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Natural Pollution By Some Heavy Metals in the Tigris River, Northern Iraq

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ABSTRACT: Twenty samples of the recent sediments were collected from the Tigris River and some of its tributaries of northern Iraq and twelve samples from the Miocene and Quaternary sediments. The study is conducted to define and assess the paleoenvironmental pollution by some heavy metals in these sediments in order to elucidate the probable source rocks and the main mineral phases suggested to be a source of such pollution by using X-ray diffraction, X-ray fluorescence and scanning electron microscopes analyses. The study indicates a polluted level of some heavy metals mainly Cr, Cu, Ni, Pb and Zn. Clay and heavy minerals may form the main sources for this natural pollution. The combined affects of mechanical attrition and chemical etching during the transportation by the river water are indicated through several morphological characteristics on the surface textures of the heavy minerals. The high concentration of some of the studied trace elements could be attributed to incorporation of such elements in the lattice of the heavy and clay minerals.

Key words: Heavy metals, North Iraq, Pollution; Provenance, Tigris River

INTRODUCTION

A study of Quaternary and recent sediments in the near shore and river settings provides the basis for defining the physiochemical and ecological settings of the natural environments and the changes caused to them by earth×s processes and anthropogenic activities (Jayalakshmi *et al.,* 2003; Karbassi, *et al.,* 2008).Sediments are considered as important indicators for the rate of pollution in rivers, seas and oceans. Suspended materials in water were highly affected by the river drainages, source of pollutant and the place of sudden pollution contributions. Therefore, many of the analysis of pollution in the suspended materials do not give the precise ratios. Finer particles in sediments are not affected by these factors. For this reason, sediments are used as a main indicator for the pollution by major and trace elements and the organic materials in the rivers (Murthy, 1977). Sediments include, rock fragments, minerals in sand, silt and mud fractions and the very fine colloidal particles.

Heavy metals are either naturally or through anthropogenic sources are introduced into river water. Metals that are naturally introduced into the river come primarily from sources such as rock weathering, soil erosion, and the dissolution of water-soluble salts. Naturally occurring metals (especially the trace metals) move through aquatic environments independently of human activities usually without any detrimental effects (Garbarino *et al.*, 1995 & Opuene *et al.*, 2008). Anthropogenic pollutants discharged from industrial, domestic and agricultural wastewater into the river water system (Ho and Hui, 2001 & Priju and Narayana, 2007). Sediment served as sinks for most of the metals in aqueous phase (Klavins, *et al.,* 1995). Monitoring of the sediment with the determination of heavy metals is fundamental to the realization of toxic pollutants in the river sediment (Hlavay, *et al.,* 1998).

Many of mineralogical, geochemical and environmental studies have revealed the presence of abnormal pollution by (Pb, Zn, Cu, Ni and Cr) in water and sediments taken from Tigris River, Northern Iraq (Jawad Ali, 1984, Al-Jumaily, 1996 & Al-Mufti, 1996). High ratios of these elements have bad effects on environment and human

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health. Many of abovementioned studies refer to the wastewater seepage to the river as a source for the pollution of those elements, in addition to the effect of erosion on different rocks cropped out along the river channel. Erosion and weathering of the rocks and minerals plays an important role in providing the river sediments with a variety of elements since many phases of clay and heavy minerals are the main holder for these elements. Study of the clastic sediments and their content of heavy and clay minerals are among the numerous processes the earth scientists adopts in understanding depositional environments, transportation processes and the physiochemical activities releasing these heavy and clay minerals. By detailed study of these minerals, particularly of the recent Tigris River and some older successions the author tend to have a picture of their distribution and to asses the main mineral phases suggested as a sources for the natural pollution by some heavy metals in the river sediments.

Iraq is divided into six physiographic provinces each with its distinctive geological environment (Berry, *et al*., 1970), (Fig. 1). North Iraq lies within the Zagros Mountains and Foothills provinces and partly within the Jezira province.The Zagros Mountains to the north consist of NW-SE trending parallel ridges of folded Upper Paleozoic and Mesozoic age limestone, and a Nappe of metamorphosed Lower Paleozoic rocks along the Iranian border. The Foothills province consists of mainly Upper Miocene and Pliocene coarse detrital sediments which are gently folded along the NW-SE axis parallel to the structural trend of the Zagros Mountains. To the west is the Jezira province that consists of relatively undisturbed Miocene and Pliocene limestone and gypsum, and poorly consolidated Pleistocene detritus (Berry, *et al*., 1970).

The main sediments which appear as outcrops range in age from Jurassic to Quaternary (Fig. 2). This zone has undergone widespread igneous activity including the intrusion of dolerite, gabbros and granite, and the extrusion of andesitic and basalt (Buday, 1980) as well as explosive pyroclastics and tuff. Rocks in this zone show a low grade of metamorphism (e.g. slates, phyllites, schist and spilites).The present stage of Tigris River has its origin in the Taurus Range in Turkey. The river travels a distance of 1710 km until its junction with the Euphrates River at Shatt Al-Arab; south of Iraq (Fig. 1). Both Rivers collect their waters from nearly the same drainage area. The Tigris River crosses the southern parts of Turkey, which is a very complex igneous and metamorphic region (Nappe Zone), while most of its tributaries have risen from NE Nappe Zone (i.e. across the Mawat-Chuwarta Ophiolite Complex area). The Nappe Zone includes the area of north and northeast Iraq, forming the high and tectonically complex Zagros Mountains which trend northwest-southeast. The headwaters of a number of rivers which drain southwestward to the Tigris River occur in this area.

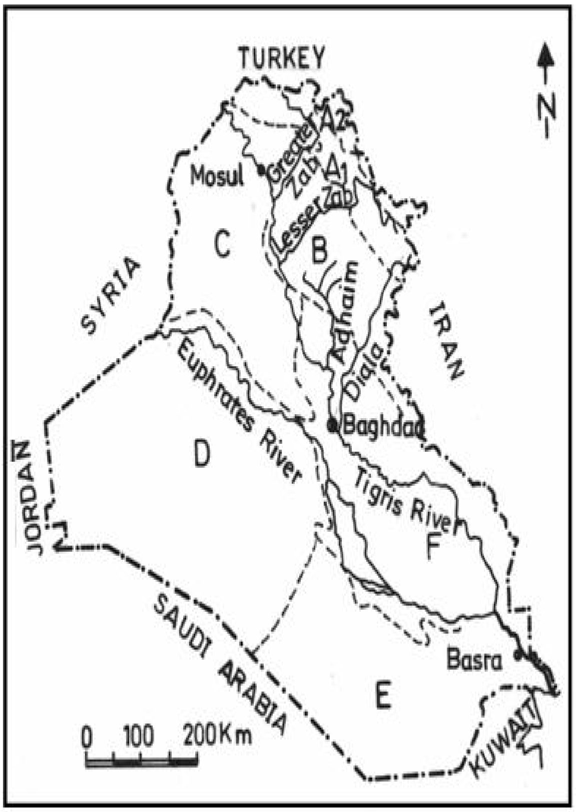

Fig. 1. The physiographic provinces of Iraq. Provinces are: Zagros mountains (A1, folded zone; A2, nappe zone), Foothills (B), Jezira (C), Northern (D) and Southern (E) deserts, and the Mesopotamian Plain (F) after Berry *et al.* (1970)

MATERIALS & METHODS

Samples were taken from the recent sediments of the Tigris River and some of its tributaries in north Iraq as well as some of the older deposits. Twenty samples were collected from the recent Tigris River sediments along the

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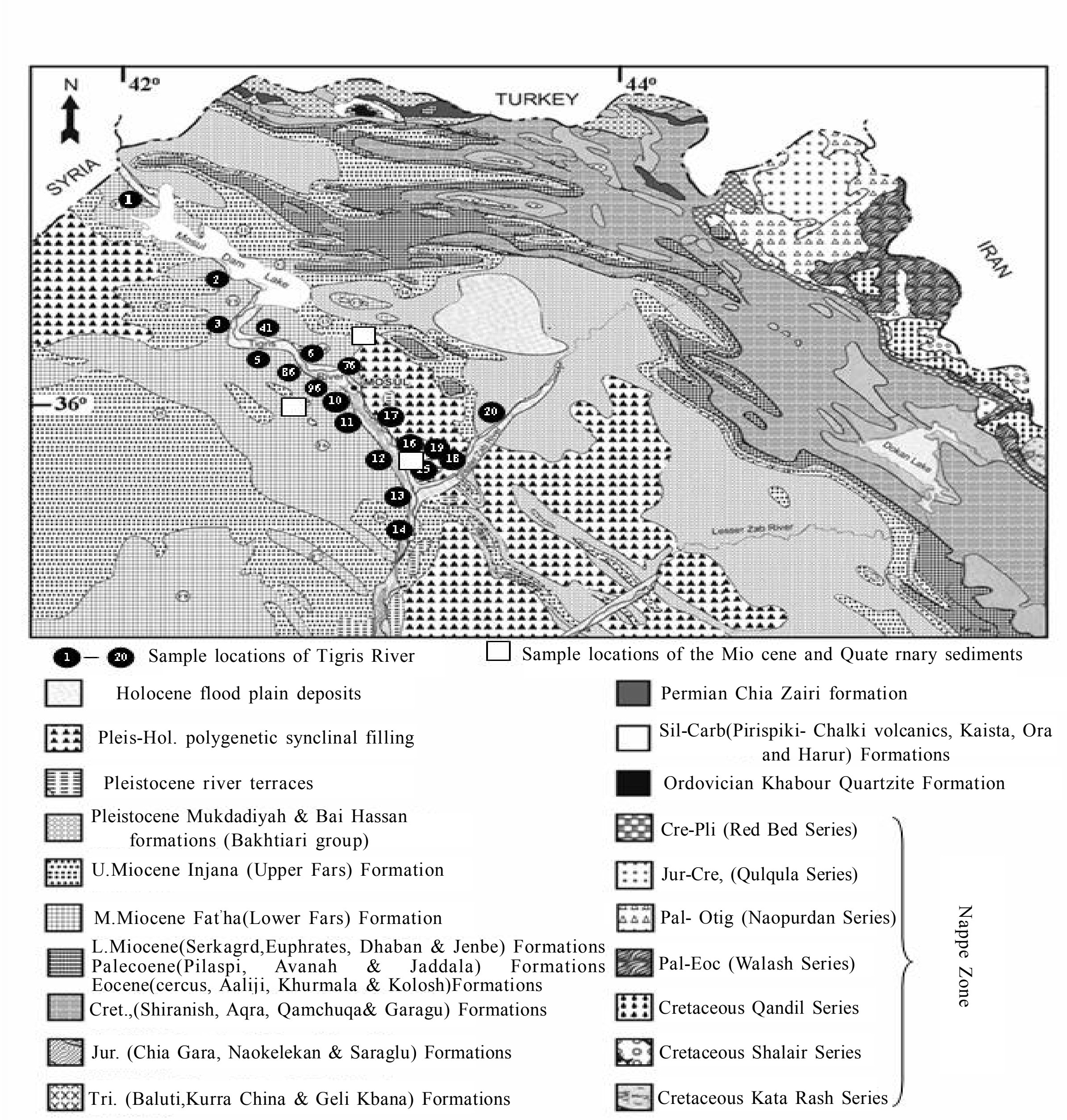

Fig. 2. Geological map of northern Iraq showing the geologic formations of the Ordovician to recent and sample locations (modified from the Geological map of Iraq, 1986)

river channel, furthermore, 12 samples were collected from the older sediments (4 samples from each of the upper clastic unit of Fatha Formation (Middle Miocene), sandstone and mudstones of Injana Formation (Late Miocene) and from the sandstone and siltstone of the Quaternary river terraces) outcropped on both sides of the Tigris River near Mosul city (Fig. 2). Fine-to medium-grained sandstones were selected for analysis of mineral assemblages. Mineral assemblages were analyzed with the petrographic microscope. Heavy minerals were separated and identified using the gravity method. Concentrations of heavy minerals were carried out by means of high density liquid, bromoform, $CHBr_3$ (specific gravity, 2.85) at 20ÚC. Heavy minerals were identified using standard petrographic procedures (Hubert, 1971 & Mange and Maurer 1992). Morphology of single grains was examined by scanning electron microscopy. A Camscan MV 2300 SEM with a calibrated energy dispersive X-ray analysis system was used.

X-ray fluorescence analysis of the major and trace element geochemistry of the samples was performed (using a Siemens automatic wavelength-dispersive XRF, type SRS 303 As). The XRF analysis was carried out at the

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Geological Institute of the Bonn University, Germany. Loss of ignition (LOI) was determined.XRD analyses were performed at the Geology Department of Mosul University, Iraq. A Philips 1730 diffract meter (copper radiation) was used to make the x-ray diffraction scans at 2°θ / min on air dried, glycolated and heated samples.

RESULTS & DISCUSSION

The recent sediments of Tigris River consist mainly of loose to fairly well-indurate deposits of sands, silts and mud. There are minor deposits of pebbles in some upstream areas and mostly of local origin, eroded from the adjacent sedimentary formations. They are fine to medium grained with moderate to poor sorted. The most significant mineralogical constituents of the Tigris sand are quartz, rock fragments predominantly sedimentary, micas, feldspars and heavy minerals (Fig. 3).

Heavy minerals are represented by opaque (magnetite, chromites and/or chromian spinels, hematite, ilmenite, goethite and pyrite) and non-opaque (epidote, garnet, amphiboles, pyroxenes, staurolite, kyanite, tourmaline, rutile, zircon and olivine). Flaky minerals (biotite, muscovite and chlorite) were exclusively observed.The study of surface texture of heavy minerals using scanning electron microscopy has revealed the combined affects of mechanical attrition and chemical etching during the transportation of mineral grains by the river water and possible effects prior to transportation in the river. The main surface textures observed throughout the present work, are the high degrees of alteration affecting unstable amphiboles, such as highly ragged cleavage planes and cockscomb structures in addition to the oriented deeply rectangular pits (Fig.4A-B). This feature is commonly observed in the recent sand samples from the Tigris River. Amphiboles are relatively unstable with respect to both chemical and mechanical abrasion. Flaky minerals of muscovite, biotite and chlorite showing variable degrees of breakdown (Fig.4C). Magnetite shows several dissolution pits, connected grooves in addition to corrosion on their borders suggesting high degree of chemical weathering in their environment of deposition (Fig.4D). Metastable heavy minerals, such as garnet, epidotes and olivine show different kinds of surface textures. Well developed facets occur on the detrital garnet studied (Fig.4E-F). These facets were commonly observed in garnets from older Miocene clastics rather than the recent sand, and it reflects the etching during digenesis and commonly in calcite cemented sandstones (Borg, 1986).

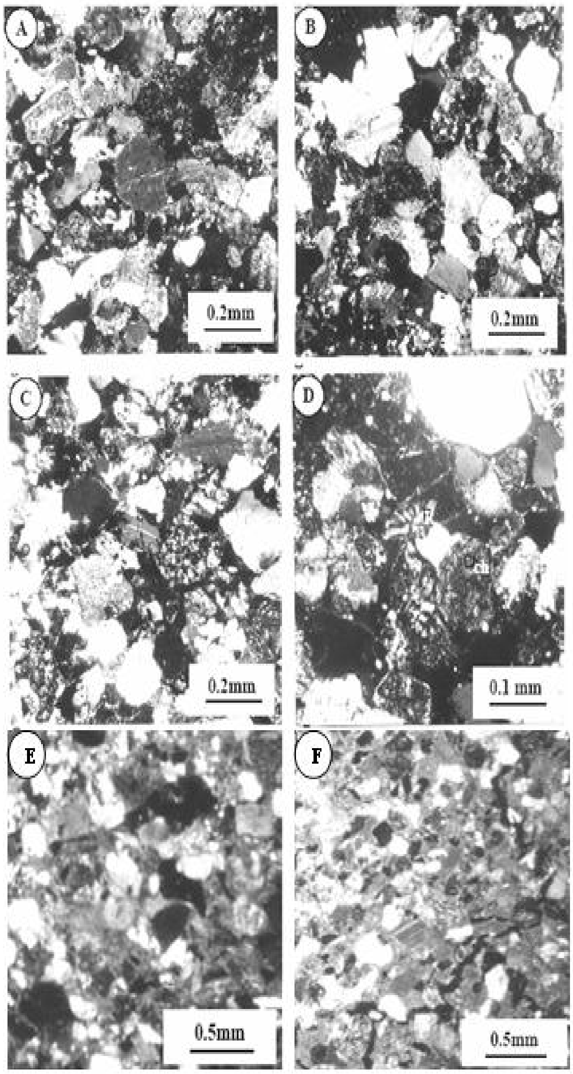

Fig. 3. Photomicrographs of selected sandstone samples of Injana Formation; (A) General view showing the common presence of carbonate lithic fragments generally rounded to sub-rounded, chert rock fragments and few poly-and monocrystalline quartz grains; (B) Both fresh and altered feldspar grains with rounded quartz; (C) Fresh feldspar, mono and polycrystalline quartz grains, chert, argillaceous and carbonate lithic fragments; (D) Chert of microcrystalline (ch) and radiolarian types (R); (E)&(F) Photomicrographs of selected sandstone samples of the Fatha Formation showing quartz, both mono and polycrystalline with chert rock fragments and little feldspar in carbonate (calcite) cement, some of calcite cement is re-crystallized

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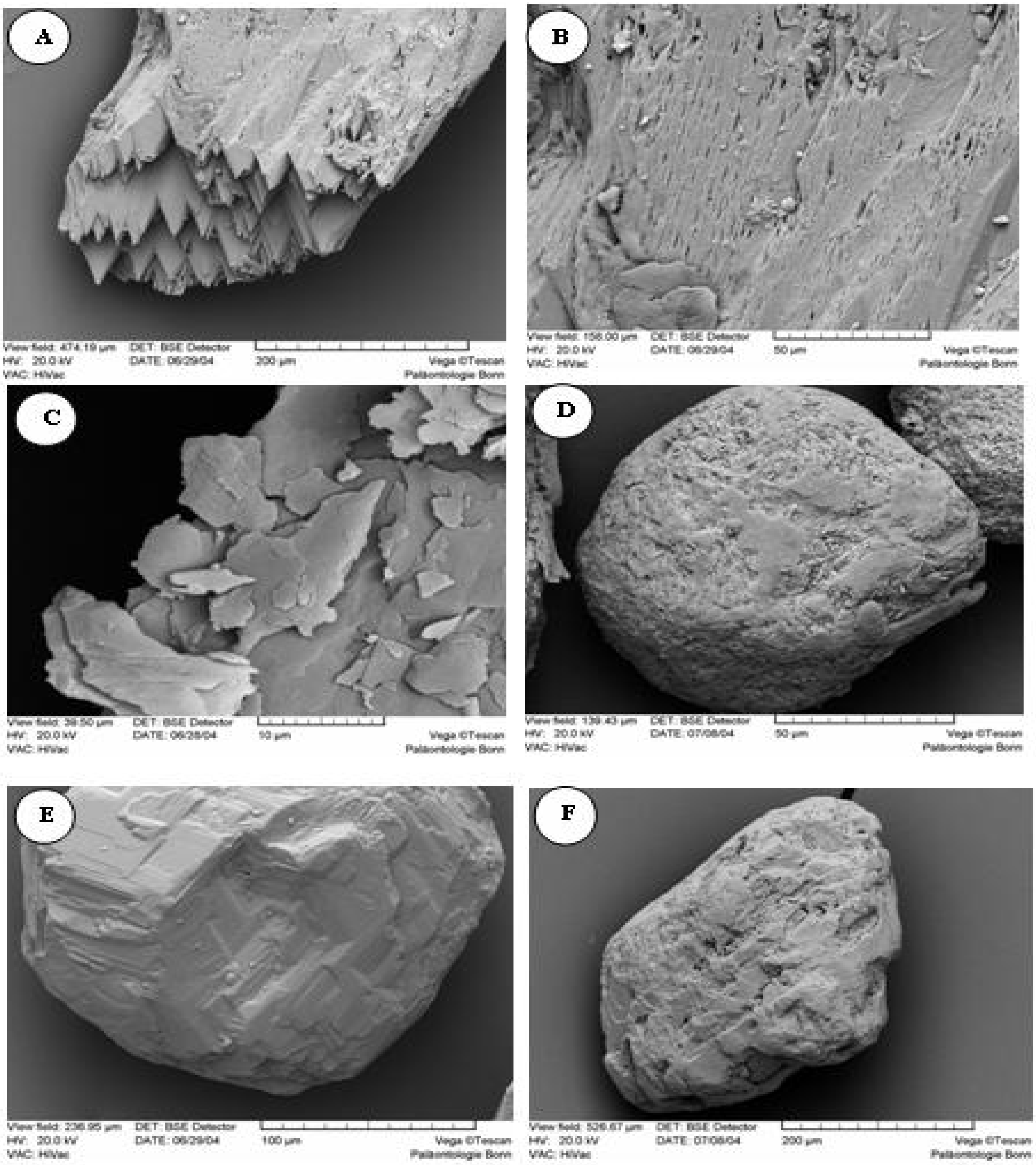

Fig. 4. Scanning electron micrographs showing surface textures of selected heavy mineral grains from recent sandy sediments of the Tigris River. A) Hornblende grain showing high degree of alteration, cockscomb shape; B) Part of hornblende grain showing large number of oriented deeply rectangular pits; C) Chlorite flakey grain showing breakdown of flakes; D) Magnetite grain showing effect of dissolution; E) Etched facets on garnet; F) Sub rounded olivine with common dissolution pits and grooves

Clay mineral analysis of the recent sediments revealed the presence of illite, chlorite, kaolinite and montmorillonite. These minerals are the same as those identified from the older Quaternary and Miocene sediments in addition to palygorskite that is recorded from the Middle Miocene Fatha Formation (Al-Juboury and McCann, 2008).Petrographic investigation of the studied clastics from the upper unit of Fatha Formation include sandstones; mainly carbonate rich and siltstones that represent bird foot, fluvial-dominated deltas (Al-Juboury *et al.*, 2001a). The studied sandstones from the Injana Formation are also carbonate-rich and have been deposited in a fluviatile environment partly affected by wave processes (Al-Juboury, 1994).

The majority of the sandstones (Table 1) have SiO_2 content ranging between 47 and 58 wt% (average 55%) for the recent sediments bulk

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Table 1. Mean percent values of the major elements analyses of the recent sediments of the Tigris River (A, bulk; B, clay-sized samples) and of the Quaternary River Terraces (C); Late Miocene Injana Formation (D, sandstone; E, mudstone); Middle Miocene Fatha Formation (F, sandstone; G, mudstone) from northern Iraq. See Fig.2 for sample locations

Sample			Major elements (%)									
	SiO_2	**TiO_2**	**Al_2O_3**	**Fe_2O_3**	**MnO**	**MgO**	**CaO**	**Na_2O**	**K_2O**	**P_2O_5**	**SO_3**	**LOI**
A	55.45	1.30	8.69	5.40	0.08	4.14	10.31	1.86	1.21	0.10	0.08	12.01
B	50.82	1.65	9.46	6.72	0.07	4.73	11.05	1.06	1.30	0.14	0.09	12.24
C	45.86	0.99	9.40	10.71	0.09	4.65	13.08	1.62	1.10	0.19	0.07	11.06
D	48.37	0.54	8.28	3.49	0.10	3.61	16.72	1.66	1.51	0.07	0.01	14.81
E	29.59	0.45	7.28	4.38	0.08	4.35	29.16	0.42	1.76	0.08	0.00	18.33
F	42.51	0.69	10.06	8.23	0.15	6.04	14.80	0.38	2.22	0.10	0.13	14.11
G	38.46	0.63	9.30	6.64	0.13	5.69	18.44	0.39	2.12	0.10	0.09	18.03

Table 2. Mean values of the trace elements analyses of the recent sediments of the Tigris River (A, bulk; B, clay-sized samples) and of the Quaternary River Terraces (C); Late Miocene Injana Formation (D, sandstone; E, mudstone); Middle Miocene Fatha Formation (F, sandstone; G, mudstone) from northern Iraq. See Fig.2. for sample locations

Sample			Trace elements (ppm)									
	Rb	**Sr**	**Ba**	**Ga**	**Cr**	**Ni**	**V**	**Cu**	**Zr**	**Pb**	**Zn**	**Co**
A	40.4	280.5	288.2	10.4	865.4	155.3	112.6	25.5	249.6	43.4	54.6	44.9
B	37.6	346.2	233.6	13.1	998.7	368.6	245.1	33.5	188.1	44.5	43.6	54.8
C	38.4	260.5	244.3	6.7	666.7	144.7	91.3	20.6	117.6	33.6	39.7	30.0
D	44.1	308.1	309.3	6.3	790.3	183.6	75.2	16.5	170.8	19.6	37.7	25.4
E	41.3	226.4	207.1	12.2	146.3	73.2	30.2	7.8	72.6	13.7	27.6	11.7
F	55.7	235.9	208.3	10.7	330.1	118.3	621.2	17.8	95.8	8.3	82.8	28.5
G	57.6	489.2	259.8	11.1	368.3	131.8	596.8	16.3	107.5	8.3	72.9	28.7

sample and 45 and 52 wt% (average 50%) for the clay sized samples. Whereas, generally it was less in the older sediments and range between 36 and 46wt% (average 45%) for the Quaternary studied sandstones; 41 and 50 wt% (average 48%); 21 and 33 wt% (average 29%); 35 and 46 wt% (average 42%); 33 and 44 wt% (average 38%) for the sandstones and mudstones from the Injana and Fatha formations respectively. Other analytical data for the major elements mean values are given in (Table 1).The distribution of the analyzed trace element for the recent and older sediments is given in (Table 2). The relations of the trace and major elements of the Tigris sediments are illustrated in (Figs.5&6).(Table 2). shows the higher concentrations of several heavy metals in the bulk and clay fractions of the recent Tigris sediments than those of older sediments. Different relations between the trace and major elements indicate the positive ratios of many of these elements with associated clay- fraction elements mainly Al and Fe as shown in (Figs.5& 6).

The metallic elements can be categorized into two groups. The heavy metals are those having densities five times greater than water, and the light metals, those having lesser densities. Well-known examples of heavy metallic elements are iron, lead, and copper. Examples of light metals are sodium, magnesium, and potassium. Humans consume metallic elements through both water and food. Some metals such as sodium, potassium, magnesium, calcium, and iron are found in living tissue and are essential to human life-biological anomalies arise when they are depleted or removed. Probably less well known is that currently no less than six other heavy metals including molybdenum, manganese, cobalt, copper, and zinc, have been linked to human growth, development, achievement, and reproduction (Friberg, *et al.*, 1979). Even these metals, however, can become toxic or aesthetically undesirable when their concentrations are too great. Several heavy metals, like cadmium, lead, and mercury, are highly toxic at relatively low

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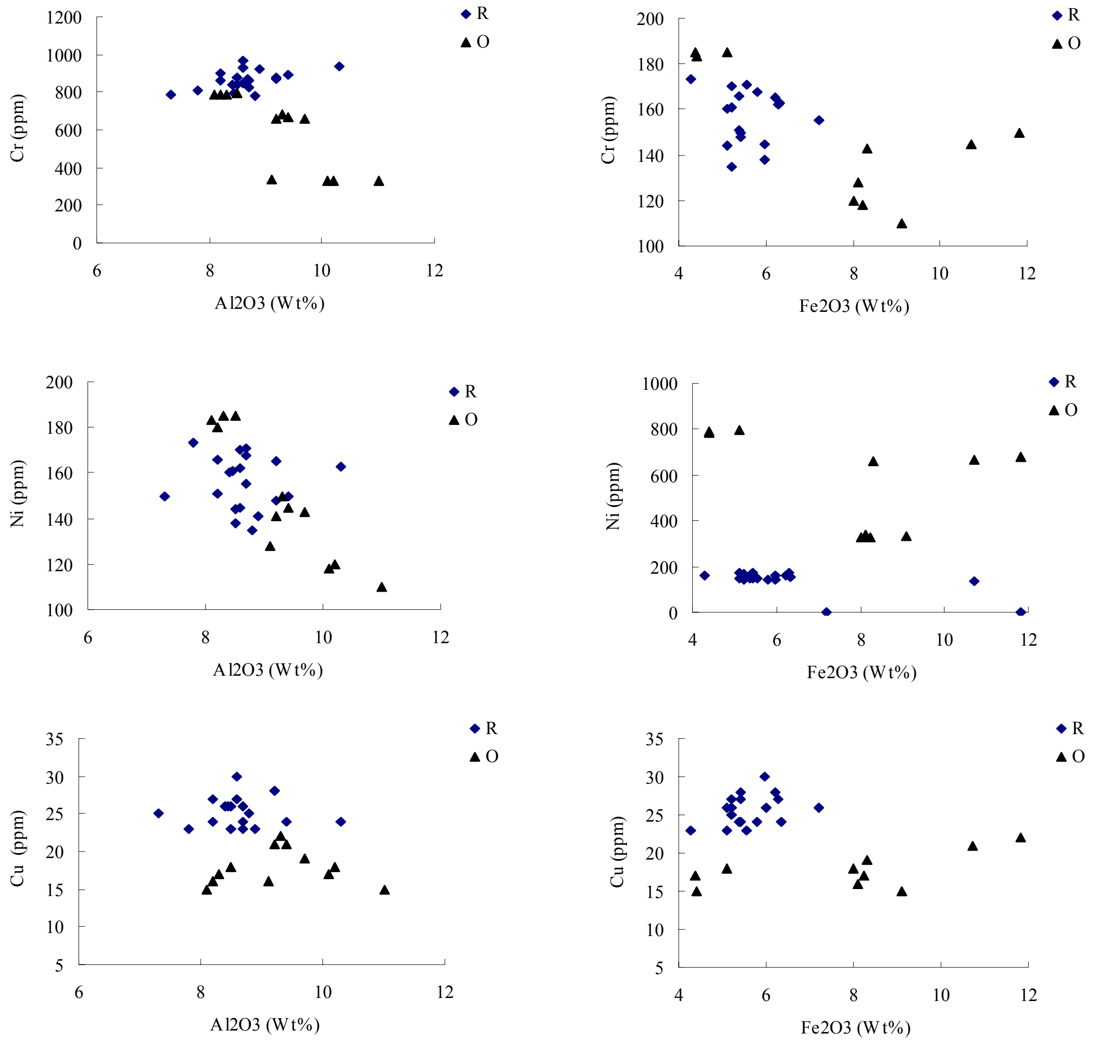

Fig. 5. The relation of Cr, Ni, and Cu trace elements against Al$_2$O$_3$ and Fe$_2$O$_3$ of the recent Tigris and older sediments R= Recent samples; O = Older sediments of Quaternary and Miocene sediments

concentrations, can accumulate in body tissues over long periods of time, and are nonessential for human health.

The analytical data of the elements (Cr, Ni, Zn, Cu and Pb) in the river sediments and their concentrations in the suspended materials show their polluted levels (Al-Mufti, 1996). The higher content of Cr may relate to the common enrichment of chromian spinels in the studied recent and older sediments, it also shows higher concentrations in the ultra basic minerals (pyroxenes and olivine), (Goldschmidt, 1964). Ni is usually incorporated within pyroxenes (Rankama and Sahama, 1950) and in chlorite which is an important weathering product of ultra basic rocks (Edel'shteyn and Suzuk, 1974). The relative higher concentrations of other trace elements suggest the effect of difference in stability of various minerals subjected to weathering and hence to differences in mobility of the chemical elements during the weathering processes. Zircon is higher in the recent river than in older sediments and is believed to have been transported chiefly as detrital zircon grains. Many trace elements may incorporate in the structure of clay minerals (e.g. Rb, Sr, Ga) and they show some increasing in the clay sized fraction of the studied sediments. Vanadium may be incorporated during weathering into developing clay minerals, due to its low solubility and relative immobility (Wedepohl, 1969). Cu like Co and V, probably entered the basin of deposition largely structurally combined in the lattice of clay minerals. Zn probably incorporated within ferromagnesian

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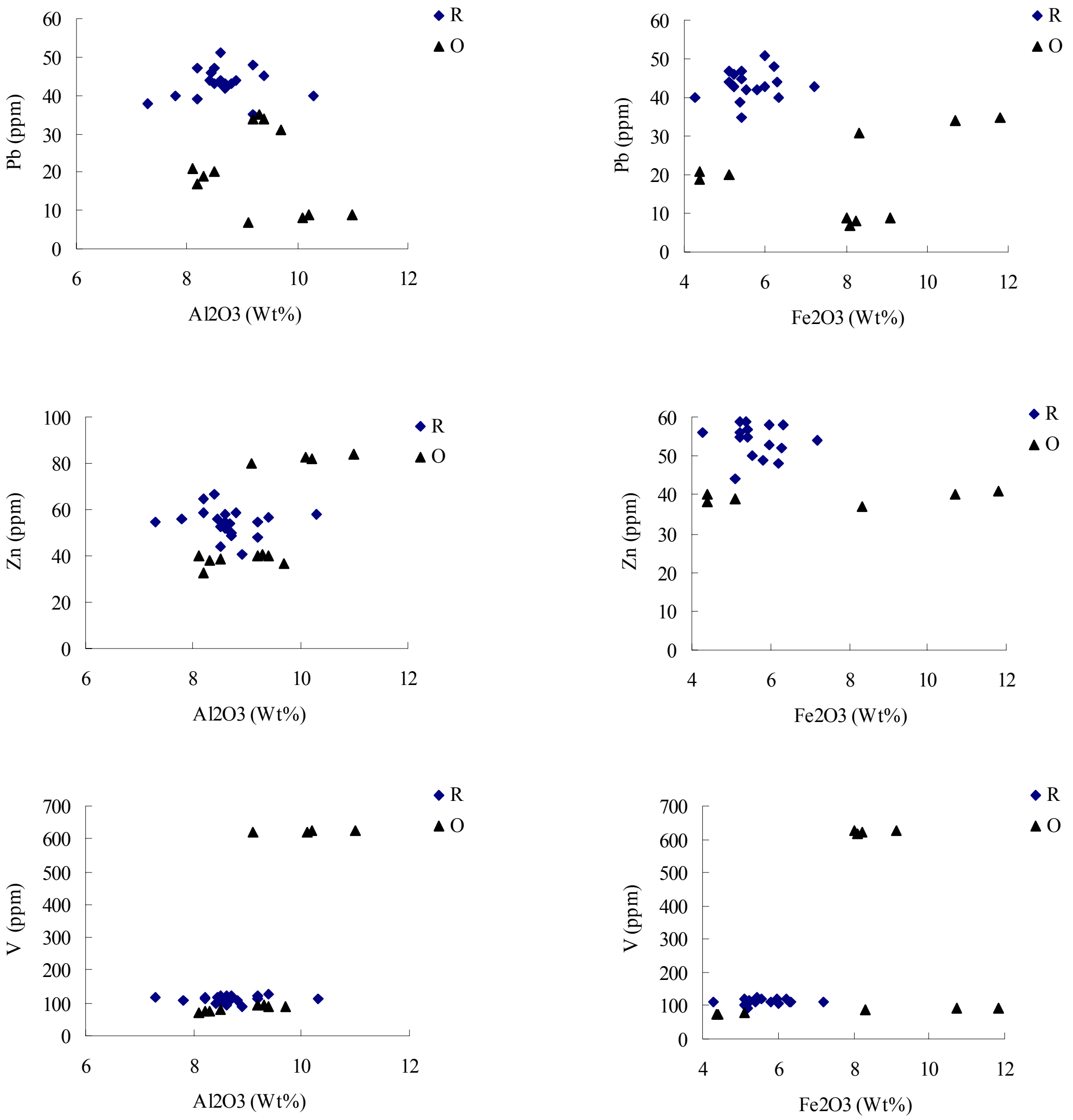

Fig. 6. The relation of Pb, Zn and V trace elements against Al_2O_3 and Fe_2O_3 of the recent Tigris and older sediments R= Recent samples; O = Older sediments of Quaternary and Miocene sediments

heavy minerals (amphiboles, pyroxenes, and biotite) as well as combination in the lattice structure of montmorillonite (Goldschmidt, 1964). All these mineral phases were abundant in the mineralogical constituents of the recent Tigris as well as older sediments. Lead (Pb) adsorbed generally within iron oxide minerals in addition to its association with Rb in the lattice structure of feldspars and mica (Goldschmidt, 1964). Barium (Ba) also associates with Rb within feldspars and biotite (Mason, 1966).

Miocene rocks and Pleistocene river terraces form the main outcrops around the Tigris River channel in northern Iraq (Fig. 2). The climate was much wetter than it is today during early Pleistocene and the Tigris River catchments area was subjected to extensive erosion producing numerous amount of sediment supplied to the river channel. The channel was choked by poorly sorted sediments which accumulated within the channel in the form of longitudinal coarse-grained bars and islands (Al-Jubouary *et al.*, 2001b).

The metamorphic provenance of the Tigris sediments is evidenced by the presence of abundant epidotes and amphiboles in addition to garnet, kyanite and staurolite. The presence of pyroxenes and olivine suggests basic igneous source rocks. This is inconsistent with the geology

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of the source area of the Tigris River sediments and its tributaries in the highlands of Turkey, Iran and the nappe zone of Iraq, which consists mainly of metamorphic and basic igneous rocks. However, some of the Tigris tributaries drain the folded zone in the northeastern parts of Iraq which consists of Eocene, Oligocene and Miocene sediments.

Opaque minerals generally forms more than one third of the heavy fraction. Most of opaque minerals of Tigris River sediments show well-developed crystals as they have been originated from the relatively nearby solid rocks in the highlands of southern Turkey. The latter are composed of a very complex basic igneous and metamorphic region (Nappe Zone) which may form the source of the opaque minerals in the studied sediments.The recent sediments of the Tigris River contain more micaceous minerals than other older sediments (Jawad Ali and Khoshaba, 1980 & Al-Juboury, 2002), and a considerable number of trace elements (e.g. Cr, Ni, V, Pb) and Fe would be incorporated within the lattice of the micaceous constituents. Generally the flaky minerals were investigated with variable degrees of breakdown (Fig. 4C&D). Such flaky and micaceous minerals (muscovite, biotite and chlorite) are unlikely to suffer complete breakdown during weathering.

CONCLUSION

The present work suggests that the clay and heavy minerals may form an important source for the natural pollution by the heavy metals in the recent sediments of the Tigris River. Although most of the pollution came from the wastewater contributed to the river. These minerals were weathered from the older clastic successions from northern Iraq which in turns come from the basic and ultra basic complexes from southern Turkey and northeastern Iraq.

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