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# $^{228}$Ra and $^{226}$Ra in the Kara and Laptev seas

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**Abstract**

The surface water in the Transpolar Drift in the Arctic Ocean has a strong signature of $^{228}$Ra. In an earlier study of $^{228}$Ra in the open Arctic we showed that the major $^{228}$Ra source had to be in the Siberian shelf seas, but only a single shelf station was published so far. Here we investigate the sources of this signal on the Siberian shelves by measurements of $^{228}$Ra and $^{226}$Ra in surface waters of the Kara and Laptev Sea, including the Ob, Yenisey and Lena estuaries.

In the Ob and Lena rivers we found an indication for a very strong and unexpected removal of both isotopes in the early stage of estuarine mixing, presumably related to flocculation of organic-rich material. Whereas $^{226}$Ra behaves conservatively on the shelf, the distribution of $^{228}$Ra is governed by large inputs on the shelves, although sources are highly variable. In the Kara Sea the maximum activity was found in the Baydaratskaya Bay, where tidal resonance and low freshwater supply favour $^{228}$Ra accumulation. The Laptev Sea is a stronger source for $^{228}$Ra than the Kara Sea. Since a large part of Kara Sea water flows through the Laptev Sea, the $^{228}$Ra signal in the Transpolar Drift can be described as originating on the Laptev shelf.

The combined freshwater inputs from the Eurasian shelves thus produce a common radium signature with a $^{228}$Ra/$^{226}$Ra activity ratio of 4.0 at 20% river water. The radium signals of the individual Siberian rivers and shelves cannot be separated, but their signal is significantly different from the signal produced on the Canadian shelf (Smith et al., in press). In this respect, the radium tracers add to the information given by Barium. Moreover, with the 5.8 year half-life of $^{228}$Ra, they have the potential to serve as a tracer for the age of a water mass since its contact with the shelves.


*Keywords:* Polar oceanography; Radium isotopes; Tracers; Sediment–water exchanges

## 1. Introduction

$^{226}$Ra and $^{228}$Ra are produced in sediments by their particle-reactive parents $^{230}$Th and $^{232}$Th, respectively. The primordial isotope $^{232}$Th is a constituent of the Earth's crust. $^{232}$Th in marine sediments is primarily of detrital continental origin

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with concentrations varying within the range of 6–24 mg $kg^{-1}$ in clays (1.5–6 dpm $g^{-1}$, Huh and Kadko, 1992). $^{230}$Th, on the contrary, is not as equally distributed. It is produced by $^{234}$U in seawater and reaches its highest activities in deep-sea sediments. As a consequence of parent distribution and of half life, the oceanographic distribution of $^{226}$Ra (1600 years half-life) is primarily controlled by release from deep-sea sediments (Broecker et al., 1967), whereas the distribution of $^{228}$Ra (5.8 years half-life) is characterized by the high activities that can be reached in the shallow water column over wide continental shelf areas (Moore, 1981; Key et al., 1985). $^{228}$Ra is therefore used as a tracer for continental shelf influence in the open ocean (Moore et al., 1986, 1995; Moore, 1987; Li et al., 1980; Rutgers van der Loeff, 1994; Rutgers van der Loeff et al., 1995; Ku et al., 1995).

Freshwater discharges constitute additional radium inputs into coastal seas. Radium is strongly adsorbed on riverine sediment and released to the dissolved state at increasing ionic strength at mid-salinities (Li et al., 1977; Elsinger and Moore, 1980; Moore, 1981; Key et al., 1985). The riverine contribution to $^{226}$Ra in the surface ocean may be as high as 30% (Li et al., 1977). This contribution is much lower for $^{228}$Ra, for which isotope the relative contribution of fluxes from the estuarine and shelf sediments is much more important than for $^{226}$Ra (Moore, 1981, 1992). $^{226}$Ra sources from groundwater discharge as reported by Moore (1996) have, to our knowledge, not been quantified for Siberian rivers. We expect them to be limited by permafrost as has been argued in case of the budget of the related element Barium (Guay and Falkner, 1998).

The release rate of $^{228}$Ra from continental shelf sediments depends on the deep mixing of these sediments. By molecular diffusion alone, only $^{228}$Ra produced in the upper about 8 cm is released (Huh et al., 1987). The high $^{228}$Ra release rate of Amazon estuarine sediments is attributed to the deep intensive mixing down to approximately 1 m (Kuehl et al., 1982). As a consequence, the $^{228}$Ra enrichment or the $^{228}$Ra/$^{226}$Ra ratio at intermediate salinities may have values specific for different estuaries or shelf regions. Although these values may be quite variable on a time scale of weeks (Moore et al., 1995), they may, averaged over a longer time scale, enable a discrimination between various sources of shelf water in the open ocean (Moore, 1992).

In a previous study in the open Arctic Ocean, we observed that Arctic surface water carries a very strong $^{228}$Ra signal. A single radium data point from the Laptev shelf was strong support for the explanation that $^{228}$Ra accumulates on the wide Siberian shelves, to be subsequently advected in the Transpolar Drift towards Fram Strait. The $^{228}$Ra decay has the potential to yield the transit time of shelf waters in the central Arctic ocean (Rutgers van der Loeff et al., 1995). On the other hand, if the various shelf regions produce a different $^{228}$Ra enrichment, the radium signal might be used to distinguish the source regions of fresh water in the open Arctic. In any case, $^{228}$Ra is a powerful tracer for shelfwaters in the Arctic Ocean.

This study is meant to identify the sources of $^{228}$Ra in the Kara and Laptev shelves. In 1993, Radium samples from surface waters were collected during expeditions with R.V. *Dmitriy Mendeleev* to the Kara Sea, and with R.V. Polarstern to the Laptev Sea. In order to explain the unexpected signals at low salinities in the Ob estuary, additional samples were collected in the Lena estuary in 1999. We present the distributions of $^{228}$Ra and $^{226}$Ra in both shelf seas, discuss the relative influence of inputs from the shelf itself and from the Ob, Yenisey and Lena rivers, and investigate whether $^{228}$Ra can be used as a tracer to distinguish sources from the Laptev and Kara seas.

## 2. Material and methods

Surface water samples were collected on two simultaneous expeditions in August/September 1993: The Kara Sea was sampled during cruise 49 of R.V. *Dmitriy Mendeleev*, which also included stations in the estuaries of Ob and Yenisey. The Laptev Sea was sampled on board R.V. *Polarstern* (expedition ARK IX/4). Three more samples were collected in the Lena River and estuary in September 1999. Station positions are given in

Fig. 1, and further details on the expedition can be found in the cruise report of ARK IX/4 (Fütterer, 1994) and in the special issue of Oceanology (English translation, 34(5), 1995) for the expedition with *Dmitriy Mendeleev*.

Dissolved radium was isolated by pumping several hundred liters of surface water over $MnO_2$-coated 1-μm pore size polypropylene cartridges, after passing an uncoated cartridge as prefilter, and leaching according to Rutgers van der Loeff and Berger (1993). Radium was isolated from the $MnO_2$-cartridge leaches as $BaSO_4$ precipitate, and after ingrowth of $^{226}$Ra daughters, the $^{228}$Ra /$^{226}$Ra activity ratio was determined by gamma spectroscopy. The 186, 295, 351 and 609 keV lines were used for $^{226}$Ra, assumed to be in secular equilibrium with its daughters $^{214}$Pb and $^{214}$Bi (Moore, 1984; Moore et al., 1985), and the 911 keV line for $^{228}$Ra. In order to obtain the absolute collecting efficiency of Ra, $^{226}$Ra was measured on separate 20-L samples of filtered seawater after coprecipitation with $BaSO_4$ and gamma spectroscopy (Rutgers van der Loeff and Moore, 1999). The efficiency of the well-type detector for the various radium decay lines was determined with radium isotopes in standard sediments (DL1A).

$\delta^{18}$O data presented here for the *Polarstern* ARK IX/4 and *Dmitriy Mendeelev* expeditions have been published by Frank (1996) and Ekwurzel (1998), respectively. The percentage river water was calculated from salinity and $\delta^{18}$O according to the 3-component model of Östlund and Hut (1984) using the end-member compositions as given by Bauch (1994). However, as the various Siberian rivers have different $\delta^{18}$O concentrations (Létolle et al., 1993), we have adjusted the $\delta^{18}$O values of the freshwater end-members: For the Kara Sea we use the value of −16.7‰ obtained for the freshwater part of the rivers Ob and Yenisey (Ekwurzel,

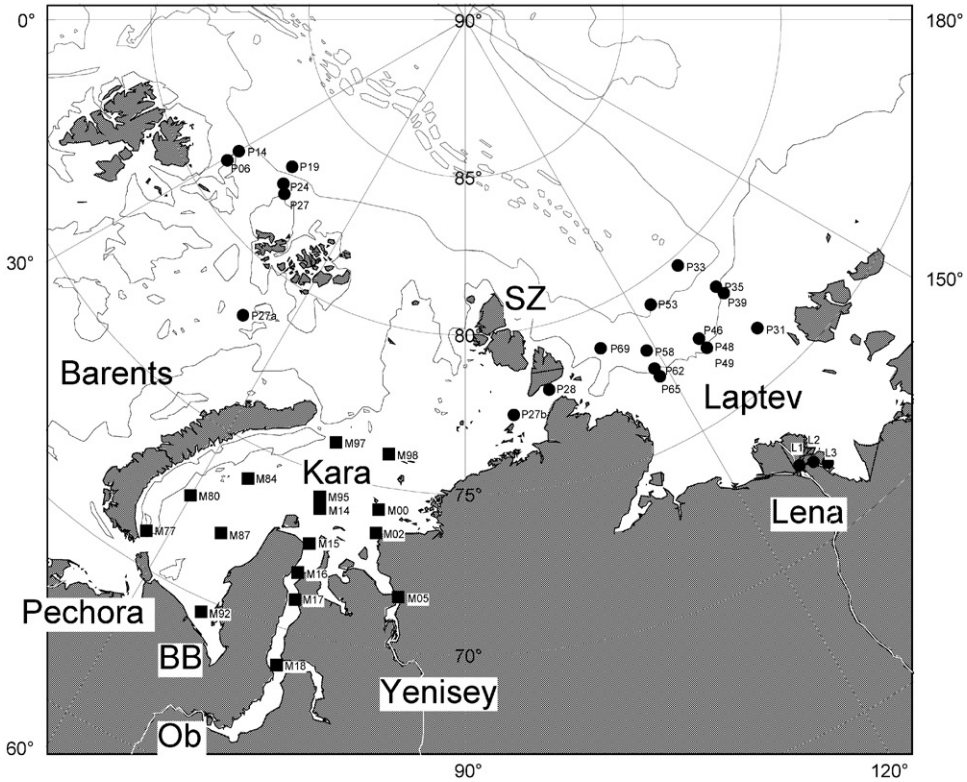


Fig. 1. Sampling stations of expeditions with R.V. *Dmitriy Mendeleev* (squares, M), R.V. *Polarstern* (P) and in the Lena estuary (L). SZ: Severnaya Zemlya, BB: Baydaratskaya Bay.

1998; Ekwurzel et al., 2001). For the Lena estuary and open ocean we use after Létolle et al. (1993) a $\delta^{18}O$ value of −21‰. Due to variability in $\delta^{18}O$, the 3-component model gives unrealistic river water percentages at very low salinities. Below a salinity of 10‰, river water percentage was therefore calculated from salinity alone.

## 3. Results

### 3.1. $^{228}Ra/^{226}Ra$ ratios

The $^{228}Ra/^{226}Ra$ activity ratios (AR) in samples with salinity >23‰ in the Barents Sea, western Kara Sea and Laptev Sea are correlated with salinity (Table 1, Fig. 2), in agreement with the relationship found earlier for surface waters in the central Arctic (Rutgers van der Loeff et al., 1995). This relationship represents a conservative mixing of an open-ocean end-member with low AR (around 0.2 in Atlantic inflow, Rutgers van der Loeff et al., 1995) and a low-salinity end-member with a source of $^{228}Ra$ and correspondingly high AR. In the southern and eastern Kara Sea and in the Vilkitsky Strait, the sea strait between Kara Sea and Laptev Sea, the AR values are also correlated with salinity, but at an approximately 50% reduced level. Two samples were taken in bottom waters of the Kara Sea. The bottom water sample at station M84 has clearly higher AR than expected from its salinity. The highest AR of 3.9 is measured at station P31 in the Laptev Sea at a salinity of 27.1. Below a salinity of 20‰, or at a percentage of river water over about 30%, the AR levels off (Figs. 2 and 3).

### 3.2. $^{226}Ra$ activity

Absolute $^{226}Ra$ activities show a clear decrease with decreasing salinity down to a salinity of 0.1 ‰ or with increasing river water percentage (Fig. 4a). This trend can be described as a conservative mixing of an Atlantic/Arctic end-member with 70 dpm $m^{-3}$ (Broecker et al., 1976; cf. Rutgers van der Loeff et al., 1995) and a freshwater end-member of 28 dpm $m^{-3}$. Only the purest river water displays a sudden increase up to 131 dpm $m^{-3}$ in the Lena and 273 dpm $m^{-3}$ in the Ob ($S = 0.01$). We have no explanation for the anomalously low value observed at station P39.

### 3.3. $^{228}Ra$ activity

As $^{228}Ra$ activities are calculated from the $^{228}Ra/^{226}Ra$ activity ratio and $^{226}Ra$ activities, the scatter in the $^{228}Ra$ data (Fig. 4b) is relatively large. $^{228}Ra$ has a strong source on the shelf with a maximum activity of 240 dpm $m^{-3}$ at a salinity of 27–29‰ (around 20% river water), reached both in the Lena estuary (station P31) and in the Baydaratskaya Bay in the SW Kara Sea (station M92). Below this salinity, $^{228}Ra$ decreases gradually to a freshwater value of 42 dpm $m^{-3}$ at a salinity of 0.1‰. As with $^{226}Ra$, high activities are observed in the freshest water with values up to 249 dpm $m^{-3}$ in the Lena and 409 dpm $m^{-3}$ in the Ob.

## 4. Discussion

### 4.1. The estuarine behaviour of $^{226}Ra$

Dissolved $^{226}Ra$ in world rivers varies between 20 and 300 dpm $m^{-3}$ (Cochran, 1992). The value observed in the Ob (273 dpm $m^{-3}$) is at the high end of this range. The best studied river concerning radium isotopes, the Amazon, has a concentration of 65 dpm $m^{-3}$. $^{226}Ra$ is released from particles to the dissolved phase during early estuarine mixing of the Amazon, yielding a maximum dissolved $^{226}Ra$ concentration approaching 250 dpm $m^{-3}$ at a salinity of about 20‰ (Key et al., 1985). Part of this release may originate in shelf sediments, but the authors estimate that most is due to desorption of riverine particulate material.

The estuarine behaviour of radium in the Ob and Lena rivers is very different from the observations in the Amazon. In Ob and Lena, $^{226}Ra$ disappears suddenly at the very first estuarine mixing. The analogy with the Lena and Ob rivers suggests that the salinity of our freshest sample from the Yenisey River (M05: 0.97‰) was

Table 1
Sampling locations. water depth. salinity. analytical results with 1-sigma counting error. $\delta^{18}O$ and percentage river water ($f_r$) for Polarstern (*P*). Dmitriy Mendeleev (M) and Lena (L) stations. All samples are surface water except B: bottom water

| Station | Name | Longitude °E | Latitude °N | Water depth (m) | Salinity (‰) | $^{228}Ra/^{226}Ra$ AR | $^{226}Ra$ (dpm m$^{-3}$) | $^{228}Ra$ (dpm m$^{-3}$) | $\delta^{18}O$ | $f_r$ |
|---|---|---|---|---|---|---|---|---|---|---|
| *R.V. Polarstern ARK IX-4* | | | | | | | | | | |
| PS27/006 | P06 | 30.60 | 81.25 | 186 | 33.57 | 0.78±0.02 | 68.9±3.9 | 56.0±3.7 | 0.05 | 1.5 |
| PS27/014 | P14 | 30.29 | 81.81 | 2750 | 32.80 | 0.56±0.03 | 90.5±4.5 | 53.1±3.6 | −0.05 | 2.1 |
| PS27/019 | P19 | 40.23 | 82.91 | 2995 | 33.50 | 0.49±0.02 | 63.8±4.6 | 32.7±2.7 | −0.03 | 1.8 |
| PS27/024 | P24 | 42.04 | 82.19 | 1005 | 33.30 | 0.59±0.02 | 70.7±4.1 | 43.9±3.1 | −0.02 | 1.9 |
| PS27/027 | P27 | 43.57 | 82.03 | 287 | 33.50 | 0.53±0.02 | 85.5±4.2 | 47.9±3.0 | 0.06 | 1.5 |
| | P27a | 53.00 | 78.30 | 230 | 31.70 | 0.86±0.05 | 74.3±4.3 | 66.4±5.6 | −0.25 | 3.3 |
| | P27b | 96.83 | 77.50 | 100 | 26.00 | 1.94±0.08 | 44.4±3.9 | 89.6±8.6 | −6.44 | 41.8 |
| PS27/028 | P28 | 102.58 | 78.00 | 165 | 27.99 | 1.68±0.12 | 61.0±4.0 | 106.4±10.2 | −3.24 | 16.5 |
| PS27/031 | P31 | 133.33 | 76.60 | 38 | 27.10 | 3.86±0.25 | 59.8±3.9 | 239.1±21.7 | −4.39 | 22.0 |
| PS27/033 | P33 | 130.55 | 79.78 | 3420 | 32.38 | 1.42±0.04 | 77.3±3.8 | 113.5±6.5 | −1.36 | 7.8 |
| PS27/035 | P35 | 133.08 | 78.46 | 2104 | 32.59 | 1.41±0.05 | 81.4±4.1 | 121.4±7.7 | −1.10 | 6.6 |
| PS27/039 | P39 | 133.52 | 78.12 | 533 | 32.17 | 1.82±0.07 | 33.9±3.0 | 65.2±6.3 | −1.29 | 7.5 |
| PS27/046 | P46 | 125.90 | 77.56 | 1740 | 32.53 | 1.53±0.06 | 79.6±3.9 | 128.7±8.0 | | |
| PS27/048 | P48 | 126.37 | 77.16 | 536 | 31.63 | 1.87±0.13 | 69.1±3.8 | 133.4±12.0 | −1.46 | 8.3 |
| PS27/049 | P49 | 126.17 | 77.15 | 952 | 31.39 | 2.03±0.10 | 72.3±3.6 | 151.6±10.7 | −1.33 | 7.8 |
| PS27/053 | P53 | 122.88 | 79.28 | 3239 | 33.74 | 0.56±0.03 | 73.0±4.7 | 43.6±3.6 | −0.41 | 3.3 |
| PS27/058 | P58 | 118.57 | 78.00 | 1930 | 33.48 | 0.94±0.05 | 62.4±3.6 | 61.8±4.8 | −0.58 | 4.1 |
| PS27/062 | P62 | 118.18 | 77.48 | 542 | 32.95 | 1.41±0.04 | 72.7±4.0 | 105.8±6.6 | −1.15 | 6.7 |
| PS27/065 | P65 | 118.70 | 77.22 | 102 | 32.76 | 1.51±0.12 | 79.2±4.3 | 126.9±11.8 | | |
| PS27/069 | P69 | 112.52 | 78.84 | 581 | 32.68 | 1.27±0.05 | 62.4±3.5 | 83.9±5.6 | −1.13 | 6.7 |
| *R.V. Dmitriy Mendeleev 49* | | | | | | | | | | |
| 4377 | M77 | 58.01 | 71.20 | 239 | 24.78 | 1.91±0.12 | 78.8±4.3 | 156.8±13.2 | −1.24±0.06 | 10.8 |
| 4380 | M80 | 60.00 | 72.77 | 92 | 31.26 | 1.88±0.18 | 70.3±3.2 | 138.0±14.3 | −1.43±0.00 | 10.5 |
| 4384 | M84B | 64.58 | 74.13 | 94 | 34.13 | 1.64±0.08 | 90.4±4.3 | 154.4±10.5 | | |
| 4384 | M84 | 64.58 | 74.13 | 94 | 10.60 | 2.41±0.13 | 28.1±3.2 | 70.6±8.8 | | |
| 4387 | M87 | 64.58 | 72.00 | 150 | 30.94 | 2.51±0.17 | 85.5±4.5 | 224.6±19.0 | | |
| 4392 | M92 | 65.95 | 69.66 | 27 | 28.86 | 3.31±0.20 | 68.2±3.0 | 237.3±17.7 | −2.20±0.03 | 15.2 |
| 4395 | M95 | 73.00 | 74.27 | 31 | 23.81 | 2.15±0.16 | 61.4±3.6 | 138.0±13.2 | −5.85±0.01 | 35.3 |
| 4397 | M97 | 73.01 | 76.19 | 140 | 14.87 | 1.11±0.22 | 48.8±3.5 | 56.3±11.9 | −9.19±0.04 | 50.4 |
| 4398 | M98 | 79.97 | 76.00 | 55 | 23.59 | 2.70±0.23 | 57.0±4.0 | 161.7±17.8 | −5.08±0.01 | 29.8 |
| 4400 | M00B | 80.00 | 74.30 | 35 | 32.75 | 1.33±0.10 | 87.4±3.8 | 121.9±10.4 | −1.95±0.06 | 12.4 |
| 4400 | M00 | 80.00 | 74.30 | 35 | 24.46 | 2.24±0.23 | 56.1±4.0 | 132.0±16.7 | −5.09±0.03 | 30.3 |
| 4402 | M02 | 79.99 | 73.61 | 38 | 9.45 | 2.69±0.26 | 45.8±4.1 | 128.9±17.1 | −13.43±0.03 | 72.9 |
| 4405 | M05 | 83.45 | 71.77 | 17 | 0.97 | 2.36±0.15 | 32.7±3.4 | 81.3±9.9 | −16.74±0.01 | 97.2 |
| 4414 | M14 | 73.50 | 74.00 | 26 | 16.88 | 2.26±0.16 | 46.1±3.8 | 108.6±11.8 | −9.18±0.05 | 62.0 |
| 4415 | M15 | 73.42 | 72.91 | 25 | 8.19 | 2.32±0.32 | 40.1±4.1 | 96.8±16.6 | −12.60±0.07 | 76.5 |
| 4416 | M16 | 73.16 | 71.90 | 20 | 1.29 | 1.68±0.24 | 23.9±3.7 | 45.8±9.7 | −15.77±0.03 | 96.3 |
| 4417 | M17 | 73.57 | 71.14 | 25 | 0.03 | 1.39±0.18 | 26.3±4.8 | 41.8±9.5 | −16.13±0.02 | 99.9 |
| 4418 | M18 | 73.68 | 68.99 | 14 | 0.01 | 1.44±0.13 | 273.1±6.1 | 409.4±37.0 | | 100.0 |
| *Lena 1999* | | | | | | | | | | |
| Lena 1 | L1 | 72.21 | 126.32 | 1 | 0.08 | 1.33±0.04 | 85.2±3.9 | 118.8±6.5 | | 99.8 |
| Lena 2 | L2 | 72.16 | 127.52 | 1 | 0.07 | 1.74±0.09 | 60.9±3.1 | 110.9±8.0 | | 99.8 |
| Lena 3 | L3 | 72.01 | 129.08 | 1 | 0.21 | 1.81±0.03 | 131.1±4.1 | 248.9±9.0 | | 99.4 |

sufficient to remove radium from solution (Fig. 4) but we have no data from lower salinity to prove this. We observed removal by flocculation even in our acidified filtered samples from Ob stations M17 and M18 (Table 2). These lowest salinity samples developed a precipitate, which we

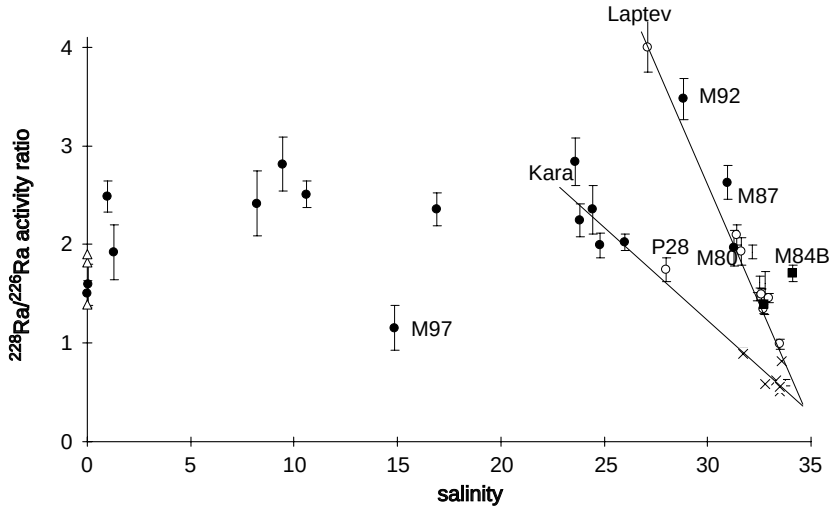


Fig. 2. $^{228}$Ra/$^{226}$Ra activity ratio as function of salinity for all surface water samples from Kara Sea (●), Laptev Sea (○), Lena estuary (△), and Barents Sea and Spitsbergen area (×). Bottom water samples from Kara Sea (■) indicated by B. Approximate mixing lines for Laptev and Kara Sea drawn by eye. M92: Baydaratskaya Bay; P28: Vilkitsky Strait.

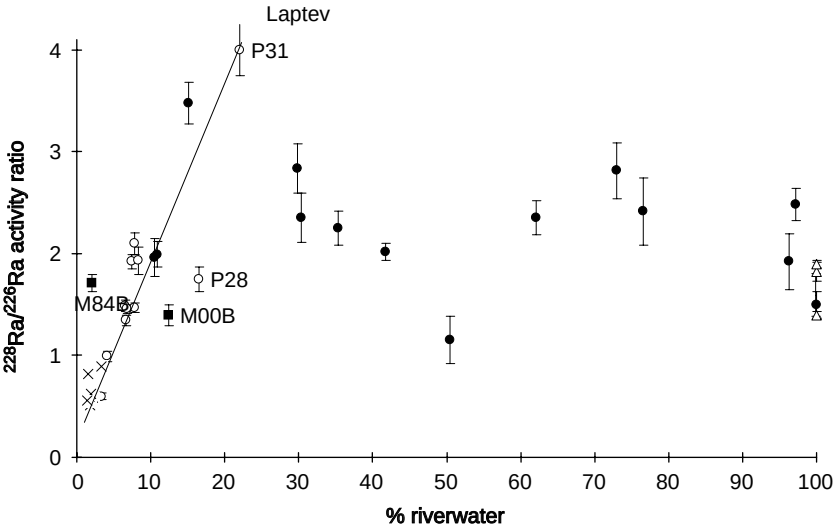


Fig. 3. $^{228}$Ra/$^{226}$Ra activity ratio as function of river water percentage. Symbols as in Fig. 2.

collected before making the $BaSO_4$ precipitate for $^{226}$Ra co-precipitation, and the two precipitates were counted separately. Twenty-eight percent of $^{226}$Ra in the freshwater sample (M18, $S = 0.01$) was in the first precipitate, showing the effectiveness of this scavenging process. Apparently, a flocculation in the early stages of estuarine mixing in the Ob and Lena rivers causes an effective removal of radium from solution.

A similar non-conservative behaviour at very low salinities has also been observed for barium, a chemical analog of radium. A parallel sample from station M18 collected for barium analysis was the only sample filtered, 3 years after collection, because it "contained a visibly high amount of suspended sediment" (Guay and Falkner, 1998). Flocculation may have removed barium from solution in this sample, which would explain why

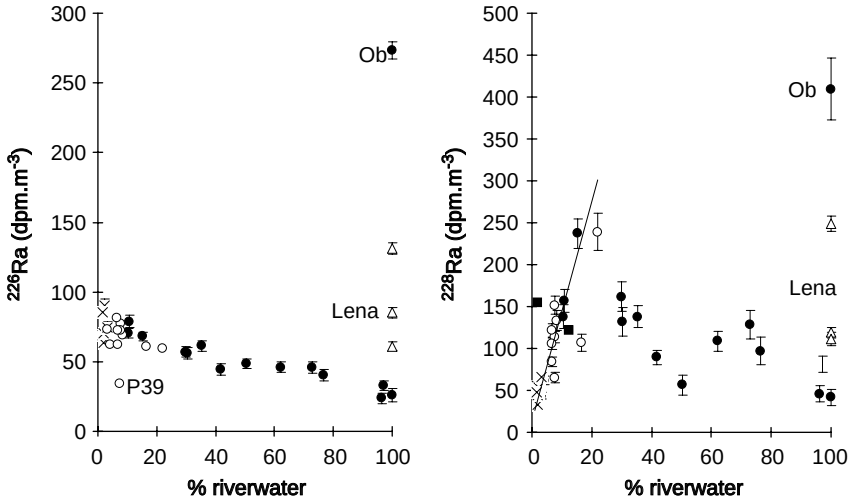


Fig. 4. Activity of $^{226}$Ra (a) and $^{228}$Ra (b) against river water percentage. Symbols as in Fig. 2.

Table 2
$^{228}$Ra in flocculated material collected from acidified filtered freshwater samples compared with remaining dissolved $^{228}$Ra collected as $BaSO_4$ precipitate

| Station | Salinity (‰) | $^{226}$Ra (dpm m$^{-3}$) | | |
|---|---|---|---|---|
| | | Flocculated | Dissolved[a] | Total |
| M17 | 0.03 | 3.6 | 22.7 | 26.3 |
| M18 | 0.01 | 73.4 | 199.6 | 273.1 |

[a] Collected by coprecipitation with $BaSO_4$.

the distribution of barium in the Ob estuary during this same expedition (Fig. 9 in Guay and Falkner, 1998) does not display the early removal we observed for its chemical analog radium (Fig. 4). A second sampling program 1 year later did show a large decrease in barium as salinity increased from near-zero to 1 (Guay and Falkner, 1998), explained by the authors by biological uptake, although they do not rule out the possibility of removal on oxyhydroxides (Coffey et al., 1997).

Flocculation of Fe, Mn and humic substances is well known in many estuaries (Sholkovitz, 1976; Boyle et al., 1977; review in Salomons and Förstner, 1984). It is due to the destabilization of organic colloids by seawater (Boyle et al., 1977) or can be directly linked to the redox cycle of Mn (Sundby et al., 1981). Estuarine removal of many particle-reactive elements has been explained by their association with this flocculating material, but Santschi et al. (1983) did not observe such a removal of radium in their microcosm experiments.

We do not know whether the flocculation in the Ob and Lena estuary is primarily organic or of an Fe- or Mn-oxyhydroxide type. The suspended load is low compared to most other world rivers including the Amazon (Lisitzin, 1972; Milliman and Meade, 1983, Rachold et al., 1996). The estuarine behaviour of trace metals has been studied in both estuaries. Removal of iron is observed during estuarine mixing in the Lena (Guieu et al., 1996) and in the Ob and Yenisey rivers (Dai and Martin, 1995). The latter study showed how flocculation of colloids removes colloidal Fe and carbon in the estuary. But this removal takes place in the salinity range until about 20‰, very different from the dramatic early removal of radium. We interpret this as an argument against iron oxides as the major phase of radium removal. In this respect Ra behaves similar to Osmium, which has also been shown to be removed at lower salinities than Fe (Levasseur et al., 2000). The flocculation observed in our acidified filtered samples (Table 2) is another argument against the role of iron oxides. Moreover, a removal on iron oxides would be expected

to cause an efficient scavenging of thorium as well. But Kuptsov et al. (1995), measuring thorium scavenging in the Kara Sea during the same Mendeleev expedition, observed little scavenging in the estuarine zone of the Ob. Appreciable scavenging did not start before sinking out of riverine particles allowed primary production to develop. Estuarine removal of thorium was also studied by Andersson et al. (1995) in Kalixälven, a Swedish river with important peatland contribution. These authors observed a removal of $^{232}$Th at low salinities, which they ascribed to settling of detrital $^{232}$Th and which may be related to the removal of Mn oxides (Pontér et al. 1992). Surprisingly, $^{230}$Th, produced in the peatlands, was removed much more slowly in the estuary, which these authors explained by stabilization as a colloidal phase.

Realizing that the evidence regarding early estuarine removal is ambiguous, we suggest that organic flocculation is the most likely removal mechanism of radium. The Lena (Lara, 1998) and Ob are rich in dissolved organic matter (Table 3) especially through the load of humic substances obtained by drainage of vast tundra and taiga areas (cf. compilation by Ludwig et al., 1996). There is additional evidence for the influence of DOC on nuclide behaviour in the Ob estuary. The adsorption efficiency of Ra to our $MnO_2$-coated cartridges, as derived from comparison with discrete 20-L samples, was only about 3% in the freshwater samples, exceptionally low compared to the 18–30% further downstream. A similar observation was made by Cochran (pers. comm.), who measured low absorption of Pu on $MnO_2$-coated cartridges in the freshwater zone of the Ob River. The adsorption efficiency of the three Lena samples on $MnO_2$-coated cartridges was better at 50–68%, probably as a result of a lower flow rate and smaller filtered volume (only 115–277 L).

Table 3
Organic carbon load of Arctic rivers in comparison with data for St. Lawrence and Amazon

| River | Discharge ($km^3$/yr) | DOC (mg/l) | POC (mg/l) | TOC (mg/l) |
|---|---|---|---|---|
| Organic carbon transport in rivers[a] | | | | |
| Yenisey | 555 | | | 7.4 |
| Lena | 505 | 6.8[b] | 2.4[c] | 9.5[d] |
| Ob | 433 | 9.09 | 0.88 | 10 |
| Pechora | 106 | | | |
| St. Lawrence | 413 | 4.9 | 0.48 | 5.4 |
| Mackenzie | 249 | 4.5 | 3.2 | 7.7 |
| Amazon | 5520 | 4.46 | 2.83 | 7.3 |

[a] Based on compilations by Telang et al., 1991 and Ludwig et al., 1996.
[b] Mean: Lara et al., 1998.
[c] Rachold and Hubberten, 1999.
[d] Telang et al., 1991.

It is interesting to consider what might happen to the radium removed from solution. It should be on particles or flocs large enough to be caught by our 1 μm cartridge filters. These particles will be transported and eventually settle out. We may have an indication of the distribution of these particles from the fate of freshwater algae. In sediments of the Laptev Sea, the present distribution of Chlorococcalalgae reaches until the 20‰ isohaline in summer (Kunz-Pirrung, 1998). These algae cannot survive salinities in excess of 8‰, and must be advected seaward through the existing gullies. The radium-enriched organic flocs could have a similar fate. However, an excess of radium in sediments has not been observed by Strobl (pers. comm.). In core PM9463-8 (74.50°N, 126.58°E) he observes a depletion of $^{214}$Bi and consequently of $^{226}$Ra with respect to $^{230}$Th, inferring a release of $^{226}$Ra to the bottom water.

### 4.2. The estuarine behaviour of $^{228}$Ra

In contrast to $^{226}$Ra, the source of $^{228}$Ra is clearly in the mid-salinity range (Fig. 4b), in agreement with the results of Key et al. (1985) for the Amazon. Apart from the two high values of the Ob River and the Lena River, the highest values are reached at a river water percentage of around 20%. Whatever the fate of the river input of radium, we know from the $^{226}$Ra results (Fig. 4a) that riverine Ra is not released at this intermediate salinity. The source of the $^{228}$Ra inputs at mid-salinity is thus from the shelf itself, not a release from riverine particles.

On the seaward side of the shelf, $^{228}$Ra displays a linear relationship with the river water content (Fig. 4b) pointing at conservative behaviour.

Radiodecay can be neglected on this time scale of mixing within the limited area studied here. The maximum $^{228}$Ra activity on the shelf is not related to a specific salinity. As $^{228}$Ra is released by the shelf, we expect that the $^{228}$Ra activity, or the $^{228}$Ra/$^{226}$Ra ratio, changes with the residence time of water over a shallow shelf, not with salinity. In the Kara Sea, the $^{228}$Ra/$^{226}$Ra ratio varies within the limits of 1.6–2.7 ($^{228}$Ra in the range 97–225) outside the Ob River ($>2$‰) with two exceptions (Figs 2 and 3). The maximum value (AR = 3.31, $^{228}$Ra = 237 dpm m$^{-3}$) is found at station M92 in the Baydaratskaya Bay in the SW Kara Sea. This Bay has little river input, but tidal resonance results here in the strongest tidal currents of the Kara Sea (Harms and Karcher, 1999). This situation favours a long residence time and strong resuspension over a shallow bay, an ideal situation for the accumulation of $^{228}$Ra. The lowest value (AR = 1.11; $^{228}$Ra = 56 dpm m$^{-3}$) is from the northern most station M97, closest to the continental shelf boundary. This sample has a river water percentage of 50% and appears to consist of a mixture of river and ocean water with very little shelf input. The neighbouring station M98 has more than twice this activity ratio. Thus, the data do not yield a single $^{228}$Ra/$^{226}$Ra ratio that might be typical for the Kara Sea. Water may thus reach the shelf edge with widely different shelf signatures.

In spring and summer there is much northward transport of river water to the central Kara Sea (Johnson et al., 1997; Harms and Karcher, 1999), and part of the river water will find its way to the shelf edge and the central Arctic through this route north of Severnaya Zemlya. But model studies of Harms and Karcher (1999) and Harms et al. (2000) show that the annual transport is governed by the eastward circulation prevailing in autumn and winter. The conclusion of an eastward circulation of Arctic river water, the traditional view of the circulation pattern, is shared by studies of river water distributions by Ekwurzel et al. (2001) and Mensch et al. (Unpublished manuscript). We conclude that a large part of the river inputs leaves the Kara Sea through the Vilkitsky Strait. The AR at the Vilkitsky Strait (1.68 at a river water percentage of 16.5%) marks the radium isotope composition of this major water input into the Laptev Sea.

In the Laptev Sea, and besides three freshwater samples, our data set is limited to salinities over 27 ‰. Here all $^{228}$Ra data can be rather well described by a single conservative mixing line between an ocean component with an AR around 0.15 in the Atlantic Inflow (Rutgers van der Loeff et al., 1995) and a low-salinity (shelf edge) component characterised by station P31 with a AR of 3.9 at a river water percentage of 20% (Figs. 2 and 3). The conservative behaviour implies that sources are negligible in the salinity range investigated, and that the major source of $^{228}$Ra is on the freshwater side of our sampling area. Indeed, except for station P31 (38 m depth), all samples are from water depths of 100 m or more where the $^{228}$Ra inputs from the sediment are dispersed in water layers in or below the halocline. Density stratification prevents such inputs to reach the surface layer. Although the sources of $^{228}$Ra and river water are far apart, they are both on the continent side of the shelf edge. On the sea side, $^{228}$Ra in surface waters closely follows the distribution of river water. In deeper water layers, sources on the slope may also contribute to the $^{228}$Ra signal (Rutgers van der Loeff et al., 1995).

As in the Kara Sea, the source of $^{228}$Ra must clearly be on the shelf. The $^{226}$Ra data for the Lena River suggest that the behaviour of radium during early estuarine mixing is similar to that in the Ob River, i.e. a removal of dissolved Ra rather than a release from riverine particles. The strong source of $^{228}$Ra apparent from the samples in the high salinity range ($>27$‰) must thus be produced on the shelf at intermediate salinities where shelf water residence times are sufficient to accumulate $^{228}$Ra from the shelf sediments (0.2–27‰; Fig. 4b).

## 5. The possible applications of $^{228}$Ra as tracer in the Arctic ocean

$^{228}$Ra closely follows a conservative mixing line off the shelf edge of the Laptev Sea. The distribution can be described as originating from a mixed Siberian continental shelf and river signal entering the Transpolar Drift, in complete

agreement with the Radium data from the central Arctic basin (Rutgers van der Loeff et al., 1995). As $^{226}$Ra is practically constant at high salinities, the $^{228}$Ra/$^{226}$Ra activity ratio can be used instead of $^{228}$Ra, reducing the analytical error.

Harms et al. (2000) expected from their model of Arctic river flow that the Siberian branch of the Transpolar Drift is dominated by inputs from the Kara sea (especially from the Ob, whose waters have the largest chance to exit the Kara Sea to the north), whereas the Canadian branch is dominated by the Lena. The two sources cannot be distinguished based on their barium content (Guay and Falkner, 1998), but it would be interesting if $^{228}$Ra or the AR would be sufficiently different between the Kara and Laptev Sea to verify this expectation. Indeed, there is a tendency in our data (Fig. 2) that the Laptev Sea is more enriched in $^{228}$Ra than the Kara Sea. However, $^{228}$Ra can be highly variable in the source area on the continental shelf, depending on the vicinity to sources like isolated shallow areas of higher residence time. Water masses with a range of radium signatures can reach the shelf edge depending on the route taken by fresh water parcels towards the open ocean. The longer residence time of Kara Sea water that transits through the Laptev Sea will accumulate more $^{228}$Ra as it crosses those shelf waters and mixes with the Laptev Sea shelf waters before moving off the continental shelf. At present we do not know whether the Kara Sea produces at its shelf edge a radium signal with lower AR values on the Eurasian side of the Transpolar Drift. The sparse $^{228}$Ra data available for the central Eurasian basin do not support that hypothesis. Three stations on a transect across the Transpolar Drift from the Gakkel Ridge to the Makarov Basin (stations 165, 173 and 176 in Rutgers van der Loeff et al., 1995) were on the same mixing line suggesting a homogenous source. We must conclude that it is unlikely that, offshore, water masses from the Kara and Laptev Seas can be distinguished on the basis of their radium signal. The $^{228}$Ra source derived from the Laptev Sea section can be considered to be representative for the joint inputs of Kara and Laptev Sea. Using the concept that water with a constant radium signal is introduced into the Transpolar Drift, Smith et al. (in press) explained their extended data set for radium in the Canadian basin to illustrate the shift of the axis of the Transpolar Drift from the Lomonosov ridge to the Mendeleyev ridge during the early 1990s (Ekwurzel et al., 2001).

Whereas the various Siberian sources cannot be distinguished from each other, there is a clear difference between the Siberian sources and the sources from the Canadian shelf where Smith et al. (in press) found $^{228}$Ra /$^{226}$Ra activity ratios of just 1.0. There exists a similar difference in the source function of Barium with higher concentrations in the Mackenzie River than in any of the Siberian rivers (Guay and Falkner, 1998). Together, these tracers can thus be used to distinguish fresh- and shelfwater sources from the two continents.

In our previous paper (Rutgers van der Loeff et al., 1995) we tentatively used the radioactive decay of $^{228}$Ra to estimate transit times of surface water masses after they detached from the shelf. This method requires first, that the source has a unique concentration. The evidence for a difference between Canadian and Eurasian sources means that the method should be applied only in areas affected by one source alone, like on the Chukchi shelf (Smith et al., in press). Second, the method requires the source to be constant in time. The release of $^{228}$Ra from sediments is mediated by resuspension. Storms or anchor ice formation may cause abrupt and seasonal changes in this release. Moreover, the large change in ice cover and the strong summer fresh water surges influence the residence time of water over parts of the shelf and consequently the local accumulation of $^{228}$Ra. It remains to be investigated whether, averaged over the several-year residence time on the shelf (Östlund, 1993; Ekwurzel, 1988; Harms et al., 2000), the shelf signatures are sufficiently constant in time to allow their use as a clock for shelf waters on the time scale of its decay.

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