8.1 Studies on plutonium speciation and radionuclide concentrations in the Ob and Yenisey estuaries and the Kara Sea under the EU project ESTABLISH

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Background and Aims

The EU Copernicus project ESTABLISH (Estuarine Specific Transport and Biogeochemically Linked Interactions for Selected Heavy metals and radionuclides) focuses on the marine-freshwater interface in the Yenisey Estuary in connection with modelling of the transport of contaminants (heavy metals and radionuclides) from inland to the open sea. This important link received little attention within related studies in the past. However, in order to understand the biogeochemical behaviour and fate of contaminants in Siberian estuarine environments, it is crucial to take this link into account. The three main issues ESTABLISH is dealing with include: (i) Short-term transport processes, conservative and sediment-bound contaminant transport, (ii) Long-term fate of contaminants in sediment deposits, and (iii) Ocean-land interaction and estuarine living resources. ESTABLISH started officially on 1 October 2000, it will last three years. The participating institutions are: The Norwegian Radiation Protection Authority (NRPA, Norway), SPA Typhoon (Russia), the Vernadsky Institute of Geochemistry and Analytical Chemistry (GEOKHI, Russia), the Institute of Oceanography at the University Hamburg (IOH, Germany), the Arctic and Antarctic Research Institute (AARI, Russia) and the Agricultural University of Norway (AUN, Norway).

The main goals in connection with the "Akademik Boris Petrov" cruise in 2001 were collection of samples:

- In the Yenisey to perform process and speciation studies regarding the behaviour of plutonium in the mixing zone
- In the open Kara Sea to have a reference outside the mixing zones
- In the Ob bay to compare the mixing zone processes in the two large Arctic estuaries
- To increase our overall knowledge of levels of radioactive contamination in the Arctic areas

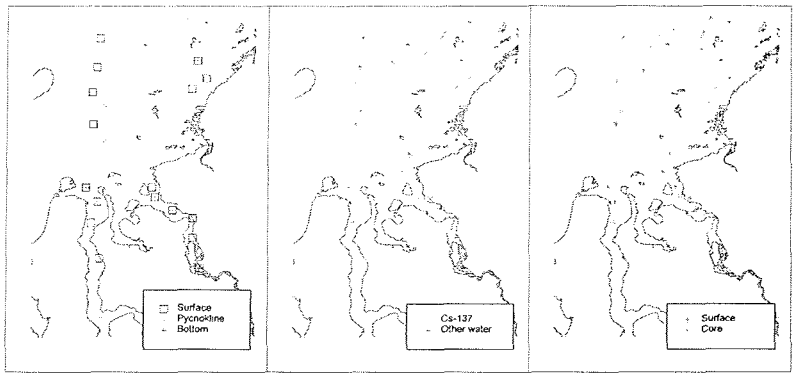
Studies in connection with samples from the "Akademik Boris Petrov" Cruise in 2000 by GEOKHI (Stepanets *et al.* 2001) and others, including the NRPA, were used as background information for the planning of the radioecological work during the cruise in 2001.

Sampling and treatment of samples

An overview on the amount and locations where water and sediment samples were obtained is given in Fig. 8.1(a-c) and Table 8.1.

Station number	Area	^{238,} $^{239+240}$Pu	^{137}Cs	^{90}Sr	^{129}I	^{99}Tc	Sediment cores	Surface sediments
2 – 21	Yenisey	13	9	4	13	4	7	13
1 and 22 – 64	Kara Sea	15	15	5	15	8	10	14
65 – 83	Ob	15	4	3	7	4	5	3

Table 8.1: Samples listed for regions and radionuclides.



a) b) c)

Figure 8.1 (a-c): Sampling localities for radionuclide measurements a) Plutonium, b) ^{137}Cs and other water samples, c) Sediment samples.

Plutonium

Being the main focus of the expedition, a major effort was undertaken to collect plutonium ($^{238, 239+240}$Pu) samples through out the mixing zones in the Yenisey estuary. To get an as complete picture as possible, water samples (200 l each) were collected from the surface, bottom and pycnokline using pumps to collect the surface water and a large-volume water sampler (batomat) for the deeper water. (Tabs. 8.1 & 8.2). Due to the geographically shorter salinity gradient, the high particle load and less time available in the Ob estuary, it was not possible to get a complete supplement in Ob to the samples collected in the Yenisey. In the open Kara Sea, the sampling density was much lower than in the rivers. However, some samples could be collected from the different water bodies in the Kara Sea. To be able to distinguish between the particulate, colloidal and dissolved plutonium (Salbu 2000), the water samples (200 l) were filtrated using a 0.45μm filter and selected samples were ultra filtrated using a 10 kDa tangential flow cassette system (Salbu & Oughton 1994; Mitchell *et al.* 1995). Then, before the alpha counting process, the water samples were reduced in volume by a chemical precipitation procedure and further processing for α-spectrometry (Chen *et*

al. 1991). Following modern radiochemical separation and purification, plutonium and americium activities and isotope ratios will be also determined by means of Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Accelerator Mass Spectrometry (AMS, see e.g. Oughton *et al.* 2000) for certain samples.

Caesium, technetium and strontium

As for plutonium measurements, water samples for measurements of ^{137}Cs (200 – 1000 l), ^{90}Sr (50 l) and ^{99}Tc (100 l) were collected using pumps for surface samples and the batomat for the deep-water samples. (Tab. 8.1). Due to time constraints, most of these samples were from surface water (Tab. 8.2). However, we anticipated achieving the largest possible geographical spread of the sampling. Samples collected for analyses of the conservative behaving radionuclide ^{99}Tc for use in long-range transport studies were not treated. For the measurements of ^{90}Sr water samples were acidified and a stable strontium carrier solution was added. Samples for ^{137}Cs (200 l) were collected by pumping water through a particle filter and an adjacent adsorbent system. The sorbents will be ashed and measured using high-purity germanium (HPGe) gamma detectors.

Sediment samples

Sediment samples were collected as sub-cores from the large box corer (GKG) for studies of vertical distribution of radionuclides (in total 22 cores), or as surface samples from the grab corer (22 samples) to be used for process studies in the laboratory (Tab. 8.1). Some sediment samples were collected to calculate *in situ* K_d values. In addition to those, samples were collected for use in process studies and for general monitoring and mapping of levels of radioactive contamination.

Analytical work

The analytical work is presently in the start-up phase. No samples are analysed yet. Measurements will be undertaken at the laboratories in Norway in Østerås and Tromsø (NRPA), in Ås (AUN), in Canberra (Australia, Australian National University) and at other foreign laboratories. Some first results are planned be reported during the conference on Arctic and Antarctic Radioecology in St. Petersburg, Russia, in June 2002.

	Station	Samples			Surface		Pyknokline	Bottom		
		Su	P	B	T	S	depth	T	S	Depth
Yenisey	3	F			4.5	10.2	5	-0.9	32.2	14.5
	4	uf			13.7	0		12.7	0	19
	8	F		uf	14.4	0		14.3	0	27.4
	11	F		uf	13	0	7.4	5.5	22.4	8.4
	16	F			13	0		13	0	27.3
	19	F	uf		9.6	6.1	3.5	-1.1	32.5	24.1
Kara Sea	28	F			1.5	24.5	18.5	-1.1	33.8	50.6
	30	F		F	2.4	27.5	8	-1.4	34.1	46.1
	34			F	1.4	29.2	19.1	-1.4	34.3	90.7
	41	uf			3.4	23.8	10	-1.1	33.9	35
	46	F			4.2	26.4	18	-0.7	34.9	300
	56	F			5.5	22	12	-0.8	34.5	170
	59			F	5.2	21.9	9.5	-1.4	34.5	170
	62	F			4.8	23.3	7	-1.3	34.3	120
Ob	65		uf		5.9	19	15	-1.4	33.7	52
	67	F			7.4	11.2	5.5	-1.4	33.3	41
	68		uf		7.8	9.9	7	-1.2	32.6	21
	70	F	uf		9.2	0.9	7	-0.5	31.1	16
	72	uf			11.4	0		11.5	0	21
	73			uf	12.5	0		12.5	0	10
	80	F			8.6	0.9	8.7	2.9	22.7	11
	82	uf		uf	6.7	9.9	6.7	-1.3	32.9	22

Table 8.2: Overview of water samples prepared for plutonium measurements (Su: Surface, P: Pyknokline, B: Bottom, F: Filtered (0.45_m), uf: Ultra filtered (10kDa), T: Temperature °C, S: Salinity. Depths in meters).