Undoubtedly, with 86.76 mg/kg of Fe for the former and 85.25 mg/kg for the latter, these two honeys have been contaminated by this element. Sample 19 is a multiflora honey (**Table 1**) identified as imported but without further information on its exact geographical origin of production. Sample 56 is also a multiflora honey but was

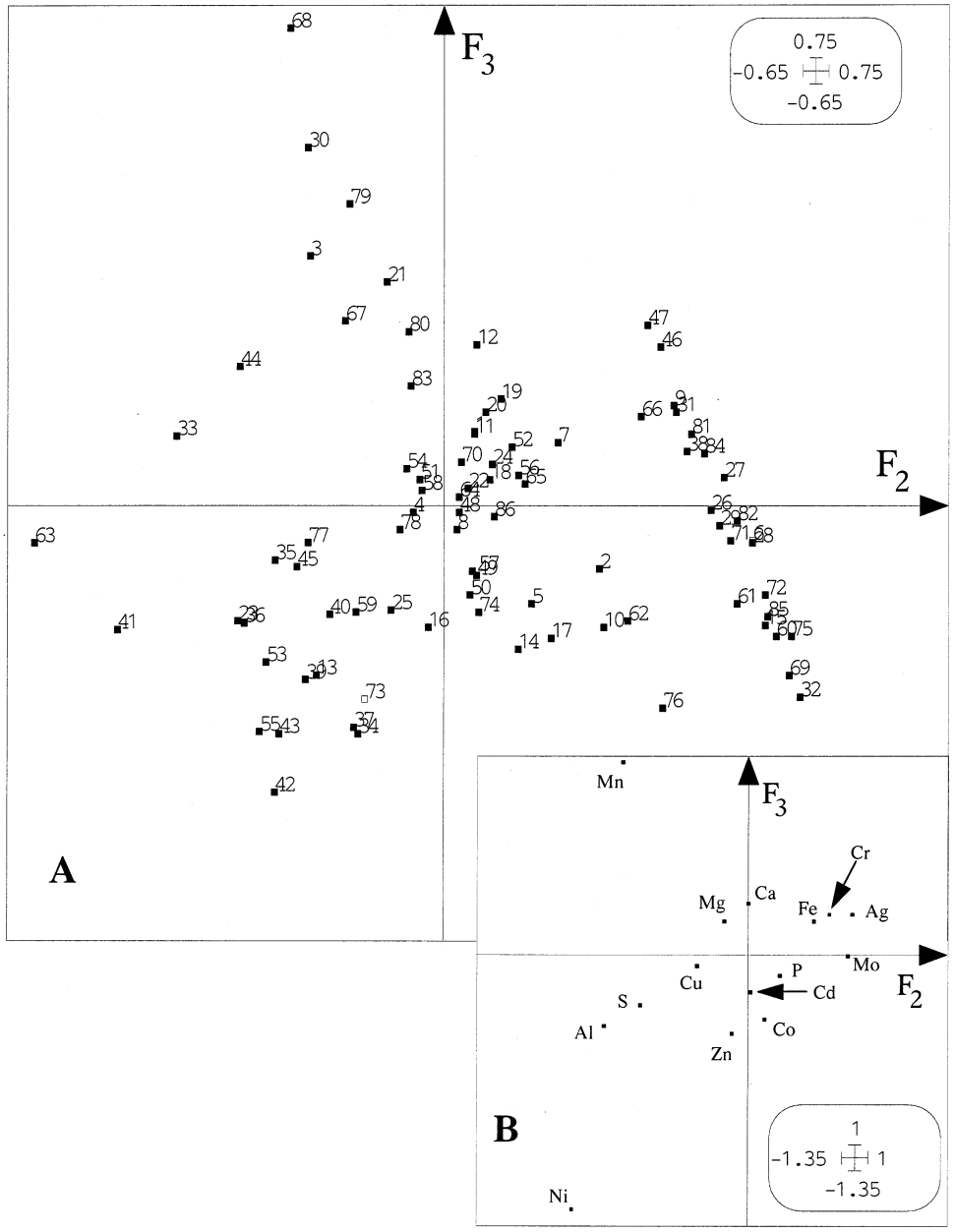

Figure 4. F_2F_3 factorial maps for the 86 honey samples (A) and the 15 elements (B).

produced in France (**Table 1**). To a lesser extent, sample 50 with 24.93 mg/kg of Fe, sample 5 with 22.71 mg/kg of Fe, and sample 18 with 21.12 mg/kg of Fe can also to be suspected of pollution by this element. This is in agreement with their location in **Figure 3B**. It is interesting to note that samples 5 and 50 are both acacia honeys coming from China (**Table 1**). In our recent study (*4*) on the elemental analysis of 150 French acacia honeys collected directly by beekeepers from hives located in areas presenting different degrees of pollution, the highest concentration found for Fe was 10 mg/kg out of 107 positive responses (**Table 3**). Sample 18 is also a multiflora honey produced in France (**Table 1**).

The specific location of samples 63 and 41 in **Figure 3B** is due to their high concentration of Al (**Figure 3A**). The former is heather honey coming from France, and the latter is a multiflora honey coming from Poland (**Table 1**).

The factorial map F_2F_3, which accounts for 33.16% of the total inertia of the system, allows us to cluster honeys without S. They are all located in the right part of **Figure 4A** (from samples 47, 46 to 69, 32). Conversely, in the left bottom part of **Figure 4A**, we can find samples with high concentrations of S. For example, honey number 63 includes 91.53 mg/kg of S but also a very high concentration of Al (9.69 ppm). This is in agreement with the location of these elements in **Figure 4B**. In the same way, samples with high concentrations of manganese are located in the top left part of **Figure 4A**. Thus, honey number 68 includes the highest concentration of Mn (42.81 ppm, **Table 2**).

HCA was also carried out on the data matrix of analytical results. An aggregative procedure using a χ^2 distance and an average linkage algorithm were selected. It is noteworthy that the results obtained with this type of multivariate method are difficult to directly compare with those produced by CFA or PCA. Indeed, with a PCA or a CFA, the different variables and objects are explained on the basis of their different factors; consequently, to draw conclusions, it is always necessary to consider different factorial maps accounting for different parts of the information. Conversely, with HCA, all the information in the data matrix is displayed through two dendrograms: one for the variables (elements) and another for the objects (sam-

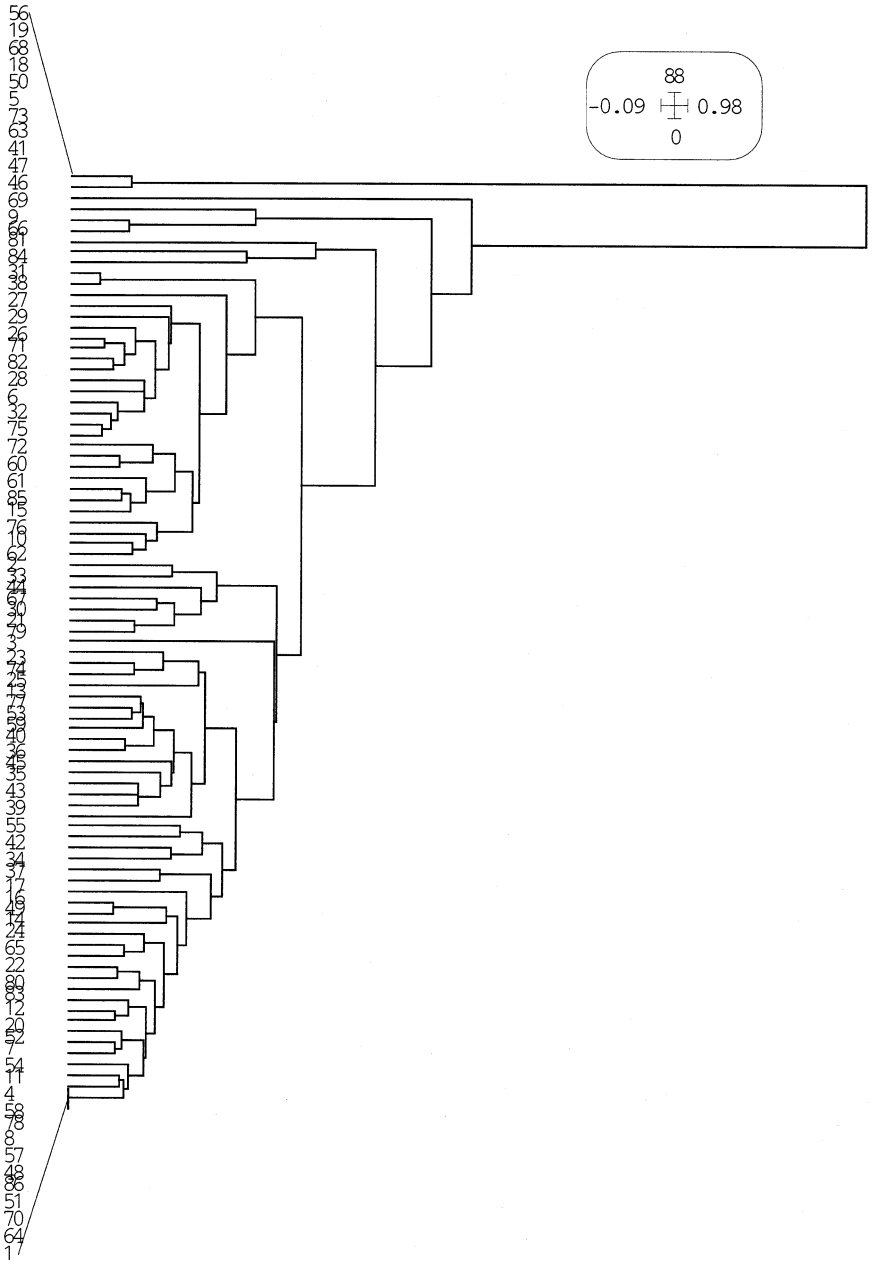

Figure 5. Dendrogram of the 86 honey samples.

ples). Therefore, the comparison of the results obtained with a PCA or CFA and a HCA is often not straightforward. Despite this, the main clusters of samples found with PCA and CFA are also detected on the dendrogram of the samples obtained with HCA (**Figure 5**). For example, samples 19 and 56 or samples 5 and 50, previously discussed, form the two strong clusters at the top of **Figure 5**. The dendrogram of the elements (**Figure 6**) confirms the ubiquity of Ca, Mg, and P in all the samples (**Table 2**). Indeed, they form a cluster in **Figure 6**. Conversely, elements such as Ni, Cr, Ag, and Fe are isolated in the dendrogram. This is proof that they originate from punctual pollutions.

Our study shows that crude relationships exist between the elemental profiles of the honeys and their botanical origin. The acacia honeys are all located in the right bottom part of **Figure 4A**, except for sample 79, which is located at the opposite side. This is due to the difference in the concentrations of calcium and manganese. Indeed, while the concentration of Ca found in honey number 79 equals 96.38 mg/kg, those found, for example, in samples 32, 60, 69, and 76 are 8.9, 16.81, 10.4, and 13.46 mg/kg, respectively. In the same way, the concentration of Mn found in the sample 79 equals 12.7 ppm, while only 0.38, 0.17, 0.18, and 0.25 ppm of Mn have been measured in samples 32, 60, 69, and 76, respectively. It is interesting to note that the acacia honey sample 79 is located in the vicinity of the three chestnut honeys, samples 3, 30, and 68 (**Table 1**), located in the top left part of **Figure 4A**. The multiflora honeys present various botanical origins and, hence, are spread out on the factorial map F_2F_3, but clusters can also be found in specific regions. For example, samples 80, 83, 12, 19, 31, 46, 47, etc. are all multiflora honeys (**Table 1**). The same situation is observed for the lavender honeys, and to a lesser extent for the "garrigue" honey, but conversely, the three heather honeys (i.e., numbers 22, 63, and 67) are spread out on F_2F_3 (**Figure 4A**).

Inspection of **Figures 1**−**6** reveals that all the different honeys can be polluted by heavy metals and related elements. However,

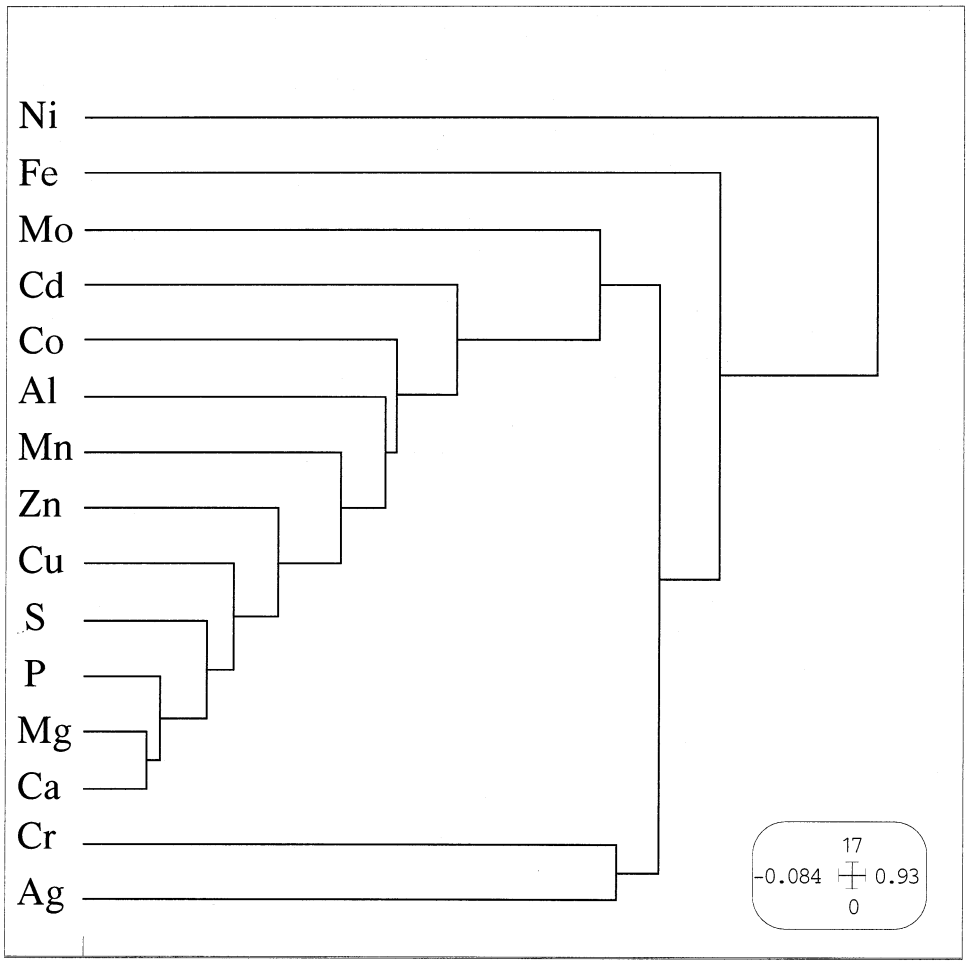

Figure 6. Dendrogram of the 15 elements.

honeys coming from trees are generally less contaminated that the honeys from other origins (**Figure 4A**). In addition, honeys coming from Mediterranean xerophytes and heather seem to preferentially concentrate heavy metals and related pollutants. This can be easily seen from **Figure 4A** and also from **Figure 2B**.

It has been impossible to find relationships between the levels of contaminations of the honeys and their country of origin, the containers in which they were stocked, or the type of marketing distribution. For example, sample 20, which originated from the biggest French conditioner and purchased in a supermarket, includes the highest concentration of chromium (i.e., 0.36 mg/kg) and 0.19 mg/kg of Cd. Sample 73, which was purchased directly from a beekeeper, selling only his own limited production, includes 1.02 mg/kg of Pb, 0.18 mg/kg of Cd, and 0.09 mg/kg of Ni.

Our analytical results and multivariate analyses clearly reveal that honeys sold in France present a very high variability in their concentrations of metallic and nonmetallic elements. Unfortunately, some of them appear to be highly contaminated by heavy metals and other industrial xenobiotics at concentrations of health concern. Broadly speaking, honeys originating from caducous trees appear less contaminated than those of other botanical origins. Aromatic plants concentrate pollutants more easily than the herbaceous ones. Our study shows that it should be necessary to reinforce the French legislation, by increasing the number of analyses and the range of elements to investigate, to ensure that beekeepers, processors, distributors, and retailers always provide consumers with safe honey.

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