Role of the sediments in scavenging inorganic contaminants in the Syr Daria River and the Small Aral Sea (Kazakhstan)

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Abstract
This study is focused on the measurement and the behaviour of the inorganic pollutants in the Suspended Particulate Material (SPM) and the Bottom Sediments (BS) of the Syr Daria River in its Kazakh course, including its outlet in the Small Aral Sea. Two field campaigns were carried out during the low and the high water period. The results display that the current Syr Daria River sediments quality is influenced by several phenomena seasonally and locally controlled: carbonate precipitation, absorption by phytoplankton, scavenging by clay mineral, upstream ground leaching during the snow melting, various discharges (drainage canals, urban and industrial waste waters), connection with delta lakes during the flooding period and impact of the geochemical background.
The study of the solid – liquid partition coefficients emphasizes the contrasted behaviour of trace elements with respect to different scavenging phases. As a result it appears that in spite of the mitigating action of the dam reservoirs, of the irrigated areas, of the overflow lakes and flood plains, the Aral Sea sediments are not completely preserved from the contamination.

Introduction

If there are still some studies concerning the Aral Sea waters and sediments, few focused on the Syr Daria River that has played, in this tragedy, a role of a target as well as a vector for the pollution [1]. Furthermore the behaviour and the impact of Suspended Particulate Material (SPM) within the main water bodies were not taken into account [2] in the Aral Sea Basin water quality assessments. Syr Daria River used to bring to the Aral Sea, before 1948, 15 millions of tons of new sediments each year [3]. This amount decreased severely with the great soviet planning of the Aral Basin but was still of around 1.7 million of tons in 2003-2004. This amount itself displays that the management of riverine sediments is a growing issue that develops in parallel with water management and soil conservation [4].
This study will attempt to answer the questions of the spatial distribution and seasonal distributions of the different inorganic contaminants in the Syr Daria River and the partition of those contaminants between the dissolved, the particulate and the settled phases. Moreover this study will be an attempt of quantification of the various fluvial filter effects [5].

Materials and Methods

Object of the study. This work will consider mainly inorganic trace elements of SPM and Bottom Sediments (BS) in some water bodies of the Kazakh Syr Daria Basin.
Sampling periods. Two field campaigns were carried out: (1) at the end of the low water season in September 2003 and (2) during the following high water (and even flooding) period in April 2004.
Samples location. Representative sampling places were selected in the Kazakh Syr Daria Basin. Along the river itself, 5 spots were selected as being supposed to play a bigger role either in the contamination or in its mitigation and sampling places were located up and downstream of them (Fig. 1): the Chardara impoundment, the Arys River confluence (a right bank tributary crossing the

industrial city of Chimkent, part of the the Arys-Turkistan irrigation system), the city of Kyzyl Orda, Chi-Ili city and its uranium processing centre, Zhalagash irrigated area, the former delta zone, its lakes and another irrigation area. Concerning other water bodies of the basin, a drainage canal (in the Zhalagash area), two lakes of the former delta (Katankol and Kamyslybas) and a "takyr" (endorheic pond) were sampled. In the Small Aral Sea, two spots were considered: one sample was taken in the Berg channel and 2 samples were collected at about 2 km offshore the Syr Daria mouth, in April, one at 0,5 m below the surface and the other 0,5 m above the 2.7 m deep bottom.

Sampling methodology. Series of 0.45µm (pore diameter) cellulose acetate filters were weighed in order to measure the amount of filtrated SPM. The BS at each sampling place were collected during the first campaign using a hand drill, made of several screwed pipes and then poured into closed plastic boxes.

Analytical means. SPM were characterised first by X-Rays diffraction. Powder X-ray diffraction (XRD) patterns have been performed on a Siemens D 5000 diffractometer using Co-K_α radiation and operating at 40 kV and 30 mA at room temperature. The scans have been recorded from 4 to 84° (2θ) with a step of 0.02° and a counting time of 1s per step.

Concerning trace elements analyses, BRGM laboratory used high performance ICP/MS (Thermo Plasma- Quad 3 model). After drying to 40°C solid samples were crushed to 80 µm and then attacked by a mixture of HNO_3 and HF acids and then again by HNO_3. All the analytical results of the contents in the solid phase samples were calculated in µg/g (mg/kg or ppm) of dry matter.

Results

The mineralogical composition of the SPM (Table 1) was determined on the samples of September. They are made up of quartz, K-feldspars, plagioclases, clay minerals, and calcite (except for calcite and clay minerals in the Syr Daria delta and for clays downstream Kyzyl Orda).

Table 1: Mineralogical composition of the SPM phase from X-Rays diffraction method

	SD1	SD2	SD3	SD5	SD6	SD8	SD9	SD10	SD13	SD14
Quartz	++	++	++	++	++	++	++	+++	++	++++
Plagio	+	+	+	+	+	+	+	+	+	+
K- Fdsp	+	+	+	+	+	+	+	+	+	+
Calcite	+	++	+	+	+	+	+	+	+	
Dolom.	(+)	(+)	(+)	(+)	(+)	(+)	(+)		(+)	
Phyllites	Chlorit	Chlorit	Chlorit	Chlorit	Chlorit	Chlorit	Chlorit		Chlorit	
	Illite &/	Illite &/	Illite &/	Illite &/	Illite &/	Illite &/	Illite &/		Illite &/	
	or mica	or mica	or mica	or mica	or mica	or mica	or mica		or mica	
		Paly.?								
		Smect.	Smect.				Smect.			
Gypsum			(+)							
Phosphat	Kon.?				Kon.?					
Pyrite		(+)								

++++: abundant - very abundant; +++: present – abundant; ++: in low concentration – present; +: traces -.**Plagio**.: plagioclase (Na-Ca)($AlSi_3O_8$); **K-Fdsp**: Microcline ($KAlSi_3O_8$); **Dolom**.:dolomite, (Ca, Mg)$(CO_3)_2$; **Chlorit.**: chlorite (Mg, Al, Fe)$_{12}$[(Si, Al)$_8O_{20}$](OH)$_{16}$; **Illite**: KAl[Si_7AlO_20](OH)$_4$; **Smect**.: smectite (Ca,Na)(Al, Mg, Fe)$_4$[(Si, Al)$_8O_{20}$](OH)$_4$.nH_2O; **Paly**.: palygorskite (Mg,Al)$_5$(Si,Al)$_8$ $O_{20}(OH)_2\cdot 8H_2O$; **Gypsum**: $CaSO_4.2H_2O$; **Kon.**: koninckite, $Fe^{3+}[PO_4]\cdot 3H_2O$

The presence of carbonates, even scarce, is noticeable and confirms the (even temporary) saturation with respect to calcite. The presence of pyrite is consistent with the high concentrations in metals that were analysed in the BS and, seasonally, in the SPM of this place. Chemical analysis of major elements confirmed the prevailing role of silica (quartz, feldspars and clays), calcium (clays and calcite) and aluminum (clays and feldspars). Furthermore the SPM contain a variable but significant part of organic carbon, mentioned as Total Organic Carbon (TOC).

Table 2: A comparison of previous analytical studies on Aral Sea BS with our data (in ppm)

	Cd	**Zn**	**Pb**	**Rb**	**Sb**	**Th**	**U**	**Cr**	**Co**	**As**	**Cs**
Aral Sea BS (this work)	*0,31*	*64.1*	*19.3*	*77.8*	*1.2*	*10.4*	*3.37*	*51.2*	*11.6*	*14.2*	*4.8*
26 samples in 1980-84 [6]		40		58	3.7	13	1.9	32	9.8	7.4	5.1
map from178 samples [7]							> 2 < 5				
160 samples in 1971 [8]			> 4 < 24								
128 samples [9]				>65 <73							6.3
128 samples [10]	> 0,012 < 0,66										

It must be stressed that the values collected from the soviet literature (Table 2) - the last analyses concerning the Aral Sea sediments - are average ones whereas we analysed a single sample. Taking this point into account the values are in relatively good agreement.

Discussion

Carbonate precipitation. As the Syr Daria River waters were controlled by the bicarbonate equilibrium [1], when temperature was supposed to reach its maximum, in September, all the samples were saturated with respect to the calcite from the Chardara impoundment to the former delta. This trend concerned Ca (-36%), Sr (-31%) and, to a lesser extent Mg (-21%) that decreased from up to downstream the reservoir. Other analysed trace elements were not significantly affected. Such a seasonal saturation in carbonate was observed in the Loire River [11]. In April the river water became under-saturated except in the former delta.

Growth of phytoplankton. The Total Organic Carbon (TOC) increases within the SPM by a factor 100 from up to downstream the Chardara impoundment in April (it was not analysed in September). In the same place, in September, the SPM content (Fig.1) increases by a factor 3.5 and then decreases again farther in the reservoir. In April, conversely, it drops continuously in the same reservoir by a factor of 111. Similar trends are observed in the lower course of the river. As a result of sunlight, summer warming and a little input of nitrate, the growth of phytoplankton explains the previous trends in the Chardara reservoir and in the former river delta, surrounded by irrigation areas and connected with lakes. Most of the mineral phases settles down in quieter waters of the reservoir or of the canals, so that the proportion of biomass, chemically expressed by the TOC, increases among the SPM. The impact of phytoplankton is confirmed by the water quality data: diurnal variation of pH and dissolved oxygen [1]. The Lake Arnasay (overflow of the Chardara reservoir) displayed the same trend of eutrophication [12]. This algal growth occurs at the expense of the dissolved silica and enables a denitrification of the Chardara reservoir and the former delta.

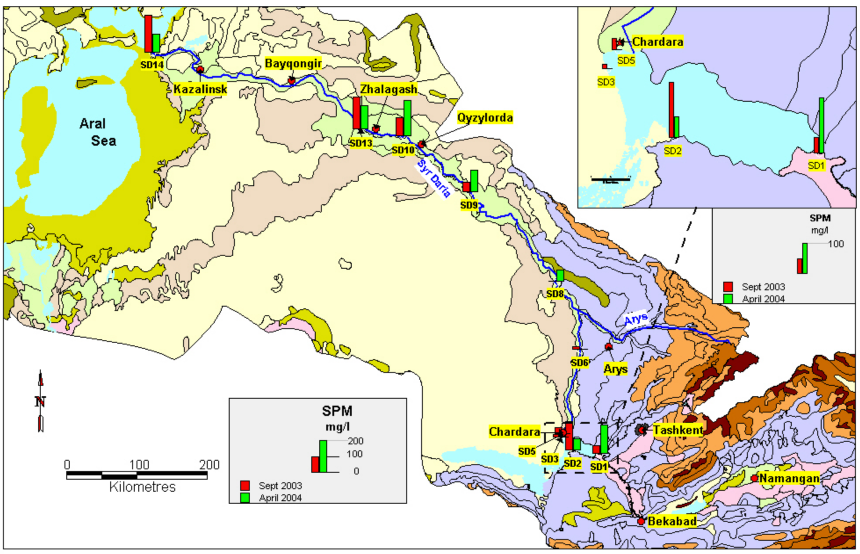
Metals scavenging. In April trace elements clearly increases vs September, in the upstream river waters (Cu: +42%; Zn: +43%; U: +3%). This increase is accentuated in the SPM (Cu: +17600%; Zn: +1216%; U: +46%) and cannot be explained only by the lowering of the inflow SPM content (-32%). Furthermore, if the presence of pyrite is confirmed, such a mining material would indicate that scavenging is not the only source of metals in the sediments. Nevertheless SPM of the Chardara impoundment better filtered the metals in September (Cu: -46%; Zn: -54%; U: -31%) than in April (Cu: -12%; Zn: -7%; U: -16%) although, at that moment, their enrichment in metals was stronger (Cu: +8307%; Zn: +2929%; U: +229%) than in September (Cu: -6%; Zn: +417%; U: -8%). This paradox results from the higher content in SPM of the September waters (16 mg/l) vs April ones

(2.2 mg/l) downstream the reservoir and it displays the seasonal change in the nature of SPM. The ability of Phytoplankton in the Chardara impoundment to accumulate heavy metals was already noticed [13] but for a same mass of particles, clays can trap metals more efficiently. Considering the trace elements contents of the BS, Chardara reservoir samples show higher values than other Syr Daria River (or Aral Sea) BS. The heavy metals content of the Chardara impoundment sediments increased from up to downstream as it was the opposite trend for the water samples [1, 17]. The reservoir acts as a selective, limited and seasonally changing trap for heavy metals.

Upstream ground leaching. An environmental review from Uzbekistan [14] indicated that uranium content in BS of the Syr Daria River, at the Kyrgyzstan and Uzbekistan border, reached 215 ppm together with high concentrations in Pb, Fe, Cr, etc. Such strong values can be explained by the mining centres in Kyrgyzstan. The lower contents of the sediments in the Kazakh part of the river, may be explained by the transit of Syr Daria through a cascade of 5 dams and their impoundments in Tajikistan and Kyrgyzstan that act as partial sediment traps and could store a large part of river contaminants. Nevertheless, the case of Chardara displays that the trap remains partial. Moreover in order to explain the relatively high contents in heavy metals of the inflows especially in April, the presence of pyrite could suggest that part of those contaminants come from the leaching of tailings upstream the dams when snow melts. The other possibility is that the pollution results from the leaching of the agricultural wastes [13] from the Tashkent oasis (an irrigated area needing formerly a discharge of 10.3 km^3/year to be compared to the mean 37 km^3/year of the Syr Daria [3]) or from the Ferghana valley (needing formerly a discharge of 17.3 km^3/year [3]).

Impact of irrigation systems. Several irrigated areas may impact the Kazakh Syr Daria course. The best period for an evaluation of the qualitative impact of the irrigation would have been when irrigation fully works. However, this impact can be studied first up and downstream the confluence with the Arys River, another right bank tributary that supplies the Arys – Turkestan irrigation system (needing formerly a discharge of 3.1 km^3/year [3]). At the end (September) or at the beginning (April) of the drainage period, waters are not significantly loaded in contaminants. Nevertheless an increase of the SPM content (higher in April) was noticed (Fig.1). Within the SPM the clayey fraction (marked by the Al content) increases and in April the heavy metals content drops significantly. Moreover, in the BS, a similar, although smoother, trend is observed (drop of all the trace elements) but with a slight increase in TOC.

Figure 1: Spatial distribution of SPM along the Syr Daria River in the Kazakh Priaralie Region



Suspended Particle Materials (SPM) in water samples of the Syr Daria River

Other irrigation areas may impact the river around the cities of Zhalagash and Kazalinsk (forming the lower Syr Daria Basin irrigated areas that needed a discharge of 7.1 km^3/year [3]). From up to downstream, in the both areas, the SPM content increases in September (likely phytoplankton) and decreases in April, the water composition does not change significantly and in the SPM phase the TOC fraction increases. In the Zhalagash area the clayey fraction increases more significantly in April leading to the same trend for the trace elements contents of the SPM and BS. In the Kazalinsk area the clayey fraction diminishes downstream (while the silica increases) as well as trace elements (except Sn and Sb for the SPM while Cs and Mo are the exceptions for the BS).

In the three irrigated areas the changes in the water composition are minor, the SPM content increases in September (Fig.1) and among the SPM, the TOC content increases from up to downstream. The clayey fraction plays a significant role in trapping the trace elements although the phytoplankton growth (marked by the TOC) is more efficient in this action (as it was noticed in the Chardara reservoir). In the former delta the sand of the dunes may be detected in the sediments as well as some signs of the geochemical background.

A drainage canal in the Zhalagash area was also studied. The mineralogical composition differs slightly of the Syr Daria sediments: a little more calcite and possibly some gypsum (its water is three times more salted). The BS are enriched in clayey fraction (Al content) and as well in trace elements in the same order than a downstream Chardara impoundment BS. Irrigated areas and reservoirs play thus a similar role for trapping the inorganic pollutants. As a result of reductive conditions at the end of the drainage period, part of the metals pass again in the dissolved phase leading to the highest concentrations in Mn, Co, Ni, Cu, Zn and U among the sampled water bodies of the Kazakh Syr Daria Basin.

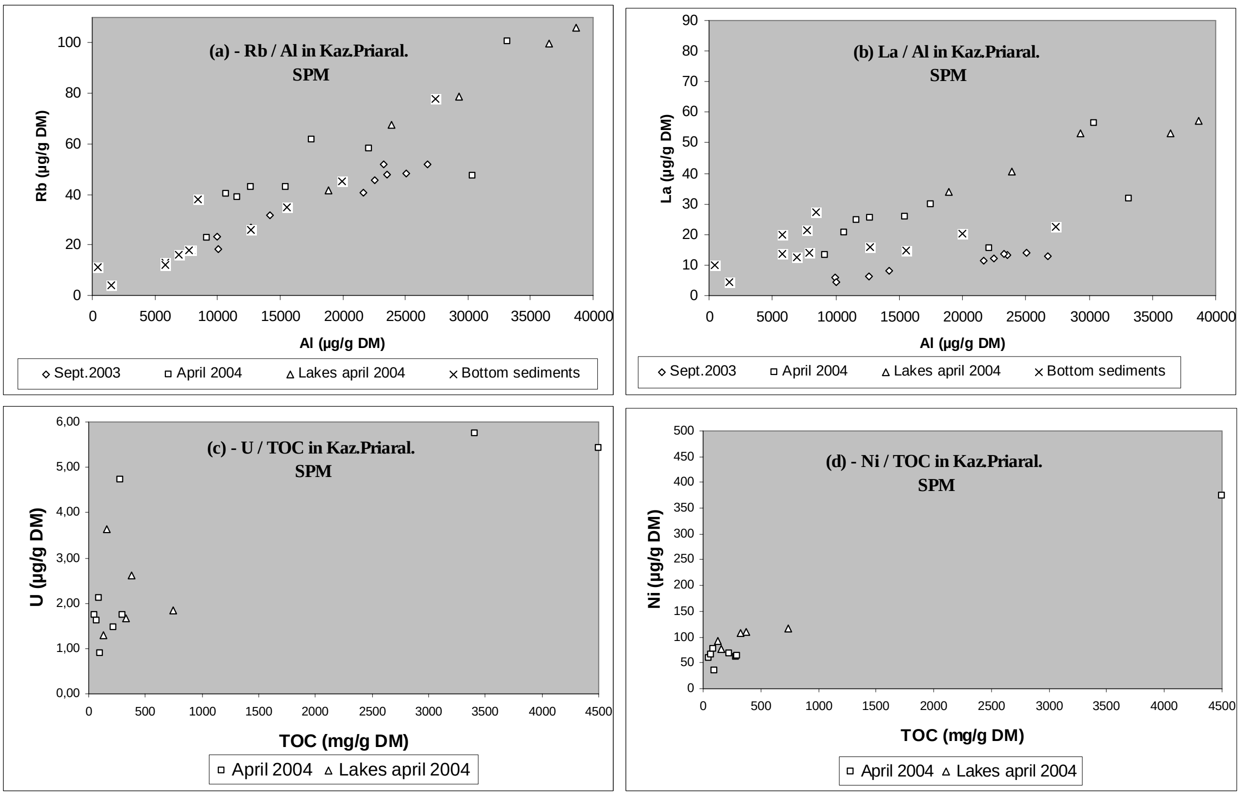
Impact of urban and industrial centres. The investigated area may be impacted also by some urban and industrial centres. Starting from upstream, the waste waters flowing from the town of Tashkent (> 2.3 M of inhab.) may contribute to the contamination of the river upstream the Chardara reservoir via the Chichik River. It may also be assumed that this contribution is mitigated by the irrigated area of the Tashkent oasis. It is also likely the case of the pollution coming from the city of Chimkent (370 000 inhab.), well known for its Zn, Pb metallurgy. The Arys River crossing the city supplies the irrigated area of Arys-Turkestan and losses there most of the possible pollutants. The city of Kyzyl Orda (180 000 inhab.) does not seem to impact significantly neither the waters nor the sediments of the Syr Daria River. A clear decrease in trace elements, TOC and clay fraction is even reported among the BS. The uranium processing centre of Chi Ili impacts only the composition of the BS downstream the city (maximum concentrations in Th, La, Ce and enrichment in other trace elements and TOC).

Other water bodies. *Delta lakes*. The Kamyslybas Lake is an overflow of the drainage system and is temporary fed by the Syr-Daria River during High Water period. Nevertheless its salinity reaches a level of brine (13 g/l). Its waters display a enrichment in Sr, Ti, and Sb (supposed signature of the geochemical background) but also in Ni, As, Mo, U that may accumulate here from the drainage system and possibly from the Syr Daria River. The SPM content is low and its composition has a spectrum similar to that of the downstream sediment of the Chardara reservoir, although in lesser contents. The lake is partly covered with reeds that bring organic matter preserving reductive conditions toward the bottom. As well as for the drainage canal and possibly the Chardara reservoir the redox conditions may give rise to re-dissolution of metals brought in the SPM. This overflow has a feedback impact to the Syr Daria bringing higher trace elements contents in the former delta part of the river waters and also some biomass (TOC content of the SPM). Flood plains are supposed to be efficient traps for riverine particulates [4] but in the case of contaminated basins, the reverse trend can be observed.

Small Aral Sea. Strong vertical (and lateral) variations characterize the salinity of the Aral Sea. Bottom water is more mineralized (+164%) although impoverished in trace elements wth respect to the surface sample. This paradox can be explained by the higher content in SPM of the bottom water sample (+36%). The bottom SPM trap a bigger part of trace elements although SPM are not so enriched in those elements. The SPM of the surface sample are enriched in almost all the trace elements (except Ti) and the TOC with respect to the Syr Daria delta sample. Eventually the sample of BS displays a general enrichment in trace elements (except B) with respect to the other sampled BS of the Kazakh Syr Daria Basin especially Ti (x34), Sn (x4.7), Sb, Th (x2), Ni, As,…In spite of successive reservoirs, drainage nets, flood plains and overflow lakes, this sample of sediments of the Small Aral Sea is more contaminated than upstream BS.

Impact of the geochemical background. It may be assumed that local background has an influence on river water composition and especially on its SPM. Some titaniferous layers in the Oligocene series are indicated at the North of the Aral Sea [15] so that a local Ti enrichment would be natural. An enrichment of the Aral Region sediments in Sb, Hg and Zn was suspected [6]. Higher contents in Sb and Ti were found in Aral Sea BS and Katankol Lake waters. The latter worked as a closed system and may reflect the geochemical background. Another sample of isolated system is given by a temporary pond (or “takyr”) sampled far from any human activity. Its alkaline water is enriched in Ti, As, Mo, Th and although its SPM content is much higher than all other sampled water bodies, the concentration in trace elements remains among the strongest and reaches the maxima for B, Fe, Co, Sn. If this takyr reflects the geochemical background, the latter is heavily loaded. The high content in Sr either in the waters (2 to 31 mg/l) or in the sediments (0,2 to 6 mg/kg) in all the studied samples, must also be stressed (Fig.3).

Figure 2: Trace elements in solid phases of the Syr Daria River and some associated water bodies: (a) Rubidium vs Aluminum; (b) Lanthanum vs Aluminum; (c) Uranium vs Total Organic Carbon; (d) Nickel vs Total Organic Carbon



Trace elements partitioning. The point is how the trace elements are distributed among the various solid phases. If we assume that Al is a good index of the clayey phase, it could be noticed that Fe, Ti, Cr, Co, Mn, La, Rb, (Fig.2 a and b) Cs are correlated with this element. Clay minerals can enhance adsorption links with metals (Co, Mn,…) but also cation exchange reactions with alkali elements (Rb, Cs, etc.). While As, Ni, U (Fig.2 c and d), Zn, Pb, Cu has a correlation with the TOC. Correlations are not exclusive. Uranium, for instance, has a better correlation with Fe but has links with Organic Carbon and Al.

The solid / liquid partition coefficient is generally used to determine how behave an element in relation to two different phases. It can be calculated knowing the respective concentrations of the given element in each phase. The following definitions are set down:

$\log Kd = \log([X]_{SPM} / [X]_{TDS})$ where

$[X]_{SPM}$ is the concentration of the element X in the particulate fraction, in µg/g of dry SPM

$[X]_{TDS}$ is the concentration of the element X in the dissolved fraction, in µg/g (or mg/l) of solution

Some significant changes occurred in April vs September concerning Mn, B, As, Mo, Cd, Sb, Th, Pb, Sr the log Kd of which increased, i.e. they were more attracted by the solid phase in spite of a bigger dilution. Furthermore the range of values of log Kd for Cu and Ni extend towards higher values, which goes in the same direction. Three factors may explain this trend (1) the lower DOC content that limits the formation of organic complexes keeping elements in the dissolved phase, (2) the lower temperature inhibiting precipitation of carbonate and hydroxides but enhancing the silica and clays, (3) the higher proportion of clay minerals in the SPM. The case of Sr is special as its concentration is abnormally high in all the studied water bodies. It behaves in compliance with Ca and seems to precipitate with carbonate in September, conversely to the case of the Loire River [11]. Nevertheless it is much more attracted by the clays and the solid phase in April (Tab.3).

Table 3: Solid / liquid distribution (Log Kd) for Pb, Zn, Rb and Sr in samples of the Syr Daria and of other rivers of the world [11]

	Pb	**Zn**	**Rb**	**Sr**
Syr Daria September 2003	4.15 – 5.2	4.1 – 5.35	3.9 – 4.5	1.6 – 2
Syr Daria April 2004	4.8 – 6.2	4.2 – 5.8	4.1 – 4.7	4.9 – 6.4
Loire	4.8	4.7	4.3	3.2
Seine	5.3	4.7 - 5		
Congo	5.8		4.3	3.6
Amazone			4.5	3.5

Conclusions

Studying the water bodies of the Kazakh Syr Daria Basin from the analysis and the behaviour of the inorganic contaminants in their sediments highlighted the combined role of different processes having natural (seasons cycle, geochemical background,…) and anthropogenic (mining works, urban, industrial and/or agricultural wastes, dam impoundments, irrigated areas, artificial lakes,…) origins. It illustrates the interaction between the river water quality and the sediments. It could contribute to explain, for instance, why the Aral Sea sediments are contaminated in spite of the successive mitigation of the Syr Daria River long course, dam reservoirs, irrigated areas, floodplains and overflow lakes. However the "contamination" has still to be qualified as the geochemical background may have a significant impact. It emphasizes the need of a specific focus on the particulate phase and the bottom sediments in order to assess the quality of the water bodies (Chardara reservoir, Zhalagash drainage canal, Kamyslybas overflow lake or even Syr Daria River itself) and to forecast what it could become. Any renewal of the Syr Daria Basin planning will have to take this aspect into account.

Acknowledgment

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