DANUBE RIVER: SEDIMENTOLOGICAL, MINERALOGICAL AND GEOCHEMICAL CHARACTERISTICS OF THE BOTTOM SEDIMENTS

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Abstract. For a better understanding of the recent pollution in the lower Romanian sector of the Danube and its relations with human activities, yearly investigations have been carried out on 50 cross sections, between km 1072 - Bazias and the Danube mouths, using R/V *Istros*. The field activity was performed on three main sections, each with different geological, morphological and hydrological features: Iron Gates I and II dam lakes, km 845 – Mile 44, and the Danube Delta area. Physical and chemical investigations were performed on bottom sediment and surface water samples. Biological samples were collected and investigated as well. The 2005 results allowed an assessment of the Danubian aquatic environment, the comparison of present and previously collected data and the identification of significantly sensitive areas. Mitigation strategies according to the EU requirements are proposed.

Key words: Iron Gates I and II, bottom sediments, geochemical standards, distributaries

INTRODUCTION

The Danube, the second longest river in Europe, plays a great role in the economic growth of the nine countries located in its basin. These countries use the river for freight transport, hydroelectricity production, water supplies, irrigation, fishing and so on. Industrial supplies, as well as agricultural products, are brought from the Carpathians and Balkans to the inner lands or downstream Germany to the Black Sea. The irrigated areas are in the lower half of the river, especially in Hungary, Bulgaria and Romania, countries with an extensively developed agriculture.

About 80 million people live in the Danube basin. The economic conditions vary greatly, the economic well being decreasing eastwards from the highly developed countries (*e.g.* Germany) to Eastern European countries with modest economies, like Romania and Bulgaria. Most of the countries of the region are in transition after recent political changes and suffer from significant economic constraints.

Yearly investigations of GEOECOMAR along the Romanian sector of the Danube River are focused on the assessment of water and sediment discharges and their quality, changes of quality under the impact of natural and anthropogenic activities and the influence of the river in the north-western part of the Black Sea basin.

The natural flow regime of the river was disturbed by anthropogenic intrusions (protection and hydroelectric dams, water supply work, navigation). The damming has strongly affected the bedload flux, favouring the scour of the riverbed, downstream the dams. After Iron Gate I and II were built, the sediment load dropped by 50 – 70% (Panin, 1996), the dam lakes acting as traps for sediments and pollutants.

The pollutant contents (nutrients, detergents, pesticides, heavy metals), located within the water column and in the bottom sediments, vary along the river. The highest values are found in the areas where the anthropogenic intrusions are strong and along the river sector dominated by fine to very fine bottom sediments (Oaie *et al.,* 1994 – 1997, Oaie *et al.,* 2005, Radan *et al.,* 1995, 1996 a, b, 2000, 2004).

Sometimes, a local influence is present at the confluences of the Danube river with its tributaries. Unfortunately, only Romanian tributaries can be sampled and studied. The influence of the Yugoslavian and the Bulgarian tributaries may be detected on the profiles located downstream their confluences with the Danube, only.

The results obtained by GEOECOMAR in 2005 are somewhat different from the previous ones, due to the water level which was very high for a long time. More intense hydrodynamic processes modified the water transparency, the suspended sediments concentration and the sediment and pollutant depositional processes.

The field and laboratory investigations focused mainly on the quality of the bottom sediments and of the surface waters (biological, hydromorphological and physico-chemical), collected from the Danube and its Romanian tributaries. The water quality characteristics were determined according to the EU Water Framework Directive. Thus, the results lead to the 2005 ecological risk assessment of the Danube in the Romanian sector.

MATERIALS AND METHODS

Superficial sediments samples for laboratory analyses have been collected with a Van Veen grab on 50 transects, between km 1072 - Bazias and the Danube mouths, on board *R/V Istros*. Each transect consisted in three sampling stations, two of them close to the banks and one in the middle of the riverbed. Superficial water samples were also collected in each station.

Once collected, the water samples were analyzed on board *R/V Istros* for a series of physico-chemical parametres including dissolved oxygen (DO), oxygen saturation, pH, Eh, conductivity and nutrients, using WTW Multiline and Hach DREL 2000 field equipments. Water samples for laboratory determinations of pesticides, TOC, cyanides and ammonium were also collected and preserved. The analyses were done according to standard methods.

Subsamples for subsequent chemical, sedimentological, mineralogical and biological analyses in the central laboratory were collected from the primary grab samples. The subsamples were preserved, where necessary. All were packed and stored under suitable conditions, according to the different kinds of analysis.

Chemical analyses of the sediment samples were performed using complex volumetric and spectrometric – AAS and XRF analytical methods (Secrieru and Secrieru, 2002).

Grain-size analysis of sediments was done by combining the dry sieving method, used for particles bigger than 4 phi (0.063 mm), with the pipetting method for particles in the size range 4 to10 phi (0.063 - 0.001 mm). Wentworth's grade scale transformed by Krumbein (1934) (phi units, with phi[T] = -log[2]d[mm]) was used to characterize the sediments grain-size. The sediment samples were classified according to the three main grain-size fractions: sand, silt, clay employing the Shepard's and/or Folk triangle diagrams.

Mineralogical analyses (heavy minerals) were done on the grain size fraction >0.125 mm using microscopic methods, after the separation of the heavy fraction with bromoform.

The benthos samples were examined according to the common analytical and data analysis methodologies (Gomoiu, 1999).

RESULTS AND DISCUSSION

Iron Gates I and II lakes. The Iron Gates I and II lakes act as traps for sediments and contaminants and are strongly influenced by variable hydrological conditions. The sedimentary environments are both fluvial, along the section km 1072 – km 1015 and km 942 – km 880, and lacustrine *stricto sensu* downstream, close to the dams.

Considering the systematic knowledge of the specific environments and depositional processes, sediment samples were collected for grain size and mineralogical analyses. Biological and control samples were collected as well. Hydrological measurements (current speed, liquid flow) and macroscopic description of the sediments were done on-board the research vessel.

The preliminary macroscopic analysis of sediments identified the following specific aspects :

- from kilometre 1072 to kilometre 1015, the bottom sediments are coarse (boulders, gravel, coarse sands), with fine sediment depositions close to the banks;
- from kilometre 1015 up to the Iron Gate I dam (km 943), the sediments are mainly silty, with a high content of clayey fraction.

The sediment grain size gives a clue to the pollution level (*e.g.* heavy metals concentrate more in finer sediments. The analyses previously performed on the sediment samples demonstrated that in the area dominated by fine sediments (km 1015 – km 943 and km 870 – km 864), the pollutant concentrations were usually higher than those in the zones characterized by coarse sediments. Thus, not knowing the sediments grain size may sometimes lead to the false conclusion that the sediments are polluted.

The Iron Gates I Dam Lake has specific characteristics due to its natural geochemical background. Here, the highest contents of the heavy metals can be explained by the geological formations with mineralizations, some of which build up deposits of economic importance.

However, severe anthropogenic intrusions are also felt. The most important area is Moldova Nouă (Fig. 1), with many ore processing and concentration mills. It is worth mentioning that many barren gangue dumps placed on the river bank, strongly inflict upon the aquatic environment (Oaie *et al*., 1996 b, Radan *et al.,* 1995, 1996 a, b, 2000). As a result, at km 1040, 1044 and 1049 Cu, especially, records extremely high levels (up to 1600 μg/g), exceeding 6 to 7 times the probable effects level for fresh water sediments (PEL = 197 μg/g – Buchman, 1999). In the same area, PEL is exceeded by zinc (PEL = 315 μg/g, actual concentrations up to 800 μg/g) and occasionally (at km 1049) by lead (PEL = 91 μg/g, actual concentration = 129 μg/g).

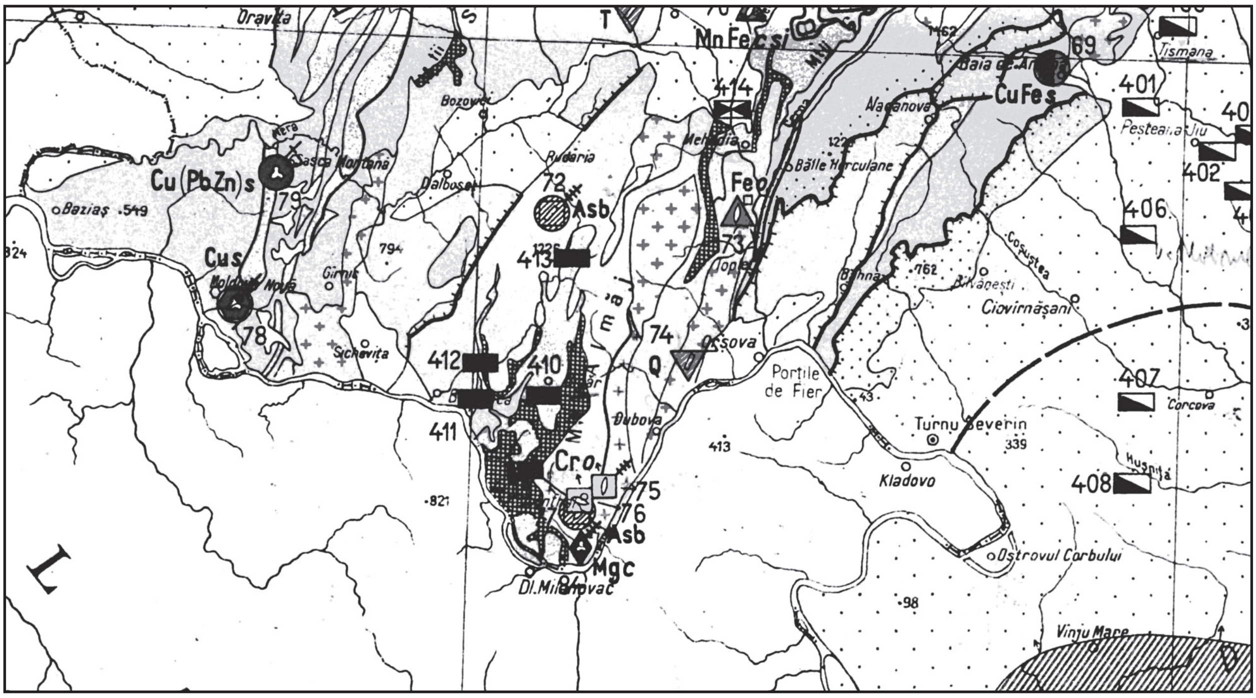

Fig. 1 The Moldova Noua area on the Romanian Upper Danube (from Map of Mineral Resources of Romania, IGG 1983).

Natural as well as anthropogenic intrusions occur along the entire sector between km 947 and km 1049, where most analyzed metals, notably Cu, Zn, Pb, Ni, Cr and Cd, exceed usually the threshold effects level (TEL – Buchman, 1999).

The nutrient analyses indicated that the NO_3, NO_2, SO_4 and detergent concentrations are within the limits stipulated by the Romanian and European quality standards and that the water quality is very good. As for the PO_4 concentrations, they exceed 0.20 mg/l on the Bazias transect (km 1072), classifying the water quality as «good», «suspected of pollution». The situation stays the same to the immediate vicinity of the Iron Gates I dam, where the water quality decreases to «average», «definite pollution», as indicated by PO_4 concentrations exceeding 0.60 mg/l. Significant phosphate loads are discharged by the river Cerna; here the PO_4 concentration is almost 1 mg/l, very close to the limit between «average» and «poor» quality, between «definite pollution» and «important pollution».

The laboratory analyses for cyanide, pesticides, TOC and ammonium indicate normal environmental conditions for the Iron Gates I dam lake.

Iron Gates II. Before the damming of the Danube, the bottom sediments from km 870 – km 863 (Ostrovu Mare dam) were mainly coarse grained. After the damming, the older sediments were covered by thin silt and silty mud deposits.

At km 866, 3 km upstream of the Iron Gates II dam, the central part of the river bed is covered with silts while near the banks the bottom of the lake is covered by medium sands with gravel elements on which *Dreissena* colonies are developing.

The physico-chemical analyses of water samples show normal values for the majority of parameters (O_2, conductivity, TDS, pH, NO_2, NO_3). The only parametre worth mentioning is the high quantity of suspended solids transported by the river (120 mg/l), explained by high waters occuring on the Danube during the field works.

The river Topolnita poses a special problem. Here the measured concentration of PO_4 exceeds 0.20 mg/l, the maximum allowable concentration for "very good" water quality. The detergent and sulphate concentrations are also increased (0.021 mg/l and 107.5 mg/l, respectively), possibly due to a waste dump on the Topolnita bank. However, the heavy metal concentrations in the bottom sediments are within the natural variation limits for silts; slightly higher Zr and TiO_2 contents indicate a medium concentration of heavy minerals. The damming diminished the river sediment load downstream Iron Gates II.

Iron Gates II dam - Mile 44 sector, the unique or braided watercourse of the river is characterized only by fluvial conditions, with important depositional and erosional areas.

According to the hydrological measurements the Danube discharge for the time interval April 3 – April 19, 2005 varied from 13 670 m^3/s at km 931 Turnu Severin to 13 564 m^3/s on the transect from km 492 Giurgiu and to 14 029 m^3/s at the entrance in the Danube Delta territory. The increase of liquid discharge greatly over the multiannual averages generated a reduction of the water transparency, as a result of the increase of the suspended solids quantity.

From km 864 (Iron Gates II dam) downstream, the bottom sediments of Danube are dominated by coarse to medium sands up to medium gravel with boulder elements and mollusc shells accumulations. Towards the banks the sediments are finer, of fine sand and silt types.

From km 375 to km 167 the river bed is covered by dominantly fine sediments, with rare occurrences of coarser ones, only at km 167 appearing coarse gravels. Between km 167 and Mile 44 (the entrance in the Danube Delta territory) are present fine, well sorted sands. From place to place these are replaced by boulders (*e.g.* Mile 54), medium gravel (Mile 44) or compact clay in the main waterway, respectively towards the banks. The appearance of such deposits is favoured by higher speeds of the water current.

The chemical analyses of water samples for NO_3, NO_2, SO_4 and PO_4 done on-board the research vessel indicated a very good quality of the water. Slightly higher concentrations, without exceeding the upper limit for very good quality, were recorded for nitrites in the Ialomita river, at Mile 54 – Isaccea and at Hm 72 Sulina, for sulphates in the Prut river while the detergents presented a somewhat higher concentration only on the transect from Mile 78.

If usually the phosphate concentrations indicate a very good quality of the water (PO_4<0.2 mg/l) on the km 247 – Mile 34 they indicate only a good quality and «suspected pollution» (PO_4 concentrations between 0.21 and 0.42 mg/l). the same situation occurs on the Arges and Ialomita rivers. On the Chilia branch the PO_4 concentrations correspond to medium water quality, with certain pollution. An exceptionally high concentration (1.44 mg/l) was measured in the Prut river, indicating «poor quality» and «important pollution» of the river. As a consequence the Prut river might negatively influence the quality of Danube waters, its discharge area being marked by significantly higher PO_4 concentration.

Some of the Danube surface water samples were analysed for cyanide, organichloride pesticides, TOC and ammonium. Cyanide concentration exceeding slightly the limit of 0.01 mg/l CN for class I waters (drinking water supplies, food industry) was identified at km 553 and in the rivers Arges, Siret and Prut, these waters being usable only for fish aquaculture and recreational activities.

The measured concentrations of ammonium and pesticides were under the limits set by the standards for class I waters. The standard does not specify TOC limits. For the Danube waters the concentrations vary from 3.84 mg/l to 6.19 mg/l. The Arges river records the highest TOC concentration – 6.95 mg/l.

Particularly notable, where the heavy metal concentration in sediments is concerned are the extremely high concentrations of Cu, Pb and Ba (the highest concentrations recorded for the entire lower Danube – 2 704 µg/g Cu, 215 µg/g Pb and 1 522 µg/g Ba), associated with high concentrations of Zn (322 µg/g), Cd (2.38 µg/g) and also Sr (466 µg/g) if compared with the very low concentration of calcium carbonate (<10%) at Km 845, close to the Timok mouth. The anomaly is persistent, being detected in previous years as well.

The metal association is characteristic to polysulfides mineralization, with barite and celestite as secondary minerals. There is no doubt that these highly abnormal concentrations are the results of mining activities in the hydrographic basin of the river Timok (Fig. 2). Here, the Bor mining and metalurgical complex (RTB Bor) consists of several mines and smelting facilities, copper processing and precious metals refining from copper ore companies, sulphuric acid plant etc. (Jovic *et al.,* 2002). These facilities produce yearly up to 395 000 t of sulphide concentrate and 24 000 000 t of flotation waste material. Due to technological failures in the closed water system wastewater from mining and processing, containing copper and other heavy metals and having a low pH, are directly discharged into the Timok tributaries Borska and Kriveljska. Through Timok river a significant quantity of these highly toxic contaminants reaches Danube, polluting the right bank of the river for some distance downstream from the Timok river mouth.

For the rest of the sector, higher concentrations of Cd, Cu, Pb and Zn, sometimes exceeding the PELs but far from the levels recorded in the upstream sectors, are occasionally met, usually at the mouths of tributaries – Vedea, Ialomita, Arges but also at km 375 and km 4.5 Macin. Although as a rule they are associated with fine-grained sediments, they still indicate some degree of local pollution related to tributaries input or other local factors (at km 4.5 Macin the concentrations may be affected by influences of complex mineralizations in the Dobruja Mountains).

In the Danube Delta area, which begins at Mile 44, the fluvial conditions are different, because the discharge is split on three meandering distributaries (Chilia, Sulina and St. George). The measurements from April 2005 indicated a liquid discharge through the three distributaries into the sea of 10740 m^3/s, an increase of almost 50% of the multiannual average discharge. The corresponding solid discharge reaches a value of 824.7 kg/s, exceeding greatly the multiannual means.

A special mention about the hydrotechnical work for cutting-off a part of natural meanders (10 cut-off sections on Sulina Canal and 6 on St. George Distributary) that altered the hydrological conditions, the grain size of the bottom sediments and their distribution. In the Danube Delta, along the Chilia, Sulina and St. George distributaries the situation is as follows:

- on the Chilia branch the dominant sediments are fine (fine sands to silts, occasionaly compacted, plastic clays – *e. g.* km 43);
- along the Sulina canal the fine sands and silts are dominant. Where the speed of the water current is great these are eroded, the bottom of the river bed being made up of compacted clays (*e. g.* Mile 33+400);

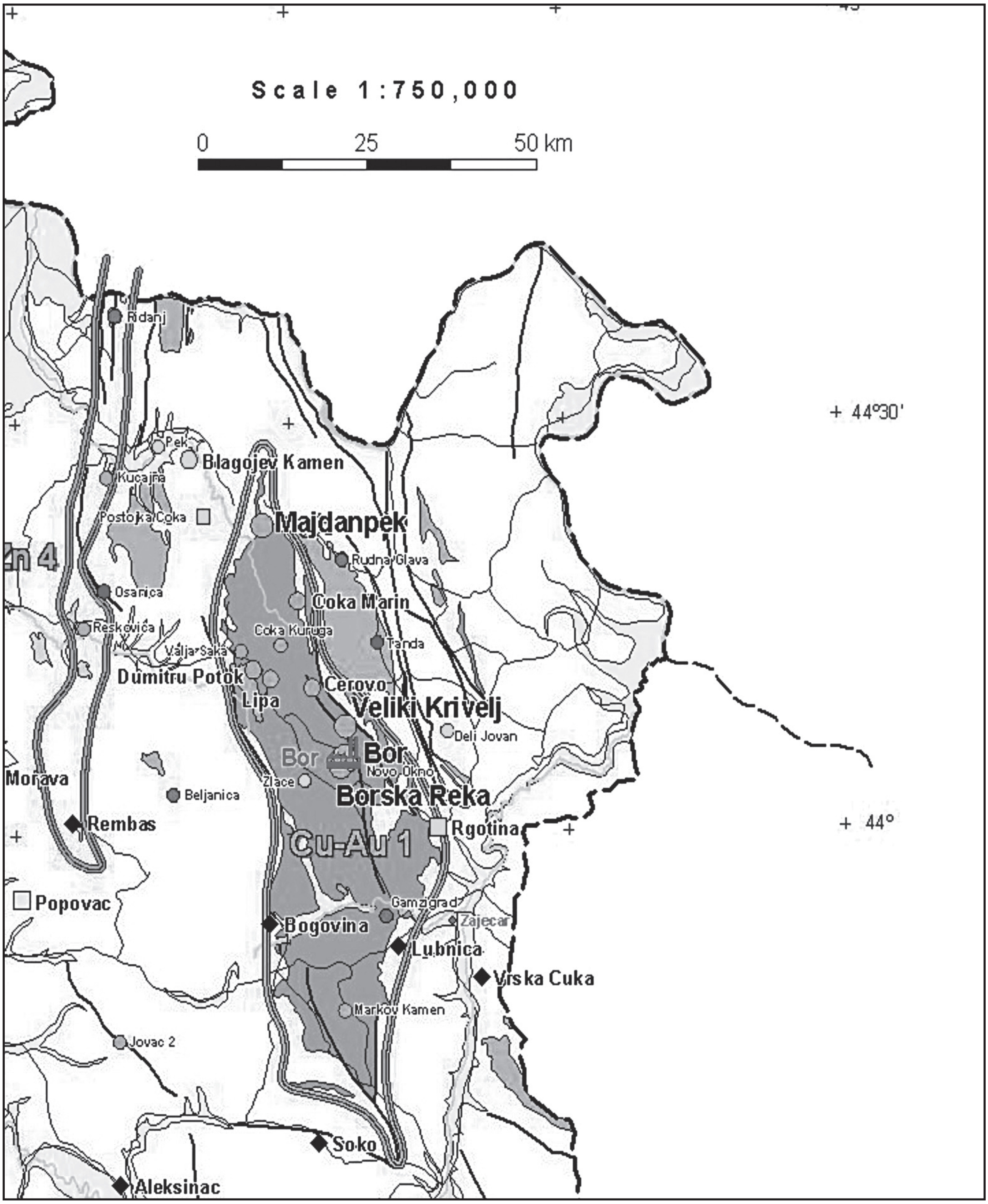

Fig. 2 The Timok Mining District (map extracted from Mineral Deposits and Mining Districts of Serbia – Compilation Map and GIS Database, Mineral Resources Division, BRGM Report – 51448, France)

- fine to medium sands and silts make up the river bed along the St. George distributary.

At the discharging mouths of the Chilia and Sulina distributaries the upper limit of 0.01 mg/l CN, admitted for class I waters, is slightly exceeded; in these circumstances the Danube water may be used only for pisciculture and recreational activities.

With two exceptions (Hm 72 Sulina and km 3 Old Stambul) the heavy metal concentrations remain in the entire sector within the natural limits of variation for fine grained sediments. At Hm 72 most toxic heavy metals (Cu, Pb, Zn, Cd) present somewhat higher concentrations, probably as a result of municipal and industrial sewage discharge from Sulina town, situated approximately 7 km upstream.

On the transect from km 3 Old Stambul, in the station close to the Ukrainian bank, appear the highest Zn, Cu, Pb and Cd concentrations from the entire Danube Delta, all exceeding the TEL levels (Buchman, 1999). As no such values were identified during the 2004 cruise it is plausible that the high concentrations of these metals are the result of a recent pollution, possibly related to the works on the new Bystroe canal.

A special mention must be made about the concentrations of chromium and nickel. Their mean and median concentrations for the entire Romanian sector of the Danube River are for chromium 87 µg/g and 82 µg/g, with a variation range from 31 µg/g up to 340 µg/g, and for nickel ≈50 µg/g and 45 µg/g, with a variation range from 13 µg/g up to 105 µg/g. For both heavy metals the mean and median concentrations are very close to the probable effects levels (90 µg/g for Cr and 35.9 µg/g for Ni – Buchman, 1999). This means that approximately 50% of the data exceed, in some cases substantially, the levels of concern, although they still are quite close to the natural backgrounds for shales (90 µg/g Cr and 68 µg/g Ni – Turekian and Wedepohl, 1961). Besides a naturally higher background, mostly as a result of the weathering of ultramafic rocks rich in both elements, characterising the Romanian upper sector of the Danube river, local enrichment mechanisms play an important part in the exceeding the TEL limits.

For chromium, the higher concentrations are always associated with heavy mineral concentrations, as demonstrated by the good linear correlations between Cr on the one hand and Zr and TiO_2, on the other hand (r_{Cr-Zr} = 0.508 and $r_{Cr-TiO2}$=0.769 for 120 samples). Extremely important from an environmental point of view is that the chromium minerals concentrating in heavy minerals accumulations are resistant to alteration and thus the bioavailability of the metal is very low.

For nickel the most important concentrating process is probably its adsorption on hydrated iron and especially manganese oxides and hydroxides, frequently enriched in the surface sediments.

The sandy fraction (0.125 – 0.250 mm) was analyzed for heavy minerals, usually present in less than 10%, the rest being represented by the light fraction. About 40 distinct heavy minerals were identified.

In the Iron Gates I and II lakes the heavy mineral fraction is characterized by the predominance of garnets and opaque minerals. In the Moldova Noua area more than 35% of the heavy fraction is represented by pyrites dumped from the ore mines. Along the sector km 864 – Mile 44 the heavy mineral associations are dominated by garnet. In the Danube Delta area ("Danubian province") the heavy mineral assemblage is characterized by garnets, amphiboles, piroxenes, epidote, ilmenite, disthene and zircon. A supplementary supply from relict deltaic sediments could be pointed out.

Close to the main Romanian distributaries' mouths, the heavy mineral association (1 – 2% per sample) is represented by garnet – amphibol – opacite – epidote, most of the sandy fraction being composed of carbonate grains and organoclasts. For Olt, Arges and Ialomita rivers, the heavy minerals associations are garnet-dominated, while for Topolnita, Vedea and Siret are opacite-dominated. Pyroxenes dominate the heavy minerals association studied in the Prut River sediments.

Clay minerals are predominant in lacustrine areas and along meandered sectors of the river. They are good absorbents for a large spectrum of contaminants (*e.g.* nutrients: NH_4, NO_3, NO_2, PO_4, SO_4; pesticides; heavy metals: Cu, Pb, Zn, Fe, Cd, As, V). Main areas dominated by silt and silty mud, are: Iron Gates I and II lakes, some sectors of the Chilia and St. George distributaries and the lowest part of the Sulina Canal.

There is a strong evidence that the Iron Gate I and II reservoirs are significant traps for sediments (Panin *et al.*, 1992, Popa, 1993; WCD, 2001). It was found that a significant amount of sediment was retained, mainly within the Lower Reach of the Iron Gate I Reservoir. The zones of sedimentation are conditioned by the hydrodynamic characteristics of the area (fluvial conditions, along the section km 072 – km 1015 and and km 942 – km 880, and lacustrine *sensu stricto* downstream, close to the dams). Along the Iron Gates I lake in some places the high velocity and *turbulence of the water are diminished*. Two zones of sedimentation have been identified, Orsova Bay and the Submerged Island before the Iron Gates I dam (Bocaniev S, 2002). Both serve as significant sediment sinks. In these zones, the water speed drops from 10-50 cm/s to almost zero, allowing the deposition of the suspended solids. These lead to the conclusion that the morphological features of the reservoirs play crucial roles for the sedimentation processes. Annually, as much as 1200000 tones of suspended solids are trapped from Danube main stream and deposited in the area of Orsova Bay and in the front of the dam (WCD, 2001).

According to ^{134}Cs and ^{137}Cs vertical distribution along the sediment cores, located in the Iron Gates I lake, rates of sedimentation vary between 1.6 – 2.3 cm/y. Using ^{210}Pb for age dating, on the same cores, the rate of sedimentation was found to be 1.9 cm/y, in agreement with the ^{134}Cs and ^{137}Cs dating (Ruzsa in Panin *et al*., 1995).

Between km 864 and Mile 44 the bottom sediments are represented by fine to medium sand and occasionally gravels. At the same time, in the Danube Delta area the sediments from the main distributaries are dominated by sand and silt. At the mouth of Musura, Sulina and Saint George distributaries the bottom sediments are fine grained (silt, mud, fine sand).

The construction of the Iron Gates I and II dams created complex hydrodynamic conditions, a possible explanation of sediment and nutrients retention. An active decomposition of the bottom organic matter (mineralization, denitrification, and methanogenesis) contribute to the nutrient retention and removal. The nutrient content of the sediments deposited in the Lower Reach of the Iron Gates I reservoirs may account for approximately 2,000 tonnes of P, 29,000 tonnes of N and 271,000 tonnes of Si (Håkanson, Jansson, 1983 in Bocaniev, 2002). Thus, the Iron Gates I Reservoir is a significant sink for nutrients.

Concerning the sediment samples collected from the Romanian tributary rivers they are mostly fine: mud rich in organic matter (Vedea, Arges), silt (Ialomita, Siret, Prut) to sandy sily and fine sand with lamina consisting of vegetal matter (Topolnita).

A notable exception is represented by the river Jiu. Here the bottom sediments are coarse, usually poorly sorted gravels. A great quantity of carbonized vegetal material originating from the coal basin localized on the upper Jiu, upstream from the sampling location, was also identified here.

High values of nutrients or/and heavy metals concentrations are present in the confluences of the Danube with some of tributaries, as Cerna, Topolnita, Jiu, Olt, Vedea, Arges, Ialomita, Siret and Prut. Chemical analysis on the Topolnita and Jiu water showed high concentrations of pesticides (0,95 – 1,12 mg/l). Arges and Ialomita waters have a low quality because NO_2^- and PO_4^{3-} exceed the standard values. However, due to the extremely limited discharge of the tributaries their impact on the Danube River is limited in space.

CONCLUSIONS

The scientific activity was focused on the study of superficial bottom sediments (grain size, mineralogy and geochemical analyses) and the water samples.

The results indicated good to very good quality of the water for most of the Romanian Danube. Locally, higher concentrations of mainly nutrients are present; as a result the water quality decreases to medium, sometimes even poor (*e.g.* Topolnita and Prut rivers).

The quality of sediments is also usually good. The highest values of the heavy metals concentrations are associated with areas of mining activity (*e.g.* km 1049 – km 995, downstream of Timok river mouth) (Fig. 3).

Fig. 3 Romanian Danube watershed – sensitive areas (Google Earth Image)

Higher concentrations are observed in areas with fine to very fine bottom sediments (Iron Gates I and II lakes), close to Iron Gates I and II dams and in the influence area of some tributaries (Vedea, Arges, Ialomita).

Because of human overuse and pollution, the Danube concentrates raw sewage from cities, chemicals from agricultural, waste from factories and oil from ships. This pollution is later washed up on the north-western Black Sea shelf area (Oaie *et al.,* 2001 a., b; Oaie, Secrieru, 2002, 2004, Radan *et al*., 2004, Project RER 2/003, 2004), spreading disease and making them unsafe for marine ecosystems and their diversity.

Chemicals in the water have killed much of the aquatic life and reduced the Danube's fishing industry. Beginning with 1989 scientists and environmentalists have been able to study the full extent of the pollution.

Reducing pollution levels on the river Danube necessitates some local, national and international rules like international and national legislation and agreements, partnerships to reducing water pollution, education at all levels, conservation activities, learning from past disasters, developing of environmental impact assessments for industry and urban planning and/or developing and promoting the use of environmentally safe technologies.

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