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Total and Extractable Heavy Metals in Indoor and Street Dust in Elbasan city, Albania

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Abstract: ***The evaluation of air pollution in the city of Elbasan from heavy metal was conducted by analyzing their content in the dust composition deposited in apartments and on streets. Dust samples were taken in 10 different streets with different traffic levels and in 6 apartments near to these streets. A soil sample was also analyzed. In the three types of samples was determined the total and extractable content (soluble) for Cu, Zn, Cr, Ni, Pb, Cd, Fe and Li. After the pretreatment of the samples with two methods, the metal content was determined by the method of atomic absorption spectroscopy. The trace values of metals in dust appear relatively low, generally within EU norms. We can notice two groups based on the proportion of concentration- level of traffic at the sampling station. The first group which included Pb, Zn and Cu, which content are correlated with the level of traffic and the degree of urbanization and are considered as "pollutant" metals originating mainly from anthropogenic sources. The second group including Cr, Ni, Fe and Li, whose content in the samples taken at stations with various level of traffic or the degree of urbanization, results in minor changes of the case. These are considered as "land" metals, originating mainly from the soil surface. The concentrations of pollutants metals in dust samples of houses are generally higher than those of the dust of the streets. The rate of extraction according to the element Zn> Pb> Cu> Ni> Cr> Li> Fe, by type of sample, almost for all metals, follows this order: indoor dust> street dust> ground dust. Correlations between Cu-Pb-Zn (r> 0.6) and Fe-Ni (r> 0.9) indicate the common origin.***

Keywords: heavy metals, indoor dust, outdoor dust, air pollution.

1. Introduction

There are two main reasons for which the knowledge of the composition of the deposited dust has become the subject of numerous studies recently:

Firstly, by analyzing the chemical composition of dusts we provide an important information about the content of toxic pollutants in the environment and their origins. Dusts are recognized as an important source of heavy metals in urban environments [Akhter, 1993]. Various studies have shown a significant dependence between the content of heavy metals in dust deposited on the street or indoor and their levels in the water or air [A. M. Ure, 1995]. Also it was found a direct dependence between the concentration of some metals (Pb, Cd) deposited in dust and the traffic level in urban environments [Charlesworth, 2003].

Secondly, the establishment of the chemical composition of the dust helps to assess their contribution to the overall population exposure to harmful substances for the human health. The deposited dusts are also known, among other things, as an important source of human exposure to some toxic metals [IPCC, 1995]. The extent of this exposure depends on the life conditions of the individual and the frequency of contacts with the environments with a high content of dust. Since that children spend more time in contact with dust deposited in their home or in the street, these are considered important source of exposure to toxic metals, especially to lead [Bolger et. al., 1991].

Other studies show a direct correlation between the content of lead in dust and its concentration in the blood of children [Laxén DPH, Rabb GM and Fulton M (1987); BP Lanphear et. al. (1995)]. Correlations were also observed between the cadmium obtained from dust and its concentration in the blood and urine of children who live in areas that present air pollution by cadmium [Butcher, JP et. al. (1983)]. The definition of heavy metals in dust deposited indoor and on streets of the city of Elbasan has as goal the indirect assessment of urban air pollution in Elbasan by heavy metals and possibly the identification of their common and specific resources.

2. Materials and Methods

2.1 Sampling

Sampling was conducted in 10 streets (R1-R10) and in 6 indoor residential (B1-B6) at each street in the city of Elbasan. The selection of sampling sites was conducted keeping in mind that they represent various areas with traffic density and urbanization rate and simultaneously cover more or less the geographical area where the city lies. The samples of dust deposited on the external environments were taken with the help of a clean plastic brush on both sides of the streets in a segment with a length of 10m and a depth of 1cm. Samples were taken in the period of June-July 2011, in terms of a dry weather.

Sampling of indoor environment for this study was conducted by the method of vacuum [IPCC, 2000]. This is one of the most used methods for this purpose because it is simple, fast and samples obtained in this way are considered fairly representative and averaged in time. This method consists in taking samples of dust from vacuum cleaner deposits in its bottom, where there are deposited the smallest particles.

Besides dust samples, for the evaluation of the natural content of metals in the ground, there were also taken ground samples. For this purpose, it was chosen a selected clean area, free of industrial and agricultural activity and

also away from roadways. Samples were taken at a depth of approximately 5 cm from the surface.

2.2 Analytical Procedures

Dust samples were purified with a forceps from relatively large impurities such as: small stones, grass, plant material, textile yarn or any other impurity. After cleaning, the sample was passed in a 500 mm pore strainer. The fraction derived passes in mortar agates and then in a 125mm sieve (strainer), the remaining part is grinded and resieved through the sieve. The milled sample is stored in a polyethylene bag before the chemical analysis. Soil samples are left in a dry room temperature and then are transferred and grinded to the 125mm sieve like the above aiming to complete the transition of the sample in the sieve. Before the chemical treatment, the samples were dried at temperatures around 100 °C for 24 hours until a constant weight. For the treatment of dust samples were used two methods [Çullaj A., Shqau K., (2000)]:

a. Total Heavy Metal Digestion (disaggregation)
In order to digest dust samples for atomic absorption spectroscopy analysis, 0.5 g of sample was placed in a covered Teflon beaker (to avoid the loss of Cd and Pb) containing a mixture of high purity HNO3(2.5 mL)/percholoric acid HClO4(2.5 mL) and allowed to remain overnight at ambient temperature. After slow evaporation to dryness, 1 mL of HNO3 was added and the solution was again evaporated to dryness, after which, the residue was extracted with 0.1 N HCl and diluted bi-distilled water, filtered on Whatman pre-washed filter paper and diluted with 1% HNO3in a 25 mL polyethylene bottle [Rashed M. N., (2008)].

b. The extraction with acetic acid (CH3COOH) 0.43M (the extractable or soluble part). In a erlenmeyer flask with lid, are weighed about 0,5g dust sample and to this is added about 20ml solution CH3COOH 0:43 M. Later the erlenmeyer flask is placed in an electric shaking and left to mix for about 16 hours with the aim of full metal extraction of acetic acid (in the absence of ultrasonic bath). The time of 16 hours was concluded after several tests to complete quantitative extraction of metals [Picolo A., G. Celano (1992)]. Through the acid treatment, is defined the full content of heavy metals in dust samples, while the method of extraction with acetic acid determines the extractable part of heavy metals which are thought to have the greatest effect on human health and the ecosystem.

2.3 Equipments and methods of measurement

The dust samples of streets and houses, treated according to the two methods, were analyzed for the content of copper, zinc, lead, chromium, nickel, cadmium, iron and lithium. The first seven metals were determined by the method of Atomic Absorption Spectroscopy (AAS) with Air-Acetylene flame (AA), while lithium was determined by the method of Atomic Emission Spectroscopy (AES) Air-Acetylene flame. Measurements were obtained with the VARIAN Spectr.AA20 + equipment.

2.4 Control Quality Analysis

For the realization of the Control Quality Analysis (QA), a certified ground sample prepared by the Joint Research Center Ispra (AQUACON Sub-Project 9 "Soil Analysis") was used as a standard reference material and it was analyzed for each set of samples considered . Additionally to this there were made parallel evaluations of dust samples for 10% of the analyzed samples. For each set of samples there were prepared and analyzed the white proof according to the same procedure as for sample preparation.

2.5 The Statistical Results Analysis

The processing and statistical analysis of the results (mean (M), Geometric mean (GM), Standard Deviation (SD) and percentiles) were provided with MS-EXCEL 2007.

3. Results and Discussion

The values of average concentrations of metals in the dust of the streets and houses analyzed with both methods of treatment are presented in Table 1 (the results of the analyzes reported in dry weight). From the review of results and their comparison with the values given in the literature (Table 2), we conclude that the concentrations of metals appear relatively low, lower than in many developed countries, especially referring to lead.

These values result very close to the values of Tirana city [Cullaj, A., 2000; Totoni, R. 2002]. Slightly higher values of metals such as Ni, Zn for Elbasan, can be explained by the influence of the metallurgical industry in the west of the city. The relatively low percentage of lead compared to some countries referred to, can be explained by the limited use of leaded gasoline **as the primary source of this metal in the urban air particles [EAA, 1998].**

Table 1: Heavy metals concentrations (mg/kg, Fe %) in street and home dust (R-street, B-home)

Element	Typesample	Sample Nr	Math. Average		Geom. Average		Percentils (disaggregate)				Percentils (Exctraction)			
			Disg.	Ekstr.	Disg.	Ekstr.	90	75	50	25	90	75	50	25
Cu	R	10	64.9	31.5	49.8	23.5	130.7	65.3	45.3	30.0	62.6	30.7	21.8	15.6
	B	6	195.1	74.8	176.7	64.8	294.7	200.6	174.5	137.4	108.3	92.4	65.0	52.6
Zn	R	10	222.2	160.8	166.6	129.4	493.0	212.4	150.9	98.7	381.3	156.4	119.9	78.7
	B	6	579.2	512.2	552.7	488.8	750.3	586.0	565.9	516.8	632.4	551.5	500.5	484.2
Pb	R	10	89.5	37.0	78.0	30.3	156.3	96.0	76.3	54.6	60.8	40.2	31.8	18.5
	B	6	110.1	73.5	105.3	65.9	152.6	107.6	102.8	101.5	112.2	84.7	69.7	67.8
Cr	R	10	33.0	6.8	31.0	6.1	50.0	32.9	30.6	23.0	9.7	7.1	6.4	4.2
	B	6	91.8	15.9	89.6	13.7	113.6	93.6	89.3	89.1	25.3	17.8	13.0	9.4
Ni	R	10	85.8	10.4	82.9	9.8	112.8	97.4	80.9	71.5	13.5	11.1	9.5	8.0

	B	6	92.1	27.5	86.4	25.0	134.0	89.2	83.4	81.9	37.8	30.2	26.2	25.6
Fe	R	10	1.3	0.09	1.2	0.08	1.6	1.3	1.2	1.1	0.12	0.09	0.08	0.07
	B	6	1.34	0.077	1.31	0.073	1.70	1.32	1.28	1.16	0.103	0.078	0.075	0.070
Li	R	10	4.3	0.93	4.2	0.87	5.2	4.5	4.1	3.8	1.52	1.13	0. 84	0.67
	B	6	5.0	0.47	4.9	0.46	5.6	5.0	4.9	4.9	0.60	0.47	0.46	0.46

Heavy metals in street dust and traffic level. We distinguish two groups of metals:

a) Metals, whose content in the analyzed dust samples, is correlated with the level of traffic and the degree of urbanization of the sampling station. In this group we would include Pb, Zn and Cu. Figure 1 shows the concentrations of these metals vary considerably from one station to another. These metals can be considered as "pollutant" metals originating mainly from anthropogenic sources.

b) b. Metals, whose content in the samples taken at various stations in terms of traffic level or degree of urbanization, results with minor changes according to the case. This group includes Cr, Ni, Li and Fe. In Figure 2 it appears that their content changes from one station to another are small. These metals can be considered as "ground" metals, originating mainly from the ground surface.

Table 2: Heavy metals concentrations (mg/kg,) in some cities (R-street, B-home)

City, country	Pb	Cd	Cu	Zn	Ni	Cr
Elbasan, R (this study)	32.7-175.2	0.45-2.43	18.2-189	68.9-482	46.3-115	19.4-49.5
Elbasan, B (this study)	75.4-183.6		82.3-335	299-858	56.6-169	62.5-127
Tirana, R [Cullaj,A. 2000]	24.5-135.2	0.11-0.27	15.3-51.9	37.2-112	54.5-78.6	42.4-61.2
Tirana, B [Cullaj,A. 2000]	90-230.5		92.3-274	498-2240	71.9-107	62.5-339
Tirana, R [Totoni,R. 2002]	21.1-141.1	1.44-2.62	23.8-233	93.4-403	68.2-101	25.3-45.8
Tirana, B [Totoni,B. 2002]	39.5-162.8		69.3-330	245-699	73.8-141	54.9-104
Turkey, R [Kartal,S. 1992]	188-1037	10-200		26.6-813	10-600	23-576
Turkey, B [Kartal,S. 1992]	120-600	11.3-62.7		15-113.8	30-190	15-274
Thessaloniki, R [Ewen,C. 2009]	206.7	4.0	269.2	462.9		
Istambul, R [Sezgin et al. 2004]	211.88	1.91	208.49	526.81		
Hong Kong, R[Xiangdong. 2001]	181	1.77	173	1450		
London, R [Thorton 1991]	1354	4.2	115	513		
Bahrain, R [Akhter M. S. 1993]	290-1250	21-129		44-1018	30-300	53-370
Bejing, R [Xingquah, Zh. 1992]	17.6-231					
Bejing, B [Xingquah, Zh. 1992]	17.5-557					
USA, UK [IPCC, 1995]	300-2500					

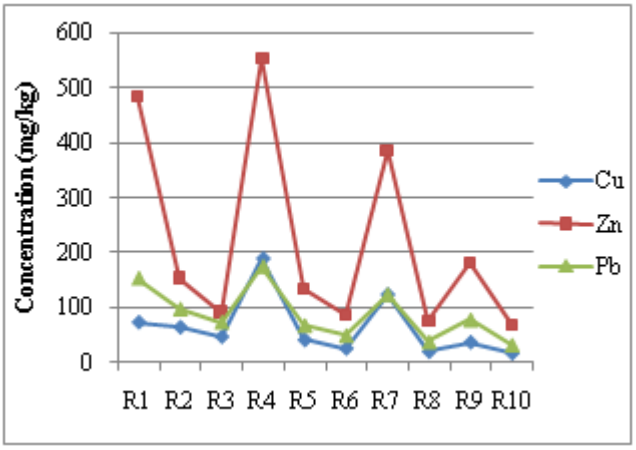

Figure 1: Concentrations of "pollutant" metals in the dust of the street (mg/kg)

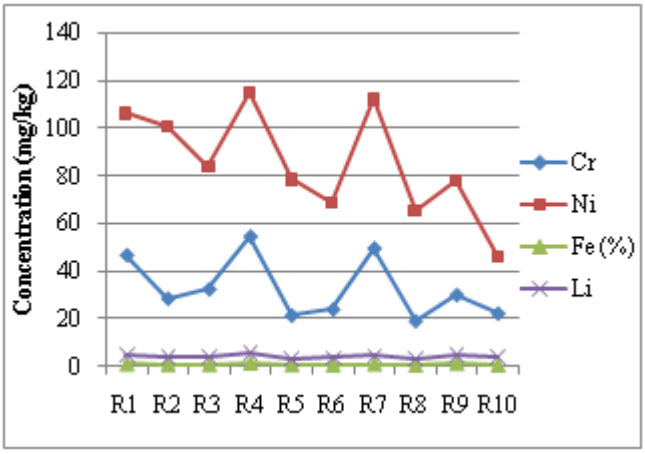

Figure 2: Concentrations of "ground" metals in the dust of the street (mg / kg; Fe (%))

Cadmium is not included in any of the groups, and this because its concentrations are very low, in the limits of detection of the SAA with flame technique. According to this, the cadmium results can be used only as a guide value for the sequence concentration and trends of changes in time and space, but not as absolute values.

Heavy metals in street and home fallen dust and in the soil. It would be interesting the interpretation of the average concentrations of metals in dust samples of the street and houses, in relation to their content in soil samples (Table 3).

Table 3: Metals concentration (mg/kg; Fe in %) in streetdust , home dust and soil through disaggregation and extration sample treatment method

		Metal concentration (mg/kg; Fe (%))						
	Sample	Cu	Zn	Pb	Cr	Ni	Fe (%)	Li
disaggregation	Street	64.9	222.2	89.5	33.0	85.8	1.3	4.3
	Home	195.1	579.2	110.1	91.8	92.1	1.34	5.0
	continental crust	31.9	126.3	16.71	104.3	165.2	2.81	10.23
exctraction	Street	31.5	160.8	37.0	6.8	10.4	0.09	0.93
	Home	74.8	512.2	73.5	15.9	27.5	0.077	0.47
	continental crust	2.65	68.19	6.12	1.20	20.14	0.02	0.32

Elements, whose content in dusts is about 3 times higher than in ground local samples, are considered as "pollutants" elements [Fishbein L. O'Neill IK, (1987)]. Based on this fact, from the analyzed metal, lead can be considered as polluting elements for both types of samples, while regarding to house dusts, except lead, zinc and copper result as "polluting" elements. Other elements: Cr, Ni, Fe and Li represent the highest concentration in ground samples than in dust samples and consequently they are considered as "ground" elements.

Based on the "pollutant" metals levels in the analyzed dust samples, the sampling stations can be classified into three groups according to the pollution degree that they denote:

High pollution level (R1, R2, R4, R7 and B1, B6), average (R3, R5, R9, and B2, B3, B4, B5) and low (R6, R8 and R10).
The grouping of sampling stations according to the concentration of "pollutants" metals, fits well with the estimates made for the sampling stations according to traffic levels and degree of urbanization.

Indoor-Outdoor Metals Concentration Relationship Pb, Cu, Zn and Cr concentrations are higher in houses dust samples compared with the street dust samples, while the content of Fe, Ni and Li is very slightly higher in house dusts, but significantly lower that ground samples (Table 2). The average content of metals in the houses dust suffers different relative increase (in%) for different elements. This increase is bigger for Cu (201 and 137%), Zn (161 and 219%), Pb (23 and 99%) and Cr (178 and 134%), respectively the total and the soluble fraction, while Ni displays a growth only the extractable part (7.4 to 169%). By metals levels comparison for both types of samples for 5 pairs of sampling stations located close (near) to each other (R1 and B1, and B2 R2, R3 and B3, and B4 R5, R8 and B5) we can see that the total content of all metals, for almost all pairs of sampling stations (except R8-B5 one), is higher in the houses dust compared to the streets one. Only Pb displays almost the same total content for both categories of samples. The chemical composition of the particles is mainly influenced by the presence of different internal resources and the penetration of external environments dust [Lebret, 1987]. The rapport contents determination of a metal in indoor environment towards its outdoors content (I / O), helps to judge the presence or not of significant interior resources of that metal [Cullaj, 2000]. The I / O> 1 rapport, indicates that internal resources are the main contributors of the content of this element in the indoor environment. However, a I / O <1 rapport does not necessarily mean a lack of internal pollution resources, keeping in mind that the efficiency of aerosols penetration from outside is less than 1. It results that for Cu, Zn, Pb and Cr, I / O <1, confirming thus the existence of internal sources of these metals (Table 3). For Ni, Fe and Li I / O ≈1, meaning that their presence in indoor environments is mainly due to external resources. In many cases in the literature is reported that the content of heavy metals in houses dust samples is smaller than the respective contents in dust streets samples, while for dust Elbasan samples results the opposite [N. C. Jones, 2000; L. Wallace, 1996; Diemel , 1981; Yokum, 1982]. We think that some of the reasons of this reversing rapport are:

a. The relatively low traffic level in the main part of the city, the industrial activity reduction as a result of the closure of several stationary emission sources factories (cement, coke, oxygen) and plants (factory 12, nickel-cobalt factory etc.) of the metallurgical plant, and the installation of capture dust filters from the remaining part. Also the wind direction generally does not favor the spreading of particulate matter from industrial zone sources towards the city. In addition to this, street dust samples in Elbasan, because of bad streets conditions, contain high amounts of solid particles originating from the ground, which "slenderize" the heavy metal content of the samples.

b. The content of metals in house dust samples increases due to additional internal resources of their emission. Among these sources, smoke can be considered as a major source of solid particles and metals in their composition of indoors environment [Krause C., Dube P., 1987]. House dust may suffer a significant enrichment with heavy metals, from the usage of various chemicals as detergents, paints, pigments, spray, deodorants, gas heaters or fire, or from such materials as carpets, rubber, plastic, rubber, house appliances, computers, etc. [B. E. Davies, 1985] .

Extraction rate (dissolution) From the results of Table 4 we can notice that the extent of metals extraction displays changes from one element to another and also for the same element in house dust samples from the streets one.

Table 4: The elements extraction rate (in%) in both types of samples.

Solublity (%)	Cu	Zn	Pb	Cr	Ni	Fe	Li
Street dust	31.4-71.2	50.8-95.3	29-51.7	13.3-28.8	9.4-15.5	4.5-8.5	12.3-38.9
Home dust	30.0-66.9	76.2-95.6	51.2-88.1	9.0-23.7	18-40.6	4.4-7.0	8.1-13.7

The most extractive element for both types of samples is Zn, followed by Pb and Cu while Fe, Li, Ni and Cr are extracted to a lesser extent. The different rates of extraction for each element show the different chemical nature of the samples taken at different stations. The variability in the extraction rate between the two types of samples (street-home), testify the different nature of pollution sources particles with metals from internal and external environments. Generally, for all elements, the extraction rate is higher in dust houses samples, compared to external environments. If we compare the dust samples (streets and houses) and ground samples, their ranking according to the elements extraction degree, it would be this: house dust> street dust> ground dust (Fig 3). The increasing extraction degree of elements in houses dust samples in relation to street dust samples, is due to that part of metals originary from the anthropogenic pollution sources. Metals with such origin apparently are extractable from this metal when is originating from the ground. So, is also important the fact that the pollution metals of dusts are connected to the smallest particles, which facilitates their extraction process.

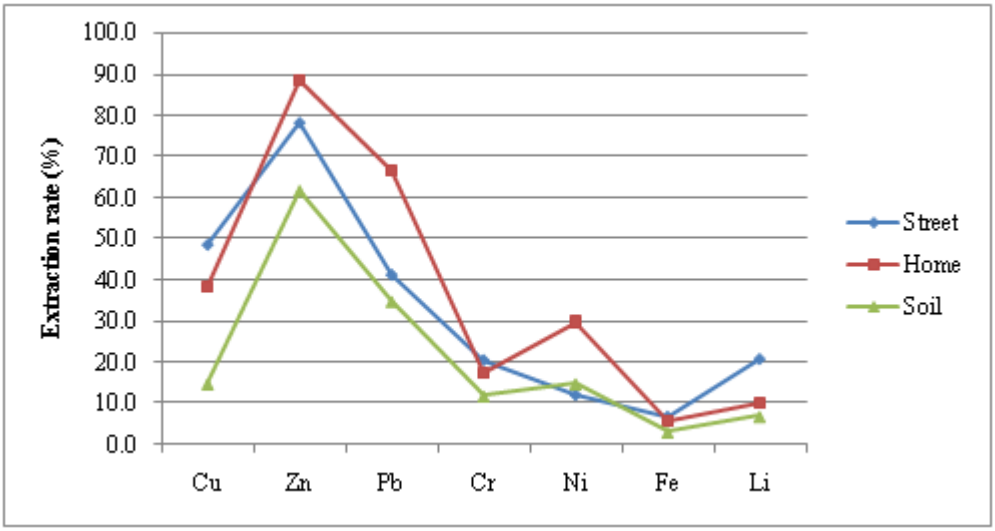

Figure 3: The rate of extraction of elements in three types of samples

4. Correlations

With the method of linear regression analysis were defined the correlations between the content of the different elements in the analyzed samples.

The data obtained by this method shows that:

For the house dust samples treated with the acid disaggregation method exists a good correlation between the "pollutant" elements Cu-Pb-Zn (r> 0.6) and between the ground elements Fe-Ni (r> 0.9). This correlation results more strong in the obtained results of the subject extraction method, namely: Cu-Zn-Pb (r> 0.8) and Cr-Zn-Pb (r> 0.7) .We notice that the elements that correlate in this case, are precisely those for whose we concluded that have internal anthropogenic resources. The high correlation of the extractable portion of these metals testifies the same sources of their emission in the indoor environments.

In the street dust samples, generally there is a lack of strong correlations between the elements, except that between the content of Pb and Zn (r = 0.65) and Ni-Cr (r = 0.81). Somewhat inherent correlations are observed between the ground elements Li-Fe-Ni-Cr. For the extractable metals part, the correlation remains high for the relation Ni-Cr (r = 0.87), while other correlations are weakened.

Date normalization, Enrichment Factors, Geo-accumulation indexes

The process of normalization of data enables us the evaluation of the contribution of anthropogenic sources of metal content in dust samples [Loring D., H., Rental R., T., (1992)]. To judge the origin of the content of metals in dust samples for each of the elements analyzed, was defined the enrichment factor and the geo-accumulation index. The evaluation of the first was conducted using as a normalization element the iron and as reference sample the ground sample, which content represents the natural content of elements in absence of pollution sources. To calculate the enrichment factor was used the formula:

$$EF = \frac{\left(\frac{C_i}{C_{Fe}}\right)_{dust}}{\left(\frac{C_i}{C_{Fe}}\right)_{ground}}$$

While the accumulation index will be calculated with the formula:

$$I_{geo} = \log_2\left(\frac{C_n}{1.5B_n}\right)$$

where Cn is the element concentration in the sample and Bn represents the concentration of this element in the reference sample (ground in our case). Based on these values, the defined metals can be classified into two groups: - "Pollutant" Metals: Pb, Cu and Zn, for whom EF> 1 and I Geo> 0; "ground" metals: Cr, Ni, Li and Fe, for whom EF <1 and Igeo <0.

The higher values of the enrichment factor among polluting elements, belongs to Pb, as for street dust (EF = 2.5-25.6) and also for house dust (EF = 4.2 - 24.1). Also Pb represents the highest values of geo-accumulation, such as road dust samples (Igeo = 0.1-2.7) also in houses dust samples (I_{geo} =

0.9-3.1). In the total of the analyzed samples, Pb displays a geo-accumulation index in the range 2 -3 to 50% of the house samples, while for Cu and Zn the values of this index goes at a maximum of 2.3 and 1.4 respectively.

The highest values of EF and I_{geo} for pollutant metals result for samples taken at stations located in areas with dense traffic, especially for Pb, although the number of gasoline vehicles that are the major source of pollution with this metal is relatively low. The annual amount of Pb deposited on the surface of our country is estimated at about 3.7mg / m2 [UNEP / WHO, (1994)]. More or less the same areas of these sampling stations are presented as the most affected one by the industrial activity, in terms of geographical proximity and influence of wind direction that with the cross-border transport, transports the suspended particles towards these areas. This is more accentuated in the case of Pb but also Cu, Zn and sometimes Ni.

Conclusions and Recommendations

1) There exists a direct link (connection) between the content of "pollutants" metal (Pb, Cu Zn) in the dust samples taken in different areas of the city of Elbasan and the traffic level, the degree of urbanization and the proximity or impact of the stationary pollution emissions sources. According to this we can mention the Steel Production Plant "Kurum", the Ferrochrome Plant, both 5km at the west of the city (former the Metallurgical Plant) and areas of urban waste disposal about 1 km in the south of the city. The difference is observed for the street dust samples as well as for the house dust.
2) The concentrations of pollutants metals in houses dust samples are generally higher than those of the street dust samples. This can be explained by two arguments: First, the stationary pollution sources are reduced and are relatively far away, and secondly, the existence of additional resources in the indoor environment. Also, the street dust samples seems to be "diluted" by the large metal particles originating from the earth's crust, the impact of which is felt even more because of the bad condition of the streets. Meanwhile houses dusts are more rich with tiny particles of anthropogenic origin from the indoor activities but also by the inter-border transport.
3) Despite the relatively low levels of metals in the samples analyzed, the large amount of dust in the city of Elbasan is an important source of population exposure to heavy metals and other harmful substances in their composition. Except the directly exposure, it can also be done indirectly through their transition into the food chain as a result of storage in the ground, water and then in the biosphere.
4) The results of this study represent a sort of base, though not complete, for evaluating the quality of urban air of Elbasan and also of the indoor houses environments. A more thorough study would require not only a time stretch and a most comprehensive coverage of the space, but also the most complete chemical analysis of the particles components, which in addition to heavy metals, it will be also interesting other in and organic pollutants. This would enable from the other hand the creation of a full picture of the pollutants, their sources and the risk of population exposure to them.

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