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Hydrochemistry of Groundwater in the North Numidian Mercurial Zone (Azzaba), North East Algéria: Effect of Mercury Contamination of Population

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Abstract: The mercury ore deposit of Azzaba is located on the northern flanc of the limestone Chain, south of Azzaba town, North East Algeria. The study area is characterized by two aquiferous. The Oligocene aquifer is composed of fissured sandstone alternating with argillaceous layers whereas the deep aquifer of Paleocene-Eocene age and sediments consisting of sandstones, limestones and conglomerates-breccias. These two aquifers are separated by the impermeable rocks of the Paleozoic and the Lutetian-Priabonian, which represents the mercurial mineralization of Cinnabar (HgS), characterized by breccias limestone, clays and limestone. Nineteen springs and wells from the two aquifers were sampled for chemical analyses. The most dominant facieses are bicarbonated calcic, sulfated calcic and chlorided sodic. Water of the Oligocene aquifer shows high Hg contents (up to 80 $\mu g.L^{-1}$) whereas in the deep aquifer, Hg contents do not exceed 7 $\mu g.L^{-1}$ though the litho-stratigraphic facies with a high Hg Clarke are located in the deep aquifer.The calculated coefficient of Hg in the area is moderate following the stability of cinnabar, compared to those of Zn and Cu wich are much higher.The increase of Hg in water of the Oligocene is not the result of the leaching of geological formations (Cinnabar). The mean urinary inorganic Hg rate in the plant worker is about 650 $\mu g.G^{-1}$ of creatinine. Clinical signs of Hg contamination within school children are compared between those of Azzaba region and those of Annaba, located 80 km East of Algeria taken as a non contaminated reference. School children of Azzaba region have a mean rate of 2.50 $\mu g.G^{-1}$ compared to 0.45 $\mu g.G^{-1}$ for those of Annaba.The mercury norm is largely exceeded in groundwater as well as in population living in the Azzaba region.

Key words: Geology, water, mercury, migration, pollution, norms

INTRODUCTION

The area of study is located in the North of Algeria on the Northern side of the numidian chain, 30 km away from Skikda and 3 to 10 km from Azzaba (Fig. 1).

The geology of the area is characterized by the presence of two structural groups, one autochthonous and the other allochtonous.

The autochtonous one is composed of deposits of the Kabyle dorsal including formations whose age varies from Trias to upper Eocene. The dislocated deposits of Paleocene are made of sandstones, limestones, micro breccia and breaches are found. The Thanetien-hypresien is made of quartzetic sandstone and limestone and Hyprésien-lutétien is made of conglomerates, limestone micro breccia and sandy limestones. The allochtonous group is formed by a series of rock scales of various age and composition, ranging from metamorphic schists of Paleozoic to clays of Oligocene (Bouarroudj, 1986).

The mercury ore deposits of the district are well located and are gathered in two mining fields. The first field, Ismaïl, is formed by three distinct ore deposits, Ras El Ma, Ismaïl and Guenicha, the horizon carrying the mercurial mineralization are the conglomerates and the limestone micro breccia of Hyprésien-luthétien. The second, Mra-Sma, is formed by Mra-Sma I, MraSma II and koudiat Sma, the carrying horizon of the mineralization being the limestone pseudo breccia of the Campanien and the sandstones of the Neocomien (Bouarroudj, 1986; Buzina *et al*., 1989; Anonymous, 2004; Charlot, 1978; Langworth *et al*., 1992; Lauverys, 1992; Mezhache, 1989).

The hydrogeology of the area is characterized by two water tables. The Oligocene phreatic water where the aquiferous rocks are the fissured sandstones alternating with beds of clay and the Paleocene Eocene deep confined aquifer, largely developed in the area, and where groundwater is associated to the sandstone, limestone, the conglomerate breccias, fissured cavernous limestones and the calcareous sandstones of the Paleocene.

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Table 1: Results of chemical analysis

No.	Deposits	Mining fields	Aquifers	Designation	pH	Dry residue ($mg.L^{-1}$)	HT (°F)	Ca^{2+} ($mg.L^{-1}$)	Mg^{2+} ($mg.L^{-1}$)	Na^{+} ($mg.L^{-1}$)	K^{+} ($mg.L^{-1}$)	HCO_3 ($mg.L^{-1}$)	Cl^{-} ($mg.L^{-1}$)	$SO4_2^{-}$ ($mg.L^{-1}$)	Hg ($\mu g.L^{-1}$)
1	Rasel Ma		e^{1-3}	85	7.0	644	40	121	24	109	5.6	329	113	70	7
2			e^{1-3}	86	7.0	588	41	129	22	131	4.4	341	95	76	7
3			e^{1-3}	90	6.9	754	45	143	22	110	3.5	450	140	68	7
4			g^2	S.O	7.8	900	21	32	30	45	5.4	152	90	35	7
5	Ismail	Ismail	g^2	350	8.0	138	74	222	45	84	17	311	156	505	80
6			g^2	210	7.0	143	74	208	53	85	19	250	184	524	80
7			g^2	398	8.9	600	38	50	61	41	35	372	89	37	4
8			g^2	325	7.8	650	15	24	18	208	16	427	82	131	4
9			g^2	260	7.8	1050	16	26	9	253	15	274	213	184	5
10			g^2	265	8.0	800	17	21	16	208	15	213	142	139	5
11			e^{1-3}	285	8.9	1170	67	206	37	74	6.6	213	163	450	5
12			g^2	S.2	7.5	1110	60	165	46	68	5.7	518	53	158	80
13			g^2	354	7.5	576	62	120	79	67	10	297	78	104	80
14			e^{1-3}	94	7.0	761	60	166	44	19	9	381	135	150	1
15	Guenicha		e^{1-3}	194	7.0	590	38	50	61	41	3.5	372	89	37	6
16			e^{1-3}	196	7.0	594	35	88	32	68	3	298	150	45	6
17			g^2	S.1	7.0	1061	56	154	43	84	6.5	402	107	194	80
18	Mra Sma	Mra Sma	e^{1-3}	91	7.8	620	38	104	29	46	1.6	311	120	34	6
19			e^{1-3}	93	7.0	820	47	138	31	67	13	300	120	50	1

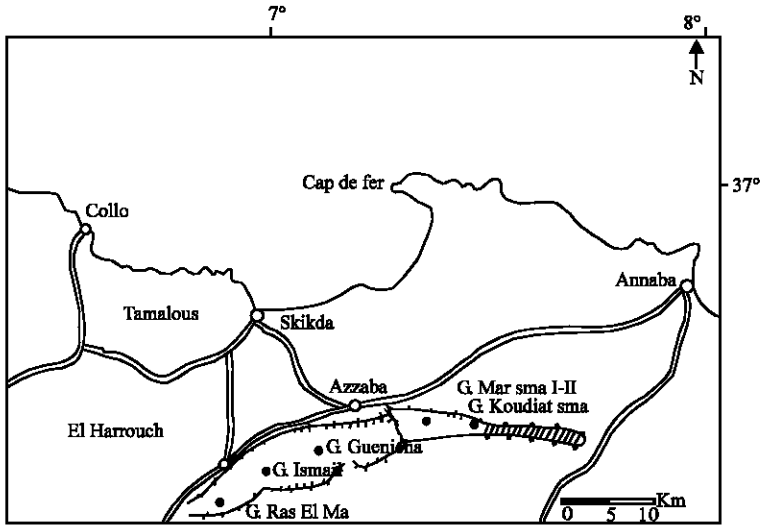

Fig. 1: Location map of the North numidian mercurial zone

The two aquifers are separated by impermeable layer of Palaeozoic and Lutetian-Priabonian. The latter are the host rocks of cinnabar mineralization (Benhamza, 1996).

MATERIALS AND MATHODS

Nineteen springs and wells of both water tables were analyzed to identify the major elements (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, HCO_3^{-} ,Cl^{-}, SO_4^{2-}), the pH, the dry residue, the hydrotimetric grade and the mercury content (Table 1).

Analyses were carried out at the laboratories of the Petrochemical Complex of Skikda and Boumerdes (Algiers).

The dosing processes are deduced from the methods of standard analyses. Total and calcic hardnesses of the samples are determined by complexometry, by titration with the acid Ethylene-Diamine-Tetracetic (EDTA). The chlorides are determined by the method of Mohr (Rodier, 1984) and sulphates by reading the absorbance with λ = 420 nm for a suspension obtained by reaction of sulphates with barium chloride. Sodium and potassium are dosed by photometry with flame emission (Telliard, 2002).

The determination of mercury is carried out in two stages. The first consists in oxidizing all kind of mercury in a bivalent state using an acid digestion to obtain elementary mercury. Mercury is extracted by bubbling dry air in the solution. The gas mixture is then sent to the cell of the atomic absorption spectrophotometer without flame.

The concentration of the sample is determined by comparison between the respective absorbance of the sample and the standard solutions. The range calibration

is between 0.1 µg.L^{-1} and 1.5 µg.L^{-1} Hg. The applicability range can be widened by carrying out suitable dilutions (Anonymous, 2004; Charlot, 1978; Langworth *et al*., 1992; Lauverys, 1992; Mezghache, 1989; Nezzal *et al*., 1994; 1996; 1991; Nixon, 1996; DNEMT, 1996; OMS, 1972; 1980; Rodier, 1984; Schoeller, 1962; Telliard, 2002).

RESULTS AND DISCUSSION

Chemical facies: The ionic balance error calculated using the results of the chemical analyses is satisfactory (< 5%). The cation-anion ratio is included within the acceptable limits (0. 95-1. 05), which makes it possible to check the reliability of the analyses and to accept the results (Rodier, 1984).

The representation of the analyses on the Piper diagram (Fig. 2) shows three chemical facies.The bicarbonated calcic one for the Paleocene Eocene water, the chlorided sodic and the sulphated calcic for Oligocene water (Rodier, 1984; Schoeller, 1962).

The bicarbonated calcic facies, the most important, accounting for 95% of the samples is explained by the presence of cavernous limestone and the calcareous sandstone of the Paleocene Eocene (Benhamza, 1996; 2005).

The chlorided sodic facies is found in sandy and clayey reservoirs of Oligocene, the chlorides are probably the result of the dissolution of sodium chlorides of the saliferous alluvium. The sulphated calcic facies is located in the sandstones and clays of the Oligocene and is explained by the dissolution of calcium sulphates contained in evaporitic inclusions and the alluvium.

The dry residue: The values of the dry residue representing the residue after water evaporation show that analyzed water samples present acceptable contents. The tolerated standard of 1500 mg.L^{-1} is not exceeded by any of the samples analyzed (OMS, 1972).

The Hydrotimetric Title (HT): According to the O.M.S standards of drinking water (Table 2), drillings whose water is average hardness to rather soft are located in the mining field of Ismail collecting the Oligocene aquifer (OMS, 1972). The analyzed points of hard water are in the field of Ras El Ma and Guenicha collecting the Paleocene Eocene aquifer. Drillings corresponding to very hard water are located in the Ismail and Guenicha fields collecting the Eocene Paleocene aquifer (Fig. 3)

Mercury contents

Geochemical Characteristics of the Northern numidian zone: Bikmeev studied the geochemical characteristics of the Northern numidian mercurial zone, using samples

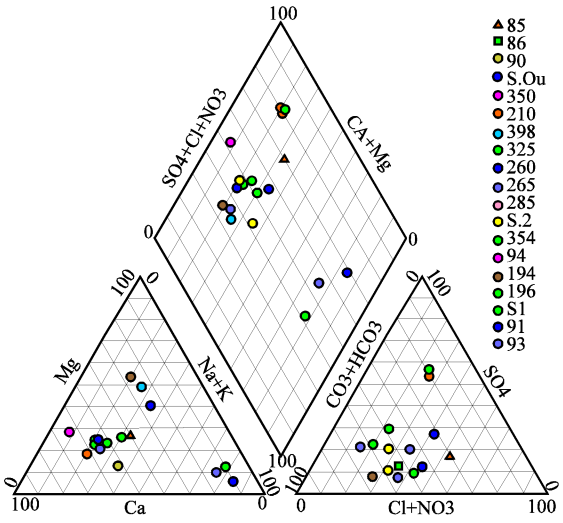

Fig. 2: Representation of the chemical analyses on the Piper diagram

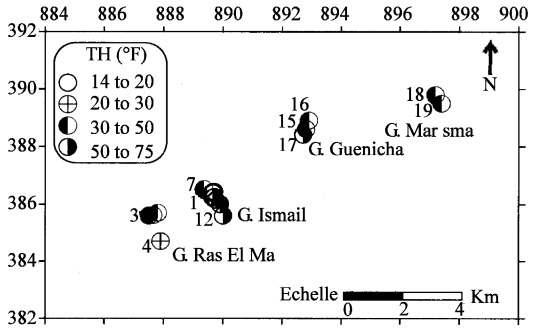

Fig. 3: Map of water hardness in the Northern Numidian zone (°F)

taken mainly from drill cores and dosed by semi quantitative emission spectrometry. The results show that litho-stratigraphic facies presenting high Hg Clarke are calcareous micro breccia of Thanétien Hyprésien (1.5 ppm), the sandstones and conglomerates of Permo Trias (1.4 ppm), clays and siltites of Luthetien Priabonien (0.9 ppm); whereas the sandstones and clays of Oligocene present low Hg Clarkes (0,6 ppm) (Bikmeev, 1972) .

Element migration: The migration coefficient of a given chemical element is defined as the ratio of its average content in water on Clarke of the rock in which this water is circulating.

$$A = (B/C) \times 10^{-4}$$

Where: A= Migration coefficient

B = The element average content in water

C = Element Clarke in the rock

Table 2: Water hardness standards O.M.S (1972)

TH (° French)	-7	7-14	14-20	20-30	30-50	50°F et plus
Hardness water	Very soft	soft fresh	average hardness	rather soft	hard	very hard
N° of springs and drillings	/	/	8-9-10	4	1-2-3-7-15-16-18-19	5-6-11-12-13-14-17

Table 3: Migration coefficient of the North numidian zone (Bikmeev, 1972)

Chemical element	No. of drillings	Migration coefficient (A)	Chemical element	No. of drillings	Migration coefficient (A)
S	30	563	Mn	30	0.15
Cl	30	973	Fe	30	0.007
Na	28	6.51	Hg	28	0.15
K	35	0.82	Zn	30	2.56
Ca	30	5.75	Pb	30	0.11
Sr	30	5.6	Cu	30	0.31
SiO2	30	0.11			

Table 4: Supergene migration of the chemical elements in the North numidian zone

	Oxidizing environment						
Intensity of migration	1000	100	10	1	0.1	0.01	0.001
Very high	Cl - S - Br						
high			Ca - Na – Mg -Sr- Zn – (As-Sb) Mo - U				
medium				Mn - Cu – Hg - Pb - Sr - K - Ni -P - (Sb-As)			
Low and Very low							Fe - Al - Ti -Noble metals

The results show that the mercury of the Northern numidian mercurial zone presents an average migration coefficient of 0.15 (Table 3) (Bikmeev, 1970; Bouarroudji, 1986) .

Classification of the elements: The calculation of the migration coefficient makes it possible to classify the elements of the area of study in four groups according to their intensity of migration (Bikmeev, 1970).

This reveals that the calcophile elements Z n and (As-Sb) are more mobile than Hg, Cu, Pb and (Sb-As) (Table 4).

Mercury content distribution: The ground water mercury distribution map of the area shows that the high values are situated in the mining field of Ismail. The maximum contents (80 µg.L^{-1}) is located in drillings 350, 210, 354 and the springs S.1 and S.2 of the Oligocene aquifer.

These values are very high according to the standard tolerated for the Hg which is 1µg.L-1 (OMS, 1980). Drillings 94 and 93 collecting the deep Paleocene-Eocene aquifer show a minimal mercury content of 1µg.L^{-1} (Fig. 4).

The contamination of Oligocene aquifer by the Hg is much higher than that of the Paleocene-Eocene, although the latter contains facies of high Hg Clarke.

Thus this contamination is not the result of leaching of the cinnabar formations, bearing in mind that Hg in the area presents a medium migration coefficient which is less mobile than the chalcophile elements (Benhamza, 1996; 2005ab; Benhamza *et al*., 2006; Bikmeev, 1970).

The distribution of mercury in groundwater in the area has thus no relationship with the high Hg Clarke

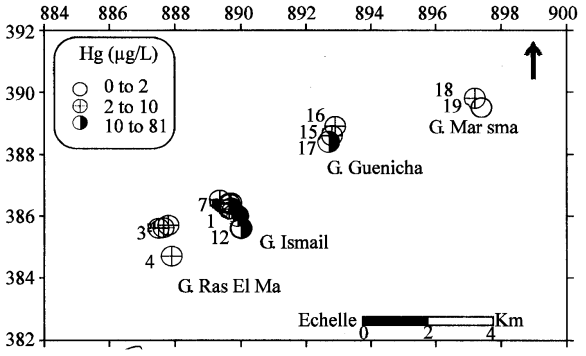

Fig. 4: Map of mercury distribution in the North Numidian zone (µg.L^{-1})

litho-stratigrahic facies. Indeed, the studies carried out in the area showed that the mercury plant contributes to both atmospheric and hydric pollutions.

A part of the mercury contents in water results from recycling of atmospheric Hg by the rainfall, the major part originating from the hydric rejection of the plant (Benhamza, 2005a; b).

The mercury plant has two tanks of decantation not adequate, where the contaminated cooling water of the process are poured. Flooding causes water to overflow the two lakes and reaches the Fendek River which feeds the Oligocene aquifer, whose roof is made of permeable sandy rocks (Benhamza, 1996; 2005a;b).

Contamination of the population by inorganic mercury: The concentration of mercury in the urines of a non exposed subject is usually of about 0.5 µg. L^{-1}, although much higher values, never exceeding 25 µg L^{-1} have been reported (Buzina *et al*., 1989).

Table 5: Inorganic urinary Hg in creatinin ($\mu g.G^{-1}$) in the plant workers, childrens of Azzaba and Annaba

	No.	min.	max.	Mean
Plant workers	88	2	13924	650
Children ofAzzaba	354	0	17	2.49
Children ofAnnaba	124	0	4	0.45

Table 6: Clinical signs among the factory workers, the school children of Annaba and Azzaba

Clinical signs	Children of Annaba (N = 124)	Children of Azzaba (N = 354)	Plant workers (N = 88)
Tremors	negative	1.29 %	50 %
Writing test	negative	0.41 %	50 %

A normal subject rejects less than 10 $\mu g.J^{-1}$ in the urines and 10 µg in excrement (Lang worth *et al*., 1992).

The issue of pollution by inorganic mercury has been the subject of several studies and publications in the world and in Algeria.

The contamination by the inorganic mercury vapors of the plant workers of Azzaba is well known (Service de toxicologie CHU Mustapha Alger, Service de Médecine du Travail CHU de Annaba -Algérie) (Nezzal *et al*., 1994; 1996).

The average inorganic urinary Hg rate in the workers of the plant mercury of Azzaba varied from 18 to 188 $\mu g.G^{-1}$ of creatinin in 1991 and from 8 to 886 $\mu g.G^{-1}$ in 1993. (Nezzal *et al*., 1994; 1996).

These results are a clear indication of the mercurial contamination of the plant workers, which at no time was observed on the population leaving around the plant.

In order to check the level of inorganic mercury contamination of the population, a study was carried out by the service médecine de travail du Centre Hospitalo-Universitaire d'Annaba.

The study includes a total of 88 plant workers, 354 school children in the area of Azzaba, around the mercury plant (1 to 7 km) and 124 school children living in the town of Annaba, situated at 80 km East of Azzaba, representing a pilot population.

As for as the choice of age is concerned, half of the population studied is in the first primary year and the second half is in sixth primary year.

The samples consist of a collection of the urines taken in the morning, in a glass tube, filled to the 2/3 to avoid breakings at the time of congelation. The tube is brought back by the children the next morning.

The samples are codified, kept in a freezer before they are carried to the laboratory of industrial toxicology of Sider El-Hadjar (Annaba-Algeria) for analysis, the time between sampling and the analysis not exceeding fifteen days.

The analysis is carried out according to the method of atomic absorption spectrophotometry (cold vapor).

In the biological samples, mercury is related to the thiols grouping, the tin chloride ($SnCl_2$) added to the samples breaks this bond and reduces mercury to the metal state. The vapors are then transmitted to a cell placed in the beam of an EDL mercury lamp. The absorbance is proportional to the concentration (Nixon *et al*., 1996).

The mercury concentration in the urines is corrected according to urinary creatinin.

The method of biological analysis is sensitive to the 1/100 $\mu g.G^{-1}$ of creatine. The results of the analyses are in Table 5.

The results of the analyses show a high average among the 88 mercury plant workers (650 $\mu g.G^{-1}$ of creatinin); giving a clear evidence of a strong impregnation, the minimum is 2$\mu g.G^{-1}$ and the maximum 13924 $\mu g.G^{-1}$ of creatinin .

The average in the 124 school children of Annaba is 0.45$\mu g.G^{-1}$, with a minimum of 0 and a maximum of 4 $\mu g.G^{-1}$, whereas the average observed among the 354 school children living in Azzaba is a 2.49 $\mu g.G^{-1}$ of creatinin, with a minimum of 0 and a maximum of 17 $\mu g.G^{-1}$.

The symptoms of this mercurial contamination (Table 6) corresponding to the clinical signs (tremors, writing test, finger nose test), which usually appear only at Hg rates higher than 50 $\mu g.G^{-1}$ of creatinin, are inexistant among the children of Annaba, are observed among those of Azzaba but are conspicious among the factory workers (Lauverys, 1992; Nezal *et al*., 1991; Nixon, 1996; DNEMT, 1996; OMS, 1972; 1980).

CONCLUSION

The underground water in the North numidian mercurial zone presents the dominating bicarbonated calcic chemical facies in relation with the hosting rocks of the Paleocene Eocene. The chlorided sodic and sulphated calcic facies result from the composition of the hosting rocks of the Oligocene.

Water of the deep aquifer of Paleocene-eocene range from hard to very hard.

The calculation of the migration coefficients of the chemical elements in the Northern numidic zone showed that mercury has an average migration coefficient of 0.15 as a result of the stability of Cinnabar.

The classification of the elements according to their migration coefficient shows that Hg, Cu, and Pb of the area are less mobile than the chalcophiles elements Zn and (As-Sb).

The limestone and the carbonated sandstone of the dorsal present basic Hg contents varying from 0.9 to 1.5 ppm, whereas the sandstone and the clays of Oligocene have a content of 0.6 ppm.

The high contents of Hg in the analyzed water, up to 80 $\mu g.L^{-1}$, are found in the wells capturing the Oligocene, whereas water of the Paleocene Eocene presents relatively low Hg contents (< 7 $\mu g.L^{-1}$).

The contamination of the phreatic water of Oligocene is thus not the result of the formations presenting a lithostratigraphic facies with high Hg Clarke.

The mercury contents found in groundwater of the area are very high and exceed the standard of 1 $\mu g.L^{-1}$, showing the contamination of the phreatic aquifer of the Oligocene.

The impregnation by the inorganic mercury of the worker workers is clear. It is weak among the school children of the area of Azzaba but is an actual fact.

The urinary mercury contents among these children are higher than those of Annaba.

The symptoms of this mercurial contamination, appearing usually at inorganic Hg rates higher than 50 $\mu g.G^{-1}$ of creatinin are not existing in the schoolboys of Annaba, are nevertheless observed among those of Azzaba and clearly marked among the plant workers.

The symptoms of mercurial contamination corresponding to the clinical signs (tremors, writing test), which usually appear only at Hg rates higher than 50 $\mu g.G^{-1}$ of creatinin, are inexistant among the children of Annaba, are observed among those of Azzaba but are conspicious among the factory workers.

The mercury norm is largely exceeded in groundwater as well as in population living in the Azzaba region.

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