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POC/^{234}Th ratios in particles collected in sediment traps in the northern South China Sea

Chin-Chang Hung*, Gwo-Ching Gong

Institute of Marine Environmental Chemistry and Ecology, National Taiwan Ocean University, Keelung 20224, Taiwan

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ABSTRACT

^{234}Th has been increasingly used as a tracer to estimate particulate organic carbon (POC) flux, based on calculation of the product of the POC/^{234}Th ratio in sinking particles and the ^{234}Th flux. Large (>50 μm) pump-collected particles are assumed to be representative of sinking particles for determining POC/^{234}Th ratios, but the basis of this assumption has not been extensively investigated. Here we present POC and ^{234}Th data for various particle size classes (1–10, 10–50, 50–150 and >150 μm) from trap-collected particles in the northern South China Sea (SCS, a subtropical region). Within the trap-collected POC pool the 1–10 μm fraction contained the largest proportion of POC (26–35%), followed by the 10–50 μm (25–29%), 50–150 μm (19–24%), and >150 μm (17–23%) fractions. The distribution pattern of ^{234}Th in the trap-collected particles was analogous to that of POC, with the smallest (1–10 μm) particles representing the largest proportion (37–54%) of the ^{234}Th flux. Our preliminary results indicate that trap-collected particles <50 μm carry most of the POC and ^{234}Th flux in the northern SCS, suggesting that the contribution of particles smaller than 50 μm to the settling flux is larger than previously thought. The results indicate that it may be necessary to simultaneously measure POC/^{234}Th ratios on size-fractionated trap- and pump-collected materials from different marine regions, including tropical and high latitude locations.

1. Introduction

The export flux of particulate organic carbon (POC) plays an important role in the transfer of atmospheric CO_2 to the ocean interior (Volk and Hoffert, 1985). The ^{234}Th/^{238}U disequilibrium has been widely used to estimate POC fluxes since Tsunogai and Minagawa (1976) proposed this concept. This is because ^{234}Th is a particle-reactive element with a short half life ($t_{1/2} = 24.1$ d), which makes this isotope suitable for tracing biologically-mediated processes and their timescales in surface waters (Coale and Bruland, 1985; Buesseler et al., 1992; Wei and Hung, 1998; Santschi et al., 2006; Forster et al., 2009). The current approach to calculating the POC flux is based on determining the product of the POC/^{234}Th ratio in sinking particles at a particular depth and the integrated ^{234}Th flux in the water column above it (Buesseler et al., 1992; Cochran et al., 1995; Bacon et al., 1996; Murray et al., 1996). Ideally, the POC/^{234}Th ratio should be determined from particles that are actually sinking (i.e. trap-collected), even though it is difficult to capture a sufficient quantity of settling particles of adequate quality (e.g. Buesseler et al., 2006, and references therein). In most studies, large pump-collected particles (i.e. >53 or >70 μm) have been assumed to be representative of the major sinking particles (Buesseler et al., 1995; Cochran et al., 2000; Moran et al., 2003). Consequently, the POC/^{234}Th ratio in such particles has been increasingly adopted for calculations of the POC flux in the euphotic zone in various marine environments (Bacon et al., 1996; Murray et al., 1996; Moran et al., 2003).

However, the ^{234}Th-derived POC method may be significantly biased because of two main factors: (1) the great variability of POC/^{234}Th ratios associated with large pump-collected particles (Fig. 1A,B; range 1–~300 μmol dpm^{-1}), and (2) large uncertainties in estimates of the ^{234}Th flux (see Savoye et al., 2006). The uncertainty associated with the former is larger than that of the latter. Despite POC/^{234}Th ratios having been determined in samples taken at the same time and at similar depths (Hung and Gong, 2007; Kawakami and Honda, 2007; Stewart et al., 2007; Buesseler et al., 2008, 2009; Lalande et al., 2008; Lampitt et al., 2008; Maiti et al., 2008; Lepore et al., 2009), most of the measured POC/^{234}Th ratios have diverged from a 1:1 ratio (Fig. 2). This generates the question what factors can cause significant difference on POC/^{234}Th ratios between pump-collected and trap-collected particles. To address this issue, two related questions need clarification. (1) Are the

* Corresponding author.
E-mail address: cchung@mail.ntou.edu.tw (C.-C. Hung).

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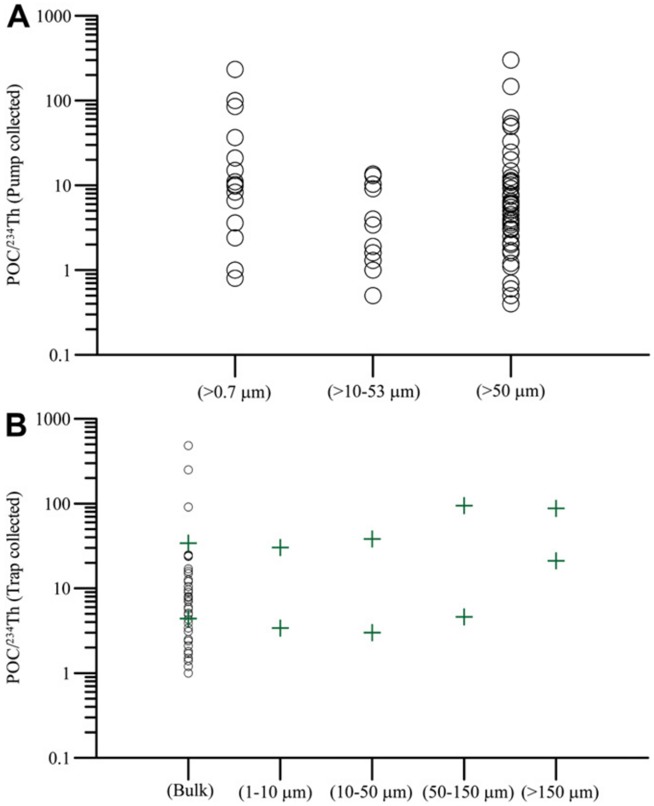

Fig. 1. Ratios of POC/^{234}Th on size-fractionated pump-collected (upper panel) and trap-collected (lower panel) particles. Data in the upper panel are derived from Amiel et al. (2002); Bacon et al., 1996; Baskaran et al., 2003; Benitez-Nelson et al., 2001; Buesseler et al., 1995, 1996, 1998, 2001, 2006; Cai et al., 2002, 2008; Charette and Moran, 1999; Charette et al., 2001; Coppola et al., 2002; Gustafsson et al., 2006; Hung et al., 2004; Hung and Gong, 2007; Kawakami and Honda, 2007; Lampitt et al., 2008; Lalande et al., 2008; Lepore et al., 2009; Moran et al., 2003; Morris et al., 2007; Murray et al., 1996; Rutgers van der Loeff et al., 1997, 2002; Savoye et al., 2008; Speicher et al., 2006; Stewart et al., 2007; Thomalla et al., 2006; Trimble and Baskaran, 2005; and Wei et al., 2009. The POC/^{234}Th ratios in bulk trap-collected particles in the lower panel are derived from Benitez-Nelson et al. (2001); Buesseler et al., 1992, 2000, 2006, 2008, 2009; Charette et al., 1999; Gustafsson et al., 2006; Hung et al., 2004,2009a,b; Hung and Gong, 2007; Kawakami and Honda, 2007; Lalande et al., 2008; Lampitt et al., 2008; Lepore et al., 2009; Maiti et al., 2008; Murray et al., 1996; and Stewart et al., 2007. The green crosses represent the POC/^{234}Th ratio range from various marine environments (Hung et al., submitted for publication). Note: high POC/^{234}Th values (in the upper panel) are often found in shallow water regions and in the Baltic Sea, which has low ^{234}Th activities because of low ^{238}U activities.

pump-collected particles (>50 μm) representative of the major settling particle assemblage? (2) Can the POC/^{234}Th ratios of such particles be used instead of average POC/^{234}Th ratios in trap-collected particles? Based on earlier results from trap-collected particles and model predictions, the sinking large particles (>100 μm), which are rare in the water column, is responsible for the majority of the downward vertical mass flux in the ocean (Fowler and Knauer, 1986; Michaels and Silver, 1988; Clegg and Whitfield, 1990). Bacon et al. (1996) subsequently questioned whether such particles are indeed representative of sinking particles generally, and suggested that the issue required clarification.

Murray et al. (1996) proposed that the POC/^{234}Th ratio of sinking particles provided optimal accuracy for estimating the POC flux from the ocean surface. Coppola et al. (2002) compared POC/^{234}Th ratios in sinking and suspended particles (>0.6 μm) in the Barents Sea, and suggested that the POC flux estimates using the POC/^{234}Th ratios from sediment trap material may be more reliable. Hung et al. (2004) reported that trap-derived POC fluxes and ^{234}Th-derived POC fluxes (based on the POC/^{234}Th ratio for large pump-collected particles e.g. >53 μm) in the Gulf of Mexico were comparable. Hung and Gong (2007) reported that in the Kuroshio Current there is good agreement between estimates of POC fluxes from the euphotic zone as measured by both sediment traps and ^{234}Th approaches (using the POC/^{234}Th ratio for medium-sized pump-collected particles e.g. 10–50 μm). Lalande et al. (2008) and Lepore et al. (2009) estimated POC export using sediment traps and POC/^{234}Th ratios in size-fractionated pump-collected particles, and found that estimates based on either approach can be biased. Savoye et al. (2008) estimated the POC flux using POC/^{234}Th ratios from the 5–210 μm size fraction and trap-collected particles. Stewart et al. (2007) investigated POC/^{234}Th ratios in particles sampled using in situ pump sediment traps and settling velocity (SV) sediment traps, and reported that in both cases there was large variation in the ratios. Szlosek et al. (2009) used SV sediment traps to separate particles and found no consistent trend in the POC/^{234}Th ratios as a function of settling velocity. More recently, Hung et al. (in press) measured concentrations of POC and ^{234}Th in size-fractionated (1–10, 10–50, 50–150 and >150 μm) trap-collected particles from the southern East China Sea and the subtropical northwestern Pacific Ocean, and found that the small-sized (1–50 μm) particles contained substantial proportion of POC (45–60%) and ^{234}Th (53–73%) in terms of both carbon and ^{234}Th content, indicating that the smaller particles may represent the majority of settling particles in the surface ocean. Based on these reports it is evident that information is required on the POC and ^{234}Th content of various size classes of sinking particles, to enable assessment of their contributions to the settling flux of POC with respect to the characteristic time scale of ^{234}Th.

The objectives of this study were to investigate the composition (carbon and ^{234}Th content) and the POC/^{234}Th ratio in various size classes of trap-collected particles. However, there is no absolute method for directly measuring POC and ^{234}Th in size-fractionated particles. To minimize the solubilization, degradation and possible disaggregation of captured particles and aggregates, we used

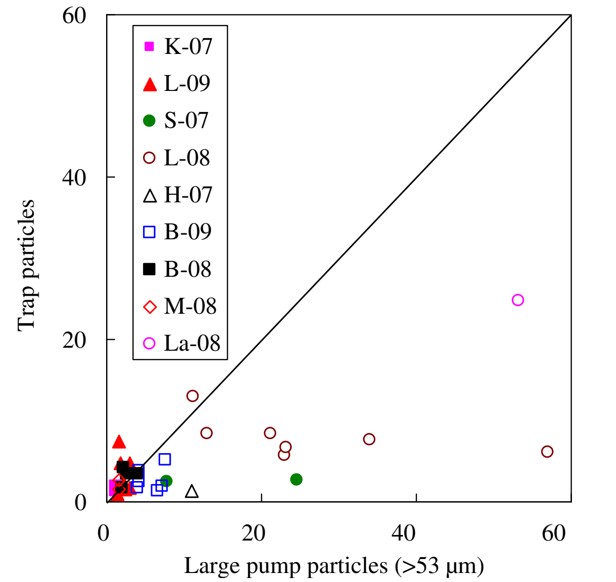

Fig. 2. Relationship of POC/^{234}Th (μmol dpm^{-1}) between trap-collected (Trap particles) and large pump-collected particles (Large pump particles) from recent studies involving similar water depths (K-07: Kawakami and Honda, 2007; L-09: Lepore et al., 2009; S-07: Stewart et al., 2007; L-08: Lalande et al., 2008; H-07: Hung and Gong, 2007; B-09: Buesseler et al., 2009; B-08: Buesseler et al., 2008; M-08: Maiti et al., 2008; La-08: Lampitt et al., 2008). Note: ratios of POC/^{234}Th >60 μmol dpm^{-1} are not shown in the figure.