Factors influencing the distribution of weapon-test plutonium alpha-emitters on the whole-basin of a lake

I. Vioque[1], G. Manjón[1], R. García-Tenorio[1,2]*

[1]Department of Applied Physics II, E.T.S. Arquitectura, Avenida Reina Mercedes 2, 41012-Sevilla, Spain.
[2]Centro Nacional de Aceleradores (CNA), Seville, Spain

Abstract

By analysing the $^{239+240}$Pu and ^{238}Pu content in a total of 32 sediment gravity cores, it has been possible to obtain quite rich information about the accumulation pattern of these radioactive pollutants on the basin of a small lake located in southwest Sweden. In fact, it has been observed that the traditional lacustrine models reflecting as expectations higher accumulation of pollutants at deeper water depths are not appropriate for describing the specific processes controlling the distribution of the Pu alpha-emitters isotopes in the analysed whole basin.

Additionally, and knowing the primary weapon-test atmospheric origin of the Pu-isotopes in the region where the lake is located, it has been possible to evaluate the magnitude of the Pu incorporated to the lake basin from the surrounding drainage area. This evaluation was based in the analysis of the Pu-isotopes profile in a twelve layers sediment core collected from the deepest part of the basin and in the comparison between the Pu cumulative depositions determined in the gravity cores and the expected direct-atmospheric inputs.

Introduction

The knowledge of the Pu-isotopes historical and integrated fallout incorporated to several compartments of the biosphere has allowed opening new fields of research and/or developing quite established tools. In this sense, new insights about the environmental behavior of these pollutants once they were incorporated to a particular environmental compartment (soils, sediments, vegetation, etc.) have been obtained, while the determination of the Pu profiles in some environmental archives (sediment and ice cores) have been used as a dating tool (Ketteret et al., 2004; Eriksson et al., 2001).
In particular, the analysis and the determination of the spatial and temporal Pu-distribution on lake basins, located in places only affected by global sources of these nuclides, can give very rich information about the fate of these pollutants in lacustrine aquatic systems and in their surroundings drainage areas. In this sense, it is well known the extremely high reactivity of the transuranic elements to the particulate matter, which, in lakes with a not very short water residence time, should imply that the great majority of the fallout Pu incorporated to the waters finishes associated to the sediment beds (Eriksson et al., 2001).
In this paper, through the analysis of one twelve layers sediment core and thirty two integrated sediment cores collected from the basin of a small Swedish lake affected by acidification, it has been performed a complete study about the behavior of the Pu alpha-emitter pollutants in a lake of this type and in its catchment. All the plutonium determinations have been performed by applying the high-resolution alpha-particle spectrometric technique with semiconductor detectors.

* Corresponding author: ivioque@us.es

Materials and Methods

Site description

For this study it was selected the Lake Härsvatten, which is located at the southwest of Sweden, near the city of Gothenburg and at about 30 km of the North Sea coast (55º 03' N, 12º 02' E) (Figure 1). The lake is at 130 m over sea level and has an extension of 0.18 km^2. The average annual precipitation in the region is about 800 mm and the lake is covered by a consolidated ice-layer during the winter. In the summer, the lake is stratified with the interface between 10 and 15 m depth. Is an acidic clear-water oligotrophic lake (pH 4.4).

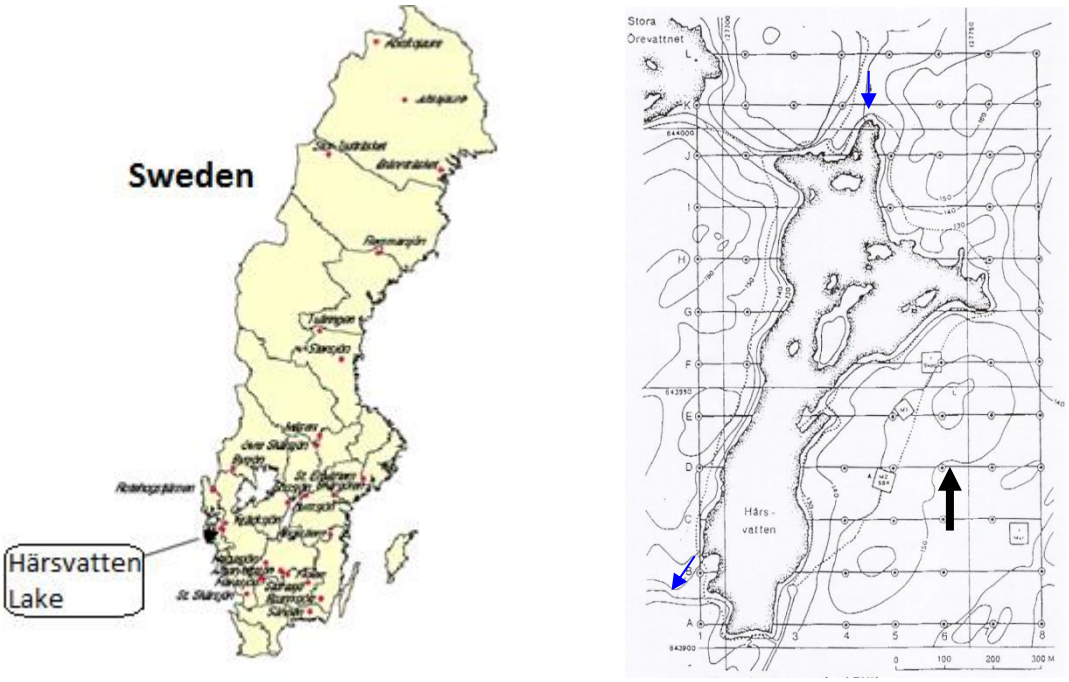

Figure 1. *Map of Sweden with the location of Lake Härsvatten and a map of the lake.*

From a bathymetric point of view, we can indicate that the lake shows a quite complex morphometry (see Figure 2). In the whole basin, it is possible to distinguish four different regions: a northern basin with a water maximum depth of 12 m, a shallow central zone with a depth ranging between 3 and 8 m, a southern basin, the largest one, with a maximum depth of 24.3 m, and an eastern basin, which is generally shallow although, in some points, it reaches a water depth of 10 m. The eastern basin is separated from the central north-south line of the lake by several islands and very shallow sills.

Sampling

A total of 32 sediment cores were collected at the places indicated in Figure 2, following this spatial distribution: 9 sediment cores were collected from the northern basin, 4 from the eastern basin, 2 from the shallow central zone of the lake and 17 from the southern basin (the largest one). Watching the southern and northern basin, the sampling strategy established allowed the collection of sediment

cores covering a wide range of water depths in order to study and analyse the use of a sediment focussing model for explaining the distribution of the Pu-isotopes cumulative depositions.). At every collection site, it was recorded the water depth from the ice surface. Once in the field, these cores where immediately sliced in two sections: the upper one corresponding to the upper 10 cm, and the lower containing the remaining part. This cutting strategy was adopted due to the knowledge gained in previous studies carried out in this lake, which indicate that the upper 10 cm of the collected integrated cores should contain all the Pu-isotopes inventories (which start to be deposited in the 50´s).

In addition to the integrated cores, it was collected in the deepest part of the south basin a twelve layers core (see Figure 2). This core had a length of 20 cm and was sliced also in the field in layers, 1 cm high each one, in order to obtain a Pu-isotope depth profile.

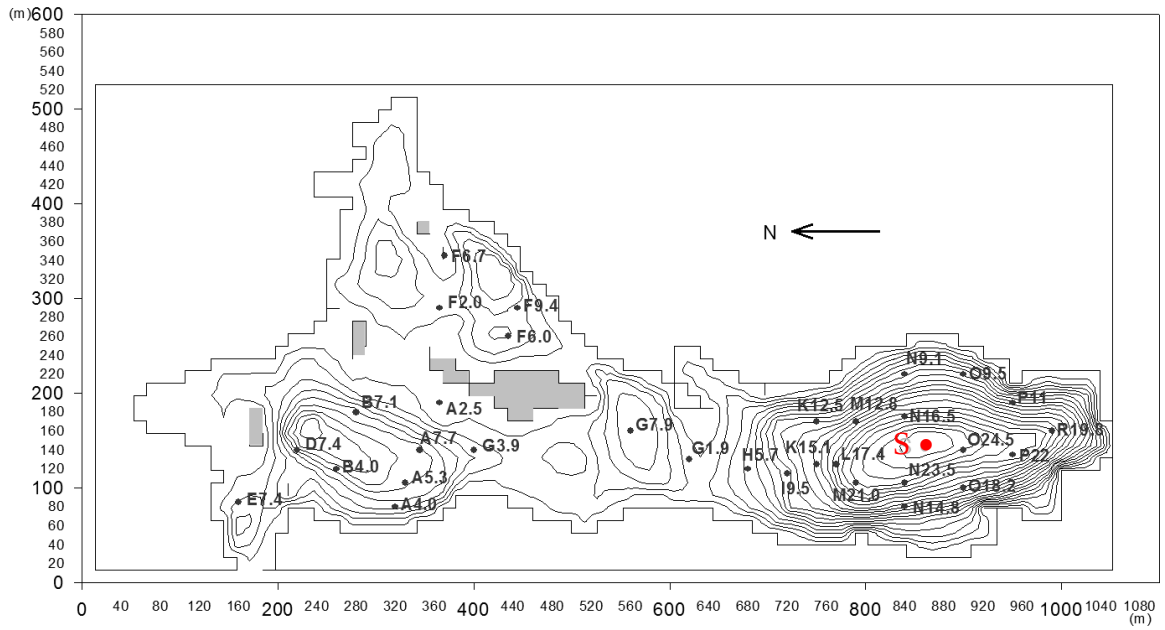

Figure 2. *Bathymetric map of Lake Härsvatten. The place of collection of the 32 integrated cores and of the high-resolution core is marked (red).*

The sediment fractions (either corresponding to the high-resolution or integrated cores), were placed in the field in a bag or container and stored at 4ºC. In the laboratory, the samples were weighed, freeze-dried, and reweighed to determine the water content and dry mass of the sediments.

Analytical methods

In order to proceed to the Pu-isotope alpha-emitters determination by alpha-particle spectrometry, a radiochemical procedure was applied to every sediment sample. According to the alpha-spectrometry requirements, plutonium was chemically isolated and deposited in thin layers to avoid interferences and to make sure a fine resolution. This radiochemical procedure can be found described in full detail in Vioque et al. (2002).

The detection system used for the measurements was an alpha spectrometer Alpha Analyst (Canberra) formed by eight independent chambers which can work in parallel. Each chamber is equipped with a 450 mm^2 passivated implanted planar silicon PIPS detector (18 keV nominal resolution), being

performed the measurements by placing the samples at a distance of 4 mm from the detector. In these conditions the counting efficiency is 0.34.

Results and Discussion

Twelve layers core

A total of 12 layers from a core, collected in the deepest part of the southern basin, were analysed for Pu-isotopes alpha-emitters ($^{239+240}$Pu, ^{238}Pu). The obtained $^{239+240}$Pu activity concentration profile (Bq kg^{-1}), with a resolution of 1 cm, is shown in Figure 3.

The shape of the profile is clearly affected by the historical evolution of the $^{239+240}$Pu concentrations in the lower layers of the atmosphere due to the weapons-tests fallout. Indeed, the maximum observed in the 5 cm depth layer could be associated with the years 1962-1963 (when the concentrations in the atmosphere of these Pu isotopes reached their maximum) as it was validated by the independent application of the ^{210}Pb dating method to the core (El-Daoushy et al., 1999). After these years the concentrations of Pu isotopes in the atmosphere decreased sharply to very low values in the 1970s and following years when weapons testing in the atmosphere decreased dramatically, fact that can be observed in the Pu sediment profile where the concentrations decrease sharply from the maximum at 5 cm until the surface. Additionally, the Pu with origin in the weapon tests fallout started to be detectable in the troposphere in the middle of the 50´s as is qualitatively reflected in the profile, because these dates correspond to the 8-9 cm sediment layer according to the ^{210}Pb dating method (El-Daoushy et al., 1999).

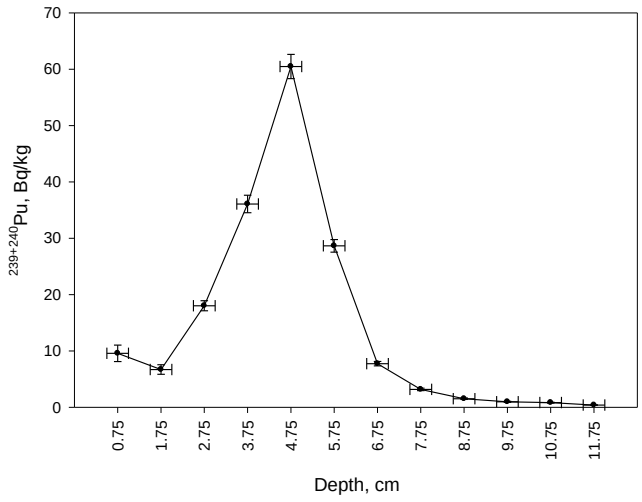

Figure 3. *$^{239+240}$Pu activity concentration profile (Bq.kg^{-1}) in the high-resolution core collected in the south basin of lake Härsvatten.*

In Figure 4, $^{239+240}$Pu deposition (Bq m^{-2}) and ^{137}Cs deposition (Bq m^{-2}), evaluated in the same samples, layers of core collected in the southern basin of lake Härsvatten, are shown for comparison. Indeed the highest value is in agreement for both elements. In the case of ^{137}Cs, the contribution of Chernobyl accident is clearly observed (El-Daoushy et al., 1999). Additionally, the $^{239+240}$Pu activity concentration (Bq kg^{-1}) is in agreement to SCP (g^{-1}) in the 12-layers core (Bindler et al., 2001). Similar patterns can be observed in Figure 5.

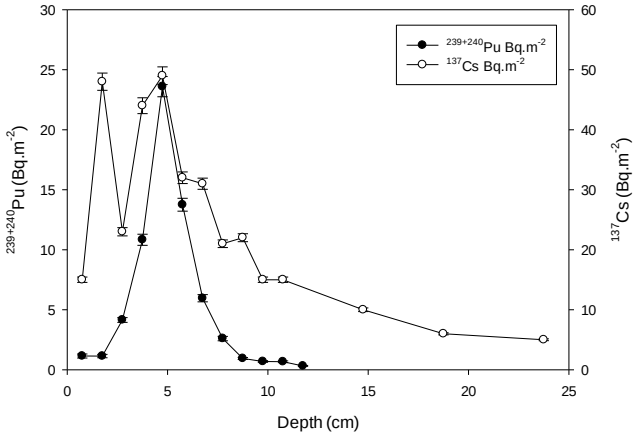

Figure 4. *$^{239+240}$Pu and ^{137}Cs deposition (Bq.m^{-2}) in the high-resolution core collected in the south basin of Lake Härsvatten.*

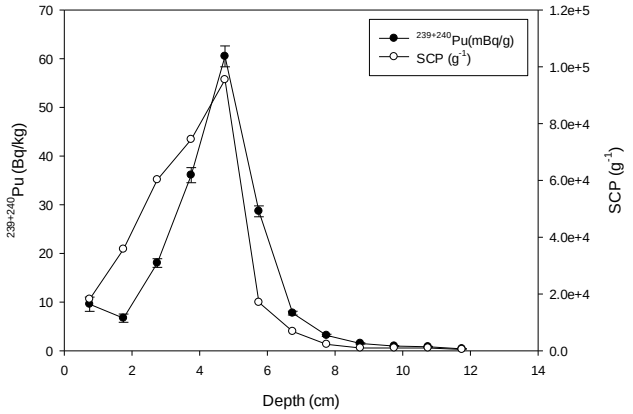

Figure 5. *$^{239+240}$Pu (Bq.m^{-2}) and SCP (g^{-1}) in the high-resolution core collected in the south basin of Lake Härsvatten.*

Once the good correlation observed between the Pu profile in the analysed twelve layers sediment core and the historical evolution of the Pu concentrations in the atmosphere was shown, several conclusions can be proposed:

a) These results reflect the no existence of physical or biological mixing processes affecting this core, at least in the upper layers of sediment.

b) By other hand, the sharp decrease observed in the twelve layers sediment $^{239+240}$Pu profile (Figure 4), from the maximum at 5 cm until the surface, as well as the comparatively low concentrations associated to these upper layers, is reflecting the quite small contributions of Pu originally deposited in the catchment area of the lake to the plutonium cumulative deposition in the core.

This last conclusion is in agreement with the fact that the integrated $^{239+240}$Pu cumulative deposition found in the sediment core (65.5 ± 1.2 Bq m^{-2}) is in a rather good agreement with the expected value of the direct weapon-test atmospheric Pu deposition in the area.

Integrated cores

In Table 1 are compiled the $^{239+240}$Pu cumulative depositions (Bq m^{-2}) determined in the 32 upper sections (0-10 cm) of the integrated cores collected at the places shown in Figure 2. These results have been arranged by dividing them into two main groups (one including the cores collected in the northern and eastern basins as well as in the central zone, the other one including the collected ones in the southern basin), and through its general analysis the following facts can be highlighted:

a) The $^{239+240}$Pu cumulative deposition, according to the data obtained at the different places of the lake bottom, is not uniform. Furthermore deposition range, 10 to 103 Bq m^{-2}, is very wide for evaluating a representative average value of cumulative deposition.

b) It can be also observed that the higher $^{239+240}$Pu cumulative deposition values are not found in sediments collected from the deepest parts of the different basins of the lake, and that the $^{239+240}$Pu cumulative depositions in sediment cores collected in some shallow zones (A2.5, F 2.0 and G 3.9, for example) are higher that the determined in another cores taken in deeper

areas. It is evident that no correlation exists between the determined $^{239+240}$Pu cumulative depositions and the depth.

c) If we amalgamate the results obtained in the northern and eastern basin (northern zone of the lake) we found that its average $^{239+240}$Pu cumulative deposition (62 Bq m^{-2}) is higher than the determined one in the south basin (44 Bq m^{-2}), in spite of the fact of the higher water depths in the southern part of the lake.

North basin, central zone and east basin		South basin	
Samples	$^{239+240}$Pu(Bq/m^2)	Samples	$^{239+240}$Pu(Bq/m^2)
A 2.5	76 ± 7	H 5.7	43 ± 2
A 4.0	78 ± 4	I 9.5	22 ± 1
A 5.3	53 ± 3	K 12.5	10 ± 1
A 7.7	46 ± 2	K 15.1	28 ± 1
B 4.0	33 ± 2	L 17.4	34 ± 1
B 7.1	77 ± 2	M 12.8	52 ± 1
D 7.4	53 ± 1	M 21.0	41 ± 1
E 7.4	97 ± 3	N 14.8	53 ± 2
F 2.0	103 ± 5	N 16.5	50 ± 1
F 6.0	56 ± 6	N 23.5	53 ± 1
F 6.7	36 ± 1	N 9.1	62 ± 2
F 9.4	27 ± 2	O 18.2	46 ± 1
G 1.9	34 ± 2	O 24.5	40 ± 1
G 3.9	81 ± 3	O 9.5	36 ± 1
G 7.9	85 ± 3	P 11	14 ± 1
		P 22	66 ± 2
		R 19.8	55 ± 2

Table 1.- *$^{239+240}$Pu cumulative depositions (Bq/m^2) determined in the 32 upper sections of the integrated cores collected in lake Härsvatten*

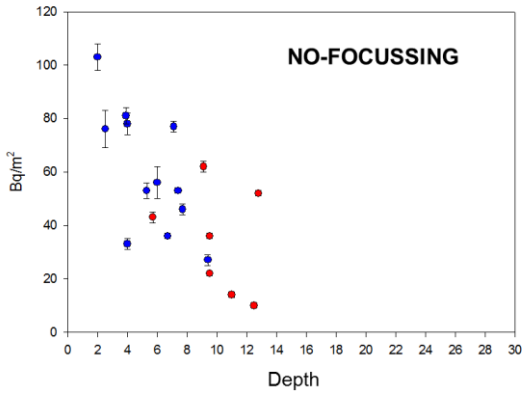

Figure 6. *$^{239+240}$Pu (Bq.m^{-2}) deposition in sampling point under 15m depth (blue, north and east basins; red, south basin).*

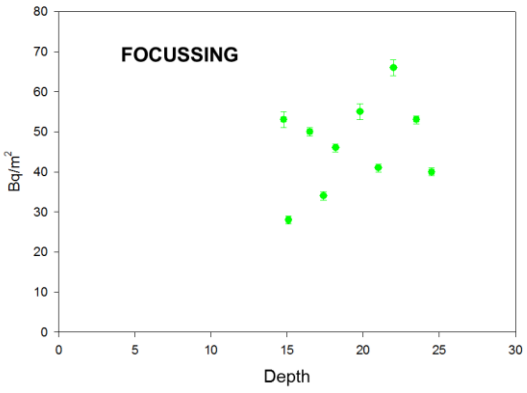

Figure 7. *$^{239+240}$Pu (Bq.m^{-2}) deposition in sampling point over 15m depth.*

The results shows that the basic model for sediment distribution and accumulation in lakes called "focussing", clearly cannot describe and explain the distribution of $^{239+240}$Pu. This model would predict a correlation between the $^{239+240}$Pu cumulative depositions and the depth, fact which is not observed at all in the lake analysed. But if we separated the deposition in the integrated cores collected at a depth under 15 m (Figure 6) and the collected over this depth (Figure 7) we can show that exits a correlation between the deposition and the depth in the samples over 15m depth (focussing) and an inverse correlation in samples under this depth (no-focussing).
As we have commented before, 15 m is the depth at were the lake is stratified in summer and it is also the depth from which it is possible to find algae mats in the bottom of the lake. The acidification of the waters can induce in this lake the expansion of algal mats on the lake bottom that prevent or interrupt sediment-focussing processes because the algae can act as efficient sediment traps, contributing to its uneven distribution across the lake bottom.

Additionally, from the data obtained through the analysis of the integrated cores some information can be ratified concerning the long-term and large-scale behaviour of plutonium in this lacustrine system. The weapon-test fallout origin of the accumulated Pu in the lake has been confirmed through the analysis of the $^{238Pu/239+240}$Pu activity ratios found in the analysed integrated cores (Figure 8). The average value of the activity ratios (0.034 + 0.008 with n=32) is in agreement with the expected value if the origin of these nuclides are the commented global ones (Hardy et al., 1973; Michel et al., 2002). There is non appreciable effect of Chernobyl (I.R.=0.47).

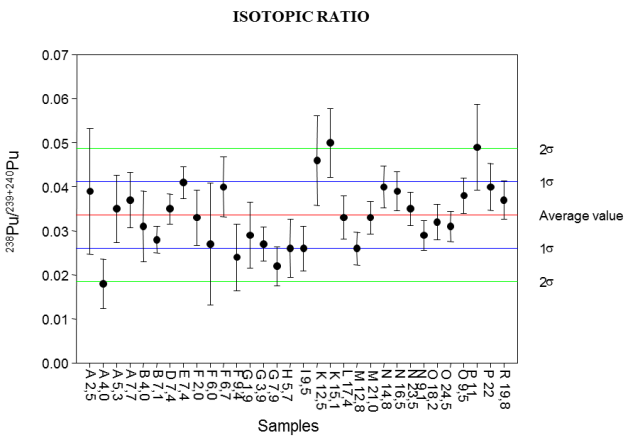

Figure 8. *$^{239+240}$Pu activity concentration profile (Bq.kg^{-1}) in the high-resolution core collected in the south basin of lake Härsvatten.*

In addition, the expected integrated atmospheric weapon-test fallout value in the region where is located lake Härsvatten can be evaluated to be in the range 45-60 Bq m^{-2} according to several data taken form the literature, which is in good agreement with the average cumulative Pu deposition on the whole lake basin (40-60 Bq m^{-2}). Indeed, being well known that the deposition of the Pu weapon-test fallout global source followed a latitudinal distribution, through the analysis of suitable soil cores, a general average deposition value of 58 Bq m^{-2} was fixed for the latitudinal band where the analysed lake is placed (UNSCEAR, 1982). Furthermore, some more refined studies performed at similar latitudes indicate that the integrated weapon-tests $^{239+240}$Pu deposition at regional scale is linearly correlated with the average annual precipitation in the different areas included in this region. Then, if we extrapolate the use of these correlations to the region of Lake Härsvatten, and we remember that the mean annual precipitation on the lake was 800 mm, the $^{239+240}$Pu cumulative weapon-test atmospheric deposition values obtained were in the range 45-60 Bq m^{-2}.

Conclusions

A detailed study about the spatial distribution of plutonium isotopes alpha-emitters have been performed on the whole basin of an acidified lake from southern Sweden, on the frame of an EU-financed project devoted to analyse the large-scale and long-term behaviour of transuranic elements in freshwaters systems and their surroundings catchment areas. The obtained results reflect the uneven distribution of this element which does not follow the pattern theoretically defined by the simple and ideal model of sediment focussing. Other factors like the lake hydrology or the influence of the acidification in the sedimentation processes can have influence in this distribution. On the other hand, through the average cumulative deposition value determined for the whole basin and through the analysis of the $^{239+240}$Pu profile in one high-resolution sediment core in the deepest part of the southern basin of the lake, it was deduced the no existence of appreciable inputs of Pu incorporated to the water body from the catchment area by erosion. In addition, the commented $^{239+240}$Pu-profile qualitatively reproduces the historical weapon-test deposition signal in the area, indicating that even under acidified conditions this signal can be well preserved in some areas of the lake bottom sediments.

References

Bindler, R., Renberg, I., Brännvall, M-L., Emteryd, O. and El-Daoushy, F. (2001). To whole-basin study of sediment accumulation using stable lead isotopes and fly-ash particles in an acidified lake, Sweden. *Limnology and Oceanography* 46, 178-188.

El-Daoushy, F. and others (1999).
"Large-scale and long-term environmental behaviour of transuranic elements as modelled through European surface water system"
European Commission, Nuclear Fission Safety, Final Report (Contract No. FI4P-CT96-0046), Uppsala University, Sweden.

Eriksson, M., Holm, E., Roos, P. and Dahlgaard, H. (2001). Plutonium in temperate and artic lakes. *Plutonium in the environment* (pp. 293 – 303). Ed. Kudo, A. In the series Radioactivity in the environment. Elsevier Science Ltd.

Hardy, E. P., Krey, P. W. and Volchek, H. L. (1973). Global inventory of fallout plutonium. *Nature*, 241, 444 – 445.

Ketteret M.E., Hafer K.M., Jones V.J. and Appleby P.G. (2004). Rapid dating of recent sediments in Loch Ness: inductively coupled plasma mass spectrometric measurements of global fallout plutonium. *Science of the Total Environment* 322, 221-229.

Michel, H., Barci-Funel, G., Dalmasso, J., Ardisson, G., Appleby, P. G., Haworth, E. and El-Daoushy, F. (2002) Plutonium and americium inventories in atmospheric fallout and sediment cores from Blelham Tarn, Cumbria (UK). *Journal of Environmental Radioactivity* 59, 127 – 137.

UNSCEAR (1982). Ionizing Radiation: Sources and Biological effects. *United Nations Scientific Committee on the Effects of Atomic Radiation, New York, United Nations*.

Vioque, I., Manjón, G., García-Tenorio, R. and El-Daoushy, F. (2002). Determination of alpha-emitting Pu isotopes in environmental samples. *The Analyst* 127, 530 – 535.

Wilson, T. A., Norton, S. A., Lake, B. A. and Amirbahman, A. (2008). Sediment geochemistry of Al, Fe and P for two historically acidic, oligotrophic Maine lakes. *Science of the Total Environment* 404, 269 – 275.