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Study of spatial distribution of heavy metals in agricultural soils of El Tarf area (Northeast Algerian)

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Abstract

The present study was conducted in both the surface horizons and during two seasons (for the wet season and the dry season) in 2014 and nine (09) sites were selected across the plain of El Tarf area (North-East Algeria. The average contents of heavy metals namely cadmium, zinc, cobalt and iron in the soil were determined using a chemical digestion process and analysis by ICP-MS to evaluate the mobility and availability to plants, also ecological characterization of the same samples: Granulometry, organic matter, pH and electrical conductivity. Analytical results show that the samples analyzed soils are characterized by an abundance of certain heavy metals especially for iron and zinc and following a seasonal pattern and are distributed according to a seasonal cycle, showing an increase taking place in the wet season, meaning dry season. Other elements cobalt and cadmium present no harm to the environment. Also the results indicate that our soils are low in organic matter, have low electrical conductivity and not salty. Soil pH has an alkaline character very alkaline, while pH_{KCl} have a neutral character. After all, the results indicate a deterioration of the environment as a result of agricultural activity intense, urban and industrial discharges. The results of the statistical analysis confirm those obtained in the laboratory and reveal variability between intra-period sites and a potential variability between the two periods.

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Introduction

A soil is considered polluted when the degradation of its quality by the toxic element intake can impair human health and/or environmental (Rodriguez Martin *et al*, 2006). The presence of a contaminant in the soil cannot be a danger (Chaussod, 1996). The risk appears when this pollutant can be mobilized and is on the environment (flora and fauna) or humans **(**Lee *et al,* 2006). The tools currently used are based on physical and chemical properties of soil, while biological parameters incorporate all environmental stresses (chemical pollution, physical condition of the soil, climatic changes, biological changes ...) and learning about the overall condition of the soil.

The trace metals in soils from different sources. Natural concentration of these elements in the soil depends on the nature of the rock, its location, its age and nature of the element (Gauchers, 1968) and can become a source of danger to the farmer, soil, plants, consumers and the environment.

However, whether or not helpful ETM to living beings, excessive presence of some of them in agricultural soils may cause toxicity phenomena in plants and in animals and humans in consume. The level of soil contamination with heavy metals depends on its physico-chemical properties (texture, clay percentage, pH, cation exchange capacity (CEC), organic content, etc.).

Soil pollution linked to poor cultivation practices is a disease that results in the reduction or sterilization of an entire region. Knowledge of the contents trace metal elements allows avoided saturation complex adsorbent by these elements that are difficult exchangeable (Benselhoub *et al,* 2015).

In Algeria, no spatial mapping of contaminated soil exists yet. In the fact of several studies in the industrial towns of the northern part of Algeria, including the determination of the total amount of heavy metals, were performed several times (Fadel *et al,* 2014).

The seasonal variation (wet and dry season) and soil structures, they are among the factors which influence of heavy metals in agricultural soil? The study tried to answer this question by assessing heavy metals (Zn, Cd, Co and Fe) in agricultural soil because usually in environmental science these are heavy metals that are associated with the concepts of pollution and toxicity.

Materials and methods

Presentation of the study area

El Tarf area, located in north-eastern Algeria, these geographical coordinates are 36 ° 42 'and 36 ° 49' N, 7 ° 54 'and 7 ° 59' E (Fig. 1). This region contains important potential water content in the Mio-Pliocene-Quaternary sediments (Djabri *et al,* 2000). In this heterogeneous complex there are three layers (Gimbert *et al,* 2006): the ground water contained in sandy clays, slick gravel and sandy soils in dune of Bouteldja which is the eastern edge of the system. The waters flowing in the free surface aquifer system are subject to the influence of evaporation and local food. We can locate in the east, recharge areas in contact with the Numidian sandstones and outlet areas with old water and busy (Chapman and Kimstach, 1996).

The region is subject Mediterranean climate warm temperate said. Precipitation is more important in winter than in summer which defined a wet period in winters 122 mm on average in January and a dry period in summer with 3 mm in July. The average temperature in the region is 18.4° C throughout the year. The summer period at an average temperature of 25.7° C which August is the hottest of the year. The coldest month of the year is that of January with an average temperature of 11.9° C. The ombrothermic chart Gaussen allows recognizing in this region two periods: one dry and warm can be long more than four months, from early May to late September, depending on the year another rainy and soft to the rest of the year (Debieche, 2002).

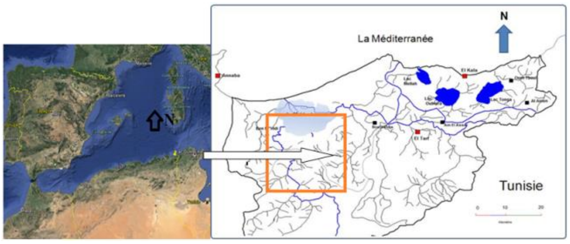

Fig. 1. Map of location of the study area.

Sampling

A sampling grid was carried out to enable the acquisition of data representative of the spatial variability of heavy metals in soils that cover the whole of the plain purpose of our study.

Two soil sampling campaigns were conducted. One at the end of the wet period (late April and early May) the other end of the dry season (late September and early October) for the dry season in 2014.

Nine (09) sites were selected across the plain, depending on their good accessibility even in winter, their locations in the valley, types of uses (agricultural land, residential or fields ...) and nature of the pollution that affects them.

A physicochemical characterization (Granulometry, organic matter, pH and electrical conductivity) was carried out on new samples from the surface horizons of agricultural land followed by a determination of heavy metals including Zinc, Iron, Cobalt and Cadmium.

Analytical Techniques

Granulometry

The granulometry analysis is used to determine the distribution of soil particle sizes. Indeed, the particle size affects soil characteristics (surface area, permeability ...) and therefore the behavior of pollutants by infiltration phenomena and retention of trace metals. It was performed according to the method of Robinson pipette (the international method (Aubert, 1978) on samples dried in air and sieved to 2 mm. The sample is previously freed of the organic matter by oxidation with hydrogen peroxide (H_2O_2).

The clay fraction was dispersed with sodium hexametaphosphate. The different size fractions are then determined by pipetting, drying and sieving.

Soil pH

The soil pH is an essential element for the existence of a mineral phase speciation and toxicity are all parameters related to the pH of the medium. The pH measurement is made frequently in an aqueous suspension, the soil mass relative to the volume of water will vary according to the methods and the texture of the medium, the most frequently encountered ratio is 1/5. The measurement is performed according to the standards (AFNOR, 1994).

Electrical conductivity (EC)

The electric conductivity is measured in a soil/water suspension of 1/5 (Mireles *et al,* 2004). The measurement is performed according to the standards (AFNOR, 1994)

Organic matter (OM)

The OM plays a very important role in soil or she is involved in soil structure (cementing particles and ventilation) and the reserve of nutrients. It comes from the activity of all organisms on the surface or inside of soil. Part of this OM is produced by living organisms: animal waste, root exudates, plant litter. The rest consists of dead plant debris, dead animals and microbial cells (Davet, 1996).

Determined by the method of Gauchers (1968) according to the following step: incineration; at 480°C, ground 5g for 4 hours. OM determining rates by the ratio of the difference between the weight of dry soil and soil weight cremated on the weight of dry soil; the latter is obtained by drying in an oven for 24 hours at 105°C.

Procedure for preparation of solid samples and extraction

The extraction method comprises introducing 0.5 ± 0.01 g of dried soil, ground and sieved in a digestion vessel (graduated polypropylene tubes) and then add 6 ml of hydrochloric acid and 2 ml of nitric acid and left in contact to allow a slow oxidation of organic matter. Cover with a watch glass stopper and reflux heat in the hot block at 95°C for 30 minutes.

If the sample is believed to have a high concentration of organic compounds, it is recommended to perform this step: Add 2 ml of 30% H_2O_2 to the welfare cool the sample. Allow exothermic reaction occurs (about 10 minutes) and place the sample in the return manifold at a temperature of 10° less than the original set point for another 30 minutes.

The reaction with H_2O_2 increases the sample temperature (H_2O_2 helps break high organic compounds in the sample, creating a more complete digestion) when completely cool. Then Bring volume of the sample to 50 ml with water UHP. Finally slowly filtered with appropriate filter paper to remove insoluble material. The filtration step is performed, with little pressure on the piston. If against excessive pressure occurs, stop the filtration and allow sediment to "settle." Apply pressure to the piston can cause sample "blow by" allowing sediment to pass through the filter in the digestat.

The calibration ranges are prepared from standard solutions of Iron, Zinc, Cobalt and Cadmium chloride-specific elements studied. The analysis was conducted using an ICP-MS (Inductively Coupled Plasma Mass Spectrometer) at the laboratory SPAR LAB of University of the West of Scotland - Paisley (UK).

It is therefore a coupling between a plasma torch and a mass spectrometer magnetic field sector. The advantages of this are ICP-MS multi-element analysis of its capabilities, its large detection power and low sample consumption (Barbante *et al,* 1998) associated with a high ability to distinguish analytes potential interference. The detection limits were 0.055 mg/l for Cd and 0.050, 0.040 and 0.248 mg/l for Co, Zn and Fe respectively (Maas *et al,* 2010).

To better interpret the results obtained in our study, a statistical study was made through the correlation matrix by using Excel software.

Results and discussion

Physic-chemical characterization of soils

The results of physicochemical analyzes of soils studied are given in Table 1.

Table 1. Results of the granulometry analysis of soils.

	Max	Min	Medium	SD	CV %
Clay %	57,69	26,15	41,13	6,692	16,269
Silt %	37,78	1,23	16,18	8,544	52,810
Sand %	69,17	18,76	42,69	11,86	27,771

The representation of results of granulometry analysis on triangular textures shows that all agricultural soils from different stations show a dominance of clay and sand fractions (41,13% and 42,69%) from the silt fraction (16,18%).

Studies have shown that heavy metals can be absorbed and immobilized by the clay minerals or also be completed by the organic matter of the soil then forming an organo-metallic complex (Lamy, 2002). More soil is rich in organic matter, the more it is able to immobilize heavy metals and prevent leaching to groundwater.

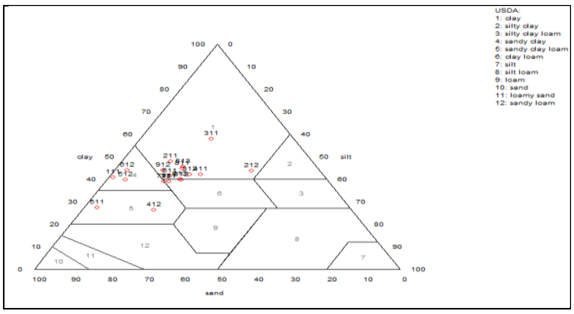

Fig. 2. Triangular classification soil samples according to their texture diagram (Campy and Meybeck, 1995).

The work exposes the surface horizons to more mineralization of organic matter, it seems more sequestered in this horizon that receives the maximum waste and crop products.

In addition, our soils are relatively rich in clay that better stabilize the organic matter providing protection that would put away the strong mineralizing activity to which expose surface horizons of cultivated soils (Albrecht, 1988).

The texture is the first property of an important ground. It gives an indication on the physical properties and the CEC. The permeability and porosity of a soil which provide good mineralization of the organic material depends on the soil texture.

The projection of results of granulometry analysis on the triangular textural (Debieche, 2002) shows that the analyzed soils are clay textures, sandy clay, sandy clay loam and clay loam with a dominance of clay textures. This is in fact essentially of fine textures. The soils studied thus have a homogeneous texture point of view. This texture promotes permeability and guarantor's soil pore water availability and nutrients needed for plants.

Table 2. Results of physicochemical analyzes of soils.

Parameters	Max	Min	Medium	SD	CV %
			Wet season		
pH	8.537	7.407	7.915	0.307	3.88
pH $_{KCl}$	7.907	6.860	7.293	0.291	3.99
EC (µS/cm²)	1288	84	201	245.081	70.65
OM %	3.251	0.090	1.108	0.911	82.21
Zn (ppm)	116.737	0.433	8.307	4.527	54.50
Fe (ppm)	6475,88	1140,510	4102,592	1293,927	31.54
			Dry season		
pH	8.097	7.010	7.628	0.396	5.19
pH $_{KCl}$	7.743	6.430	7.176	0.368	5.13
EC (µS/cm²)	300	70.333	129.68	50.83	39.20
OM %	4.647	0.019	1.983	1.397	70.64
Zn (ppm)	19.066	2.090	10.297	4.906	47.65
Fe (ppm)	6370.002	2660.811	4410.866	1071.836	24.30

Cd and Co are no detected

The electrical conductivity reflects the ability of an aqueous solution to conduct electric current. It is directly proportional to the amount of mineral salts dissolved in water.

The results of the electrical conductivity, show that, by the standards given by the Publications of the Ministry of Cooperation, almost all (98%) soils in the study area is low salinity (EC less than 500 µS/cm²) and only 2% of the soils are saline (electrical conductivity greater than 1000 µS/cm²) in the site 9 and those in the wet period. Comparing the seasonal concentrations reveals a little stronger in summer and winter, this difference is due to the dilution by rainwater.

The pH plays an important role in the biological activity of the soil and supply of mineral nitrogen to plants while a synthetic reflection of assessing the chemical soil fertility.

Soil pH affects the efficiency of the growth of a culture in a sol; it affects the availability of nutrients, the activity of microorganisms and is related to the risk of toxicity. The activity of the soil, as well as availability of most nutrients depends on pH. It corresponds to the concentration of H^+ ions available, existing in the soil solution.

During the wet season, the pH varies between 8.537 and 7.407 with a mean of 7.293 ± 0.396, while during the dry period varies between 8.097 and 7.010 with a mean of 7.628 ± 0.396.

According to the standards given by the Publications of the Ministry of Cooperation, the studied soils are classified as neutral to slightly alkaline. Such pH is due to the presence of limestone in the soil. In fact, according to Baize and Jabiol (1995) calcareous soils have pH between 7.3 and 8.5.

The temporal evolution shows no significant difference in levels of the wet season compared to the dry season.

pH $_{KCl}$ is represented by H^+ ions exchangeable set by colloids, existing in the soil solution. It is always less than the pH of water (except in some lateritic soils and sodic soils or is equal). During the wet season, it varies between 6.860 and 7.907 with a mean of 7.293 ± 0.291 and between 6.430 and 7.743 averaging 7.176 ± 0.368 and this during the dry period.

Soil pH also plays an important role in the mobility of heavy metals and this is another important factor influencing the solubility of the metal and therefore its toxicity (Moore *et al,* 2006) admit that these substances are not leached and have a reduced bioavailability due to the richness in organic matter of soil and their basic pH (between 6.9 and 8.6).

The analysis of total organic matter in the different samples, to distinguish a change in rates, from site to another, in the same period. A great variability is observed with 11 samples (dry season) and 14 samples (wet period) which exceed the average and coefficient of variation (82.21% and 70.64%) correlates well with this observation.

The rate of OM varies between 0.090% and 3.021% with an average of 1.108% ± 0.911 during the wet season. While increases during the dry season with an average of 1.983 ± 1.397% and varies between 0.019% and 4.647%.

The OM of the sites is currently at relatively low levels with almost always lower levels of 2%, a situation that may expose this soil degradation by water erosion when the slopes become strong and stimulate the effective runoff. This organic matter due to its low content in soils of the sites is not involved in the cation exchange process that remains mainly governed by the mineral fraction (Benselhoub *et al,* 2015).

The work in soil exposes the surface horizons to more mineralization of organic matter, it seems more sequestered in this horizon that receives the maximum waste and crop products.

In addition, our soils are relatively rich in clay that better stabilize the organic matter providing protection that would put away the strong mineralizing activity to which expose surface horizons of cultivated soils (Albrecht, 1988).

Tables 3 and 4 show the correlation matrix. A significant negative correlation was observed between the different elements of the granulometry: clay/sand and silt/sand respectively with correlation coefficients equal to -0.828 and -0.723.

Note that the organic matter seems no direct link between them and with the other elements. A strong positive correlation was observed between the couple Zn/Fe with a correlation coefficient equal to 0.912 and 0.877 for the wet season and the dry season.

Regarding the wet season, some parameters such as silt, organic matter and water appear pH without direct connection between them and with other variables; their degrees of correlation with other variables are not significant. Same observation in the case of dry season with the parameters: silt and organic matter.

A significant correlation was inscribed between the electrical conductivity and the clay content and sand during the wet season respectively with a correlation coefficient equal to 0.653 and -0.614 and between the couple CE/pH$_{KCl}$ with a degree equal to 0.601. While during the dry season, the pair CE/Fe correlates well with a correlation coefficient equal to 0.695.

Assessing the level of soil contamination is based on the standards of the European community and the Algerian recommendations (Bendjama *et al,* 2011).

The results of the four heavy metals obtained for samples from the surface horizons of agricultural land in both seasons of 2014, we conclude that our study area is highly polluted or contaminated by iron that exceeds accepted standards, and it varies between 6475.88 ppm and 1140.510 ppm with an average of 4102.592 ppm ±1293.927 during the wet season. While during the dry season, it varies between 6370.002 ppm and 2660.811 ppm with an average of 4410.866 ppm ± 1071,836 (Fig. 3).

Table 3. Correlation matrix of different parameters of the wet season.

	Clay	Sand	Silt	Zn	Fe	OM	pH	pH_{KCl}	EC
Clay	1								
Sand	-0,828	1							
Silt	0,211	-0,723	1						
Zn	0,000	0,012	-0,022	1					
Fe	-0,081	0,029	0,049	0,912	1				
OM	-0,125	0,055	0,059	-0,356	-0,201	1			
pH	-0,022	0,175	-0,279	-0,177	-0,301	-0,267	1		
pH_{KCl}	0,582	-0,329	-0,145	-0,006	-0,143	-0,370	0,570	1	
EC	0,653	-0,614	0,266	-0,006	-0,058	-0,300	0,070	0,601	1

Table 4. Correlation matrix of different parameters of the dry season.

	Clay	Sand	Silt	Zn	Fe	OM	pH	pH_{KCl}	EC
Clay	1								
Sand	-0,828	1							
Silt	0,211	-0,723	1						
Zn	0,387	-0,255	-0,033	1					
Fe	0,490	-0,348	0,003	0,877	1				
OM	0,006	0,215	-0,382	0,373	0,213	1			
pH	0,370	-0,299	0,065	0,639	0,613	0,126	1		
pH_{KCl}	0,513	-0,371	0,014	0,601	0,623	0,099	0,899	1	

It is noteworthy that the highest values far exceed the 1000 ppm limit values defined by (AFNOR, 1994 ; Baize, 1997) are observed during the summer season. These excessive levels may be due to possible industrial units discharges, not listed by the ABH (Hydrographic Basin Agency).

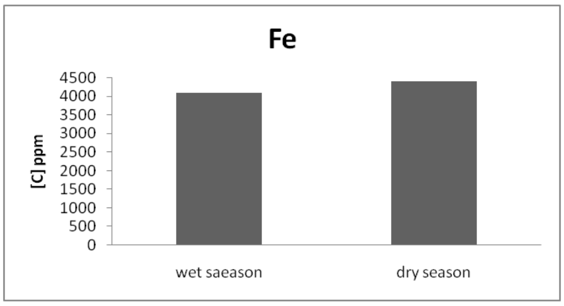

Fig. 3. Average levels of Iron in both seasons.

Regarding the soil of our study area, the mean levels of Zinc (Fig. 4), although lower eligible concentrations in soil (300 ppm: Ben Hassine *et al,* 2016) are also inferior to those which are considered normal in soils.

For other metals (cadmium and cobalt) considered for the study, the results raise concerns or they suffered a slight contamination of the surface horizon related to agriculture, allowing to state that our area is not polluted these elements and that during the two seasons and shows no contamination by these two metal elements.

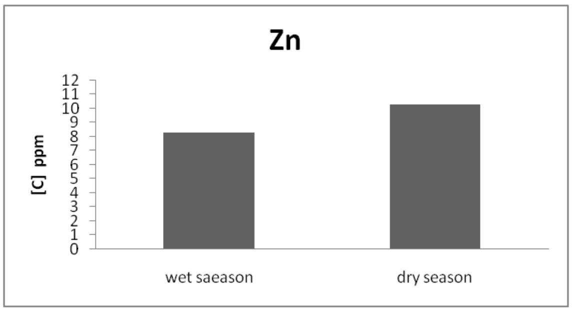

Fig. 4. Average levels of Zinc in both seasons.

In this study, we looked for the presence of heavy metals in surface horizons and for two seasons in the El Tarf area (Northeast Algeria).

Comparing the results obtained spreads the effect of particle size variations. In fact, clayey muds easily retain metals. This suspended in water that is not the case for the quartz fraction (sand), (Claisse, 1995).

For each analyzed metal element, the contents generally vary from site to another. This would be related to sedimentary facies and hydrogeology of lake ecosystems.

The analysis of heavy metals quarters measured in surface soils of the study area brings up a contamination above accepted standards for iron, while lower for zinc. It is known that contaminated soil containing a large number of non-silicate iron is adsorbed a greater absolute amount of zinc than non-contaminated soil (Baize, 1997).

Other Cd and Co elements present no harm to the environment. Moreover, the levels of these heavy metals are markedly elevated during low water in times of flood.

The very abundant element iron shows high levels compared to other metals. It is noteworthy that the highest values are observed during the summer season. These high iron values are mainly due to erosion during floods, fumes and dust from steel Mittal Steel steelworks (El Hadjar) 60 km as the crow flies from the El-Tarf area, further promoting the fixing of heavy metals (Larba and Soltani, 2014).

The high iron contents may be related to hydrodynamic and physicochemical conditions and especially the precipitation of iron oxides. In fact, during floods, firstly by the strong stirring currents promotes good oxygenation increasing precipitation of iron oxides, facilitating the formation and aggregation of organic and mineral particles, according to Claisse (1995) and secondly of obtaining the material in the colloidal state, it constitutes a support to the phenomena of adsorption, complexation and precipitation of metals (Viard-La Rocca, 2004).

This can be explained by the fact that the amount of organic matter does not plays a leading role in adsorption process, as qualitative composition of organic and mineral formations as carriers of sorption capacity (Ben Hassine *et al,* 2016).

Conclusion

The results indicate that our soils are low in organic matter, not salty, have low electrical conductivity and an alkaline character very alkaline, while pH $_{KCl}$ have a neutral character.

We note that at 9 sampling sites, the contents of four heavy metals are characterized by either a decrease, or an increase. However, the contents of the analyzed metals (Zn and Fe) follow a seasonal trend and are distributed according to a seasonal cycle, showing an increase taking place in the direction wet season, dry season. The contribution of heavy metals inputs are of varied origins: soil, fertilizers, pesticides, sewage sludge, irrigation, urban and industrial pollution ... etc. In fact, phosphate fertilizers are known for their high ETM, particularly those of Algerian origin. Only cadmium and cobalt levels are not alarming as they show no contamination.

The results of the statistical analysis confirm those obtained in the laboratory and reveal variability between intra-period stations (inter-site), and a potential variability between the two periods (inter-period variability). This variability is due course to the characteristics of each site and to human pollution the effect that season. The findings in this study could play a key role to effective assessment of soil pollution with heavy metals in the study area.

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