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Concentration ratios to aquatic plants at and near Olkiluoto repository site

V. Kangasniemi[1], J. Helin[2], T. Kirkkala[1] and A.T.K. Ikonen[2]

[1] *Pyhäjärvi Institute, Sepäntie 7, 27500 Kauttua, Finland*
[2] *Posiva Oy, Olkiluoto, 27160 Eurajoki, Finland*

Abstract. In Finland, Olkiluoto Island on the western coast has been selected as a repository site for spent nuclear fuel disposal. With the approaching licensing steps (application for the nuclear construction licence in 2012), the biosphere assessment demonstrating the long-term safety of the repository is developed into more and more site specific. At the present coastal site, lakes will form in the future millennia due to the post-glacial crustal rebound, i.e. land uplift, which at least eventually will outrun the anthropogenic sea level rise. Both the brackish bays of Baltic Sea and the future lakes can be primary recipients of releases from the deep underground repository, and the aquatic plants can form a major pool of radionuclides with a rather rapid turnover. In some cases the aquatic plants are a relevant part of wildlife food web and possibly also a resource to human. To provide the biosphere assessment models with site-relevant input parameter data, samples of typical aquatic plants were collected from the sea area at the site and from two nearby lakes analogous to those expected to form at the site during the future millennia. This contribution presents water-to-plant concentration ratios of stable elements of high relevance to the biosphere assessment of the Olkiluoto spent fuel repository.

1. INTRODUCTION

In Finland, Olkiluoto Island on the western coast has been selected as a repository site for spent nuclear fuel disposal. With approaching licensing steps (application for nuclear construction licence in 2012), the biosphere assessment demonstrating the long-term safety of the repository is developed into more and more site specific. In the case of the Olkiluoto spent nuclear fuel repository, the nuclides contributing most to the doses of the most exposed persons, and of the other public, were in the recent assessment [1] I-129 and Cl-36, with C-14 having a high contribution in some release scenarios. Other elements (corresponding to the released nuclides) of high relevance include Se, Mo, Ni, Nb and Cs, and possibly Ag. One factor to be taken into account in the safety analyses is the postglacial land uplift (approximately 6 mm/y [2]), which affects the hydrogeochemical and biological systems of Olkiluoto and its surroundings. The properties of the future lake ecosystems surrounding Olkiluoto Island can be forecast and radionuclide transport models applied based on the properties of present lakes. Currently, however, there are no lakes on the island. Lakes of various successional stages were selected within a larger geographical area as analogues of those expected to form at the Olkiluoto site, see [3] and the paper by Haapanen et al. to the present conference.

To initialise the collection of the site-specific data on transfer to aquatic plants, common macrophytes were sampled from the Olkiluoto site and from two lakes chosen to represent those expected to form at the site within a few millennia. Sampling procedures were trained in June 2010, and comprehensive sampling took place in August 2010. More samples were taken in May 2011 but as these were for a longer-term study on the growth rates of the plants, only the August results are yet meaningful to be reported. Water samples were collected with a Limnos sampler from about one metre above the bottom close-by the plant sampling locations (Fig. 1) in March, July and October 2010 at Olkiluoto and in March and August 2010 from the lakes.

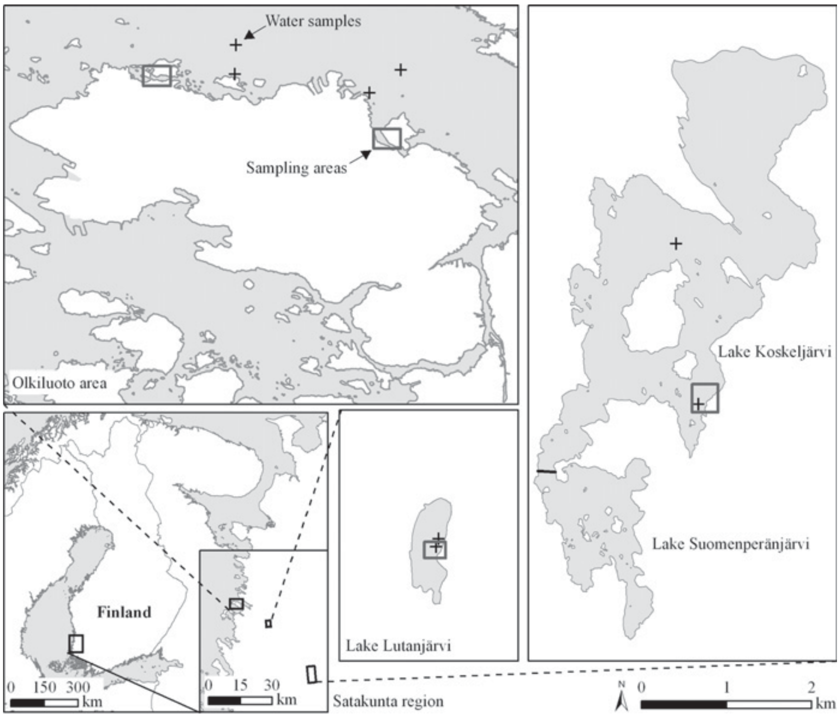

Figure 1. Aquatic plant sampling areas and locations of water samples used in this study. Background map: topographic database by the National Land Survey of Finland, permission no. 41/MYY/11. Map layout by Jani Helin/Posiva Oy.

2. MATERIAL AND METHODS

2.1 Sampling locations

The Olkiluoto area is rather exposed part of the brackish-water Bothian Sea coast (Fig. 1) with a mean depth of less than 10.0 m and the deepest depressions to 15.5 [4]. Two rivers, with a combined mean discharge of 12.0 m^3/s and a catchment area of 1700 km^2, have their mouths in eastern part of the area [5]. The catchment area is covered by clays field, till and rocky forest and mires. From the northern shore of Olkiluoto Island, two sampling areas of reed bed were selected and from each tree quadrats were harvested both above and below waterline. Typically to the coastline, the reed beds were growing on a very stony bottom with muddy sediment between the stones. There were also occasional observations of a sandy layer at a depth of 20 cm as the bottom was probed with a 1" metal tube.

Koskeljärvi is a complex-shaped drainage lake (Fig. 1) of 657 ha with mean depth of 1.2 m (max. 3.2 m). Its catchment area, 65 km^2, is covered by till and rocky forests and mires [5]. In Posiva's biosphere assessment the lake has been chosen to represent an overgrowing, larger lake. An underwater area with three quadrats was selected here, together with sedge area (five quadrats) and a reed bed with three quadrats both above and below waterline. The water at the sedge area was very shallow but the depth was difficult to measure due to the very soft muddy bottom with stones at depths of 30–70 cm. In the reed bed the bottom was of detritus mud in between of the dense root system, occasionally 70–90 cm but mostly over 1 m to the hard bottom (rock or bedrock). The submerged vegetation area was located on a fluffy, muddy bottom.

Lutanjärvi is an oblong headwater lake (Fig. 1) of 40 ha with a mean depth of 1.6 m (max. 6.5 m). Till and rocky forests and clay fields cover most of its 5.0-km^2 catchment area [5]. The lake has been

Table 1. Characteristics of the vegetation sampling areas; the height of dominant vegetation above the water surface is given as arithmetic mean (minimum-maximum, standard deviation, number of observations).

	Water depth, cm	Height of vegetation, cm	Dominant species
Olkiluoto coastal area			
- reed bed, west	0–33	208 (147–285, 42, 18)	*Phragmites australis* [1]
- reed bed, east	0–44	179 (122–258, 43, 18)	*Phragmites australis*
Koskeljärvi Lake			
- reed bed	0–15	216 (184–297, 32, 15)	*Phragmites australis* [2]
- sedge	∼10	83 (-, 17, 10)	*Carex* spp.
- bur-reed	80	0	*Sparganium gramineum*
Lutanjärvi Lake			
- reed bed	∼ 0	210 (160–288, 35, 15)	*Phragmites australis* [3]
- bulrush	0–10	130 (114-150, 13, 6)	*Typha angustifolia* [4]
- bur-reed	100	6 (3-11, 2.4, 10)	*Sparganium gramineum*
- water lily	40	0	*Nuphar lutea*
- pondweed	90	0	*Potamogeton natans*

[1]Other species: *Carex* spp., *Stellaria fennica, Potentilla palustris, Peucedanum palustre, Aegopodium podagraria, Plantago maritima, Cirsium* spp., *Silene* spp., *Myriophyllum alterniflorum*; next to the quadrats also *Filipendula ulmaria, Lysimachia vulgaris, Parnassia palustris.*
[2]Other species: *Carex rostrata, Potentilla palustris, Sphagnum* spp.
[3]Other species: *Potentilla palustris, Calla palustris, Lysimachia vulgaris, Sphagnum* spp., *Carex* spp., *Warnstorfia* spp., *Stellaria palustris.*
[4]Other species: *Carex rostrata, Potentilla palustris, Lysimachia vulgaris, Stellaria fennica, Sphagnum* spp.; next to the quadrats also *Peucedanum palustre.*

chosen as an analogue of a small headwater lake. Two sampling areas were selected from the waterline: a bulrush plot with two quadrats and reed bed area with similar layout to the other reed sampling areas. Moreover, three underwater areas of three quadrats were included in the study. The bottom in the reed bed was clayed mud over 1 m thick with occasional smell of sulphides. The bulrush area was on a soft muddy bottom too, as well as the submerged vegetation quadrats.

All aquatic plant sampling areas, located with expert judgement, represent typical conditions of the water body – their characteristics are presented in Table 1. Water samples were collected within another sampling campaign close by the plant sampling sites (Fig. 1) and additional water samples next to the plant sampling areas were taken from the lakes in August 2010.

2.2 Sampling and pre-treatment

From the sampling quadrats ($0.5 \times 0.5\,m^2$, except in case of the sedge plot of Koskeljärvi a ring of $0.035\,m^2$) all plant material was carefully collected into clean plastic bags. In the laboratory, the samples were separated into different species and dead plant matter that were weighed for their contribution to the biomass. The samples were then further divided into subsamples for laboratory analyses (section 2.3), and for archive, when allowed by the sample size. All plant samples collected above the water surface were rinsed in distilled water in order to remove the possible atmospheric deposition component. In cases of logistical delay in pre-treatment samples were stored in a fridge (only short periods) or a freezer. The prepared samples were all frozen for transport to the analytical laboratory or for archive. Dry matter content (DM) was determined from subsamples by drying at 105°C for at least 48 h.

2.3 Chemical analyses

All plant samples were analysed by the commercial laboratory ALS Scandinavia AB using a total elemental analysis (ICP-SFMS) after drying at 50°C and digestion with HNO_3/HF (trace). Also the water samples were analysed by the same laboratory: The samples of October were filtered using an ACROR 32 Supor™ 0.2-μm filter. The other water samples were analysed unfiltered. The samples were acidified with 1 ml of ultra high-purity nitric acid per 100 ml of sample. For selenium the samples were digested with hydrochloride acid in autoclave (120°C, 30 min.) and for silver the samples were preserved with HCl. Chloride was analysed by liquid ion chromatography (based on CSN EN ISO 10304-1 and CSN EN ISO 10304-2). The other elements were analysed from the water samples with ICP-AES/SFMS.

2.4 Derivation of concentration ratios

Concentration ratio (CR) approach is a simple and the most widely used method of quantifying radionuclide transfer in the environment, especially in relation to human radiological protection [6]. The approach involves the assumption that an organism is in biogeochemical equilibrium with its surroundings [6]. The CR is the concentration in part of the organism is divided by the concentration in its surrounding medium. In this report, only concentrations on the basis of dry weight are used.

Due to their chemical characteristics, most of the water samples had concentrations of Cs and Ag below the limit of quantification (LOQ), a similar situation to Mo in the lake water and to Nb in the sea. In such cases, a value of LOQ/2 has been used as a surrogate to enable estimates of CR. The averaged concentrations in the water, as used in the concentration ratio calculations, are presented in Table 2.

CR calculations are generally based on filtered water samples, but in this paper data of unfiltered samples was used; no significant differences were observed in comparison between the paired filtered and unfiltered water samples taken in October 2010 from the Olkiluoto area. It was also observed that the concentration of I is usually higher and that of Nb usually lower in the filtered samples than unfiltered ones because of element specific properties; using the data from unfiltered samples for I appears to give conservative CR estimates at least in our case.

All water concentration values of lakes and Olkiluoto area are mean values of above mentioned sampling dates and the two closest locations available to the plant sampling area in question.

3. RESULTS

The main results are presented in Table 3. Full dataset of dry matter concentrations and water-to-plant concentration ratios also to other than the dominant species at each sampling area is presented in the conference poster, which is available from the authors upon request.

The standing living biomass varies at the sampling areas from 65–2100 g_{dw}/m^2, bur-reed showing the minimum and sedge the maximum (mainly, 60%, contributed by a dense mat of aquatic mosses), both in the Koskeljärvi Lake. The biomass of the reed beds is somewhat higher at the coastal site, Olkiluoto (780-940 g_{dw}/m^2), than at the lakes (460–510 g_{dw}/m^2). However, the internal variation of the sampling quadrats in the reed beds is rather high depending on the small-scale characteristics of the growth site. Lake Lutanjärvi seems to be higher in the biomass of aquatic plants in general than Lake Koskeljärvi, and also the concentrations of especially Cl, Mo and Cs are higher, possibly also the nutrients. As an example of more detailed but unfortunately sparse data on biomass distribution within the plants, water lily exhibits in our samples the mass ratio of roots vs. above-bottom part (assuming all roots have been harvested) being 2.3 (std. 2.0, N = 3), on the dry-weight basis. A single water lily individual had an inflorescence of 8%, the leaves 52% and the stems 40%, measured by the dry weight.

The concentration ratios are roughly the same order of magnitude between the different plant species and sampling sites, except for Cl in which case the CR in the coastal site is something completely

Table 2. Temporally and spatially averaged concentrations (μg/l) from water samples as used in the derivation of the concentration ratios of Table 3.

	I	Cl	Se	Mo	Ni	Nb	Cs	Ag
Olkiluoto	0.48	3 000 000	0.10	13.1	1.58	0.0084	1.70	0.08
Koskeljärvi	0.15	2 900	0.05	5.4	0.15	0.0063	0.69	0.18
Lutanjärvi	0.15	12 300	0.08	20.9	0.66	0.0059	4.46	0.16

Table 3. Standing live biomass on the sampling areas, share of the visually dominant species (dom.) of the biomass, dry matter content (DM) of the biomass (average) and of the dominant species, and concentration ratios from the water to the bulk biomass of the sampling area.

	Biomass, g_{dw}/m^2		DM, g_{dw}/g_{fw}		CR, (μg/kgdw)/(μg/l)							
	all	dom., %	all	dom.	I	Cl	Se	Mo	Ni	Nb	Cs	Ag
Olkiluoto coastal area												
- reed bed, west	940	71	0.48	0.47	28	1.6	45	60	240	320	280	320
- reed bed, east	780	80	0.42	0.39	8.0	1.9	130	150	300	650	380	650
Koskeljärvi Lake												
- reed bed	510	73	0.17	0.44	59	520	1300	370	550	2300	1300	320
- sedge	2100	40	0.09	0.25	120	160	1600	630	1300	14000	8100	660
- bur-reed	65	24	0.11	0.18	560	370	13000	6600	5200	170000	31000	4300
Lutanjärvi Lake												
- reed bed	470	39	0.25	0.43	52	150	790	90	260	3500	320	190
- bulrush	630	39	0.21	0.18	28	370	530	50	390	4000	360	460
- bur-reed	440	34	0.14	0.11	340	54	8300	1100	4400	160000	11000	2500
- water lily	270	95	0.11	0.11	42	520	1200	680	860	23000	2500	690
- pondweed	150	96	0.12	0.12	45	250	800	1100	2300	15000	5800	600

different from the lake values. Also with the other elements similar trends can be observed but not consistently. Nb exhibits the opposite behaviour to Cl. In general, bul-reed appears to be a generally effective accumulator in the aquatic ecosystems.

4. CONCLUSIONS

Typical aquatic macrophytes were sampled from Olkiluoto, a spent nuclear waste repository site in Finland, and from near-by lakes selected as representatives of those forming at Olkiluoto with the proceeding post-glacial crustal rebound. Variation in dry matter content of sampled plants follows the physiological differences between the plant species; the structure of submersed plants (e.g. water lily) is less dense and water content is higher compared to helophytes (e.g. common reed). Same differences can be also found between helophytes (e.g. bulrush vs. common reed). There is little available literature from the concentrations of and even less on the concentration ratios to aquatic plants from similar conditions. The coarse comparison between somewhat similar classes of plants in our study and in [7] shows an agreement within an order of magnitude. However, the concentration ratios for Nb seem anyway rather high. This should be investigated further in future studies, including the reliability of analytical methods and possible contamination sources. Overall, our results exhibit similar elemental compositions of the different plant species at least in most cases – an observation similar to that made in [8].

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