Rev. Int. Contam. Ambient. *10* (2), 77-82, 1994

HEAVY METALS IN ROCKS AND STREAM SEDIMENTS FROM THE NORTHWESTERN PART OF BAJA CALIFORNIA, MEXICO

Leticia ROSALES HOZ, Arturo CARRANZA EDWARDS and Susana SANTIAGO PÉREZ

Instituto de Ciencias del Mar y Limnología, Universidad Nacional Autónoma de México, Ciudad Universitaria, Coyoacán 04510 D.F. México

(Recibido enero 1994, aceptado agosto 1994)

Keywords: heavy metals, sediment pollution, Cu, Cd, Zn, Ni, Cr, Ti

ABSTRACT

In order to assess the amount of heavy metals apported by natural weathering to the coastal area of the northwestern part of Baja California, México, rocks from outcrops, rock fragments and sediments from the Guadalupe River and other streams that drain towards this area were studied. Major elements (Si, Al, Na, K, Ca, Mg, P, Fe, Mn and Ti) and some trace elements (Cu, Cd, Zn, Ni, Cr and Pb) were evaluated in the samples studied. The Guadalupe River sediments show the greatest weathering and the San Antonio stream sediments show the lowest values of the index of chemical alteration. Heavy metal concentration in stream sediments were: 0.02 to 0.17% for MnO, 0.07 to 1.49% for TiO_2, 10 to 20 ppm for Cu, 10 ppm for Cd, 30 to 140 ppm for Zn, 30 to 40 ppm for Ni, non-detected to 150 ppm for Cr. All the reported concentration are within values reported for non-contaminated soils, except Cr whose upper limit has been established at 75 ppm in Switzerland and 100 ppm in Germany.

RESUMEN

Con el objeto de evaluar la cantidad de metales pesados suministrados por procesos naturales de intemperismo en el área costera del noroeste de Baja California, México, se estudiaron rocas que afloran en las cuencas y fragmentos de rocas y sedimentos de los ríos y arroyos que drenan hacia esta área. Se evaluó la concentración de elementos mayores (Si, Al, Na, K, Ca, Mg, P, Fe, Mn y Ti) así como algunos elementos traza (Cu, Cd, Zn, Ni, Cr y Pb). Los sedimentos del Río Guadalupe son los que muestran el mayor grado de alteración, en tanto que los sedimentos del arroyo de San Antonio el menor grado. La concentración de metales pesados en los sedimentos estudiados fue de: 0.02 a 0.17% de MnO%, 0.07 a 1.49% de TiO_2, 10 a 20 ppm de Cu, 10 ppm de Cu, 10 ppm de Cd, 30 a 140 ppm de Zn, 30 a 40 ppm de Ni, no detectado a 150 ppm de Cr. Las concentraciones evaluadas en los sedimentos están dentro de los límites establecidos para suelos no contaminados, con excepción del Cr cuyo límite máximo permitido en Suiza es de 75 ppm y de 100 ppm en Alemania.

INTRODUCTION

The industrialization and development of the coastal zone has contributed to an increased pollution of estuaries and coastal marine waters with heavy metals. Most of the heavy metals in estuaries and coastal waters come from freshwater runoff and the atmosphere.

Heavy metals in coastal areas may have a natural origin, for example from weathering of rock outcrops that compose the basin, or they are anthropogenic. A significant fraction of heavy metals in coastal waters originates from domestic and industrial wastes; an example of this would be substantial amounts of copper, lead and zinc released from pipes and tanks in domestic systems which eventually enter natural waters (Kennish 1992).

Some studies realized about watershed weathering (e.g. Lush 1984) have shown that the rate of loss of major nutrient elements (e.g. Ca, Mg, Na, K) and trace elements depend on the type of minerals exposed to weathering, the physiography of the basin, the climate to which the watershed is exposed, the nature of the vegetative cover and the natural or anthropogenic events ocurring within the watershed.

Recently regional geochemical surveys have been undertaken in different areas to predict an environmental impact. The study of the regional distribution of a particular element tries to pinpoint anthropogenic sources of industrial, urban or agricultural chemicals that may be contributing to a real or perceived environmental degradation. An initial sampling will show background levels. These levels may then be forecast to change with time in a predictable manner.

In the past few years the development of the coastal zone in the northern part of Baja California (México) has increased considerably. The Port of Ensenada has augmented its activity as well and the urban areas of the cities of Ensenada and Tijuana have grown tangibly. In the present study, rocks from outcrops, rock fragments and sediments from streams that flow towards Todos Santos Bay and the coastal zone in the northern part of Baja California México, were chemically studied in order to evaluate the natural aluminum, titanium, manganese, copper, cadmium, zinc, niquel, lead and chromium sources which are ultimately apported to the coastal area.

STUDY AREA

The study area is located on the northern portion of Baja California, México, from 31° 30' to 32° 15' north and 116°00'to 117°00' west. 6 rock fragments, 15 rock outcrops and 11 sediment samples were collected on basins and streams flowing towards Todos Santos Bay and the northern part of the bay, in a region of diverse lithological nature **(Fig. 1)**. On the highlands the climate is temperate with winter rains, whereas on the lowlands it is dry (Tamayo 1991).

The fluvial system of the area **(Fig. 2)** is mainly formed from north to south by the Guadalupe River, whose source is in the Sierra de Juárez. Its basin area covers 2,484 km^2 and drains through schist, undiferentiated volcanic material, basalts, granodiorites and gabbros before reaching the sea. Towards the south a series of small streams are present, for example: San Miguel, Sauzal, Ensenada and El Gallo. Their total basin area covers 780 km^2. The most important of these streams is the stream of Ensenada, which is approximately 28 km long and goes through volcanic material, tonalites and granodiorites, gneisses and alluvion. South of El Gallo, is the San Carlos stream, whose source is in La Sierra Juárez which has an altitude of 1860 m. This stream has a basin area covering 729 km^2 and it runs through volcanic material, tonalites, gabbros and alluvion, before reaching the sea 12 km south of Ensenada. The San Antonio or Las Animas stream, flows 75 km before reaching the sea 18 km south of Ensenada. Its basin area covers 940 km^2. It flows through: gabbro, granodiorites, gneisses, tonalites, undifferentiated volcanic material conglomerates and alluvion. The Santo Tomás stream with its source on El León Hill 1,700 m high, is 70 km long and covers a basin area of 707 km^2. It flows through gneisses, tonalites, undifferentiated volcanic material, conglomerate and alluvion (Pozos-Salazar 1985).

According to Pozos-Salazar (1985), the highest apport

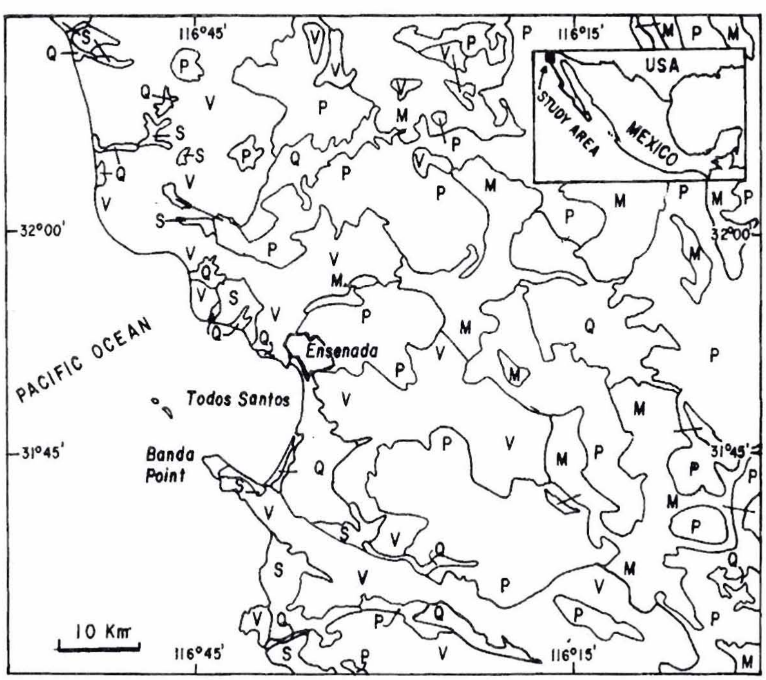

Fig. 1. Study area and sinthetized lithologic map modified from INEGI (1982)

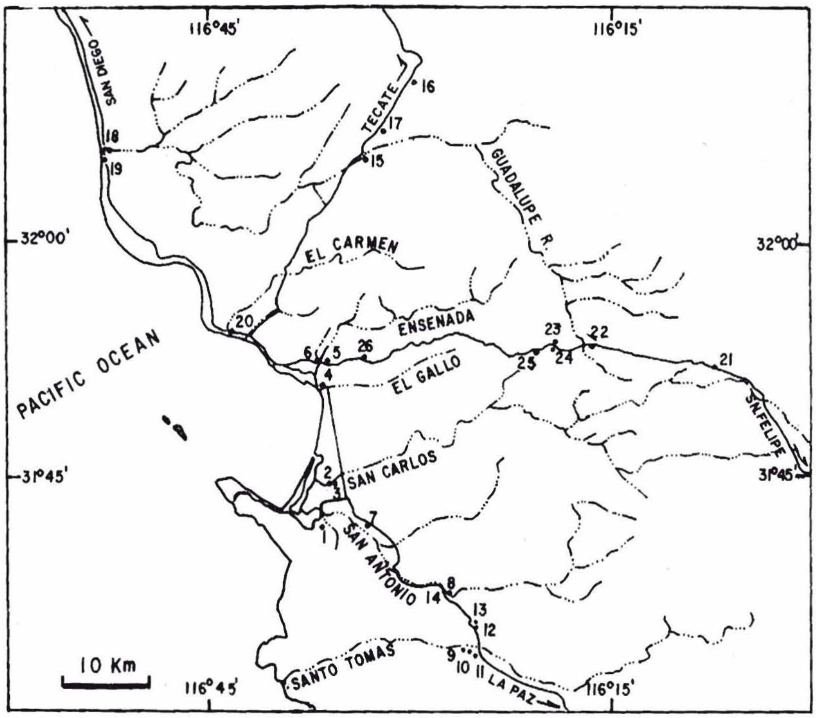

Fig. 2. Location of the sampling sites and fluvial net

of sediment to the coastal area comes from the Guadalupe River, with an average apport of 834,889 tons per year followed by the San Antonio stream with 230,995 tons/year. The San Carlos stream apports 210,080 tons/year, San Miguel, Sauzal, Ensenada and El Gallo 172,524 tons/year and Santo Tomás 164,054 tons/year.

MATERIALS AND METHODS

Rocks from outcrops, rock fragments and sediments from streams were collected in the study area. Rocks and rock fragments samples were ground in an electrical mill. Sediments were dried in an oven at 105°C and ground manually. Major element analysis (Si, Al, Na, K, Ca, Mg, P, Fe, Mn and Ti) were made by XRF. Samples were fused with lithium tetraborate at 1150°C and compared with geological standards on a Philips PW-1050/25 difractometer with a chromium anticatode at 60 Kv and 20 mA. Trace metals were extracted by the digestion of the dried sample for 30 minutes in a CEM microwave oven with H_2O:HNO_3:HF:HCl (10:5:4:1). Extracts were diluted to 25 ml with a saturated boric acid solution and analysed on a Varian Spectra AA-10 Plus Atomic Absorption Spectrophotometer.

Variation coefficient on trace metal evaluation was on an interval of 0.95 to 4.38%. The accuracy of the method was established by using a SD-N-1/2 sediment sample (IAEA 1985) and all metals reported were within the established confidence interval at a 0.05% significance level.

RESULTS AND DISCUSSION

Chemical analysis of rocks from outcrops, rock fragments and sediments collected on the streams studied are shown in **Table I.**

The rate of rock weathering is controlled by a number of variable factors, with climate and rock composition being some of the most important. Aluminum is almost immobile during chemical weathering, whereas other oxide components are lost, resulting in a rising concentration of aluminum as weathering progresses (Faure 1991). The index of chemical alteration (CIA) (Ibbeken and Schleyer 1991), based on aluminum, calcium, sodium and potassium concentration, $[(Al_2O_3/Al_2O_3 + CaO + Na_2O + K_2O) \times 100]$, was used to evaluate the degree of weathering. In the present study the rocks and the sediments from the Guadalupe River have the highest CIA values, which suggest that they are highly weathered. The San Antonio stream sediments show the lowest CIA values. Based on Pozos-Salazar (1985) data, the amount of sediments apported by each of the basins studied is correlated with the basin area, and not with the type of rocks present and their CIA values.

The analysis of major elements demonstrates a higher occurrence of intermediate type rocks (SiO_2 between 52 and 66%) and felsic type rocks (SiO_2 greater than 66%).

San Antonio and Santo Tomás basin outcrops show the rocks with the highest Fe_2O_3 and TiO_2 content; these values are reflected on the sediment sample collected

TABLE I. CHEMICAL ANALYSES OF ROCK (R) AND SEDIMENT (S) SAMPLES COLLECTED IN DIFFERENT BASINS AND STREAMS
(oxides express in % and elements in ppm)

Sample	Type	SiO_2	Al_2O_3	Na_2O	K_2O	CaO	MgO	P_2O_5	Fe_2O_3	MnO	TiO_2	LOI	Cu	Cd	Zn	Ni	Cr	CIA
SANTO TOMÁS STREAM																		
10-1	RF	69.27	17.58	4.84	1.95	3.06	0.00	0.13	2.52	0.09	0.18	0.40	10	10	30	30	100	64.09
10-2	RF	52.36	20.17	3.28	0.36	6.30	7.50	0.33	7.96	0.15	1.09	0.50	60	10	70	50	50	67.00
10-3	RF	59.69	16.97	2.37	0.39	2.77	7.31	0.22	8.77	0.24	0.98	0.38	10	10	170	40	50	75.42
10-4	RF	70.67	18.44	2.68	4.33	1.52	0.66	0.11	0.99	0.04	0.32	0.25	10	10	20	30	100	68.38
10-5	RF	67.65	16.95	0.93	5.35	2.10	1.54	0.21	4.07	0.04	0.46	0.34	20	10	70	40	50	66.90
10-6	RF	64.04	20.41	4.13	0.63	1.91	2.29	0.17	4.96	0.19	0.75	0.53	30	10	770	40	200	75.38
11	R	62.61	21.49	4.23	1.63	3.07	0.62	0.22	4.10	0.11	0.58	1.32	20	10	50	40	300	70.63
9	S	60.95	18.05	2.44	1.08	4.93	1.54	0.27	8.40	0.16	1.49	0.68	20	10	70	40	50	68.12
SAN ANTONIO STREAM																		
12	R	50.02	21.31	4.22	0.21	1.83	1.06	0.30	14.60	0.49	1.46	4.49	20	10	60	40	50	77.29
13	R	50.18	22.31	3.88	4.59	4.01	1.78	0.28	8.63	0.18	1.07	3.10	20	10	60	50	50	64.12
14	R	56.46	24.78	3.13	1.33	5.62	2.10	0.35	4.99	0.07	0.79	0.37	100	10	80	50	150	71.09
7	R	68.22	16.54	3.96	1.35	3.04	0.57	0.24	4.19	0.11	0.55	1.23	10	10	70	30	100	64.96
8	S	64.20	18.98	2.84	0.85	4.88	1.70	0.25	4.85	0.09	0.80	0.57	10	10	40	40	50	68.89
1	S	68.01	16.70	3.64	1.07	4.88	1.40	0.20	2.91	0.09	0.45	0.67	10	10	40	30	0	63.53
SAN CARLOS STREAM																		
21	R	55.39	27.39	2.85	1.46	5.41	1.58	0.26	4.20	0.09	0.47	0.89	20	10	70	40	0	73.81
23	R	63.22	22.68	3.11	1.35	2.91	0.66	0.12	1.32	0.03	0.17	4.43	20	10	20	40	0	75.46
25	R	71.41	15.87	0.54	1.50	5.22	0.96	0.02	2.51	0.10	0.27	1.59	40	10	40	60	0	68.61
26	R	59.36	26.17	2.91	1.25	4.35	1.08	0.16	1.82	0.09	0.41	2.40	40	10	50	50	0	75.46
2	S	72.36	14.33	2.82	3.45	3.40	0.77	0.10	1.80	0.04	0.27	0.65	10	10	40	20	0	59.70
3	S	49.83	21.70	2.28	1.41	2.95	1.96	0.25	7.39	0.17	0.86	12.13	50	10	140	40	0	76.57
24	S	70.86	21.80	3.16	2.86	0.72	0.05	0.05	0.45	0.02	0.09	0.81	20	10	20	30	0	76.38
EL GALLO STREAM																		
4	S	71.86	16.28	3.16	1.82	3.69	0.65	0.12	1.56	0.04	0.28	0.55	10	10	40	30	0	65.27
ENSENADA STREAM																		
6	R	67.22	16.57	3.29	2.79	3.35	0.96	0.13	3.52	0.07	0.48	1.62	20	10	30	30	0	63.72
5	S	74.09	15.38	2.82	1.23	3.58	0.63	0.09	1.30	0.03	0.18	0.77	20	10	30	30	0	66.84
EL CARMEN STREAM																		
20	S	68.21	19.86	2.19	1.81	2.29	0.66	0.08	2.20	0.05	0.32	2.34	30	10	50	40	0	75.94
GUADALUPE STREAM																		
22	R	47.55	26.88	0.75	0.55	10.43	5.65	0.48	6.89	0.17	0.31	0.34	70	10	70	160	350	69.62
16-1	R	61.62	26.53	1.09	3.85	0.67	0.39	0.18	4.47	0.05	0.58	0.58	40	10	120	70	100	82.55
16-2	R	77.11	15.42	2.97	0.75	1.01	1.01	0.14	0.93	0.04	0.31	0.31	30	10	110	40	250	76.53
17	R	60.31	27.14	3.71	2.35	0.64	0.64	0.11	2.85	0.08	0.35	1.83	20	10	50	30	0	80.18
19	R	54.32	26.95	3.45	1.98	0.82	0.82	0.33	5.72	0.04	1.33	4.25	60	10	60	60	100	81.18
15	S	68.53	22.63	2.50	2.57	1.61	0.20	0.05	0.49	0.01	0.07	1.34	20	10	30	40	150	77.21
18	S	66.43	20.35	3.48	1.08	1.43	1.43	0.15	3.41	0.08	0.56	1.62	20	10	40	40	50	77.26

R: rocks from outcrops, RF: rock fragments from streams, S: sediments near the rock outcrops rich in these elements. Samples from Santo Tomás show the highest Na_2O values as well as the lowest K_2O values. A rock sample from the Guadalupe River shows the highest MgO and the lowest SiO_2 content; significant amounts of Cr_2O_3 are normally present in these types of samples, which represents an extreme in the chemical composition of common igneous rocks. Sample 22 from Guadalupe River shows these characteristics. It contains 5.65% of MgO and 350 ppm of Cr, the highest values observed in all the analysed samples, as well as 47.55% of SiO_2, the lowest value measured in this study.

Cu, Cd, Zn, Ni, Pb and Cr trace elements were studied, in order to evaluate if the concentration levels present in sediments were originated by natural weathering of the rock outcrops of the area. The reported concentration of Cu in the earth's crust is approximately between 50 and 90 ppm in igneous rocks and between 20 and 30 ppm in soils (Wedepohl 1991). In the present study a Cu concentration from 10 to 100 ppm was found in rocks and 20 to 50 ppm in sediments. A rock sample from the San Antonio stream shows the highest Cu value (100 ppm), but the sediment from this area averages 10 ppm. Cu is usually concentrated in the clay mineral fraction with slight enrichment in clays rich in organic carbon. This is the case for sediment sample 3 located on the San Carlos stream, which has the highest concentration of Cu (50 ppm) and organic matter (2.13%) among the samples studied.

The average Cd concentration of the earth's crust is estimated to be about 0.1 mg/Kg (Wedepohl 1991). Cd resources are linked to the presence of zinc. In the present study a homogeneous value of 10 ppm for rocks and sediments was observed. Zinc concentration of non-polluted soils varied from 10 to 300 ppm and were comparable with their rocky subsoils. On the average, levels of 20 ppm are found in non-polluted soils (Wedepohl 1991). The continental crust contains average concentrations of 69 ppm, with values of 50 ppm in granitic rocks, 65 ppm in gneisses, mica and schists, 23 ppm in limestones and 105 ppm in greywackes. In the present study the highest zinc values **(Table I)** correspond to rock fragments from the Santo Tomás stream. These values of 770 ppm belong to a sample with high Na values as well, which could be attributed to the presence of sodium amphibole reported to contain up to 4.7% of ZnO (Rankama and Sahama 1962). However, the Zn values in sediments are in the 30 to 40 ppm range, well within non-polluted soil values.

Niquel concentration average in the continental crust is 45 ppm and the average content in basaltic rocks is 134 ppm; at pH 6.5 or lower most Ni compounds are soluble and acid rains may therefore lixiviate Ni from rocks and soils (Wedepohl 1991). In polluted areas organic Ni compounds in rivers are absorbed by silica particles, with the gradual accumulation of Ni in the upper layers of mud. In the present study, Ni values in rocks are in the 20 to 160 ppm range, with 20 to 40 ppm values in sediments.

The average chromium concentration in the continental crust is 88 ppm, but values up to 168 ppm are found in basaltic rocks (Wedepohl 1991). The danger of environmental pollution by Cr depends on its oxidation state. Its hexavalent form is 100 to 1000 times more toxic than the most common trivalent compounds (Merian 1991). In the present study Cr values in rocks range from non-detected to 350 ppm and the concentration in sediments from non-detected to 150 ppm. Rocks and sediments from the

TABLE II. CORRELATION COEFFICIENT OF CHEMICAL ELEMENTS ON ROCK AND SEDIMENT SAMPLES STUDIED

	SiO_2	Al_2O_3	Na_2O	K_2O	CaO	MgO	P_2O_5	Fe_2O_3	MnO	TiO_2	Cu	Zn	Ni	Cr
SiO_2	1.00	-0.63	-0.01	-0.21	-0.38	-0.50	-0.80	-0.77	-0.60	-0.65	-0.45	-1.09	-0.51	-0.14
Al_2O_3		1.00	0.00	0.02	-0.05	-0.02	0.33	0.15	0.03	0.18	0.43	0.03	0.33	0.10
Na_2O			1.00	-0.21	-0.28	-0.21	0.00	0.09	0.21	0.19	-0.26	0.13	-0.52	0.00
K_2O				1.00	-0.35	-0.40	-0.29	-0.29	-0.42	-0.25	-0.22	-0.22	-0.17	-0.17
CaO					1.00	0.53	0.56	0.24	0.16	0.06	0.42	-0.11	0.59	0.19
MgO						1.00	0.57	0.51	0.40	0.37	0.35	0.21	0.44	0.20
P_2O_5							1.00	0.72	0.48	0.64	0.53	0.07	0.59	0.42
Fe_2O_3								1.00	0.89	0.87	0.18	0.17	0.27	0.09
MnO									1.00	0.66	0.00	0.26	0.16	0.09
TiO_2										1.00	0.21	0.19	0.06	0.01
Cu											1.00	0.08	0.61	0.38
Zn												1.00	0.03	0.29
Ni													1.00	0.56
Cr														1.00

Significative correlations with 99% confidence: values above 0.449
Significative correlations with 95% confidence: values above 0.349

stream of San Carlos, El Gallo, Ensenada and El Carmen did not demonstrate the presence of Cr. Rocks from the stream of Santo Tomás and San Antonio show values ranging from 50 to 300 ppm Cr and from 0 to 50 ppm in sediments. The Guadalupe River rocks show values ranging from 50 to 350 ppm and from 50 to 150 ppm in sediments. The rock with the highest value (sample 22 with 350 ppm Cr) is located in the Guadalupe River basin, on a small gabbro outcrop which can be observed on the California Geological map of Gastil *et al.* (1971). This was confirmed by the observation of a thin section of this rock. The accessory minerals of gabbro are magnetite, ilmenite, chromite and apatite, which explains the high Cr, Ca, P, Fe and Mg concentrations observed in this sample. Sediment values in this area are slightly higher given that the allowed Cr concentration in soils in Switzerland is 75 ppm and in Germany 100 ppm (Morgan 1991).

An analysis of the correlation of the elements studied shows **(Table II)** the highest values for Fe when correlated with Mn, Mg and Ti; this could be explained by the presence of minerals such as ilmenite ($FeTiO_3$) whose formula can be more realistically expressed as (Fe, Mg, Mn)TiO_3. Ilmenite is a common accesory mineral in igneous rocks and may be massively present in gabbros, diorites and anorthosites, these minerals are commonly the source of a large percentage of ilmenite recoverd from beach sand (Klein and Hurlbut 1985). The high correlation for Ca and Mg can be explained by the presence of dolomite ($CaMg(CO_3)_2$), and for Ca and P by the presence of apatite which is widely disseminated as an accesory mineral in all classes of rocks (Klein and Hurlbut 1985). The high association of Ni to Ca and P suggests that this metal is trapped around the apatite crystals.

Of the elements studied, Cu, Cd, Zn and Pb are reported as atmophile elements (Morgan 1991), with interference factors (IF) [(total anthropogenic emisions/natural emisions) X 100] considerably higher than 100%. The IF reflects the importance of anthropogenic fluxes. The values found in the present work, show that the concentration levels found correspond to natural values. Elements such as Al, Ti, Mn and Cr are lithophile. Their mass transportation by streams exceeds their transportation by the atmosphere. The results evaluated in the present work show that their concentration is within the natural levels expected in the area, which at present do not demonstrate any anthropogenic effects.

CONCLUSIONS

The degree of weathering changes for the different material sampled in the streams studied. The samples from the Guadalupe River show the highest CIA values, because it has the highest degree of weathering in the area. The material collected in the stream of San Antonio shows the lowest CIA values. The metals studied show concentration levels in sediments in the following order of magnitude: 14.33 to 22.63% for Al_2O_3, 0.01 to 0.17% for MnO, 0.07 to 1.49% for TiO_2, 10 to 50 ppm for Cu, 10 ppm for Cd, 30 to 140 ppm for Zn, 30 to 40 ppm for Ni, 0 to 150 ppm for Cr; Pb was not detected. The metal concentration in sediments is within expected natural levels based on reported values in literature and is in agreement with the composition of rocks in the areas studied. The only metal with slightly higher values is Cr, whose maximum allowed concentration in soils in other countries varies from 75 to 100 ppm.

ACKNOWLEDGEMENTS

We thank J. M. del Moral Briones for his assistance in the collection of the samples. Thanks are also due to the authorities of the Instituto de Geología, UNAM, for allowing us the use of the XRF equipment, to R. Lozano Santa Cruz for his help in grinding rock samples and to P. Altuzar Coello for her technical assistance in FRX analysis.

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