THE DISTRIBUTION OF DISSOLVED AND PARTICULATE MERCURY IN THREE SIBERIAN ESTUARIES AND ADJACENT ARCTIC COASTAL WATERS

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Abstract. Dissolved and particulate mercury distributions were determined in the three largest Siberian rivers and in adjacent Arctic coastal waters during two cruises. Water samples were collected in the Lena River and its mixing zone in the Laptev Sea in September 1991, and in the Ob and Yenisei Rivers and the adjacent Kara Sea in September 1993. Average total dissolved Hg concentration was 5.0 pM in the Lena River, 2.8 pM in the Ob River and 1.5 pM in the Yenisei River. Mercury content of suspended particulate matter was low, averaging 0.17 mg kg^{-1} in the Lena and 0.05 mg kg⁻¹ in the Ob and Yenisei Rivers. These concentrations are lower than those observed in other world rivers affected by local input of man-made origin. In the estuarine mixing zones, higher concentrations of dissolved and particulate Hg which may originate from the spring flood were found. The carbon cycle is apparently a driving mechanism for Hg distribution in Arctic coastal waters. Particulate Hg content was positively correlated with the content of organic matter of the particles. In the Kara Sea, uptake by phytoplankton is suspected to be responsible for the increase in particulate Hg levels. Mercury fluxes from the three rivers to the Arctic Shelf are estimated and compared to direct atmospheric inputs.

1. Introduction

Arctic regions have long been considered to be pristine. However, volatile elements and substances, especially those from anthropogenic origin, have been shown to be subject to long range atmospheric transport. As a result of their volatilization and subsequent condensation, such chemical compounds may accumulate in polar regions (Barrie *et aL,* 1992). Mercury, which cycle is highly affected by anthropogenic emissions, is one of these compounds. Therefore, we suggest that the drainage basins of Siberian rivers flowing into the Arctic Ocean are collectors of atmospheric Hg deposition. Even though most of the deposited mercury may be trapped in the tundra, we hypothesize that part of it is transferred to the adjacent marine environment.

Owing to climatological conditions and logistics difficulties, Hg distribution in these major estuarine systems remains unknown. We report the concentrations of Hg in three Siberian estuaries during the summer season and discuss the fate of Hg during the fresh water - sea water mixing and the subsequent flux of Hg to the Arctic Ocean.

2. Study area

Three main rivers drain the Eurasian continent: Ob, Yenisei and Lena (Fig. 1). These rivers are the largest rivers draining to the Arctic Ocean in terms of water discharge.

Together, they provide more than 50% of the annual water discharge and about 36% of the annual suspended particulate matter (SPM) export from the Eurasian Arctic basin into the Arctic Ocean (Gordeev *et al.,* 1993). The climate in the Siberian area is characterized by long cold winters and short cool summers. These rivers and the surface waters of the mixing zone have a thick ice cover during eight months. The temperature of the bottom waters of the Laptev Sea and the Kara Sea remains always below 0° C *(Martin et al.,* 1993). The rivers discharge about 60% of the annual water discharge and more than 70% of the annual SPM discharge during the flood period (May-July) (Gordeev *et aL,* 1993).

Fig. 1. The Arctic coast of northern Siberia.

The Lena River delta is located on the coast of the Laptev Sea which belongs to the East Siberian Basin (Fig. 1). The Lena represents the second largest river (after Yenisei) discharging to the Arctic Ocean and ranks first with regard to the total suspended matter export (Gordeev *et al.,* 1993) (Table I). It drains the Siberian forest (taiga) and tundra, and is characterized by a low particulate content as compared with other major world rivers, and by 'black' waters enriched in organic matter (Martin *et al.*, 1993).

TABLE I Mean annual water and suspended matter discharge into the Arctic Ocean (Telang *et aL,* 1991; Gordeev *et aL,* 1993)

River	Drainage area	Length	Mean annual	Suspended solids
	$(x10^6 \text{ km}^2)$	(km)	water discharge $(km^3 a^{-1})$	discharge $(x10^6 \text{ ta}^{-1})$
Lena	2.49	4,337	525	17.6
Ob	2.55	3,650	429	16.5
Yenisei	2.59	3,844	620	5.9

The Ob River is formed by the confluence of mountain streams Biya and Katun, both of them originating in the Altai Mountains. Ob represents the third largest river discharging into the Arctic Ocean (Gordeev *et al.,* 1993) (Table I). It flows through taiga forest then through the forest tundra zone and tundra (Telang *et al.,* 1991).

The Yenisei is the largest river of the Eurasian Arctic basin (Gordeev *et al.,* 1993) (Table I). The main hydrochemical characteristics of the Yenisei River are a low turbidity and a weak mineralization (Telang *et al.,* 1991). This is because the river flows across the mountains and the permafrost zones. The amount of SPM transported from Yenisei into the Arctic Ocean is the lowest of the three rivers (Table I).

In general, the low average turbidity observed in Laptev and Kara Sea basin rivers can be related to the wide-spread permafrost and the small thickness of the active layer on the drainage basins of these rivers and to the very short time with above freezing temperature (Telang *et al.,* 1991).

3. Sampling and analytical methods

3.1. SAMPLE COLLECTION

Sampling was undertaken as part of a Russian-French cooperative program. Water samples were collected in the Lena River and the adjacent Laptev Sea from September 4 to 21 1991, and in the Ob and Yenisei Rivers and in the Kara Sea from September 15 to 29 1993. The location of the sampling stations is shown in Fig. 2.

Fig. 2. Location of the sampling sites in the Laptev Sea and the Kara Sea.

For the Lena River, station L1 was located upstream of the divergence of the delta branches. River station L16 was established near the mouth of the Bykovskaya branch which, on average, accounts for 27% of the river discharge to the sea (Létolle *et al.*, 1993). The Laptev Sea is shallow (generally less than 25 m) and subsurface samples were collected in the open brackish surface plume. Maximum salinity of the samples collected reached 19.6 and 32.6 in surface and deep waters, respectively, at the northernmost station (C30).

Two sampling transects were carried out in the Kara Sea, the first one at the Yenisei Estuary, from 76° N southward until we reached freshwater (stations 10 to 12), and the second one along the Ob Estuary, reaching fiver water at stations 26 and 27. Subsurface water samples were collected in the two estuaries. In the Kara Sea, subsurface samples were collected as well as samples from the halocline layer. The highest salinity obtained in subsurface samples was only 25.1, and deep water samples (salinity up to 34.1) were collected at four stations to provide sea water references.

Sub-surface samples were collected in 5 1 Teflon bottles using a Teflon pumping system including an all-Teflon diaphragm pump (Asti, model PCS-2) and Teflon tubing. The tubing was maintained distant from the ship with the help of a 5m long pole. At two sites in the Kara Sea, surface samples were also collected from a small boat directly by hand in the 5 1 Teflon bottles wearing arm-length polyethylene gloves, and the similarity of the Hg concentration measurements with pumped water samples was verified. Samples at depth greater than 10 m were collected with Teflon coated 5 1 Go-Flo bottles (General Oceanics, FL, U.S.A.) fixed on a Kevlar hydrowire. Sample collection was performed under a laminar air flow hood and polyethylene gloves were used for handling operations to avoid sample contamination. All Teflon and plastic-ware was washed and stored according to Cossa *et aL* (1994).

3.2. ANALYTICAL METHODS

Samples were analyzed for total dissolved Hg ($[Hg_T]_D$) and total particulate Hg ($[Hg_T]_p$). For the separation of dissolved and particulate mercury species, water samples were filtered on board ship under a laminar flow hood through combusted (500° C) and acidcleaned quartz fiber filters (Whatman QM-A, $0.8 \mu m$) held in polypropylene filter holders. Filters were stored at -18^oC in tightly closed polystyrene Petri dishes before analysis. Filtered water was transferred into acid-cleaned Teflon bottles and acidified with concentrated HCl (0.5% v/v , Suprapur Merck). The bottles were tightly sealed using pliers. Analysis were performed at the 1FREMER laboratory. All Hg species were detected by cold vapour atomic fluorescence spectrometry after transformation to $Hg⁰$ (Bloom and Fitzgerald, 1988) using a Merlin instrument (PSAnalytical). Concentrations of $(Hg_T)_D$ were determined after reduction by NaBH₄ and double gold amalgamation (Gill and Bruland, 1990). Concentrations of $(Hg_T)_p$ were measured after HNO_3/HCl (9:1) digestion of the particles in Teflon reactors, using Suprapur (Merck) acids, and reduction with $SnCl₂$ (Cossa and Fileman, 1991).

Detection limits, defined as three times the standard deviation of the blank expressed per unit sample analyzed, were: 0.7 pM for $(Hg_T)_{D}$ and 0.02 mg kg⁻¹ for $(Hg_T)_{P}$. Method accuracy was routinely checked using available reference material (mercury in water 1641b from the U.S. National Bureau of Standards; marine sediments BEST-1 from the National Research Council of Canada). Precision, defined as a coefficient of variation of

Measurements of salinity, SPM concentrations, particulate organic carbon (POC) and chlorophyll pigments were performed using standard procedures.

4. Results and Discussion

4.1. HYDROLOGY

Progressive mixing of the river waters in the Laptev Sea and Kara Sea results in the formation of large brackish surface plumes extending several hundred km northward, over more dense and saline waters. Examination of CTD (conductivity, temperature, depth) profiles in the Laptev Sea and the Kara Sea shows that a three-layer stratification could often be defined, with a thick brackish surface plume overlying an intermediate layer and a bottom water mass. According to these observations, tentative identification of the water masses from which samples were taken is presented in Table II.

In the Lena area. SPM content varies in subsurface waters from 29.6 mg 1^{-1} in the river to 0.6 mg l^{-1} at the northern station in the Laptev Sea (Table II). For the Ob and Yenisei area, highest SPM content are also found in the river end members: 135.6 mg $1⁻¹$ in the Ob and $20.4 \text{ mg } l^{-1}$ in the Yenisei. In the surface water plumes, SPM content decreases as salinity increases to about 0.4 mg $1⁻¹$ at the seaward extremity in the Kara Sea. The organic carbon content of SPM increases with the decrease of suspended load. Thus, higher POC content is found in particulate matter of the mixing zones than in the rivers. Such a relation between SPM and POC content of particles was previously described in several rivers by Meybeck (1982) and in the Lena (Gordeev and Sidorov, 1993).

4.2. MERCURY IN RIVERS

River water concentrations are represented by the freshwater data obtained for subsurface samples: stations L1 and L16 in the Lena, stations 26 and 27 in the Ob, and stations 10, 11 and 12 in the Yenisei. In these cases, a straight vertical profile of salinity was found showing the absence of stratification. Results of dissolved and particulate Hg analysis are given in Table II.

Total dissolved Hg concentrations in the rivers are comparable for the three rivers. Average (Hg_T)_D concentrations range between 1.5 and 5.0 pM. These average values are low compared to the concentration range measured in other world rivers (Table III). Only the Krka (Croatia) and the Loire (France), which have no local sources of Hg contamination in their upper course, have comparably low levels. We restricted the comparison to a small number of recent credible data obtained in rivers draining climatologically and geologically diverse catchments.

Average Hg content of particulate material was 0.05 mg kg^{-1} in the Ob and Yenisei and 0.12 mg kg^{-1} in the Lena. The levels measured in the Ob and Yenisei are comparable to the lowest levels measured in non contaminated sites, for instance in deep marine sediments (Cox and McMurtry, 1981). Our values are lower than those of other world rivers (Table III).

TABLE II

Particulate and dissolved concentrations in the Lena, Ob and Yenisei Rivers and adjacent seas	

Location	Station number	Water mass	Sample depth(m)	Salinity	SPM $(mg1^{-1})$	POC (%)	(HgT)D (pM)	(Hgr)p $(mgkg^{-1})$
	16a	surf.	8	9.18	3.4	4.6	1.7	0.07
	16b	surf.	8	8.30	$\qquad \qquad \blacksquare$	$\qquad \qquad \blacksquare$	1.1	0.06
	17	surf.	5	18.38	6.5	3.4	1.2	0.11
Kara Sea	$\boldsymbol{2}$	surf.	8	21.32	0.42	10.2	6.7	0.11
	3	surf.	8	25.07	0.48	18.8	1.3	0.12
	$\overline{\mathbf{4}}$	surf.	9	23.64	0.40	11.3	0.7	0.03
	6	surf.	9	21.60	1.1	9.5	9.5	0.06
	18	surf.	0.2	16.14	1.3	7.3	2.0	0.09
	18	surf.	$\overline{\mathbf{4}}$	16.00	1.1	13.0	1.5	0.07
	19	surf.	5	23.66	0.36	7.6	1.2	0.11
	20	surf.	9	17.24	0.45	14.1	2.2	0.12
	21	surf.	9	22.07	0.31	11.2	6.5	0.25
	$\mathbf{1}$	interm.	9	27.64	0.29	4.1	$\hat{}$	0.11
	4	interm.	15	29.18	\blacksquare	2.3	12.1	
	5	interm.	8	27.34	0.55	8.0	1.1	0.09
	7	interm.	9	31.40	1.4	7.4	4.6	0.06
	21	interm.	15	24.01	9.6	5.6	3.9	0.42
	22	interm.	9	27.11	0.20	8.8	5.1	0.24
	24	interm.	9	31.27	3.6	7.3	2.9	0.05
	3	deep	45	34.02	0.57	2.3	2.2	0.08
	$\overline{\mathbf{4}}$	deep	30	33.92	1.3	2.6	17.0	0.07
	20	deep	100	34.10	0.07	10.5	1.5	0.67
	21	deep	30	33.63	9.6	3.1	11.1	0.03

TABLE II (continued)

TABLE III

Comparison of average concentrations of dissolved and particulate Hg in the suspended matter of the Lena, Ob and Yenisei Rivers and other world rivers

(1) Cossa *et al.,* 1994; (2) Coquery (1994); (3) Cossa and Martin, 1991; (4) Mikac *et al.,* 1989.

Concentrations measured in the Ob and Yenisei Rivers are more than one order of magnitude lower than heavily industrialized rivers like the Seine or the Rhône (France). **Thus, in both the dissolved and the particulate phase, mercury concentrations are low in these Arctic rivers at the time of sampling. These low levels may result from a restricted human activity over the drainage basins and from the low temperature for most of the year, which limits the processes of erosion and weathering in the river drainage basins.**

Particulate Hg represents the main fraction of mercury transported by the rivers: 64% in the Lena, 79% in the Ob and 59% in the Yenisei.

4.3. MERCURY IN THE MIXING ZONE

LENA

Examination of concentration variations in the mixing zone **is restricted to the** surface **plume for the** Lena, as mainly subsurface water **samples were** collected (Table II). Concentrations of $(Hg_T)_D$ are presented along the salinity gradient (Fig. 3a, b). Mercury concentrations are low in **the river** and for low salinity **samples which** correspond to recently formed brackish waters. Then, we observe a **steep increase** of Hg concentrations, from 5 pM to more than 13 pM, with higher salinity (above 5) that shows the $(Hg_T)_D$ enrichment in the mixing zone, away from the delta mouth.

Fig. 3. Distribution of Hg concentrations (a) (Hg_T)_D in the Laptev Sea; (b) (Hg_T)_p in the Laptev Sea; (c) (Hg_T)_D in the Kara Sea; (d) $(Hg_T)_p$ in the Kara Sea.

Concentration of $(Hg_T)_D$ in the deep marine water sample is lower (4.0 pM) and comparable to the two river values (5.4 and 4.5 pM, Table II).

Although scattered, particulate Hg content in the mixing zone is generally higher than in the river samples and increases up to 1.88 mg kg^{-1} in the mixing zone (Fig. 3b).

The dissolved Hg represents the main part of Hg in the Laptev Sea (average 69% for salinities above 5) and this partitioning is easily explained by the lower SPM content in the mixing zone than in the river samples.

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Concentrations of Hg along the salinity gradient are showed in Fig. 3c, d. The distribution patterns for dissolved and particulate Hg are similar to those observed in the Lena Delta and Laptev Sea. Concentrations of $(Hg_T)_D$ are lower than 5 pM at low salinity and increase for higher salinities. In the Kara Sea, $(Hg_T)_D$ reaches up to 12 pM for salinities greater than about 20; whereas in the Laptev Sea, maximum concentrations were observed for intermediate salinities. This difference in the position of the maximum (Hg_T) concentrations relatively to salinity suggests that the observed distribution is not a result of the formation of highly soluble chlorocomplexes as predicted by thermodynamic models (Morel, 1983)

Mercury concentrations in particulate matter are low in the rivers and greater in the mixing zone of the Kara Sea. As for Laptev Sea, $(Hg_T)_D$ in Kara Sea waters represents the major fraction of Hg (average 68%), according their low SPM load.

The deep marine water samples exhibit quite variable Hg concentrations. However, $(Hg_T)_D$ represents always more than 85% of the total Hg in these samples. The relatively high SPM content of some deep samples indicates that the marine end-member is probably not unique and sea-water masses of different age or/and origin may reach the mixing zone.

4.4. RELATION OF HG WITH PHYTOPLANKTON AND OTHER PARTICULATE ORGANIC MATERIAL

The Hg enrichment in particulate matter measured both in the Laptev Sea and in the Kara Sea is due to the well known affinity of Hg to organic matter. The significant positive correlations found between $(Hg_T)_p$ and POC (Fig. 4) are consistent with similar observations made in other marine environment (Lindberg and Harass, 1974; Bartlett and Craig, 1981; Rae and Aston, 1982).

In the Kara Sea, where chlorophyll was measured, the significance level of the correlation between $(Hg_T)_p$ and chlorophyll is better than with POC considered as a whole (Fig. 5). At the time of sampling, the chorophyll content of riverine particles was low; we can conclude that Hg concentrations in SPM are mainly controlled by the mixing of a Hg-poor river material, probably dominated by inorganic components, with a Hgrich marine organic material consisting of autochthonous phytoplankters.

Albeit the incorporation of Hg into particles due to biological uptake would usually result in low dissolved fraction in water, we observed an enrichment of the dissolved Hg fraction (Fig. 3c). This enrichment of dissolved Hg in the mixing zone may result from the input of Hg-rich organic material by the rivers floods and the melting of the ice seacover during the warmer season.

Fig. 4. Relationships between $(Hg_T)_p$ and POC (a) Laptev Sea; (b) Kara Sea.

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Fig. 5. Relationship between $(Hg_T)_p$ and total chlorophyll in the Kara Sea.

During the flood, the steep increase of water discharge takes place due to the melting of snow and to atmospheric precipitation. Large amounts of dissolved and colloidal humic substances as well as POC reach the river waters from surrounding forests, tundra, bogs and lakes (Telang *et al.,* 1991). It is possible that the water masses from the spring flood enriched in organic matter and in Hg (dissolved and particulate) are temporarily trapped in the mixing zone and still influence the carbon and Hg levels at the time of sample collection. Actually, the presence of such "old" water masses, enriched in organic carbon, was shown previously in the southern part of the Laptev Sea by Létolle et al. (1993).

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4.5. MERCURY FLUXES TO THE ARCTIC SHELF

Inputs of Hg from the Lena, Ob and Yenisei Rivers to the Arctic Shelf were calculated based on the average Hg concentrations measured in freshwaters in the present study and on the average annual water and SPM discharges recently re-evaluated (Gordeev *et al.*, 1993). Results of the calculation are shown in Table IV. These estimations of the gross particulate Hg inputs are based on the assumption that our Hg measurements, from September 1991 in the Lena and September 1993 in the Ob and Yenisei, are representative of the yearly Hg concentration averages. For the dissolved fluxes, we took into account the large difference in Hg concentrations between the flood and the dry season. During the period from August to April, we calculated the dissolved Hg fluxes assuming that the water discharge constitutes 40% of the total annual discharge; taking for $(Hg_T)_D$ the average of our measurements in the rivers. For the flood period (May-July), which flushes about 60% of the total water discharge, (Hg_T) concentrations used in the calculations for the three rivers were taken as three times those of the dry period. This estimation was based on the extrapolation to zero salinity of the dilution line between Hg concentration and salinity (7.1 - 32.6) obtained in the Laptev Sea (Fig. 3a).

	(Hgr)D (10^3 kg a^{-1})	(Hgr)p (10^3 kg a^{-1})	Hg Total (10^3 kg a^{-1})
Lena River	1.15	2.9	4.0
Ob River	0.53	0.82	1.3
Yenisei River	0.41	0.31	0.7
Total Laptev Sea [*]	1.6	4.1	5.7
Total Kara Sea*	1.3	1.7	3.0
Total Eurasian Arctic Rivers* Atmospheric deposition **	3.9	11.3	15.2
Laptev Sea			2.0
Kara Sea			2.6 .

TABLE IV Mercury fluxes to the Arctic Shelf

* Extrapolations on the basis of Hg fluxes of Lena, Ob and Yenisei rivers and average total annual water and SPM discharges from Eurasian Arctic rivers (Gordeev *et al.,* 1993).

** Surface areas: 662 10³ km² for the Laptev Sea and 883 10³ km² for the Kara Sea (Gorshkov, 1983).

Results were extrapolated to estimate the gross Hg inputs to the Laptev and the Kara Sea, and the total Hg flux from all Eurasian Arctic rivers which reaches 15.2×10^3 kg a^{-1} .

The direct Hg atmospheric deposition to the Laptev Sea and the Kara Sea was estimated using the lowest wet deposition rates of total Hg recently measured in northern Norway (3 μ g m⁻² a⁻¹) (Iverfeldt, 1991). Direct atmospheric inputs of Hg appear to be of **the same order of magnitude as those from the rivers (Table IV). Mercury evasion from sea surface remains to be assessed.**

Mercury concentrations measured in summer in these three Siberian rivers are among the lowest ever measured in freshwaters. However, according to the elevated Hg concentrations measured in the mixing zones, Hg concentrations in the rivers during flood are probably much higher. These Arctic estuarine systems are influenced by anthropogenic inputs originating from atmospheric deposition; most of the Hg being released during flood time.

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