Deposition of ^{237}Np in Finland

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^{237}Np in environment

^{237}Np and its behavior in environment is less investigated than other transuranium nuclides.

Still,

- long physical half-life (2.14 Ma)
- several, easily changeable oxidation states
- more mobile than Pu in environment
- possible enrichment in terrestrial food chains (greater uptake of Np to some plants than of Pu and Am)
- concentration in environment is constantly increasing due to nuclear test fallout from1950-1960's, nuclear fuel reprocessing, accidents such as Chernobyl
- direct ^{237}Np emissions and from decay chain

^{241}Pu ($t_{½}$ = 14.35 a) → ^{241}Am ($t_{½}$ = 432.2 a) → ^{237}Np ($t_{½}$ = 2144000 a)

Sources of ^{237}Np in Finland

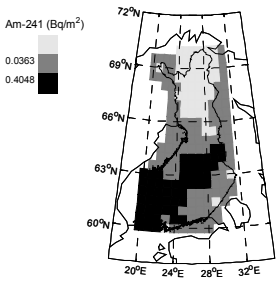
- from both nuclear weapons testing fallout in 1950-1960´s and Chernobyl accident
- ingrowth from ^{241}Pu (and ^{241}Am) more significant source than direct ^{237}Np fallout

Objectives of this study

- To complete Chernobyl-derived transuranium deposition study of peat in Finland, previously Pu-, Am- and Cm-concentrations have been determined from same peat samples
- as far as is known, the first attempt for separating Np from environmental samples in Finland

General problems with analysis of ^{237}Np

- concentration of ^{237}Np in environment is remarkably lower than with other transuranium nuclides
- low concentration → large sample mass is needed (20-100 g of peat) → interferences due to sample matrix
- adjusting and stabilising the oxidation state during chemical separation
- activity concentration determination by alpha spectrometry: long counting time is needed (weeks)
- Mass concentration determination by (HR)-ICP-MS: ^{238}U disturbs the ^{237}Np (238-peak tailing over 237-peak), therefore chemical separation has to be more efficient than with alpha spectroscopy
- AMS: ^{238}U is not a problem, lower D_L than with ICP-MS, but poor availability
- choice of tracer for yield determination: ^{239}Np, ^{235}Np, ^{236}Np and ^{242}Pu can be used



Deposition of Chernobyl-derived ^{241}Am.

Salminen, S., Paatero, J., Jaakkola, T., Lehto, J., 2005. Americium and curium deposition in Finland from the Chernobyl accident. Radiochimica Acta 93, 771-779

Samples

- peat samples were collected immediately after the Chernobyl accident (12.-14.5. 1986) in Southern and Central Finland from peatlands in production
- 20-100g of peat was utilised in analysis

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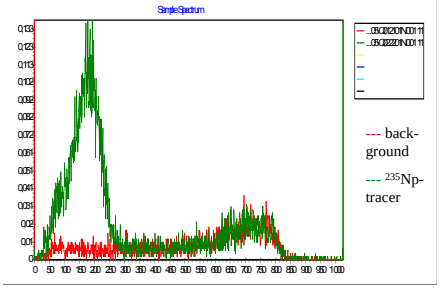
Separation method for ^{237}Np

- separation method of ^{237}Np from peat includes ashing, wet-ashing, Ca-oxalate co-precipitation and TEVA®-separation (Eichrom Technologies)
- mass concentration of ^{237}Np was measured with HR-ICP-MS
- recovery was determined by adding ^{235}Np-tracer before wet-ashing the sample

^{235}Np-tracer

+relatively long half-life (1.08 a) compared to often used ^{239}Np (2.36 d)
+decays with EC, X-rays of UL_{α} and Auger electrons can be measured with LSC or low-energy gamma detector
+prepared at Laboratory of Radiochemistry, *via* reaction ^{236}U (p, 2n) ^{235}Np
+recovery from TEVA-separation of tracer was ~100%, with peat samples the median recovery was 79%
-always contains small amount of ^{237}Np as impurity
-decay data of X-rays and Auger electrons is somewhat inconsistent (intensities and energies vary, mother and daughter nuclides are mixed up)
-success of irradiation and chemical separation (concentrations of ^{235}Np and ^{237}Np in tracer) can be concluded as late as six months after irradiation, when short-lived Np isotopes have decayed



The spectrum of ^{235}Np measured by Quantulus 1220. The peak at energy region 12–21 keV consists of L-shell transitions producing x-ray and Auger electron radiation. Counting efficiency was 33%.

Problems with separation method

•large samples were difficult to handle because of disturbing matrix (often iron) and high reagent amounts

•amount of ^{235}Np-tracer used in analysis had to be minimised because of ^{237}Np-impurity in tracer

•liquid scintillation cocktail OptiPhase Hi'Safe 3 was found to contain higher activity level than ca. 5 years ago, background count level is elevated both at low-energy (β, X-ray) and high energy (α) regions → recovery could not be determined for some samples

•uranium concentrations were higher than expected from test results of selected samples → generally two TEVA® separations are needed for reducing uranium concentration to acceptable level

→ these factors together with ^{235}Np-tracer decay while solving the problems caused loss of many samples

Activity concentration of ^{237}Np and fraction of Chernobyl-derived ^{237}Np (%) of total ^{237}Np in peat samples in Finland.

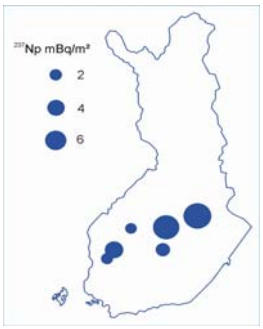
Peat bog	Location	A ^{237}Np (mBq/kg)	Chernobyl-Np/total Np (%)	^{237}Np/^{239,240}Pu activity ratio
21 Löyniönsuo, Hankasalmi	62.2 °N, 26.3 °E	0.73±0.02	0.5	0,00255±0,00011
11 Viherperä, Kankaanpää	61.7 °N, 22.8 °E	0.48±0.01	13.3	0,00145±0,00009
95 Korpisalonneva, Vimpeli	63.1 °N, 24 °E	1.97±0.04	0.6	0,00286±0,00010
144 Kulvesuo, Rautavaara	63.5 °N, 27.6 °E	3.37±0.08	5.4	0,00211±0,00040
148 Kumpusensuo, Pielavesi	63 °N, 26.8 °E	3.84±0.08	2.3	0,00263±0,00012
99 Korvaneva, Jalasjärvi	62.3 °N, 22.9 °E	1.81±0.04	4.6	0,00226±0,00010

Fractions of Chernobyl-^{237}Np/total-^{237}Np has been calculated using following literature values for activity ratios

A ^{237}Np/A ^{239,240}Pu$_{\text{global fallout}}$ **0.0037**

A ^{237}Np/A ^{239,240}Pu$_{\text{Chernobyl}}$ **0.000125**

- Beasley, T. M., Kelley, J. M., Maiti, T. C., Bond, L. A., 1998. ^{237}Np/^{239}Pu atom ratios in integrated global fallout. Journal of Environmental Radioactivity 38, p. 133-146
- UNSCEAR 2000 Annexes C and J.



Deposition of Chernobyl-derived ^{237}Np in peats in Finland. Activity values of Northern and Southern Finland were below MDA.

Transuranium nuclides from Chernobyl accident in peat in Finland, compared to total transuranium deposition in peat in Finland

$\frac{^{241}\text{Pu}_{\text{Ch}}}{^{241}\text{Pu}_{\text{total}}}$(%)	$\frac{^{239,240}\text{Pu}_{\text{Ch}}}{^{239,240}\text{Pu}_{\text{total}}}$(%)	$\frac{^{241}\text{Am}_{\text{Ch}}}{^{241}\text{Am}_{\text{total}}}$(%)	$\frac{^{237}\text{Np}_{\text{Ch}}}{^{237}\text{Np}_{\text{total}}}$(%)
10-99	0-100	0.9-100	0.5-13

Varying portions of old nuclear test fallout have been skimmed off the peat before sampling in 1986, therefore at some peat bogs all Pu and/or Am originates from the Chernobyl accident. All ^{244}Cm in peats was Chernobyl-derived.

Conclusions

- ^{237}Np in peat in Finland originates mainly from global nuclear test fallout
- compared to other transuranium nuclides, the fraction of Chernobyl-derived deposition of ^{237}Np was low
- concentration of ^{237}Np, both Chernobyl-derived and from older global fallout, varies depending on location
- analytical method needs further development: large sample mass (up to 100g), high U concentration in peat and problems with liquid scintillation cocktail set challenges for separation procedure