Fluxes and residence times of particles in the water column, derived from radionuclides measurements

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Introduction

The specific objectives of the Laboratoire des Sciences du Climat et de l'Environnement in OMEX II-II are:

- to determine particle residence times (**Task III.5.3**)
- to characterise seasonal variations in the concentration and composition of suspended particles in relation to its source, pelagic biogeochemical behaviour and fate, using natural radionuclides (**Task III.11.2**)
- to estimate the delivery efficiency of surface production to the "carbon depocentre" (**Task III.11.3**)

Powerful tools for calculating the intensity of chemical scavenging are the different isotopes of the natural radioactive decay series (Cochran, 1992). In particular ^{234}Th ($t_{1/2}$ = 24.1 d), ^{228}Th ($t_{1/2}$ = 1.9 y) and ^{210}Pb ($t_{1/2}$ = 22 y) are being continuously produced *in situ* by uranium and thorium decay in the water column. These daughter products are rapidly scavenged onto biogenic particles. Such partition between dissolved and particulate phases may be used to constrain scavenging efficiency of particles, and to estimate residence times for particle fallout and its variations in the water column. Usually, except in euphotic layer where ^{234}Th, with its short period of 24.1 days, is widely used, the longer life thorium, ^{228}Th (1.9 years) and ^{230}Th (75,200 y) are more suitable to follow processes occurring in open ocean water column (Bacon and Anderson, 1982). Nevertheless, as processes occurring in intermediate and bottom nepheloid layers over the Iberian Margin are supposed to occur on time scale of the order of months, ^{234}Th: ^{238}U and ^{228}Th: ^{228}Ra are the most appropriate pairs to study particles interaction on such short time scale. The principal objective are: - to determine how radionuclides are partitioned between dissolved and particulate forms and how this partitioning is affected by the presence of intermediate/deep nepheloid layers in the water column, and - to determine particle residence times.

Vertical profiles collected during OMEX cruises will provide an instantaneous view of the seasonal radionuclide distribution in the water column. Besides, synchronous measurements conducted on trapped samples will supply a continuous recording of radionuclide behaviour and a check of trap derived sedimentation rates.

1. Sampling

LSCE participated in 6 OMEX cruises, with contrasting success, mainly due to bad weather in winter:

CD105 (June 1997): meso-scale study of ^{234}Th distribution in the upper 50-m layer, 23 stations
PE109 (summer 1997): profiles of ^{234}Th/^{238}U and ^{228}Th/^{228}Ra in intermediate (1) and bottom (3) nepheloid layer
CD110 (January 1998): ^{234}Th/^{238}U and ^{228}Th/^{228}Ra in surface waters (8 underway stations) and in the 0-200 m layer (3 CTD casts).
PE121 (August 1998): ^{234}Th/^{238}U in 6 underway samples
M43/2 (January, 1999): 11 samples for ^{234}Th/^{238}U and ^{228}Th/^{228}Ra determination
PE138 (May 1999): 30 samples for ^{234}Th/^{238}U and ^{228}Th/^{228}Ra in clear water and nepheloids layers.

2. Method

a. Water samples

20 - 60-liters of seawater are sampled from the CTD rosette. Immediately after sampling, the seawater sample is vigorously mixed and filtered though a 0.45-µm (142 mm) Millipore membrane to separate the dissolved and particulate phases. Within three weeks after the collection, $^{234/228}Th^{p}$ are directly measured on the membrane with low background-high efficiency well type γ-detectors located in the underground laboratory of Modane (French Alps) [Reyss *et al.*, 1995].

The isolation and purification of $^{234/228}Th^{d}$ from seawater is performed using the anion-exchange procedure described by [Schmidt *et al.*, 1992]. Due to the short half-life of ^{234}Th and to avoid significant ingrowth corrections, the separation of ^{234}Th from its ^{238}U parent is carried out on board within 24 hours after seawater collection. For analysis, ^{229}Th yield tracer and 120 mg Fe (as $FeCl_3$) are added to the dissolved sample after acidification to pH 2. After spike equilibration, $Fe(OH)_3$ is precipitated by adding NH_4OH to pH 7. After recovery of the precipitate, the separation of ^{234}Th and ^{238}U is obtained by passage through an anion exchange column (Dowex 1x8, 100-200 mesh) under 8N HCl conditions.

During the two first cruises (*CD105-PE109*), we have improved our radiochemical procedure. The previous procedure implies time consuming work at the onshore laboratory: a further purification, by anionic exchange, an extraction with TTA, an α-counting for the determination of ^{229}Th (chemical yields) and ^{228}Th, then a γ-counting of ^{234}Th. The improved procedure is based only on a single γ-counting [Schmidt *et al.*, in preparation].

Uncertainties in the activity of dissolved $^{234/228}Th$ are calculated for each sample by propagation of the statistical errors in γ counting. As a result precision estimates are variable, reflecting the count rate of each sample (dependent on the chemical efficiency and the decay, between 30 to 80%) and the detector used: they ranged between 5 to 20% for the particulate phase, and between 5 to 15% for the dissolved phase.

b. Sediment trap samples

In the laboratory, trap samples, received from partner 17, were centrifuged, rinsed with deionised water and then were dried at 60°C, to be conditioned for radionuclides investigations. We measured the activities of radionuclides on dried samples by non-destructive gamma spectrometry. The standards used to calibrate the γ detectors are a mock-up of sediment and U-Th U.S. standard from the NBS. The use of small samples considerably reduces self-absorption effects for the detection of low-energy gamma rays, such as the 46 keV ray of ^{210}Pb.

3. Pre-upwelling conditions

^{234}Th data of *CD105* cruise have been interpreted in collaboration with partner 26. See also report of Partner 26 (I. Hall *et al.*, U.Camb.) for the discussion of the influence of particles on ^{234}Th scavenging and of POC - PON export flux derived from ^{234}Th.

Scavenging rate constants for dissolved ^{234}Th are relatively high: as a comparison k_1 presents lower values, from 0.002 to 0.015 yr^{-1} in the subtropical/equatorial Atlantic Ocean (Charette and Moran, 1999). The average residence time for dissolved ^{234}Th in the surface water is 68 days. Although a pre-upwelling situation is observed during *CD105* with low phytoplankton biomass and export fluxes, particle scavenging is more efficient over the margin, than observed in open ocean regions.

Scavenging constant rates and particle residence times, estimated from ^{234}Th, are presented Table 1.

	k_1 yr^{-1}	T^p y		k_1 yr^{-1}	T^p y
P100	0.044	8.3	N100	0.013	5.4
P200	0.013	27.5	N220	0.010	8.8
P1000	0.017	14.9	N1600	0.013	21.6
P2000	0.015	24.7	N2300	0.012	17.7
P2250	0.023	27.5	N2000	0.028	7.2
P2900	0.021	11.9	N3300	0.013	26.2
S130	0.014	12.6	V75	0.013	8.1
S200	0.018	16.6	V160	0.008	19.5
S1000	0.022	5.3	V1150	0.015	14.6
S2550	0.011	44.9	V2200	0.010	196.3
			V2800	0.016	22.1
			V3100	0.019	27.5

Table 1 - Average near-surface (0-50 m) scavenging rate constants for dissolved ^{234}Th and particle residence time, estimated from ^{234}Th deficiency, against water column depth. Stations were sampled. Each label corresponds to a station, along transects N, P, S and V, and its depth.

4. Winter conditions

In winter (*CD110*), total ^{234}Th activities are greatly variable, and both deficit / equilibrium situations are observed. The mean deficit of ^{234}Th in January 1998 is very limited, about 10% compared to 25% in June 1997. The reduced deficit may indicate low export fluxes. Moreover, in the shelf waters, $^{234}Th^P$ presents higher levels when compared with the summer results, which may be due to higher resuspension of shallow sediments. As a consequence, the average residence time for particle, derived from ^{234}Th, is significantly longest in winter, around 40 days, when compared to the summer situation (26 days in June 1997).

5. Intermediate/Bottom nepheloid layers

Most of nepheloid layer samples have been sampled in May 1999, during the *PE138* cruise and are in treatment. The Figure 1 presents profiles of ^{234}Th/^{238}U and ^{228}Th/^{228}Ra sampled on the Iberian shelf (*PE109* - station 11). Both thorium present deficit relative to their parents, more pronounced in the bottom nepheloid layer (200, 190 m), than in clear water (100m).

The simplified model of ^{234}Th, used to calculate particle residence for surface water data, is not relevant for the treatment of ^{234}Th and ^{228}Th in the water column. Because of the important scavenging of thorium, we adapted, as a first approach, the reversible scavenging model, previously described by Bacon *et al.* (1989), to evaluate scavenging rate and particle residence time from this profile. Particle residence times are relatively short, and increase from 8-10 days in the deepest bottom nepheloid layer to 15-18 days in the clear water.

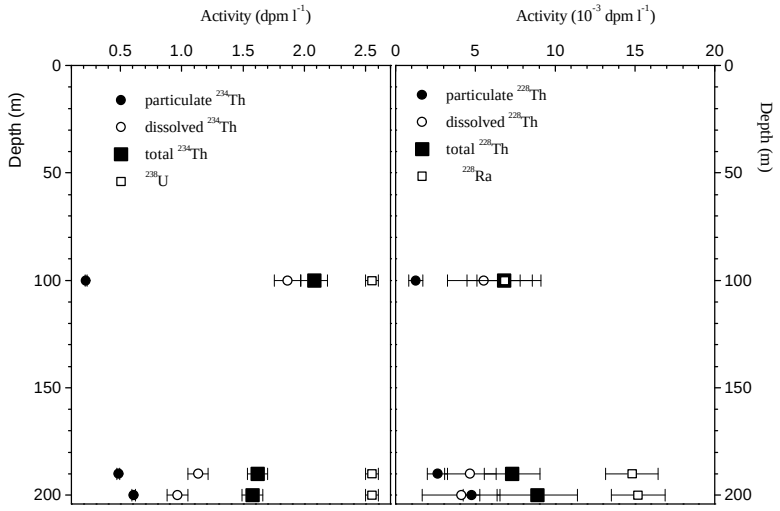

Figure 1 - Profiles of ^{234}Th and ^{228}Th in the nepheloid (190, 200 m) and clear (100 m) waters at the station 11 (42°20N - 9°15W) of the *PE109* cruise

6. Sediment trap fluxes:

Radionuclide measurements onto trap samples are in course. For instance we have only measured particle samples from the IM3 mooring at 600 m. ^{210}Pb specific activities vary from 30 to 100 dpm g^{-1} with the highest values associated with the lowest mass fluxes. As expected from production ratio in the ocean, ^{228}Th specific activities are lower, from 4 to 10 dpm g^{-1}.

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