

Particulate organic carbon : natural radionuclide ratios in zooplankton and their freshly produced fecal pellets from the NW Mediterranean (MedFlux 2005)

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Abstract

To discern controls on particulate organic carbon (POC) : natural radionuclide (RN) ratio variability in order to enhance the accuracy of water column radionuclide-based carbon flux estimates, ²³⁴Th, ²¹⁰Po, and POC were analyzed in seven size classes of mixed micro- and mesoplankton (1–1,500- μ m size range), in larger zooplankton from different taxa (salps, euphausiids, copepods, pteropods), and in freshly produced feces from zooplankton collected during spring in the NW Mediterranean. POC : RN ratios in zooplankton ranged between 120 and 11,600 and between 89 and 9,200 μ mol dpm⁻¹ for ²³⁴Th and ²¹⁰Po, respectively. In fecal pellets, POC : RN ratios were one to three orders of magnitude lower for ²³⁴Th and 3-fold to 30-fold lower for ²¹⁰Po; the only exception was euthecosome pteropods, which had a higher POC : ²¹⁰Po ratio in their pellets than in their whole bodies. Significant increases in POC : RN ratios with organism size were best described by a power relationship for POC : ²³⁴Th ($p < 0.0006$) and a saturation exponential equation for POC : ²¹⁰Po (with a constant POC : ²¹⁰Po ratio above 70 μ m; $p < 0.004$), suggesting that the observed trend most likely results from surface adsorption processes for ²³⁴Th and food chain bioaccumulation for ²¹⁰Po. This inference is further supported by the observation that, for the >33- μ m size classes, ²¹⁰Po specific activity correlates negatively with the surface : volume ratio, while ²³⁴Th correlates positively with it ($p < 0.004$ and $p < 0.001$, respectively). POC : RN ratios vary greatly among species and to a lesser extent among fecal pellet types, most probably due to differences in zooplankton feeding strategies. Partial removal of most zooplankton “swimmers” from trap samples would not likely confound assessment of ²³⁴Th flux; however, it could considerably bias similar measurements of ²¹⁰Po flux as well as those of POC : RN ratios.

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Given the interest in climate change and the potential role of the oceans in sequestering carbon, ²³⁴Th and to a minor extent ²¹⁰Po have been increasingly used to assess particle fluxes, especially particulate organic carbon (POC) out of surface waters (e.g., Kim and Church 2001; Friedrich and Rutgers van der Loeff 2002; Murray et al. 2005). When using natural radiotracers such as ²³⁴Th and ²¹⁰Po to study carbon export and cycling in the oceans, it is necessary to determine both the activity concentration of the radionuclides in the water column and their ratio to carbon in the particles responsible for carbon sequestration and export (Moran et al. 2003).

Although information on POC : ²³⁴Th ratios in a variety of particles from different size classes and regions is available (for review, see Buesseler et al. 2006), similar data on POC : ²³⁴Th and POC : ²¹⁰Po in zooplankton and fecal pellet material are extremely sparse (Fowler and Fisher 2004). This is somewhat surprising, considering that (1) many studies show that POC : ²³⁴Th variability is much greater in surface waters than at depths below the euphotic zone (Buesseler et al. 2006), suggesting a major role of biological processes in governing the POC : ²³⁴Th ratio, and (2) zooplanktonic organisms and their excretion products play a major role in radionuclide and trace-element

partitioning, and thereby represent one of the key vectors for carbon uptake and sequestration in the sea (e.g., Fowler 1977; Krishnaswami et al. 1985; Fowler and Knauer 1986).

The accuracy of the radionuclide tracer approach in estimating carbon flux is highly dependant on the assessment of POC: ^{234}Th (^{210}Po) ratios in particles that are representative of the sinking particulate flux (Moran et al. 2003; Buesseler et al. 2006; Smith et al. 2006). In this respect, future improvements in the use of the ^{234}Th and ^{210}Po flux approach necessitate, among other factors, a collection of representative types of sinking particles, e.g., fecal pellets, for POC: ^{234}Th (^{210}Po) ratio analysis (Buesseler et al. 2006). It is noteworthy that, although fecal pellets represent only one component of the vertical particle stream, they have been widely invoked as having a major effect on the mobilization and vertical transport of a variety of constituents in seawater (Fowler and Knauer 1986; Carroll et al. 1998).

Recent observations have indicated how different processes can influence particle characteristics at depth and, in particular, how the distribution of particulate ^{234}Th and ^{210}Po with particle size depends upon different parameters, including sorption, particle aggregation, disaggregation, repackaging by organisms, remineralization, and settling velocity (Savoie et al. 2006; G. Stewart unpubl. data). Future models of ^{234}Th (and ^{210}Po) export may thus have to include new parameters such as size, settling velocity, and lability in order to be more realistic.

Zooplankton accidentally caught in sediment traps ("swimmers") can affect the ability to accurately measure particle flux in the sea. Indeed, their presence in sediment traps influences both the quality and quantity of the particles in the traps and represents a major problem in measuring flux especially at shallow depths (Buesseler et al. 2006). Although swimmers are reported not to significantly affect use of the ^{234}Th method for determining trap collection efficiency because they normally have a much lower ^{234}Th content compared to detritus caught in sediment traps (Buesseler et al. 2006), no published data are available on POC: ^{234}Th (^{210}Po) ratios in zooplankton swimmers. This is somewhat surprising when considering that the value of POC: RN ratios in sediment trap material has recently been indicated as one of the parameters recommended for use in making radionuclide-based POC export estimations (Buesseler et al. 2006). Moreover, since the activity: mass ratio may be species dependent (Rodríguez y Baena et al. 2006a) and some species of zooplankton, especially cryptic swimmers such as gelatinous zooplankton, may be difficult to distinguish and thus separate from the rest of the detritus (Buesseler et al. 2006), the knowledge of organism-specific POC: ^{234}Th (^{210}Po) ratios would provide essential information on the potential biases associated with the presence of these organisms in sediment traps.

The overall objective of our study was to obtain a better understanding of POC: ^{234}Th (^{210}Po) variability and causes thereof, and in particular to: (1) refine our knowledge of POC: ^{234}Th (^{210}Po) ratios in large, fast-sinking fecal pellets of known source, which are vectors for carbon export from the euphotic and mesopelagic zones;

(2) determine ^{234}Th and ^{210}Po activity concentrations and their ratios to POC in different types of zooplankton that can enter sediment traps as "swimmers" and potentially confound direct measurements of particulate ^{234}Th and ^{210}Po flux; and (3) examine the role that plankton composition and organism size play in governing ^{234}Th and ^{210}Po concentrations and POC: ^{234}Th (^{210}Po) ratios in biogenic particles. This work reports natural radionuclide concentrations and POC: RN ratios in the most complete and varied set of zooplankton taxa (salps, pteropods, euphausiids, and copepods) and their freshly produced fecal pellets available to date.

Methods

During a spring MEDFLUX cruise (08–14 March 2005) onboard R/V *Endeavor*, we collected a set of different zooplankton species in the NW Mediterranean Sea. The sampling site (DYFAMED station: 43°25' N, 7°52' E) is located 52 km off the coast of Monaco in the Ligurian-Provençal Basin, seaward of the Ligurian-Provençal Current, which flows southwest along the coast through the Gulf of Lions toward the Strait of Gibraltar. This well-defined longshore current forms an effective physical barrier between coastal regions and the basin interior, thus providing relatively open-ocean conditions at this site (Andersen and Prieur 2000).

Sample collection, sorting, and incubation—Samples were collected by means of horizontal net hauls (either 50- or 480- μm nominal mesh size) taken at night from several depths (20–60 m). Two of the catches from the 50- μm mesh phytoplankton net were sorted using sieves of different mesh sizes in order to obtain seven size-fractionated subsamples (1–10; 10–33; 33–70; 70–200; 200–300; 300–600; and 600–1,500 μm). Particles much smaller than the nominal mesh size were also routinely collected in a qualitative way due to initial adhesion to larger organisms, particle aggregates, and the net mesh, which impeded their passage through mesh openings. Other tows made with a zooplankton net ($n = 3$, 480- μm mesh) were used to isolate selected live organisms. These samples were sorted by taxa: (1) salps (*Salpa maxima*); (2) euphausiids (*Meganyctiphanes norvegica*); (3) copepods (consisting primarily of *Euchirella rostrata*, *Calanus helgolandicus*, and *Heterorhabdus* sp.), and (4) thecosome pteropods (euthecosomes, consisting mainly of *Cavolinia gibbosa* and *Euclio pyramidata*, and a single pseudothecosome species, *Cymbulia peroni*). The selected organisms were then incubated separately for several hours in specially designed fecal pellet collectors containing filtered seawater (La Rosa 1976), and their freshly produced feces were concentrated onto a 43- μm mesh. Onboard, both organism and fecal pellet samples were cleaned of any extraneous foreign material, briefly rinsed with Milli-Q water, and immediately frozen. Upon return to the laboratory, small aliquots of the samples preserved in buffered formalin were examined microscopically, and the frozen samples were freeze-dried. The dried samples were then homogenized in an agate mortar, and weighed aliquots were analyzed for

carbon using a CHN analyzer and for ^{234}Th and ^{210}Po by a combined radiochemical procedure.

Radionuclide determination—In order to optimize the often-limited amount of sample available for analysis (viz. only ~3 to 50 mg dry weight of fecal pellet samples were available for radionuclide measurements), the procedure described by Rodriguez y Baena et al. (2006b) was combined with that of Flynn (1968). Accordingly, after radiochemical purification with UTEVA[®] columns, all feed and rinse solutions containing species not retained by the resin from 4 mol L⁻¹ HNO₃ (which include ^{210}Po and ^{210}Pb ; Horwitz et al. 1992) were analyzed for ^{210}Po and ^{210}Pb as described by Flynn (1968), rather than being discarded as proposed by Rodriguez y Baena et al. (2006b).

Briefly, after the addition of appropriate yield tracers (^{236}U , ^{230}Th , stable Pb, and ^{209}Po) and digestion with concentrated HCl, HNO₃, and HClO₄, the Th and U isotopes were separated from the Pb fraction by extraction chromatography using UTEVA[®] resin, and the feed solution was stored for later analysis of Po and Pb. The U and Th isotopes retained by the resin were separated by sequential elution steps with 0.6 mol L⁻¹ HCl and 6 mol L⁻¹ HCl, respectively, and finally coprecipitated with NdF₃ to obtain two distinct sources (Rodriguez y Baena et al. 2006b). The first source was used for alpha-spectrometry of the U isotopes (silicon surface barrier detector; EG&G Ortec), and the second source was used to estimate Th recovery through alpha measurement of the ^{230}Th yield tracer, and then for ^{234}Th determination via beta counting (gas-flow proportional beta counter; RISO National Laboratories) of its highly energetic daughter $^{234\text{m}}\text{Pa}$ (Rodriguez y Baena et al. 2006b).

Po contained in the feed solution was plated onto a silver disc and measured by alpha-spectrometry (Flynn 1968). The plating solution was further cleansed of Po traces by replating Po onto Ag foil, in order to ensure ^{210}Pb purification in the solution, and an aliquot was removed for Pb recovery determination. The remaining solution was then re-spiked with ^{209}Po , stored for at least six months, and finally plated a second time for determination of ^{210}Pb through ^{210}Po ingrowth.

POC determination—Particulate organic carbon (POC) was analyzed by high-temperature combustion using a Vario-El CHN microanalyzer, and carbonate was removed from the samples prior to combustion with 1 mol L⁻¹ orthophosphoric acid (Miquel et al. 1994). Sample values were blank-corrected, and the analyzer was calibrated daily using an acetanilide primary standard. The overall accuracy of the POC measurements, expressed as relative percent difference (RPD) between expected and mean measured value, was <3%.

Data analysis—Decay of ^{234}Th was followed by beta counting each sample four times during a six-month period after collection. As described by Rodriguez y Baena et al. (2006b), the initial ^{234}Th activity estimated from beta counting was corrected for beta detector efficiency, tracer

activity uncertainty, chemical yield (from alpha-counting), and sample weight, and then back-corrected to the sampling date considering ^{234}Th decay and ingrowth from ^{238}U . Overall ^{234}Th -propagated analytical uncertainties and between-sample variability (1σ) for these calculations averaged ~7%, with a range between 4% and 25% (the wide range was due to very low ^{234}Th signals in the pseudothecosome pseudoconchs and euthecosome fecal pellets, and occasionally to large between-sample variation).

The ^{210}Po activity values were blank and decay-corrected from the time of collection to the mid-point of counting. Po chemical recoveries (estimated by counting the samples in alpha detectors calibrated using a ^{209}Po standard source) were generally >80%, whereas the chemical yields for ^{210}Pb were always >90% (with the exception of one lower value, 45%, in a sample that was partially lost due to mishandling), as determined by stable Pb recovery checks with atomic absorption spectrometry (AAS) (Varian SpectrAA 220 Fast Sequential Flame-AAS). Overall ^{210}Po -propagated uncertainties (1σ) ranged between 5% and 90% (the wide range resulted from very low ^{210}Po signals for both first and second plating of the fecal pellet and pseudoconch samples, and occasionally to large between-sample variation).

For the size-fractionated mixed micro- and mesoplankton samples, regression analyses were carried out on the radionuclide specific activities and POC:RN ratios versus size and corresponding surface:volume ratios (based on spherical geometry). Changes in RN specific activities and POC:RN ratio with size were fitted by either a single-component exponential equation with an additional constant term (Eq. 1), a saturation exponential equation (Eq. 2), a direct power equation (Eq. 3), or a single linear regression equation (Eq. 4):

$$\text{RN(POC:RN)}_{s(\text{sv})} = y_0 + ae^{-bs(\text{sv})} \quad (1)$$

$$\text{RN(POC:RN)}_{s(\text{sv})} = a \left\{ 1 - e^{[-bs(\text{sv})]} \right\} \quad (2)$$

$$\text{RN(POC:RN)}_{s(\text{sv})} = a s(\text{sv})^b \quad (3)$$

$$\text{RN(POC:RN)}_{s(\text{sv})} = y_0 + as(\text{sv}) \quad (4)$$

where RN represents ^{234}Th or ^{210}Po specific activities (dpm g⁻¹ dry wt), POC:RN represents POC to ^{234}Th (^{210}Po) ratios ($\mu\text{mol dpm}^{-1}$), s represents size (mean mesh size, μm), sv is the surface to volume ratio (μm^{-1}), and y_0 , a , and b are constants.

Model constants and their statistics were estimated by iterative adjustment of the model and Hessian matrix computation using the nonlinear curve-fitting routines in the Sigmaplot[®] software. Best-fitting regression models were selected according to the highest determination coefficients and an examination of residuals.

Results

As can be seen in Table 1, both ^{234}Th and ^{210}Po activity concentrations vary greatly among the different zooplank-

Table 1. Average POC (% dry weight), RN concentrations, and POC:RN and $^{210}\text{Po} : ^{210}\text{Pb}$ ratios for zooplankton samples collected between 09 and 11 March 2005 in the NW Mediterranean Sea and their freshly produced fecal pellets (f.p.). All values are expressed as mean $\pm \sigma$. n = number of samples analyzed; σ = propagated error taking analytical uncertainty and between sample variation into account.

Sample	n	POC (%)	^{234}Th (dpm g^{-1})	POC : ^{234}Th ($\mu\text{mol dpm}^{-1}$)	^{210}Po (dpm g^{-1})	POC : ^{210}Po ($\mu\text{mol dpm}^{-1}$)	$^{210}\text{Po} : ^{210}\text{Pb}$
Zooplankton samples							
Salps	3	1.87 \pm 0.18	13.0 \pm 4.0	120 \pm 20	1.5 \pm 0.2	1,000 \pm 200	4.0 \pm 1.7
Euphausiids	3	39.7 \pm 0.4	3.1 \pm 1.4	11,600 \pm 4,200	3.61 \pm 0.44	9,200 \pm 1,200	20 \pm 13
Pseudothecosomes (body)	3	20.4 \pm 1.2	37 \pm 16	530 \pm 240	29 \pm 3	590 \pm 70	2.0 \pm 0.6
Pseudothecosomes (cartilaginous pseudoconch)	3	4.6 \pm 1.8	1.6 \pm 0.4	6,600 \pm 2,300	0.07 \pm 0.01*	64,000 \pm 13,000	2.6 \pm 0.6
Euthecosomes	2	17.3 \pm 0.9	41 \pm 3	380 \pm 50	164 \pm 19	89 \pm 8	46 \pm 12
Copepods	3	40.5 \pm 1.8	13 \pm 2	2,700 \pm 500	11.7 \pm 0.8	2,900 \pm 130	20 \pm 2
Fecal pellet samples							
Salp f.p.	3	6.20 \pm 0.06	1,980 \pm 170	2.61 \pm 0.16	45 \pm 5	115 \pm 11	0.74 \pm 0.16
Euphausiid f.p.	3	15.3 \pm 0.4	1,300 \pm 100	10.0 \pm 0.9	60 \pm 50	300 \pm 200	1.0 \pm 0.2
Pseudothecosome f.p.	3	6.5 \pm 0.8	1,920 \pm 170	2.8 \pm 0.5	34 \pm 3	160 \pm 30	0.70 \pm 0.08
Euthecosome f.p.	2	13.0 \pm 1.4	600 \pm 300	18 \pm 7	69 \pm 9	170 \pm 30	2.1 \pm 1.0
Copepod f.p.	3	16.5 \pm 0.4	1,000 \pm 90	13.4 \pm 1.8	37 \pm 3	370 \pm 60	1.4 \pm 0.4

* Pool of three samples.

ton classes (1.6–41 dpm g^{-1} dry wt for ^{234}Th ; 1.50–164 for ^{210}Po), and to a lesser extent among fecal pellet types (600–1980 dpm g^{-1} dry wt for ^{234}Th ; 34–69 for ^{210}Po). Fecal pellets were always highly enriched in ^{234}Th , and to a lesser extent ^{210}Po , compared to the organisms that produced them, the single exception being ^{210}Po in the euthecosome pteropods. The latter displayed an extremely high ^{210}Po specific activity (164 dpm g^{-1} dry wt), which, as shown by dissection, was mainly (96% on a dry wt basis) associated with the organisms' soft parts, (Rodriguez y Baena et al. unpubl. data from MEDFLUX 2006). This value is in good agreement with previous measurements of other authors (viz., 224, 246, and 676 dpm g^{-1} dry wt for three euthecosomes samples; M. Heyraud and R. Cherry unpubl. data) and approaches the highest ^{210}Po concentrations reported in the literature for pelagic marine species, viz. a value of approximately 300 dpm g^{-1} dry wt measured in whole midwater shrimp (Cherry and Heyraud 1982). Interestingly, the pseudoconch of *Cymbulia*, i.e., the separate transparent cartilaginous cover surrounding its body, contained the lowest specific activities for both ^{234}Th (1.6 dpm g^{-1} dry wt) and ^{210}Po (0.07 dpm g^{-1} dry wt). POC:RN ratios generally displayed a greater variability than those observed for RN specific activities, with values ranging in zooplankton from 120 to 11,600 $\mu\text{mol dpm}^{-1}$ for ^{234}Th and 89–9,200 $\mu\text{mol dpm}^{-1}$ for ^{210}Po , and from 2.61 to 18 $\mu\text{mol dpm}^{-1}$ ^{234}Th and 115–370 $\mu\text{mol dpm}^{-1}$ ^{210}Po in fecal pellets. POC:RN ratios were one to three orders of magnitude higher for POC: ^{234}Th in organisms relative to fecal pellets, and 3- to ~30-fold higher in the case of ^{210}Po , with the exception of the euthecosomes, for which the POC: ^{210}Po ratio in fecal pellets was higher than that in organisms.

The $^{210}\text{Po} : ^{210}\text{Pb}$ ratios in fecal pellets (range between 0.7 and 2.1; Table 1) were found to be at least 3-fold lower

than those measured in the zooplankton that produced them (range between 2.0 and 46; Table 1); these ratios are of the same order of magnitude as those reported in previous studies (Beasley et al. 1978; Higgo et al. 1980; Stewart et al. 2005). Interestingly, the ratios in both organisms and fecal pellets increased from lowest in strict filter-feeders (i.e., salps and pseudothecosomes) to highest in carnivorous species (i.e., euthecosomes).

Microscopic analysis of the size-fractionated, mixed plankton samples indicated that the smaller-size classes (<33 μm) consisted of a mixture of phytoplankton and detritus, and to a lesser extent microzooplankton, whereas the larger size fractions (>33 μm) were composed mainly of heterotrophic organisms. Table 2 reports the POC concentrations, RN specific activities, POC:RN and $^{210}\text{Po} : ^{210}\text{Pb}$ ratios in size-fractionated, mixed micro- and mesoplankton. Regression analyses of the RN specific activities and POC:RN ratios with respect to size and corresponding surface:volume ratios are shown in Figs. 1–3. When considering the entire data set, the variation of ^{234}Th specific activity with size was best fitted using a single-component exponential equation with an additional constant term (Fig. 1A). The ^{234}Th values showed a strong relationship with size ($p < 0.0001$, adjusted $R^2 = 0.93$). As shown in Fig. 1B, the same equation may be used to fit the variation of ^{210}Po specific activity as a function of particle size ($^{210}\text{Po} = 24 + 1460e^{-0.6164s}$); however, this model does not reliably explain the data pattern, since the entire exponential decay term is driven only by the activity concentration measured in the <10- μm sample. In addition, this equation suggests that the ^{210}Po concentration would then be virtually constant (24 dpm g^{-1}) over the 33–1500- μm size range, which is not consistent with an examination of the relationship at a higher-resolution scale. Indeed, when considering only the >33- μm size

Table 2. Average POC, RN concentrations, and POC:RN and $^{210}\text{Po} : ^{210}\text{Pb}$ ratios for size-fractionated micro- and mesoplankton collected on 10 and 11 March 2005 in the NW Mediterranean Sea. These organisms are the presumed food of the larger zooplankton groups shown in Table 1. Symbols are as in Table 1.

Size fraction (μm)	<i>n</i>	POC (%)	^{234}Th (dpm g^{-1})	POC : ^{234}Th ($\mu\text{mol dpm}^{-1}$)	^{210}Po (dpm g^{-1})	POC : ^{210}Po ($\mu\text{mol dpm}^{-1}$)	$^{210}\text{Po} : ^{210}\text{Pb}$
600–1,500	1	38.9 \pm 0.4	42 \pm 3	780 \pm 60	23.2 \pm 1.2	1,400 \pm 80	18.7 \pm 1.3
300–600	1	42.85 \pm 0.09	32 \pm 2	1,130 \pm 80	29.8 \pm 1.6	1,200 \pm 70	25 \pm 2
200–300	2	36.7 \pm 0.5	85 \pm 5	360 \pm 60	27.0 \pm 1.1	1,140 \pm 160	10.6 \pm 1.1
70–200	2	33.41 \pm 0.16	86 \pm 20	340 \pm 150	26.2 \pm 3.5	1,100 \pm 100	18 \pm 12
33–70	2	25.0 \pm 0.2	235 \pm 70	100 \pm 50	14.4 \pm 1.2	1,400 \pm 400	1.7 \pm 0.2
10–33	2	21.2 \pm 0.3	700 \pm 80	25 \pm 3	22 \pm 3	810 \pm 140	0.9 \pm 0.4
1–10	2	31.64 \pm 0.13	770 \pm 120	35 \pm 10	60 \pm 20	500 \pm 300	2.5 \pm 0.5

classes, which were mainly composed of heterotrophic organisms, the variation in specific activity of ^{234}Th and ^{210}Po with the surface:volume ratio was best fitted using a simple linear regression equation (Fig. 2A,B), which resulted in an opposite trend for the two radionuclides. In this case, ^{234}Th showed a strong positive correlation with the surface:volume ratio ($p < 0.001$, adjusted $R^2 = 0.84$), whereas ^{210}Po correlated negatively with surface to volume ratio ($p < 0.004$, adjusted $R^2 = 0.74$). As shown in Fig. 3A and B, the corresponding variations of POC:RN ratios with size were both significant and best described by a direct power relationship for POC : ^{234}Th ($p < 0.0006$, adjusted $R^2 = 0.69$) and a saturation exponential equation for POC : ^{210}Po ($p < 0.004$, adjusted $R^2 = 0.64$). These regression equations strongly suggest that ^{234}Th uptake behavior is essentially driven by adsorptive processes (POC : ^{234}Th increases continuously with increasing size), whereas internal bioaccumulation processes are the basis for the trend observed for ^{210}Po . For example, the observed steady state in ratio variation with size (Fig. 3B) indicates that an increase in ^{210}Po is limited by the relative POC content, which in turn is proportional to organism volume rather than to its surface.

The $^{210}\text{Po} : ^{210}\text{Pb}$ ratios in the smaller size classes ($< 70 \mu\text{m}$) of the fractionated plankton ranged between 0.9 and 2.5, and were always > 10 in those mainly composed of heterotrophic organisms.

Discussion

Although information on ^{234}Th and ^{210}Po specific activities in zooplankton and fecal pellets is sparse, our results, given in Tables 1 and 2, were generally in line with the findings of previous studies (see Table 3).

Zooplankton fecal pellets have been frequently indicated as representing one of the major components of biogenic particle fluxes (e.g., Ramaswamy et al. 2005), and in particular have been shown by Carroll et al. (1998) to account for a large percentage (annual average: 18%; spring and winter seasons: 35%) of the total POC flux at the DYFAMED site investigated here. In addition, fecal pellets, especially those of salps, owing to their reactivity and high settling velocities (up to 2700 m d^{-1} ; Bruland and Silver 1981), represent a primary source of fast-sinking, natural radionuclide-enriched biogenic particles in the oceans (Beasley et al. 1978; Higgs et al. 1980; Schmidt et al. 1990). It is in fact noteworthy that intact salp fecal pellets have been regularly found in sediment traps at 900–1000 m (Iseki 1981; J. C. Miquel unpubl. data). In this respect, our results on POC:RN ratios in different zooplankton fecal pellets of known source may be coupled with site-specific data on plankton abundances in order to predict the impact of changing food web processes on regional POC:RN variations, as has been suggested by Buesseler et al. (2006). Furthermore, it is important to

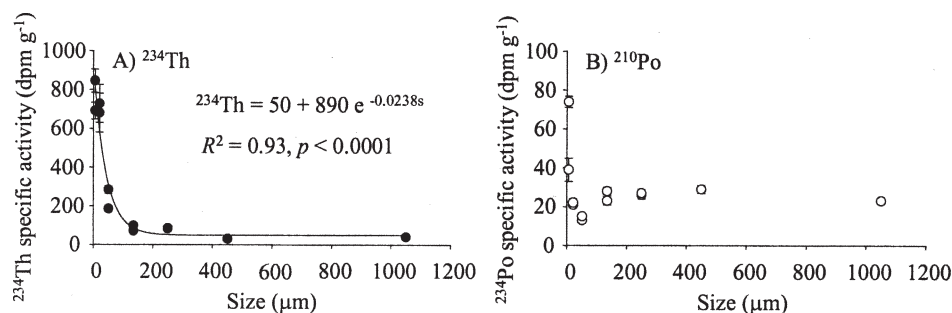


Fig. 1. Relationship between RN specific activities (dpm g^{-1}) and particle size (mean mesh size in μm) in the individual samples of fractionated micro- and mesoplankton collected on 10 and 11 March 2005 in the NW Mediterranean Sea (entire data set reported in Table 2) for (A) ^{234}Th and (B) ^{210}Po . Error bars indicate 1σ propagated analytical uncertainty.

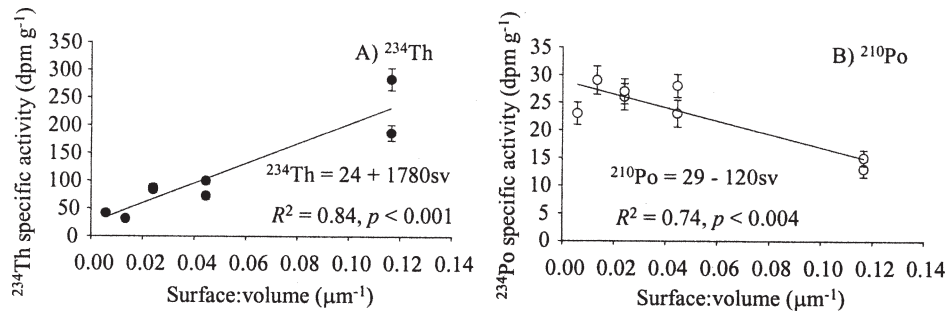


Fig. 2. Relationship between RN specific activities (dpm g^{-1}) and surface:volume ratios (based on spherical geometry, μm^{-1}) in the individual samples of fractionated mixed micro- and mesoplankton collected on 10 and 11 March 2005 in the NW Mediterranean Sea ($>33\text{-}\mu\text{m}$ classes, see Table 2) for (A) ^{234}Th and (B) ^{210}Po . Error bars indicate 1σ propagated analytical uncertainty.

better understand POC:RN variability in a variety of particle types in order to improve our estimates of carbon export and sequestration in the sea. In other words, data on POC:RN ratios in fecal pellets may be more effectively used as both a predictive and comprehensive tool within the framework of radioisotope-based particle flux studies.

As can be seen in Table 1, among the different types of pellets analyzed, salp fecal pellets display the lowest POC:RN ratios (2.61 $\mu\text{mol dpm}^{-1}$ for ^{234}Th ; 115 $\mu\text{mol dpm}^{-1}$ for ^{210}Po), as well as very low $^{210}\text{Po}:^{210}\text{Pb}$ ratios (0.74). This is probably linked to the fact that strict, filter-feeding organisms like salps play a major role in packaging phytoplankton dispersed in the water column and thus produce fecal pellets that are particularly rich in small phytoplankton cells and fine detritus, both of which are characterized by generally lower POC: ^{234}Th and $^{210}\text{Po}:^{210}\text{Pb}$ ratios (e.g., Stewart et al. 2005) compared to larger particles. Furthermore *Cymbulia*, a filter-feeding pseudothecosome that has a feeding strategy similar to salps, produces ^{234}Th -enriched fecal pellets with low POC: ^{234}Th (2.8 $\mu\text{mol dpm}^{-1}$) and $^{210}\text{Po}:^{210}\text{Pb}$ (0.70) ratios exactly like salps. These observations can serve as a practical example of the predictive and comprehensive potential of our POC:RN fecal pellet data. For instance, because POC:RN ratios are low in fast-sinking salp and pseudothecosome fecal pellets compared to those from crustacean zooplankton, they may be used (1) to predict the occurrence of low POC:RN ratios in particles during

certain seasons, and (2) to better comprehend why, for example, Szlosek et al. (2006) observed that during a sampling period in the NW Mediterranean characterized by a particulate flux dominated by fast-sinking salp fecal pellets, POC: ^{234}Th ratios in sinking particulates increased slightly with decreasing settling velocity, i.e., they behaved differently to what would have been expected if particle size rather than particle origin were the dominant control on POC: ^{234}Th ratios. In fact, an earlier field study undertaken near the DYFAMED site found very good correlation between salp biomass and the ^{234}Th deficit with respect to ^{238}U in seawater and suggested that salps play a major role in exporting ^{234}Th from surface waters to depth (Schmidt et al. 1992).

Results reported in Table 1 show that zooplankton organisms have much lower ^{234}Th specific activities than those in their fecal pellets and in sinking particulate matter in general (Buesseler et al. 2006). In this respect, our data support the suggestion of Buesseler et al. (2006) that incomplete removal of most swimmers from sediment trap samples will not significantly affect measurements of ^{234}Th fluxes in traps. However, this contention would not hold in the case of direct measurement of ^{210}Po trap fluxes given that the specific activity values for this radionuclide were found to range only from approximately 3- to 30-fold lower in organisms compared to their fecal pellets (Table 1). Moreover, in the case of euthecosome pteropods, ^{210}Po concentrations in whole organisms were roughly 2.4 times

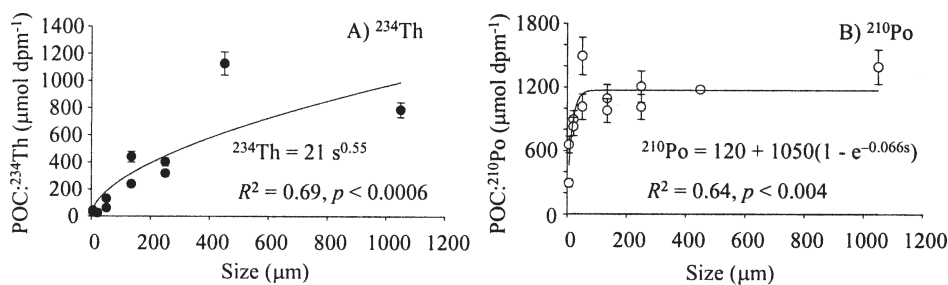


Fig. 3. Relationship between POC:RN ratios ($\mu\text{mol dpm}^{-1}$) and particle size (mean mesh size in μm) in the individual samples of fractionated plankton collected on 10 and 11 March 2005 in the NW Mediterranean Sea (entire data set reported in Table 2) for (A) ^{234}Th and (B) ^{210}Po . Error bars indicate 1σ propagated analytical uncertainty.

Table 3. Summary of published ^{234}Th and ^{210}Po specific activities in zooplankton and fecal pellets. No published data on POC : ^{234}Th (^{210}Po) ratios in these types of samples were found.

Sample type	^{234}Th (dpm g ⁻¹ dry wt)	^{210}Po (dpm g ⁻¹ dry wt)	Reference
Salps	1	1	Coale 1990
Mixed zooplankton swimmers	560	130	
Phyto-aggregates	1670	—	Dunne et al. 2000
Foraminifera	1160	—	
200–500 μm net tow	550	—	
200–500 μm net tow	530	—	
Copepods	310	—	
Euphausiids	84	—	
Chaetognaths	<20	—	
Shrimps (Caridae)	<20	—	
Gelatinous material	<20	—	
Copepods	—	7.5	Higgo et al. 1980
Euphausiids	—	2.4	
Euphausiid fecal pellets	—	54.4	
Mixed plankton, mainly calanoid and cyclopoid copepods	—	31–114	Kharkar et al. 1976
Salps (average value)	47	15.6	Krishnaswami et al. 1985
Salp fecal pellets	750	39.5	
Copepods (<i>Anomalocera patersoni</i>)	18.7	37.7	
	NM	20.9	
Mixed copepods (600–2,000 μm)	11.6	28.2	
	18.6	18.0	
Crab larvae	5.7	17.5	
Mixed surface zooplankton fecal pellets	300–600	—	Schmidt et al. 1990
Zooplankton swimmers	25–126	—	Murray et al. 1996

NM: not measured.

higher than in the fecal pellets they produced, suggesting that inclusion of euthecosome swimmers in trap samples could significantly perturbate the measured concentration of ^{210}Po in trapped particles. In fact, based on a ^{210}Po concentration in the NW Mediterranean waters of approximately 0.05 dpm L⁻¹ (Stewart et al. 2006), the euthecosomes in our study have a wet weight concentration factor (calculated using an experimentally derived dry wt : wet wt ratio of 0.28 ± 0.04 ; $n = 8$) of roughly 9×10^5 , one of the highest values recorded for zooplankton species, which typically display ^{210}Po concentration factors nearly two orders of magnitude lower (IAEA 2004). Furthermore, concentrations of both radionuclides were generally higher in zooplankton fecal pellets than in the micro- and mesoplankton food that these organisms were ingesting at that time (Table 2). This observation indicates that these natural radionuclides are not assimilated by the zooplankton to the same degree as carbon and other nutrients, which in effect leads to an overall enrichment of the radionuclides along with other nonassimilable material in the ejected fecal pellets.

The 3- to 22-fold higher $^{210}\text{Po} : ^{210}\text{Pb}$ ratios found in all zooplankton species compared to their excretion products (Table 1), as well as the observed increase in $^{210}\text{Po} : ^{210}\text{Pb}$ ratios with trophic level (following the order filter-feeding salps and pseudothecosomes, omnivorous euphausiids and copepods, and strictly carnivorous euthecosomes), are perfectly coherent with the suggested buildup of ^{210}Po along food chains and its preferential biological uptake compared to that of ^{210}Pb (Stewart et al. 2005). These additional data confirm our earlier supposition that

differences in zooplankton feeding strategies may represent one of the primary causes of the observed POC : RN ratio variability in fecal pellet samples; therefore, feeding behavior is likely to affect POC : RN ratios in suspended and rapidly sinking particulate matter.

Due to the “amplification” effect driven by the POC numerator in the ratio and the lower specific activities in organisms compared to their fecal pellets, POC : RN ratios in zooplankton were found to be much higher (one to three orders of magnitude for ^{234}Th and 3- to ~30-fold for ^{210}Po , with the exception of the euthecosome pteropods) than those measured in their fecal pellets (Table 1). This confirms the importance of completely removing swimmers from those sediment trap samples selected for the determination of the POC : RN ratios to be used in radionuclide-based carbon export models. Otherwise, RN-based POC fluxes may be significantly affected by positive biases.

Results reported in Fig. 1A and B show an inverse relationship between organism size and ^{234}Th and ^{210}Po activity concentrations when considering the entire range of size-fractionated plankton examined. The decrease of activity concentration with increasing particle size suggests an adsorption-driven behavior for both radioisotopes; this observation is consistent with previous findings for ^{234}Th (Fisher et al. 1987), but it is in contrast with the presumed bioaccumulative behavior of ^{210}Po in zooplankton food chains (Stewart and Fisher 2003). However, as indicated already (Results section), in the case of ^{210}Po , this inverse relationship is entirely due to the higher specific activities measured in the 1–33- μm size fractions, and therefore it

cannot be considered as representative of the actual behavior over the entire size range. Since the smaller size fractions ($<33\ \mu\text{m}$) were largely made up of phytoplankton cells and detritus, for which radionuclide accumulation is considered to be mainly driven by adsorption processes (Fowler and Fisher 2004), we looked for a possible bioaccumulative pattern in the size classes mainly containing heterotrophic organisms ($>33\ \mu\text{m}$). As shown in Fig. 2A and B, we found that RN specific activities in the $>33\text{-}\mu\text{m}$ size classes were significantly correlated with the surface:volume ratio; i.e., ^{234}Th correlated positively with the surface:volume ratio, suggesting a behavior triggered by adsorption even in this more-limited size range, whereas ^{210}Po was negatively correlated with the surface:volume ratio, suggesting bioaccumulation as the driving factor affecting changes in radionuclide specific activity with size.

The comparative examination of several studies indicating a general increase (or no change) in POC: ^{234}Th ratio with particle size in the 0.6–210- μm range (see Buesseler et al. 2006) appears to hold for the large biogenic particle range (1–1,500 μm) analyzed in our study (Table 2). Indeed, we found a significant direct power relationship between POC: ^{234}Th ratios and particle size (Fig. 3A). The power coefficient (0.55) was ~ 0.6 , viz. the ratio of the squared increase of surface area to the cubic increase of volume, suggesting that the increase in POC: ^{234}Th ratios is actually driven by surface adsorptive processes. Although this finding seems encouraging for modeling purposes, the value of the power coefficient for the relationship should be considered with caution given that the data in the larger size fractions are not numerous; hence, additional data are needed to confirm both this power trend and the accuracy of the coefficient, as well as to explore the eventual applicability of this relationship to other plankton classes.

To the best of our knowledge, no published information on POC: ^{210}Po ratios in size-fractionated plankton is available in the literature. Our data in Fig. 3B indicate that the POC: ^{210}Po ratio increases steeply with particle size up to 70 μm , and thereafter was relatively constant at a ratio of approximately 1,050 $\mu\text{mol dpm}^{-1}$. This observation is in accordance with the suggested bioaccumulative behavior of ^{210}Po as was noted for Fig. 2B. Indeed this constancy in the ratio demonstrates that ^{210}Po concentration increases proportionally with POC content, which for organisms is proportional to their body weight. The very steep increase in POC: ^{210}Po ratio in small size fractions up to 70 μm stresses the need to carefully assess this ratio for computing ^{210}Po -based flux estimations, because any small variation in particle size of the samples selected for a POC: ^{210}Po ratio assessment may have dramatic effects on the related carbon flux estimate. Similarly, the high and constant POC: ^{210}Po ratio found in larger particles (i.e., heterotrophic micro- and mesoplankton organisms) compared to smaller biogenic particles indicates that great care should be taken in removing all zooplankton (i.e., swimmers in the case of sediment traps) from samples dedicated to a POC: ^{210}Po ratio assessment for carbon flux estimation purposes.

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