

An Introduction to the Application and Future Use of ^{234}Th in Aquatic Systems

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ABSTRACT

^{234}Th is an extremely useful radiotracer of particle dynamics in aquatic systems. Its use, however, is still limited to a relatively narrow field of investigators. The reasons for this may in part be due to a lack of knowledge of how this nuclide has been used in the past as well as how and where ^{234}Th might be profitably applied in future research. The purpose of this paper, then, is to examine the variety of ^{234}Th applications that have been used to understand natural aquatic processes in four major areas: vertical transport, particle cycling, horizontal transport, and sediment dynamics. We provide a general overview of the possible applications of ^{234}Th in the hopes of provoking an increased interest in the inherent potential and future application of ^{234}Th in these systems. We end this paper with a discussion of future research avenues in the context of three specific regimes: i) the upper 1000 m of the open ocean, ii) coastal sediment/water processes and iii) large freshwater lakes.

1. Introduction

Thorium-234 was first discovered in 1900 by Sir William Crookes at 7 Kensington Park Gardens, London (Crookes, 1900; Merritt, 1903) and it can be argued, without too much exaggeration, that this discovery was key to our eventual understanding of the cause and nature of radioactivity itself (Rutherford and Soddy, 1902; Edwards et al., 2003).

In recent decades, ^{234}Th has become one of the most actively used tracers in oceanography. The primary reasons for ^{234}Th 's popularity as a tracer are three fold. First, ^{234}Th is radioactive and differences between its measured activity in the field and its expected activity based on known rates of production and decay can be used to determine the rate of the nuclide's removal by any particular process in an aquatic system. Second, ^{234}Th is particle reactive while its parent, ^{238}U , is generally non-reactive and conservative in oxic systems. ^{234}Th , therefore, can be used to trace the temporal and spatial dynamics of particles and other particle reactive species. Moreover, uniform, *in situ* production of ^{234}Th by a conservative parent greatly simplifies the interpretation and modeling of ^{234}Th activity data and its implied association with any particular process in an aquatic system. Finally, the half-life of ^{234}Th is only 24.1 days. This makes ^{234}Th particularly suited for studying biologically mediated and other relatively fast (physical) processes that occur on time-scales of days to weeks, which are typical in aquatic systems (Figure 1).

In spite of the popularity of ^{234}Th as a tracer, its use (paradoxically) is still limited to a relatively narrow field of investigators. The reasons for this may in part be due to a lack of knowledge of how this nuclide has been used in the past and how and where ^{234}Th might be profitably applied in future research. The purpose of this paper, then, is to look at the variety of ^{234}Th applications that have been successfully used to understand a broad spectrum of natural processes in aquatic systems. A synthesis of the whole is then used to suggest where and how ^{234}Th may still be fruitfully employed in the future.

Details of ^{234}Th speciation, methodology or modeling will not be focused on here but can be found in the companion papers of Santschi et al. (2005), Rutgers van der Loeff

et al. (2005), Buesseler et al. (2005) and Savoye et al. (2005) in this issue. Excellent reviews on the use of other uranium and thorium series radionuclides have already been written (e.g., Goldberg, 1954; Broecker and Peng, 1982; Cochran and Masqué, 2003; and Swarzenski et al., 2003) and readers are encouraged to examine these sources as well.

2. ^{234}Th Metadata

In 1967, Bhat et al. (1969) were the first to measure the activity of ^{234}Th in a marine system. By 2004, nearly 240 papers dealing with the application of ^{234}Th in aquatic environments were published in refereed journals. A timeline showing the evolution of ^{234}Th applications in aquatic systems is shown in Figure 2. It is interesting to note that as many ^{234}Th papers have been published in the last three years (2002 – 2004) as were published in the first 22 years (1969 – 1990) following Bhat et al.'s original study.

All of the ^{234}Th studies shown can be broken down into four distinct themes or “super-groups”: 1) *vertical transport*, 2) *particle cycling*, 3) *sediment dynamics* and 4) *horizontal transport* studies (Figure 2). Of the 237 papers included in this survey, 43% can be classified as vertical transport studies, or studies which focus on the downward flux of particles in the water column. Particle cycling studies, which look at rates of particle transformation, account for 19% of all ^{234}Th application papers. Studies looking at the horizontal transport of particles (and particle-reactive elements) account for nearly 10% of all ^{234}Th application papers, and studies of sediment dynamics, which look at transport within the upper sediment layers and particle resuspension in bottom waters,

account for the remaining 28% of the papers in this survey. Of the four major themes, the vertical transport studies have clearly driven the rapid increase in the number of ^{234}Th related publications in recent years.

The four themes or super-groups have been further divided into 12 categories. *Vertical transport* is divided into: 1) mass scavenging and flux, 2) mass component flux and 3) sediment trap calibration. The theme of *particle cycling* is divided into: 4) reaction rates, 5) colloids and 6) biological processes. *Horizontal transport* is divided into three categories by area: 7) open and coastal system, 8) estuary and river and 9) groundwater. Finally, the theme of *sediment dynamics* is divided into: 10) sediment deposition/accumulation, 11) sediment mixing and 12) sediment resuspension. A chronography or catalog of all studies within each of the 12 categories, and a reference to each study, is presented in Figures 3 and 4 and Table 1. It is interesting to note that, on this scale, the influence of individual scientific programs on research direction and productivity is clearly demonstrated (e.g., the Joint Global Ocean Flux Study [JGOFS, 1988–2003] and its effect on the number of vertical flux studies, Figure 3a).

3. ^{234}Th Applications in Aquatic Systems

This review is intended to serve as a primer on the application of ^{234}Th in aquatic systems. In the section that follows, each of the 12 process topics or categories identified in Figures 3 and 4 are briefly examined with regard to how ^{234}Th has helped us to further understand each of these processes. The objectives and sampling requirements of a small

number of studies in each category are given as an example of what can be done and the breadth of ^{234}Th research currently in the literature.

Issues of modeling and measurement methodologies are not critical here, as we are more interested in the potential of ^{234}Th applications in aquatic systems, not their current limitations. Also absent are any critical assessments of this body of literature (i.e., the papers listed in Table 1). Instead, critical assessments of any particular ^{234}Th study can often be found in the successive literature of its theme or category as shown in Figures 3 and 4.

3.1. Vertical Transport

Vertical transport here refers to the vertical movement of particles in the water column. As such this group is comprised of studies that examine total mass and specific component (e.g., particulate organic carbon [POC]) scavenging and flux. Also in this group, are those studies which have used ^{234}Th to calibrate sediment traps, which are passive collectors of sinking particles.

Bhat et al.'s (1969) seminal paper on ^{234}Th demonstrated that ^{234}Th activities were lower near the continental margins and reached secular equilibrium (i.e., equal activity) with its long-lived conservative parent, ^{238}U , at relatively shallow depths (50-200m). ^{234}Th was subsequently widely used as a particle “scavenging” tracer to examine sinking particle flux (Bacon and Anderson, 1982; Kaufman et al., 1981; Santschi et al., 1979; Tsunogai et al., 1986). In the simplest application of ^{234}Th , one can use the difference between the expected activity distribution of ^{234}Th in the water column, which is assumed

to be in secular equilibrium with its soluble parent ^{238}U , with what is actually measured. The difference in expected versus measured activity must be due to that portion of ^{234}Th that is removed by scavenging onto sinking particles. Numerous models that include or neglect temporal and/or physical terms have been used to examine this removal in detail (see Savoye et al., 2005 in this issue).

It wasn't until the 1980's that the link between biological processes and the extent of ^{234}Th removal was clearly demonstrated in a series of papers by Coale and Bruland (Coale and Bruland, 1985; Coale and Bruland, 1987). They proposed that a relationship existed between ^{234}Th removal rates and primary and new production in the open ocean.

During the US JGOFS North Atlantic Bloom Experiment (NABE), a clear relationship was found between the onset of the spring bloom, the subsequent drawdown of nutrients and carbon dioxide, and the net removal of ^{234}Th (Buesseler et al., 1992). It was postulated that if the ^{234}Th export rate could be quantified (from the ^{234}Th activity balance), then POC or PON export fluxes via sinking particles could be determined simply by multiplying the ^{234}Th export rate by the measured ratio of ^{234}Th to POC or PON on sinking particles. Since then, this empirical approach has been applied in numerous systems (see review by Buesseler, 1998; and Buesseler et al., 2005 in this issue) and applied to other elemental fluxes, such as particulate inorganic carbon (Bacon et al., 1996), polycyclic aromatic hydrocarbons (PAHs) (Gustafsson et al., 1997a), polychlorinated biphenyls (PCBs); Gustafsson et al., 1997b), biogenic silica (Buesseler et al., 2001; Rutgers van der Loeff et al., 2002) and trace metals (Weinstein and Moran, 2005).

Comparisons between the thorium derived particle flux method and fluxes determined using particles collected with shallow (i.e., 100 – 200 meter deep) sediment traps, demonstrated that there was a difference between the two techniques (e.g., Buesseler, 1991). Although ^{234}Th as a flux tracer has many assumptions and issues (see, e.g., Santschi et al., 2005; Rutgers van der Loeff et al., 2005; Buesseler et al., 2005 and Savoye et al., 2005 in this issue), it has been regarded by many to be a more robust estimate of particle flux than sediment traps (Buesseler et al., submitted), as traps may suffer from hydrodynamic biases (e.g., Gust et al., 1996) and concerns regarding the alteration of material within the trap itself (e.g., Gardner, 2000; Buesseler et al., submitted). As a result, ^{234}Th has continued to be used in concert with sediment traps to calibrate sediment traps for under or over collection of sinking particles from the upper ocean.

In essence, one can determine if sediment traps are over-collecting or under-collecting material by comparing the ^{234}Th flux determined from water column $^{234}\text{Th}/^{238}\text{U}$ disequilibria with the ^{234}Th flux captured and measured directly in the sediment traps. If water column derived ^{234}Th fluxes are higher than those measured in the sediment trap, then one can assume that the sediment traps are under-collecting material and vice versa. Sediment trap fluxes can then be corrected assuming that there is no preferential capture or loss of particles carrying ^{234}Th relative to other elements. One must also make sure to make the flux comparison over the same depth interval (i.e., one would compare the integrated $^{234}\text{Th}/^{238}\text{U}$ disequilibria over a depth interval of 100 meters with sediment traps placed at 100 meters depth). Moreover, one must realize that sediment traps and ^{234}Th integrate flux over different time scales and that there are several inherent assumptions in

the steady state model used to derive ^{234}Th water column fluxes (see Savoye et al., 2005 in this issue).

While many open ocean studies have calculated the flux of ^{234}Th out of the euphotic zone, a few studies have also considered the distribution of the corresponding excess ^{234}Th activity in deeper waters, which can be used to calculate rates of remineralization. This excess activity may be dispersed over such a wide depth interval that the excess is too small to differentiate from the background ^{234}Th activity. However, in some cases the remineralization of settling particles has been observed as excess ^{234}Th activity at intermediate depths in the water column (e.g., 300 – 1000 meters; Murray et al., 1989; Usbeck et al., 2002; Savoye et al., 2004).

3.2. Particle Cycling

Given the particle reactive nature of thorium, and the presence of multiple thorium isotopes (e.g., ^{234}Th , ^{228}Th , ^{230}Th , ^{232}Th), ^{234}Th has been used to examine particle cycling within the water column in detail (see Savoye et al., 2005 in this volume). In 1975, Matsumoto used a simple 1-box scavenging model that described the activity of ^{234}Th in the water column as a balance between ^{234}Th production from ^{238}U , decay and scavenging, using a first order rate constant, k . The inverse of k is hence, the residence time of ^{234}Th with respect to scavenging. Matsumoto (1975) used this model to determine a residence time of thorium in the Pacific Ocean of ~ 150 days. Results were consistent with other thorium isotopes (e.g., $^{228}\text{Th}/^{228}\text{Ra}$) and subsequent ^{234}Th studies that estimated thorium residence times of less than 10 days in the nearshore to 300 days

in surface waters of the open ocean (Broecker et al., 1973; Kaufman et al., 1981). Unfortunately, while these simple models provide estimates of total thorium residence times, they give no information on sorption kinetics or particle dynamics explicitly. Because each thorium isotope has a different half-life, however, differences in the distribution of multiple isotopes between the dissolved and particulate phases or between various particle class-sizes can be used to constrain estimates of particle dynamics via a system of equations.

The development of two box models, one comprised of the dissolved phase and the other of large particles, significantly advanced the understanding of particle dynamics within the ocean (Krishnaswami et al., 1976; Nozaki et al., 1981). In 1982, Bacon and Anderson used multiple thorium isotopes (^{234}Th , ^{228}Th , and ^{230}Th) to examine the distribution of thorium in the water column. They found that the distribution of thorium between dissolved and particulate phases was only consistent if reversible exchange between solids and solution occurred (e.g., a release of particulate thorium to the dissolved phase).

The idea of reversible exchange of thorium has since been supported by other investigations, and more complex models have been developed which incorporate not only multiple particle boxes of varying size, but variations in aggregation/disaggregation, adsorption/desorption, remineralization, sinking rate and so on (e.g., Bacon et al., 1985; Clegg and Whitfield, 1991; Burd et al., 2000). The mechanism of interaction between dissolved and particle phases is usually classified as adsorption and desorption, whereas the interaction between small and large particles are typically classified as aggregation and disaggregation. Increasing model complexity increases the number of model

unknowns and while inclusion of multiple thorium isotopes will help to constrain rate constants, more often simplifying assumptions must be made.

One of the conundrums of these early particle aggregation models was that the scavenging rate constants derived from field studies were significantly larger (0.2 -1.2 y^{-1}) than those predicted in the laboratory ($<0.1 y^{-1}$, Bacon and Anderson, 1982; Honeyman et al., 1988). This led to the recognition that colloids – small particles ranging in size from 1 nm to 1 μm that are typically not collected using standard water filtration techniques – may play an important role in the scavenging of metals.

In the Brownian Pumping model (Honeyman and Santschi, 1989; Burd et al., 2000), dissolved thorium rapidly scavenges onto colloidal particles that randomly collide and aggregate to form larger and larger particle size classes. Although measurement of colloids is difficult and requires careful methodological evaluation (e.g., Buesseler et al., 1996), a number of studies have used ^{234}Th to examine the relative distribution and residence time of metals in the colloidal phase. For example, Baskaran et al. (1992) and Moran and Buesseler (1992, 1993) determined that colloids comprised a substantial portion of the particulate ^{234}Th pool with a residence time of ~ 10 days when using a 3 box (i.e., dissolved ($<10\text{-kD}$), colloidal ($>10\text{-kD} - 0.45 \mu\text{m}$) and particulate ($>0.45 \mu\text{m}$)) model with irreversible exchange. These short residence times have since been confirmed by further studies (e.g., Santschi et al., 1995; Guo et al., 1997), suggesting that colloids do act as reactive intermediates in metal adsorption.

Several studies with ^{234}Th imply that colloidal interactions may be even more complex than previously thought. For example, Baskaran et al. (1992) proposed that only a small portion of the colloid pool actively participates in scavenging. Niven et al. (1995)

and Dai and Benitez-Nelson (2001) have argued that colloids are comprised of a range of chemical components and that ^{234}Th may trace only the biologically “active” colloidal pool. Quigley et al. (2001, 2002) suggest that ^{234}Th more strongly complexes with colloidal organic matter than with minerals and that the reversible scavenging implied from earlier studies is due to colloidal aggregation and disaggregation (see review by Santschi et al., 2005 in this volume).

In addition to simple aggregation and disaggregation, biological processes play a significant role in particle formation, specifically via grazing, and biologically mediated remineralization. In the pelagic environment, Krishnaswami et al. (1985) measured a suite of metals and excess ^{234}Th activities in salps, salp fecal pellets and several other zooplankton species in order to determine how important these filter feeding species were in repackaging small particles into large rapidly sinking fecal pellets. Schmidt et al. (1992) looked at dissolved and particulate ^{234}Th activities and phyto- and zooplankton biomass to determine what correlation there was between the density of zooplankton and the ^{234}Th -based export of particles from the upper water column. More recently, several studies have looked at the effects of animal grazing on trace metal cycling. For example, Barbeau et al. (2001), used ^{234}Th (in conjunction with ^{59}Fe , and ^{14}C) in the laboratory to measure the transformation and fate of bacterial carbon and particle bound metals ingested by several different protist species.

In benthic environments, ^{234}Th has been used to examine food quality and as an analog for trace metal and contaminant uptake (see section 3.4 below). Lauerma et al. (1997) measured excess ^{234}Th and ^{210}Pb in sea cucumbers, sediment trap material, seafloor detrital aggregates and surface sediment. The radio chronometers were then

used to model the apparent age of material within each sea cucumber gut as well as what percentage of the total vertical mass flux the species could process at population densities observed within the study site. In a similar study, Miller et al. (2000) measured chlorophyll *a* and excess ^{234}Th activity in surface sediment, sediment trap material and the gut contents of a number of benthic deposit feeding species and examined the correlation between feeding selectivity and species mobility as well as the percentage of the vertical mass flux consumed by these deposit feeders. Based on the high levels of excess ^{234}Th activity typically found in the gut contents of deposit feeders, Demopoulos et al. (2003) went on to look at potential tracers of deposit feeder food quality and measured correlations between excess ^{234}Th activity and chlorophyll *a*, amino acids, adenosine triphosphate, total organic carbon and nitrate in surface sediments.

3.3. Horizontal Transport

^{234}Th has also been used to study the horizontal transport of materials in systems where rapid advective flow or differences in end member chemistries present new challenges and opportunities for the use of radionuclide tracers. On the simplest level, Gustafsson et al. (2000) has measured the extent of $^{234}\text{Th}/^{238}\text{U}$ disequilibrium in water samples collected along the course of a brackish estuary and argued that if only a small disequilibrium existed at each of the stations along the estuary, then there could be little or no net removal of mass and other biogeochemically important materials either.

More complicated studies of horizontal transport have examined ^{234}Th inventories in both the sediments and overlying water column. In the absence of horizontal

advection and diffusion, water column deficiencies in ^{234}Th activity should in theory equal the inventories of excess ^{234}Th found in the underlying sediments (where excess ^{234}Th refers to that which is not supported by ^{238}U decay within sediments). Differences between the inventory of excess ^{234}Th in the sediment and the deficiency of ^{234}Th activity in the water column indicate that lateral transport processes must have occurred (Aller et al. 1980). For example, McKee et al (1984) found that ^{234}Th inventories in the sediment on the Yangtze continental shelf exceeded the possible supply from the overlying water column by an order of magnitude, indicating sediment focusing onto the shelf. Hirschberg et al. (1996) repeatedly measured excess ^{234}Th inventories in surface sediments at a set of stations along the major axis of an estuary and compared the inventories to the amount of ^{234}Th produced in the overlying water column. Fluctuations in the ratio of ^{234}Th production to deposition at each station served as an indicator of the temporal variability of short-term sediment deposition and transport along the major axis of the estuary.

When ^{234}Th is used alone, however, it is difficult to distinguish sediment focusing (i.e., the actual movement of sediment from one area to another) from the advection of dissolved ^{234}Th into areas of high particle loads and subsequent scavenging (i.e., boundary scavenging.). For this reason (and others), more complicated studies of horizontal transport have involved the use of multiple tracers. In an estuary contaminated by a point source of anthropogenic radionuclides, Mudge et al. (1997) measured $^{234}\text{Th}/^{137}\text{Cs}$ and $^{234}\text{Th}/^{239, 240}\text{Pu}$ ratios in surface sediments and were able to identify areas within the estuary where sediment deposition occurred only during low river flow, areas of intermittent deposition between tidal cycles as well as areas of more permanent

sediment deposition. Other studies have paired ^{234}Th measurements with measurements of ^7Be ($t_{1/2} = 53.3$ days) or ^{210}Pb ($t_{1/2} = 22.3$ years) which both enter the ocean predominantly via the atmosphere (Turekian et al., 1983). In combination, the means by which these nuclides arrived on the seafloor can be identified. For example, if sediment inventories of ^{234}Th and ^7Be differ from that measured in the water column, but have constant ratios, then one can assume either sediment winnowing (depletion relative to source) or sediment focusing (excess relative to source). In contrast, if the ratio of $^7\text{Be}/^{234}\text{Th}$ in sediments increases substantially over that measured from the water column, then boundary scavenging has occurred. For example, in Casco Bay in the Gulf of Maine, Gustafsson et al. (1998) measured water column and sediment activities of ^{234}Th , ^7Be , and ^{210}Pb and found that sediment inventories exceeded the overlying source waters by as much as 300 % as one moved from the center of the Gulf towards the coast. Sediment ratios of $^7\text{Be}/^{234}\text{Th}$ more than tripled. Gustafsson et al. (1998) concluded that boundary scavenging was the dominant transport mechanism of these radionuclides to the inner shelf sediments over short timescales. They further used this data to calibrate their water column transport models, which examined the dispersion of particle-reactive contaminants (e.g., hydrophobic organics) away from point sources along the coast (Gustafsson et al., 1998).

Because water column ^{234}Th activities decrease with decreasing salinity as one moves inland (due to decreasing activities of ^{238}U), while atmospherically derived ^7Be inputs over the same area remain relatively constant, measurements of ^{234}Th and ^7Be in suspended and bottom sediments can also be used to evaluate the relative importance of local sediment resuspension as well as the direction of sediment advection through an

estuary. When Feng et al. (1999a,b) measured ^7Be and ^{234}Th in the Hudson River estuary and determined high $^{234}\text{Th}/^7\text{Be}$ ratios in the upper estuary during low discharge, they reasoned that lower estuary particles (higher salinity) are transported up estuary (lower salinity) by as much as 10 to 20 km, comparable to the tidal excursion. In contrast, during high discharge, $^{234}\text{Th}/^7\text{Be}$ activity ratios were found to decrease, indicating either down-river transport of particles or enhanced resuspension. Furthermore, similarities between the $^{234}\text{Th}/^7\text{Be}$ activity ratios of suspended and bottom sediments suggested that as much as 30 % of the resuspended particles in the turbidity maximum were derived from particles transported during the flood-tide.

In other multiple tracer studies, Corbett et al. (2004) measured ^{210}Pb , ^{137}Cs , ^7Be and ^{234}Th activities on suspended solids and surface sediments in a deltaic region of a river and, by observing differences in the nuclide activity ratios over time, was able to determine the distance that river-borne materials travel before initial deposition as well as the residence time of these materials before they are resuspended and advected further offshore. Porcelli et al. (2001) explored the possibility of utilizing measured ratios of $^{234}\text{Th}/^{230}\text{Th}$ in river water to determine the transit time of thorium (and by inference, other particle reactive materials) through an entire watershed.

Only a few measurements of ^{234}Th in groundwater have been made. However, these few studies have yielded much information. Because ^{234}Th is particle reactive, a calculation of its residence time (in the dissolved state) in groundwater can be used as a proxy for the mobility or migratory behavior of other particle reactive radionuclides, metals, or toxins. Measured ^{234}Th activities in groundwater tend to be significantly less than that expected from in situ production via the radioactive decay of dissolved ^{238}U

with $^{234}\text{Th}/^{238}\text{U}$ activity ratios typically less than 0.3. This would correspond to a mean dissolved ^{234}Th residence time of less than 5 days if dissolved ^{238}U were the only source (Hussain and Krishnaswami, 1980; Krishnaswami et al., 1982; Baskaran et al., 1986; Tricca et al., 2001). However, Krishnaswami et al. (1982) utilized ^{222}Rn as a proxy to estimate the recoil supply rates of ^{234}Th and other uranium series radionuclides to groundwater. Their results suggested that the direct recoil supply of ^{234}Th from solid phase ^{238}U was the main contributor of ^{234}Th while decay from dissolved ^{238}U was negligible. The residence time of ^{234}Th calculated using an irreversible adsorption model with recoil as the major source was found to be significantly less (i.e., on the order of several minutes) than that determined when using dissolved ^{238}U as the major source of ^{234}Th (see also Kigoshi, 1971; and Tricca et al., 2001).

3.4. Sediment Dynamics

An understanding of sediment dynamics, particularly in the coastal ocean, is essential for elucidating sediment accumulation rates and the uptake and release of particle reactive contaminants to and from the overlying water column. In the simplest sense, ^{234}Th is often used in studies of sediment dynamics to pinpoint recent sedimentation or the depth to which recent particle reworking has occurred (either through bioturbation or physical resuspension and mixing). The short half-life of ^{234}Th (i.e., 24.1 days) implies that excess activity in the sediments must be associated with freshly (re)deposited material (i.e., less than several months old).

Excess ^{234}Th (see above) is most easily characterized in coastal waters, where ^{234}Th : ^{238}U disequilibrium and particle scavenging rates are high relative to offshore sites (Bhat et al., 1969; Kaufman et al., 1981). Profiles of excess ^{234}Th typically show mixing rates in the upper cm of the seafloor ranging from ~ 1 to $50 \text{ cm}^2 \text{ y}^{-1}$ in estuarine and slope sediments (Aller and Cochran, 1976; Aller et al., 1980; Sun et al., 1994; Green et al., 2002) to $>10 \text{ cm}^2 \text{ y}^{-1}$ in the deep sea (Aller and DeMaster, 1984). It has been hypothesized that since ^{234}Th scavenging often occurs with increased biological activity (e.g., Buesseler, 1998), it tends to be associated with fresh, and presumably more labile organic material. Thus, it is possible that particles containing ^{234}Th are preferentially consumed by benthic organisms (e.g., Smith et al., 1993; Miller et al., 2000). If true, then ^{234}Th may be used as a tracer of organic matter utilization and particle processing. This needs to be examined in more detail (see end of section 3.2).

Early measurements of ^{234}Th in the coastal environment showed that ^{234}Th was rapidly removed from the lower water column near the seafloor (Aller and Cochran, 1976). Santschi et al. (1979) presented a model where this removal was explained by the continuous resuspension of bottom sediments. The link between the depletion of ^{234}Th in bottom waters and the enrichment of ^{234}Th in surface sediments was described in a model integrating the resuspension cycle with bioturbation (Rutgers van der Loeff and Boudreau, 1997). Other studies have used ^{234}Th to document sediment resuspension in areas such as the Yangtze (McKee et al., 1984) and Amazon estuaries (McKee et al., 1986), the Amazon Shelf (Aller et al., 2004), the Irish (Kershaw and Young, 1988) and Laptev Seas (Kuptsov et al., 1999) and the Fram Strait (Rutgers van der Loeff et al., 2002b) to name a few. Initial results implied that there is a continuous cycling of

resuspension and settling, with fine suspended particles having relatively long residence times of ~50 days (Rose, 1994; Kershaw and Young, 1988; Bacon and Rutgers van der Loeff, 1989). In the Laptev sea, Kuptsov et al. (1999) determined ^{234}Th settling rates and concluded that particles were resuspended 2.3 times before settling. An extreme case of this settling and resuspension cycle is found in the Amazon Shelf where Aller et al. (2004) found a deep penetration of various tracers including ^{234}Th in the fluid mud layer. They attributed this to the rigorous exchange between the suspended sediments and the fluid mud layer, which they described as “a massive suboxic batch reactor”.

In the studies above resuspension was assumed to be a steady state process. Smoak et al. (1996) studied time variations in ^{210}Pb and ^{234}Th in the Amazon shelf and suggested that the exchange between the seabed inventories and water column inventories is controlled by the spring-neap cycle. Indeed, resuspension is often an episodic or even a catastrophic process where major events are caused by severe storms (Green et al., 1995). Giffin and Corbett (2003) used the changes with time of sediment inventories of ^{234}Th and ^7Be to quantify the effect of such resuspension events. The results of both tracers were mutually supportive. In other areas this approach could be complicated by changes in the inventory by other reasons like changing plankton input which has been invoked to explain large fluctuations in ^{234}Th inventories in the Iberian margin (Schmidt et al., 2002a).

4. Future Applications of ^{234}Th in Aquatic Systems

The previous sections have provided a brief overview of the research that is ongoing within the ^{234}Th community. Predicting exactly where and how ^{234}Th will be used in the future is no easy task. As such, we have chosen to focus on a general overview of current questions in three aquatic systems, with the understanding that the processes that work in one system may generally apply to other systems as well. These three systems are: i) the upper 1000 m of the open ocean, ii) the benthic zone of the coastal ocean and iii) large freshwater lakes. All have been the subject of recent workshops convened to identify future research needs. A sampling of key research questions and how ^{234}Th might be used to help answer these questions is presented below.

4.1. Open Pelagic Ocean

Much of the ^{234}Th work that has been conducted in the open ocean has been done in relation to or through the auspices of the JGOFS program, which recently concluded after nearly two decades of research. Among the primary goals of the Joint Global Ocean Flux Study (JGOFS) Program was the need to “determine and understand on a global scale the processes controlling the time-varying fluxes of carbon and associated biogenic elements in the ocean” (Bowles and Livingston, 1997). While significant progress has been made toward this goal, a number of key questions remain.

One area of interest concerns the mechanisms that control particle decomposition or preservation in the water column. The bioavailability of organic matter for heterotrophic organisms controls the rates at which bioactive elements are regenerated.

While freshly produced biomass is readily digested by marine heterotrophs, more aged organic material accumulates in both dissolved and particulate pools, which exhibit a refractory character (e.g., Druffle et al., 1992; Benner, 2002). The temporal evolution of these pools has been empirically modeled, but the fundamental biological and chemical mechanisms that control the observed rates of decomposition remain enigmatic. Since ^{234}Th is often associated with sinking organic matter both on particle surfaces as well as within particles, examining the rates at which ^{234}Th is remineralized to the water column relative to organic C and other tracers may help us understand the possible roles of inorganic surfaces in protecting otherwise labile organic constituents from remineralization (Mayer, 1994) or how the chemical and physical formation of organic aggregates (e.g., marine snow; Alldredge and Jackson, 1995) affects particle flux.

Inherent in this understanding is the role that particles play in mediating behavior of organic and inorganic constituents in seawater. Recent evidence suggests many trace elements, in addition to iron, have an active role in regulating primary production and species composition (Morel and Price, 2003). ^{234}Th has already provided insight into the role of organic complexation (e.g., Quigley et al., 2002) and how colloids modify particle coagulation (see section 3.2). Further study of ^{234}Th complexation may elucidate how specific ligands modify metal behavior (see Santschi et al., 2005 in this volume).

Finally, ^{234}Th , to date, has most often been used to determine surface ocean export of POC from the base of the euphotic zone at ~100 to 150 m depth (e.g., Buesseler, 1998). Future studies should also quantify the export of other elements (e.g., N, Si, Fe and other trace metals), which play a great role in the global ocean and, ultimately, climate regulation. While future work of this sort is valuable, particularly with regard to

examining episodic phenomena (e.g., storms, dust events, mesoscale eddies) in controlling surface ocean export, ²³⁴Th may also provide insight into processes that occur deeper in the water column (i.e., in the mesopelagic “twilight zone” at ~100 to 1000 meters depth). Several studies have already begun to investigate this research avenue (see section 3.1) and we feel that this area may be fertile ground for examining rates of particle formation and remineralization at depth once particulate and dissolved size classes are measured at high resolution.

4.2. Coastal Benthic Processes

Land-sea exchanges at the ocean margins influence biogeochemical cycles to an extent much more than their areal coverage might imply. Processes that occur disproportionately in margin environments, such as organic matter decomposition and burial, mineral formation, and denitrification affect the oceanic balances of many elements. Because of the proximity of sediments to the euphotic zone, these environments are also unique in the sense that sedimentary processes can influence pelagic processes. In 2004, the Coastal Ocean Processes (CoOP) Program conducted a workshop in Coastal Benthic Exchange Dynamics (CBED) to discuss pressing research needs in the coastal benthic zone and to formalize and prioritize research questions for the future (Reimers et al., 2004). This workshop identified a number of areas of future research including horizontal transport of material, sediment resuspension, groundwater and pore water flow. ²³⁴Th may be used to help answer some of these pressing research questions. Although studies are limited, ²³⁴Th has been shown to be an effective tracer of

horizontal sediment transport, particularly in combination with other radionuclides (e.g., ^7Be and ^{210}Pb , see section 3.3). The power of using a particle reactive radionuclide is that these processes are averaged over the radioisotope's mean life, which is equivalent to ~ 35 days for ^{234}Th . Thus, with proper time-series spatial measurements, ^{234}Th could be a useful tool for examining horizontal sediment transport on seasonal and event (e.g., storm) driven timescales (e.g., Gustafsson et al., 1998; Giffin and Corbett, 2003; Kersten et al., 2005).

A specific example of where ^{234}Th may be particularly useful is in examining the role of the estuarine turbidity maximum (ETM) in adsorbing and or releasing carbon, pollutants and other constituents. The ETM is an area of temporary sediment accumulation with diurnal and semidiurnal tidally forced resuspension events. Thus, a time series of ^{234}Th inventories of the seabed and water column could be used to understand the residence time of these materials and to even investigate the adsorption/desorption processes occurring during resuspension events (see section 3.4).

An important aspect of understanding coastal ecosystem nutrient dynamics focuses on the ultimate source of nutrients released from coastal sediments: is it from the recycling of organic matter deposited from the overlying water column, or is it from the outflow of groundwater through permeable sediments?

One mechanism for delineating the source of nutrients is an assessment of whether the sediment contains fresh organic material. In productive coastal systems (i.e., where water column $^{234}\text{Th}/^{238}\text{U}$ activity ratios < 1), sediments with excess ^{234}Th can be assumed to contain material deposited relatively recently (within a few months) and nutrient efflux may be related to organic matter remineralization, groundwater outflow,

or a combination of both. If, on the other hand, nutrient efflux is observed from sediments with little to no excess ^{234}Th , this would make a groundwater source more likely.

It should also be noted that a fresh supply of organic matter is not limited to the particle rain on accumulating sediments or to the ingestion of particles by suspension feeding organisms. Permeable coastal sediments are ventilated by currents and tides (Riedl et al., 1972; Shum and Sundby, 1996) providing a mechanism for the introduction of fresh material into the sediment bed (Huettel et al., 1996), a process that can make the sediment into a biocatalytic sand filter (Huettel et al., 1998; Huettel 2002) even in the absence of net sediment accumulation. The importance of this process, which is theme of a current EU Project COSA (Coastal Sands as Biocatalytical Filters), has been demonstrated with ^{210}Pb (Bacon et al., 1994). It has also been observed for ^{234}Th (Epping and Van Raaphorst, unpublished results), which is better suited to study this process on a seasonal time scale.

If pore water and groundwater flow is important, then understanding the timing of this input is essential. Moore et al. (2002) and Moore and Wilson (2005) have suggested that tides cause significant fluid exchange in the upper several meters of the seabed. Recent studies of ^{234}Th in groundwater suggest that ^{234}Th can be used to estimate residence times on the order of hours to days (see section 3.3). Thus, rapid time series measurements of ^{234}Th within sediment pore waters and groundwater may provide insight into ventilation pathways and on how groundwater flow and pore water ventilation, and possibly chemical fluxes, vary over short timescales.

4.3. Large Lakes

Large lakes are in many ways oceanic in scale, and yet much easier to constrain. Large lake process rates are also in many cases much higher than those found in marine systems and are thus easier to measure with finer resolution. In spite of this potential, the application of ^{234}Th to study process rates in large lakes, and freshwater in general, is an underdeveloped area. In 2003, a workshop was held to discuss the status of large lake research, which resulted in the Science of Freshwater Inland Seas (SoFIS) Report (Johnson, 2003). The report outlined the need for research in three major categories: i) circulation dynamics, ii) biology and chemistry, and iii) geology. Although ^{234}Th activities can be more difficult to measure when ^{238}U activities are low, methods currently exist for measuring ^{234}Th in almost all freshwater systems (Rutgers van der Loeff et al., 2005 in this volume). Thus, ^{234}Th has great potential for advancing current knowledge in many of the same areas discussed above as well as in the three major areas outlined at the SoFIS Workshop.

In terms of circulation dynamics and the effect they have on large lake processes, ^{234}Th can be used to great benefit in at least three areas of study: thermal bars, mesoscale eddies, and episodic events. Thermal bars develop along the shore of high latitude lakes before the onset of summer stratification. While the general circulation patterns of these thermal structures are understood, the magnitude of vertical transport associated with thermal bar convection is unknown. Thus, ^{234}Th inventories in the water and lakebed may be used to quantify the thermal bar's effect on benthic mixing (see section 3.4), as well as changes in productivity and vertical export (see section 3.1) (e.g., Klump et al.,

2003). The recent discovery of mesoscale eddies in Lake Superior (Ralph, 2002) may have major implications for nutrient dynamics and biological productivity in large lake systems. We suggest that ^{234}Th could be further used to examine the vertical particle and elemental flux associated with eddy age as well as define how mesoscale eddies affect annual productivity and the carbon budget in large lakes (Bidigare et al., 2003; Sweeney et al., 2004). The effects of storms and other episodic events on large lake transport and productivity have only recently been studied (CoOP EEGLE Program in Lake Michigan). ^{234}Th -based estimates of basin-wide offshore sediment transport (Waples et al., 2004) are in good agreement with $^{210}\text{Pb}/^{137}\text{Cs}$ -based estimates of sediment accumulation in the central basin of Lake Michigan. ^{234}Th -based estimates of alongshore sediment transport (Klump et al., in progress) also appear to be in good agreement with transport estimates based on water column transmissometer and current meter data (see section 3.3). Other applications of ^{234}Th might include lagrangian studies of productivity within resuspended sediment plumes (see section 3.1).

There is currently no routinely used short timescale particle-reactive radio-chronometer in freshwater science. In the fields of biology and chemistry, many of the ^{234}Th applications that have already been successfully used in marine systems (see e.g., sections 3.1 and 3.2) have yet to be tried in a freshwater setting. For example, particle scavenging rates of the exotic invasive zebra mussel (*Dreissena polymorpha*) seem particularly suited for study with ^{234}Th (Kryger and Riisgard, 1988; Fahnenstiel et al., 1995; Vanderploeg et al., 2002). By constraining estimates of carbon cycling in the photic zone (see section 3.1) and the age of labile colloidal carbon (see section 3.2), ^{234}Th might also help to resolve the apparent paradox of net heterotrophy in large lakes and the

role of allochthonous DOC in large lake metabolism (Cole, 1999; Del Giorgio et al., 1999; Biddanda and Cotner, 2002).

In terms of geological processes, many large tectonic lakes have sediment deposits over 4 km deep. These sediments provide a unique record of both regional and global climate forcing over periods that range from orbital (glacial-interglacial) to seasonal timescales. ^{234}Th -based studies linking process dynamics in the water column with the sedimentary strata should include measurements of trace metal flux from the water column (Weinstein and Moran, 2005, see section 3.1) as well as measurements of surface sediment mixing or storm related disturbances (e.g., Schmidt et al., 2002a,b; Giffin and Corbet, 2003; see section 3.4). ^{234}Th inventories could also be used to study the fate and transport of river-borne materials as they enter the lake or to measure rapid sedimentation rates resulting from hypopycnal flows (e.g., Walsh and Nittrouer, 2003; see sections 3.3 and 3.4).

5. Conclusions

Over the past several decades, ^{234}Th has proven to be an extremely useful tracer of particle dynamics and particle flux in the open ocean. However, this radionuclide has the potential to help us understand many other processes as well. In an age when many believe that further advances in our understanding of aquatic systems will rest primarily with autonomous instrumented arrays, satellite sensors, and more powerful models (e.g., Wurtsbaugh et al., 2002), it is our belief that the field of environmental radiochemistry and the use of ^{234}Th in particular should not be overlooked or underestimated.

Here, we have shown how ^{234}Th may be used to advance our understanding of the kinetics of a wide variety of biological, chemical and physical aquatic processes. We have broken down our discussion of some of the key applications of ^{234}Th along four major themes: a) vertical transport, b) particle cycling, c) sediment dynamics and d) horizontal transport. Each theme is further subdivided into twelve categories with specific references to the measurement of a1) mass scavenging and flux; a2) mass component flux; a3) sediment trap calibration; b4) particle reaction rates; b5) colloid dynamics; b6) biological processes; horizontal transport within an c7) open and coastal system, c8) estuary and river and c9) groundwater; d10) sediment /deposition/accumulation; d11) sediment mixing and d12) sediment resuspension. Suggestions on how these ^{234}Th applications can be used to help us better understand some of the outstanding questions in the open ocean, coastal benthic environment and large freshwater lakes are also given with the understanding that new applications of ^{234}Th are possible in nearly every aquatic system.

The effort involved in measuring ^{234}Th (see Rutgers van der Loeff et al., 2005 in this issue) can be large (especially in freshwater systems), however, the information gathered is, in many instances, unattainable by any other means. ^{234}Th is still the fastest (i.e., shortest-lived) natural particle reactive chronometer in general use. Nanotechnology may perhaps someday replace ^{234}Th with a better substitute (i.e., a “smart” easily detected surface specific chronometer), but that day is not likely to come soon. In the mean time, improvements in sample handling, resolution and throughput are increasing the scope of opportunities where ^{234}Th can yield important information on the process kinetics of our planet’s aquatic systems.

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FIGURE CAPTIONS

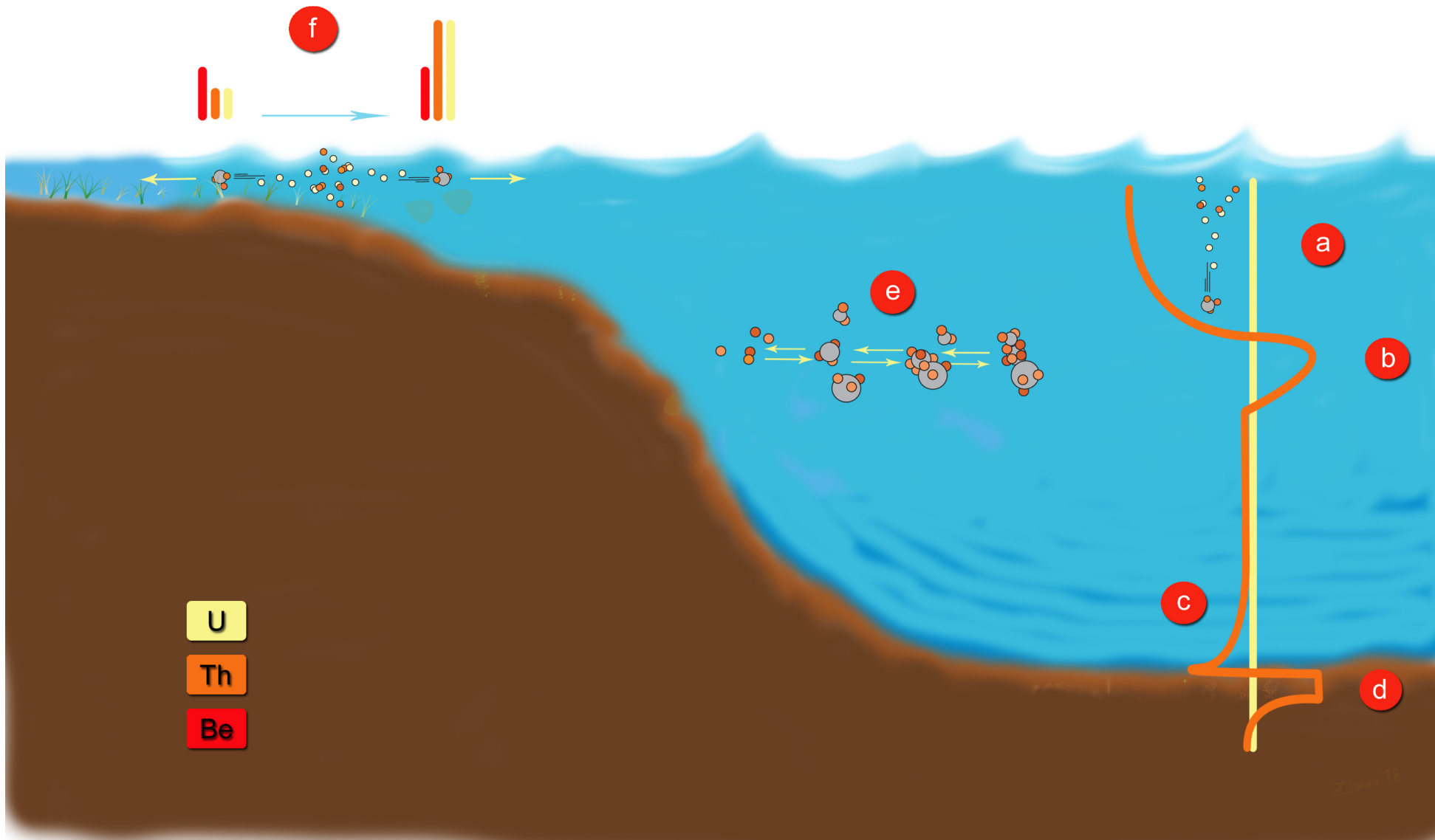
Figure 1. An overview of some of the major applications of ^{234}Th in aquatic systems: a) particle reactive ^{234}Th is removed from the upper water column by sinking particles. The mass flux out of the upper water column or the flux of any component of the total mass (e.g., carbon, nitrogen, silica, trace metals, PAHs, PCBs, etc.) can be modeled based on the extent of disequilibrium between ^{234}Th and its parent ^{238}U and the ratio of ^{234}Th to mass or any component of the total mass on the sinking particles. The calculated export of ^{234}Th can be used to calibrate sediment traps; b) Excess ^{234}Th in the mesopelagic zone can be modeled to determine rates of particle remineralization; c) near the benthic boundary layer, the scavenging of (or in some instances, excess) ^{234}Th can be used to model the rate of sediment resuspension; d) profiles of excess ^{234}Th in surface sediments can be used to model physical or biological mixing rates as well as rates of rapid sedimentation; e) by measuring the distribution of multiple thorium isotopes (e.g., ^{234}Th , ^{228}Th and ^{230}Th) on particles of differing sizes (e.g., large, small, colloidal), estimates of both adsorption/desorption and aggregation/disaggregation can be modeled; f) in riverine or estuarine environments, where there are differences in end member concentrations of ^{238}U (and production of ^{234}Th), sediment inventories of ^{234}Th can be compared with inventories of ^{234}Th depletion in the water column, and measured in tandem with a more uniformly deposited tracer (e.g., atmospherically derived ^7Be) to differentiate between sediment focusing and boundary scavenging as well as to derive rates of particle advection in both upstream and downstream directions.

Figure 2. A chronology of applied ^{234}Th studies in aquatic environments. A literature search revealed a total of 237 ^{234}Th application papers published in (primarily) refereed journals between the years of 1969 and 2004 (inclusive). ^{234}Th application papers are here defined as studies that apply the measurement of ^{234}Th to understand some natural process in an aquatic system. Book chapters, abstracts, technical reports and other “grey” literature were excluded from this search as were papers dealing primarily with methods and modeling, self-referencing papers (i.e., ^{234}Th minutiae for the ^{234}Th community) and secondary studies (e.g., a discussion of processes that may have been measured using ^{234}Th). Many of these excluded papers will be referenced in the companion papers of this special issue; however, it is implicit that some papers may have been accidentally overlooked. The division of all 237 studies into four process-related super-groups was based on the primary theme of each paper. If a single study devoted equal emphasis to several different processes, a subjective classification was made.

Figure 3. A chronology of ^{234}Th studies in aquatic environments: a) vertical transport studies are divided into categories of: i) mass scavenging and flux, ii) mass component flux and iii) sediment trap calibration; b) particle cycling studies are divided into categories of: i) reaction rates, ii) colloids and iii) biological processes. Numbers associated with each study box in the figure correspond to a similarly numbered citation in Table 1.

Figure 4. A chronology of ^{234}Th studies in aquatic environments: a) horizontal transport studies are divided into categories of: i) open and coastal systems, ii) estuaries and rivers

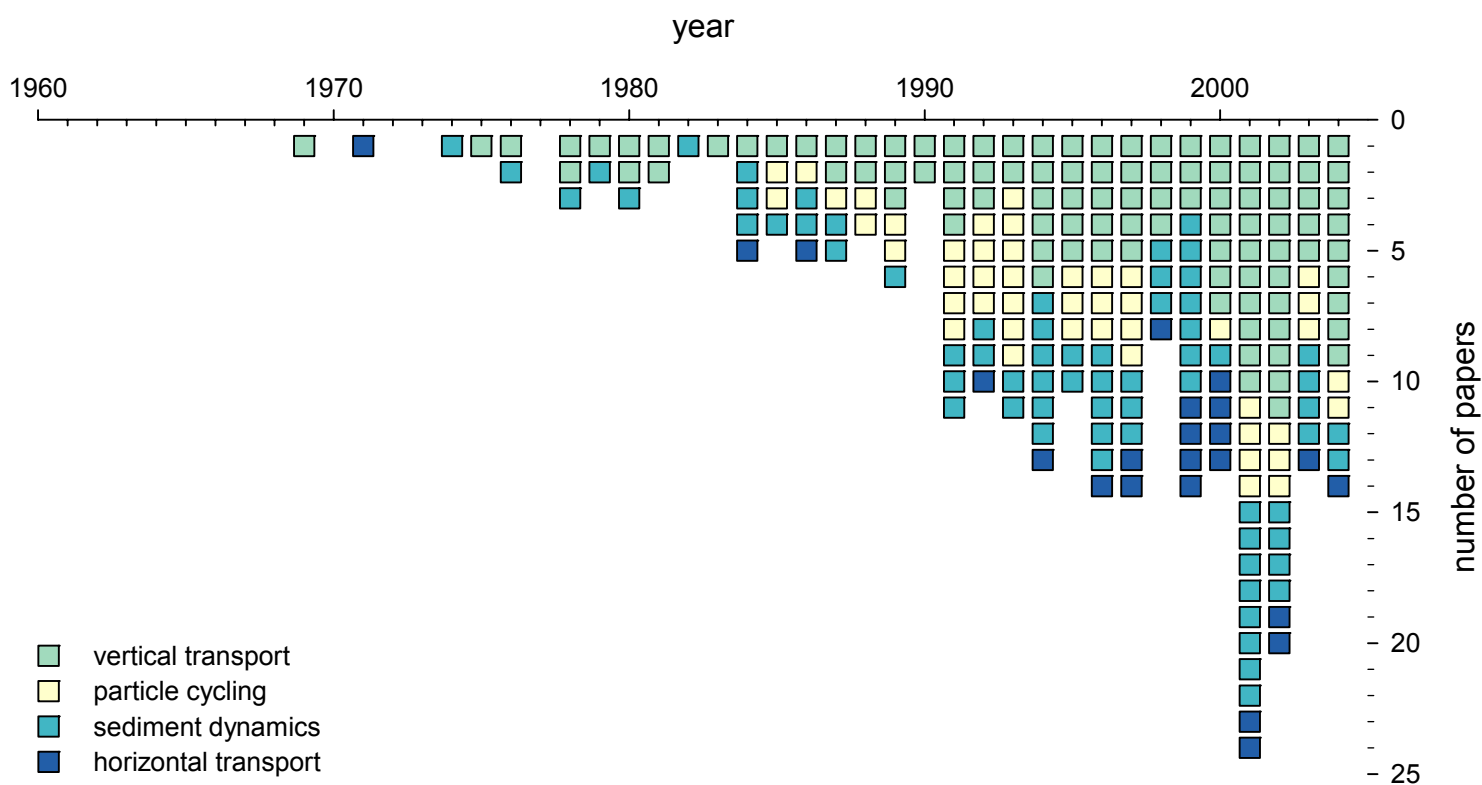
and iii) groundwater; b) sediment dynamics studies are divided into categories of: i) sediment deposition/accumulation (sedimentation), ii) sediment mixing and iii) sediment resuspension. Numbers associated with each study box in the figure correspond to a similarly numbered citation in Table 1.



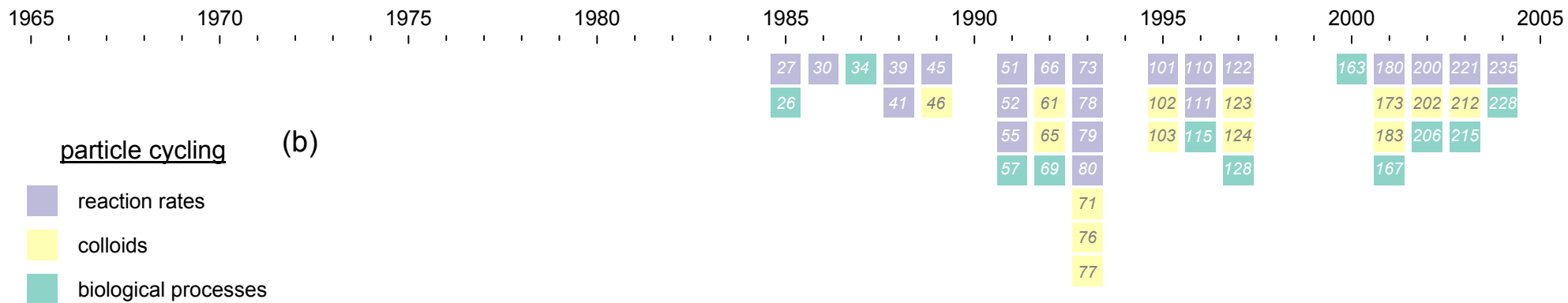
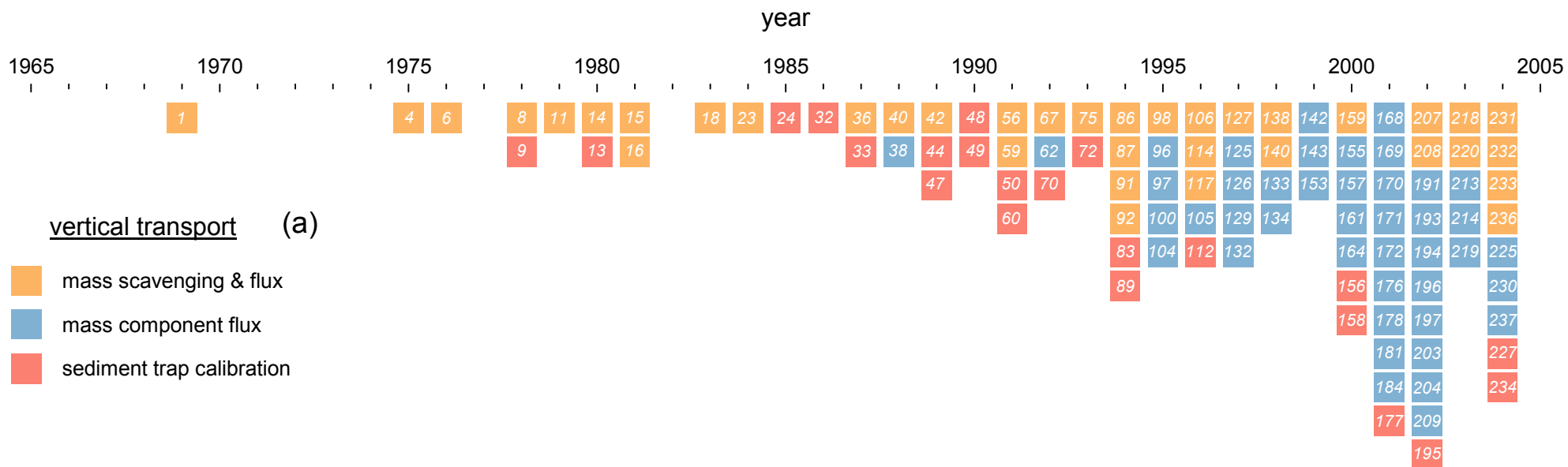
WAPLES FIGURE 1

Future Applications of ^{234}Th in Aquatic Environments

^{234}Th application papers: a catalog of the work completed: 1969 - 2004

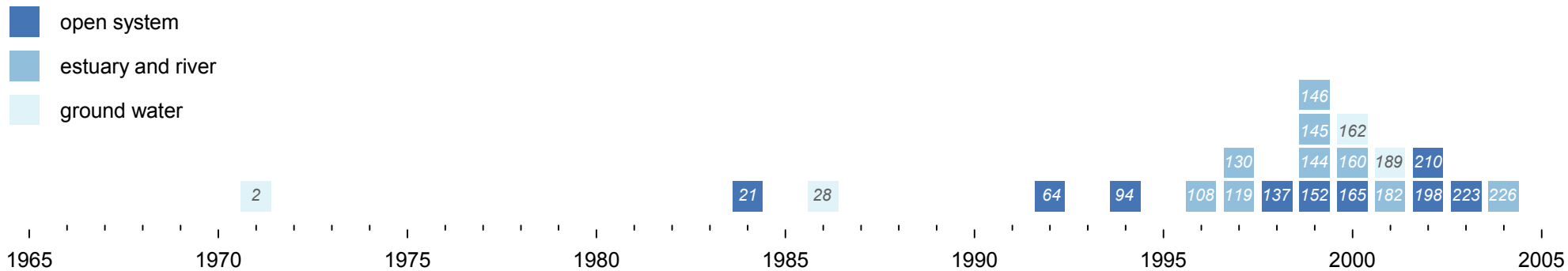


WAPLES FIGURE 2

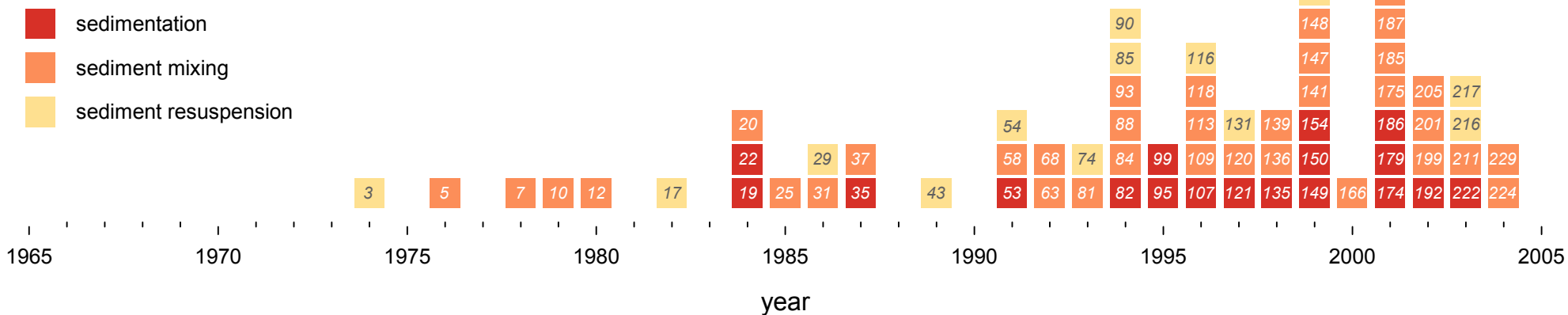


WAPLES FIGURE 3

horizontal transport (a)



sediment dynamics (b)



WAPLES FIGURE 4

TABLE 1. ²³⁴Th Application papers: 1969 to 2004*

1. Bhat et al. 1969
2. Kigoshi 1971
3. Amin et al. 1974
4. Matsumoto 1975
5. Aller and Cochran 1976
6. Krishnaswami et al. 1976
7. Aller 1978
8. Knauss et al. 1978
9. Spencer et al. 1978
10. Cochran and Aller 1979
11. Santschi et al. 1979
12. Aller et al. 1980
13. Brewer et al. 1980
14. Minagawa and Tsunogai 1980
15. Kaufman et al. 1981
16. Nozaki et al. 1981
17. Bacon and Anderson 1982
18. Tanaka et al. 1983
19. Aller and DeMaster 1984
20. Krishnaswami et al. 1984
21. McKee et al. 1984
22. Nittrouer et al. 1984
23. Santschi 1984
24. Coale and Bruland 1985
25. DeMaster et al. 1985
26. Krishnaswami et al. 1985
27. Moore and Hunter 1985
28. Baskaran et al. 1986
29. DeMaster et al. 1986
30. McKee et al. 1986
31. Silverberg et al. 1986
32. Tsunogai et al. 1986
33. Coale and Bruland 1987
34. Fisher et al. 1987
35. Huh and Beasley 1987
36. Huh et al. 1987
37. Martin and Sayles 1987
38. Fisher et al. 1988
39. Honeyman et al. 1988
40. Kershaw and Young 1988
41. Moore and Millward 1988
42. Bacon et al. 1989
43. Bacon and Rutgers van der Loeff 1989
44. Dominik et al. 1989
45. Honeyman and Santschi 1989
46. Moran and Moore 1989
47. Murray et al. 1989
48. Schmidt et al. 1990
49. Tsunogai et al. 1990
50. Buesseler 1991
51. Clegg et al. 1991
52. Clegg and Whitfield 1991
53. DeMaster et al. 1991
54. German et al. 1991
55. Lavelle et al. 1991
56. Lee et al. 1991
57. Levin et al. 1991
58. Sun et al. 1991
59. Rutgers van der Loeff and Berger 1991
60. Wei and Murray 1991
61. Baskaran et al. 1992
62. Buesseler et al. 1992
63. Keafer et al. 1992
64. McCartney et al. 1992
65. Moran and Buesseler 1992
66. Moran and Moore 1992
67. Sarin et al. 1992
68. Schaff et al. 1992
69. Schmidt et al. 1992
70. Wei and Murray 1992
71. Baskaran and Santschi 1993
72. Clegg and Whitfield 1993
73. Cochran et al. 1993
74. German and Sparks 1993
75. Huh et al. 1993
76. Lee et al. 1993
77. Moran and Buesseler 1993
78. Murray et al. 1993
79. Niven and Moore 1993
80. Rudnicki and Elderfield 1993
81. Smith et al. 1993
82. Bollhofer et al. 1994
83. Buesseler et al. 1994
84. DeMaster et al. 1994
85. Kadko et al. 1994
86. Kuptsov 1994
87. Kuptsov et al. 1994
88. Legeleux et al. 1994
89. Michaels et al. 1994
90. Rose et al. 1994
91. Sarin et al. 1994a
92. Sarin et al. 1994b
93. Sun et al. 1994
94. Thunell et al. 1994
95. Allison et al. 1995
96. Buesseler et al. 1995
97. Cochran et al. 1995a
98. Cochran et al. 1995b
99. Dukat and Kuehl 1995
100. Gulin et al. 1995
101. Guo et al. 1995.
102. Huh and Prah 1995
103. Santschi et al. 1995
104. Shimmield et al. 1995
105. Bacon et al. 1996
106. Baskaran et al. 1996
107. Bentley et al. 1996
108. Hirschberg et al. 1996
109. Hughes et al. 1996
110. Moore et al. 1996
111. Murnane et al. 1996
112. Murray et al. 1996
113. Pope et al. 1996
114. Sarin et al. 1996
115. Smith et al. 1996
116. Smoak et al. 1996
117. Vogler et al. 1996
118. Wheatcroft and Martin 1996
119. Assinder et al. 1997
120. Bentley and Nittrouer 1997

TABLE 1. ²³⁴Th Application papers: 1969 to 2004 (continued)

121. Bou-Rabee and Bem 1997
122. Dunne et al. 1997
123. Greenamoyer and Moran 1997
124. Guo et al. 1997
125. Gustafsson et al. 1997a
126. Gustafsson et al. 1997b
127. Langone et al. 1997
128. Lauerman et al. 1997
129. Moran et al. 1997
130. Mudge et al. 1997
131. Rutgers van der Loeff and Boudreau 1997
132. Rutgers van der Loeff et al. 1997
133. Buessler et al. 1998
134. Buessler 1998
135. Feng et al. 1998
136. Gerino et al. 1998
137. Gustafsson et al. 1998
138. Kersten et al. 1998
139. Smith et al. 1998
140. Wei and Hung 1998
141. Bentley and Nittrouer 1999
142. Charette and Moran 1999
143. Charette et al. 1999
144. Feng et al. 1999a
145. Feng et al. 1999b
146. Feng et al. 1999c
147. Fornes et al. 1999
148. Fuller et al. 1999
149. Jaeger and Nittrouer 1999a
150. Jaeger and Nittrouer 1999b
151. Kuptzov et al. 1999
152. Santschi et al. 1999
153. Smoak et al. 1999
154. Sommerfield et al. 1999
155. Benitez-Nelson et al. 2000
156. Buessler et al. 2000
157. Cochran et al. 2000
158. Dunne et al. 2000
159. Gulin 2000
160. Gustafsson et al. 2000
161. Hall et al. 2000
162. Luo et al. 2000
163. Miller et al. 2000
164. Moran and Smith 2000
165. Smoak et al. 2000
166. Turnewitsch et al. 2000
167. Barbeau et al. 2001
168. Baskaran 2001
169. Benitez-Nelson et al. 2001
170. Buessler et al. 2001
171. Cai et al. 2001
172. Charette et al. 2001
173. Dai and Benitez-Nelson 2001
174. Eckman et al. 2001
175. Fornes et al. 2001
176. Gustafsson et al. 2001
177. Hernes et al. 2001
178. Kim and Church 2001
179. Mulder et al. 2001
180. Muller et al. 2001
181. Nodder et al. 2001
182. Porcelli et al. 2001
183. Quigley et al. 2001
184. Sambrotto 2001
185. Santschi et al. 2001
186. Schmidt et al. 2001
187. Shull 2001
188. Sommerfield et al. 2001
189. Tricca et al. 2001
190. Turnewitsch and Springer 2001
191. Amiel et al. 2002
192. Anschutz et al. 2002
193. Cai et al. 2002
194. Chen et al. 2002
195. Coppola et al. 2002
196. Foster and Shimmield 2002
197. Friedrich and Rutgers van der Loeff 2002
198. Frignani et al. 2002
199. Green et al. 2002
200. Guo et al. 2002
201. Levin et al. 2002
202. Quigley et al. 2002
203. Schmidt et al. 2002a
204. Schmidt et al. 2002b
205. Schmidt et al. 2002c
206. Shull and Mayer 2002
207. Somayajulu et al. 2002
208. Usbeck et al. 2002
209. Rutgers van der Loeff et al. 2002a
210. Rutgers van der Loeff et al. 2002b
211. Alexander and Venherm 2003
212. Baskaran et al. 2003
213. Bidigare et al. 2003
214. Chen et al. 2003
215. Demopoulos et al. 2003
216. Giffin and Corbett 2003
217. Klump et al. 2003
218. Miller and Svaeren 2003
219. Moran et al. 2003
220. Radakovitch et al. 2003
221. Santschi et al. 2003
222. Walsh and Nittrouer 2003
223. Yamada and Aono 2003
224. Aller et al. 2004
225. Buessler et al. 2004
226. Corbett et al. 2004
227. Gustafsson et al. 2004
228. Ishikawa et al. 2004
229. Jahnke and Jahnke 2004
230. Palm et al. 2004
231. Roberts and Santschi 2004
232. Savoye et al. 2004
233. Smith et al. 2004
234. Smoak, et al. 2004
235. Trimble et al. 2004
236. Waples et al. 2004
237. Yang et al. 2004

*Numbers correspond to study ID numbers in Figures 3 and 4.