# 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 utility, however, has yet to be realized by many within the aquatic science community. 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.
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## 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 sys-

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tems. $^{234}$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 (Fig. 1).

In spite of the popularity of $^{234}$Th as a tracer, its use is still limited to a relatively narrow field of investigators and its utility has yet to be realized by many within the aquatic science community. 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 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. (this volume), Rutgers van der Loeff et al. (this volume), Buesseler et al. (this volume) and Savoye et al. (this volume) 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; 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 Fig. 2. It is interesting to note that as many $^{234}$Th papers have
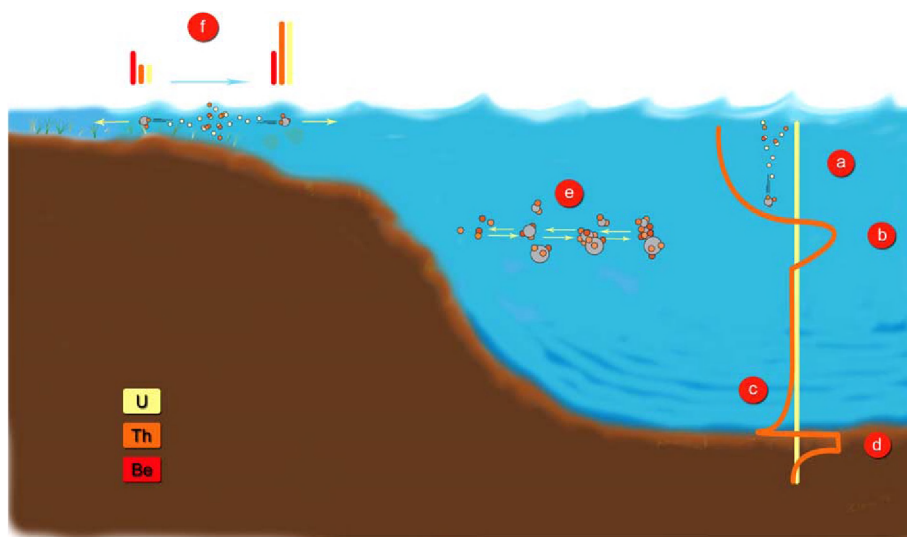


Fig. 1. An overview of some of the major applications of $^{234}$Th in aquatic systems. (a) Particle reactive $^{234}$Th is removed from the upper water column by sinking particles. The mass flux out of the upper water column or the flux of any component of the total mass (e.g., carbon, nitrogen, silica, 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 $^{7}$Be) to differentiate between sediment focusing and boundary scavenging as well as to derive rates of particle advection in both upstream and downstream directions.

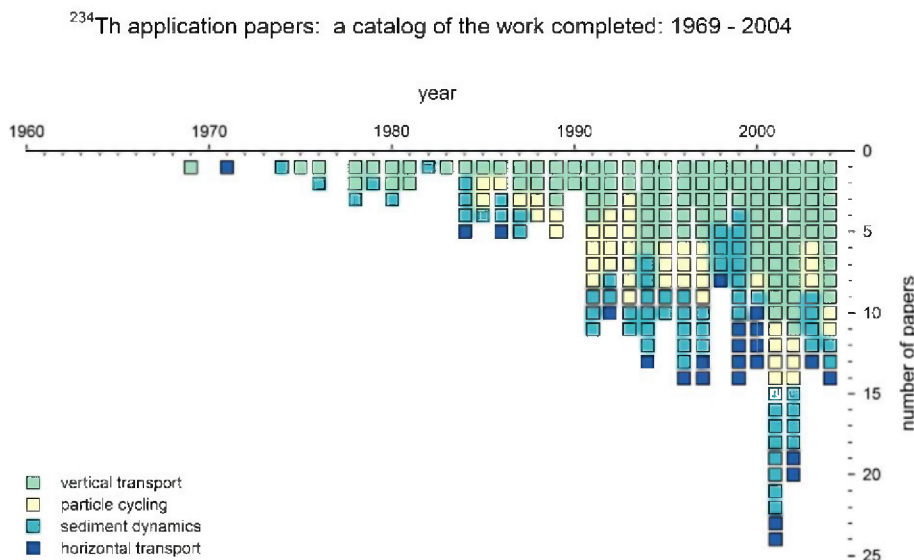## Future Applications of $^{234}$Th in Aquatic Environments

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



Fig. 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 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.

been published in the last 3 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 (Fig. 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 Figs. 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, Fig. 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 Figs. 3 and 4 are briefly examined with
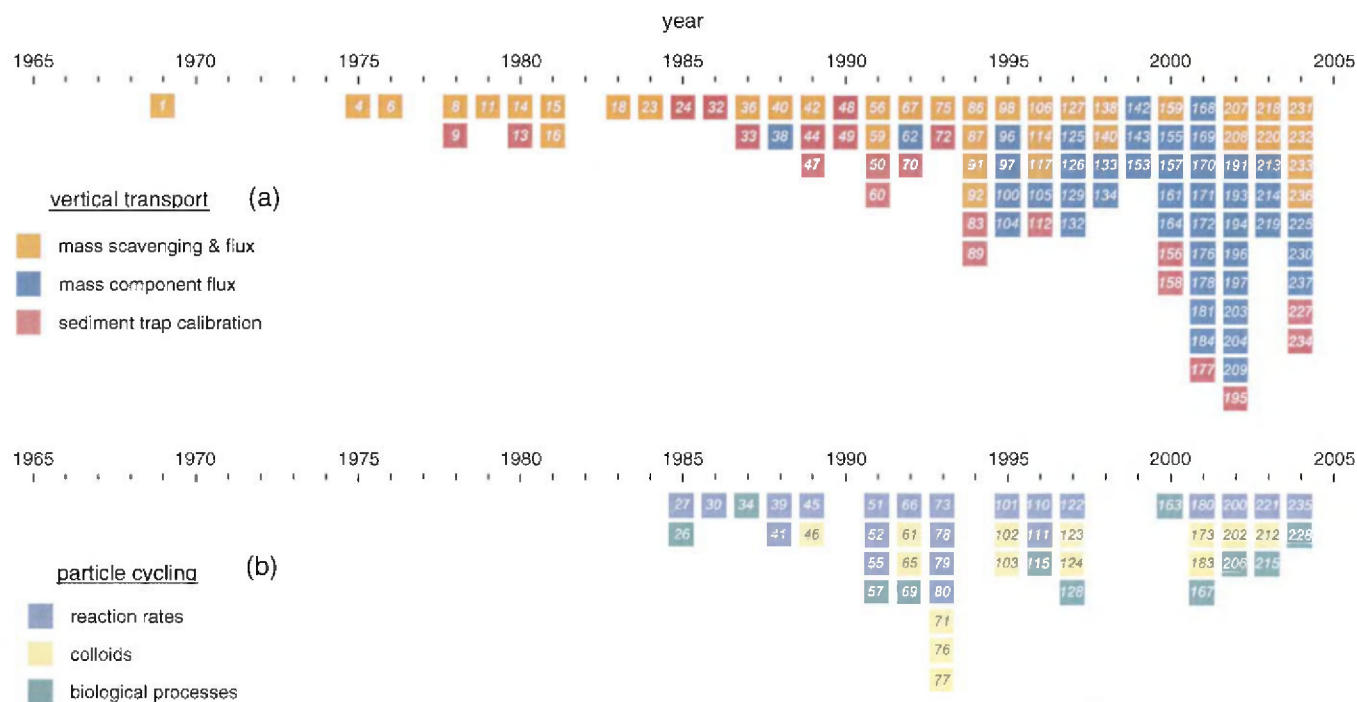
Fig. 3. A chronology of [234]Th studies in aquatic environments: (a) vertical transport studies are divided into categories of: (i) mass scavenging and flux, (ii) mass component flux and (iii) sediment trap calibration; (b) particle cycling studies are divided into categories of: (i) reaction rates, (ii) colloids and (iii) biological processes. Numbers associated with each study box in the figure correspond to a similarly numbered citation in Table 1.
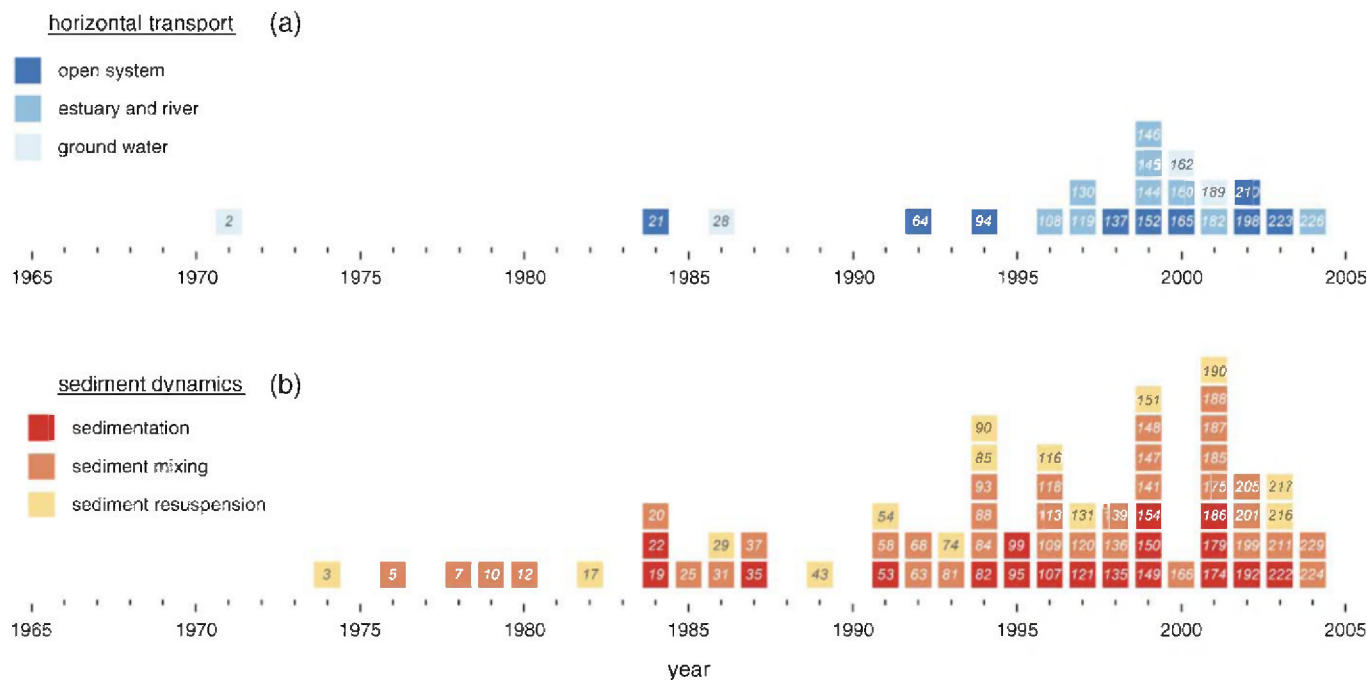
Fig. 4. A chronology of [234]Th studies in aquatic environments: (a) horizontal transport studies are divided into categories of: (i) open and coastal systems, (ii) estuaries and rivers and (iii) groundwater; (b) sediment dynamics studies are divided into categories of: (i) sediment deposition/accumulation (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.

Table 1
$^{234}$Th application papers: 1969–2004*

| | |
|---|---|
| 1. Bhat et al., 1969 | 61. Baskaran et al., 1992 |
| 2. Kigoshi, 1971 | 62. Buesseler et al., 1992 |
| 3. Amin et al., 1974 | 63. Keafer et al., 1992 |
| 4. Matsumoto, 1975 | 64. McCartney et al., 1992 |
| 5. Aller and Cochran, 1976 | 65. Moran and Buesseler, 1992 |
| 6. Krishnaswami et al., 1976 | 66. Moran and Moore, 1992 |
| 7. Aller, 1978 | 67. Sarin et al., 1992 |
| 8. Knauss et al., 1978 | 68. Schaff et al., 1992 |
| 9. Spencer et al., 1978 | 69. Schmidt et al., 1992 |
| 10. Cochran and Aller, 1979 | 70. Wei and Murray, 1992 |
| 11. Santschi et al., 1970 | 71. Baskaran and Santschi, 1993 |
| 12. Aller et al., 1980 | 72. Clegg and Whitfield, 1993 |
| 13. Brewer et al., 1980 | 73. Cochran et al., 1993 |
| 14. Minagawa and Tsunogai, 1980 | 74. German and Sparks, 1993 |
| 15. Kaufman et al., 1981 | 75. Huh et al., 1993 |
| 16. Nozaki et al., 1981 | 76. Lee et al., 1993 |
| 17. Bacon and Anderson, 1982 | 77. Moran and Buesseler, 1993 |
| 18. Tanaka et al., 1983 | 78. Murray et al., 1993 |
| 19. Aller and DeMaster, 1984 | 79. Niven and Moore, 1993 |
| 20. Krishnaswami et al., 1984 | 80. Rudnicki and Elderfield, 1993 |
| 21. McKee et al., 1984 | 81. Smith et al., 1993 |
| 22. Nittrouer et al., 1984 | 82. Bollhofer et al., 1994 |
| 23. Santschi, 1984 | 83. Buesseler et al., 1994 |
| 24. Coale and Bruland, 1985 | 84. DeMaster et al., 1994 |
| 25. DeMaster et al., 1985 | 85. Kadko et al., 1994 |
| 26. Krishnaswami et al., 1985 | 86. Kuptsov, 1994 |
| 27. Moore and Hunter, 1985 | 87. Kuptsov et al., 1994 |
| 28. Baskaran et al., 1986 | 88. Legeleux et al., 1994 |
| 29. DeMaster et al., 1986 | 89. Michaels et al., 1994 |
| 30. McKee et al., 1986 | 90. Rose et al., 1994 |
| 31. Silverberg et al., 1986 | 91. Sarin et al., 1994a |
| 32. Tsunogai et al., 1986 | 92. Sarin et al., 1994b |
| 33. Coale and Bruland, 1987 | 93. Sun et al., 1994 |
| 34. Fisher et al., 1987 | 94. Thunell et al., 1994 |
| 35. Huh and Beasley, 1987 | 95. Allison et al., 1995 |
| 36. Huh et al., 1987 | 96. Buesseler et al., 1995 |
| 37. Martin and Sayles, 1987 | 97. Cochran et al., 1995a |
| 38. Fisher et al., 1988 | 98. Cochran et al., 1995b |
| 39. Honeyman et al., 1988 | 99. Dukat and Kuehl, 1995 |
| 40. Kershaw and Young, 1988 | 100. Gulin et al., 1995 |
| 41. Moore and Millward, 1988 | 101. Guo et al., 1995 |
| 42. Bacon et al., 1989 | 102. Huh and Prahl, 1995 |
| 43. Bacon and Rutgers van der Loeff, 1989 | 103. Santschi et al., 1995 |
| 44. Dominik et al., 1989 | 104. Shimmield et al., 1995 |
| 45. Honeyman and Santschi, 1989 | 105. Bacon et al., 1996 |
| 46. Moran and Moore, 1989 | 106. Baskaran et al., 1996 |
| 47. Murray et al., 1989 | 107. Bentley et al., 1996 |
| 48. Schmidt et al., 1990 | 108. Hirschberg et al., 1996 |
| 49. Tsunogai et al., 1990 | 109. Hughes et al., 1996 |
| 50. Buesseler, 1991 | 110. Moore et al., 1996 |
| 51. Clegg et al., 1991 | 111. Murnane et al., 1996 |
| 52. Clegg and Whitfield, 1991 | 112. Murray et al., 1996 |
| 53. DeMaster et al., 1991 | 113. Pope et al., 1996 |
| 54. German et al., 1991 | 114. Sarin et al., 1996 |
| 55. Lavelle et al., 1991 | 115. Smith et al., 1996 |
| 56. Lee et al., 1991 | 116. Smoak et al., 1996 |
| 57. Levin et al., 1991 | 117. Vogler et al., 1996 |
| 58. Sun et al., 1991 | 118. Wheatcroft and Martin, 1996 |
| 59. Rutgers van der Loeff and Berger, 1991 | 119. Assinder et al., 1997 |
| 60. Wei and Murray, 1991 | 120. Bentley and Nittrouer, 1997 |

Table 1 (*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. Buesseler et al., 1998
134. Buesseler, 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. Buesseler 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. Buesseler 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. Buesseler 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 Figs. 3 and 4.

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 Figs. 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 (50–200 m). $^{234}$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., this volume in this issue).

It wasn't until the 1980s 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 (1985, 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; Buesseler et al., this volume) and applied to other elemental fluxes, such as particulate inorganic carbon (Bacon et al., 1996), polycyclic aromatic hydrocarbons (PAHs) (Gustafsson et al., 1997a), polychloronated biphenyls (PCBs) (Gustafsson et al., 1997b), biogenic silica (Buesseler et al., 2001; Rutgers van der Loeff et al., 2002a,b) and trace metals (Weinstein and Moran, 2005) (Fig. 1a).

Comparisons between the thorium derived particle flux method and fluxes determined using particles collected with shallow (i.e., 100–200 m 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., this volume; Rutgers van der Loeff et al., this volume; Buesseler et al., this volume; Savoye et al., this volume 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 for publication), 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 for publication). 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 $^{234}$Th/$^{238}$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 $^{234}$Th/$^{238}$U disequilibria over a depth interval of 100 m with sediment traps placed at 100 m 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 one-dimensional model that is commonly used to derive [234]Th water column fluxes (see Savoye et al., this volume, 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 (Fig. 1b). 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 m; 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., this volume in this issue). 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]Th/[228]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 (Fig. 1e).

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 Ander-

son 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 year$^{-1}$) than those predicted in the laboratory (<0.1 year$^{-1}$, 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 kD), colloidal (>10 kD–0.45 μm) and particulate (>0.45 μm)) 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., this volume, in this issue).

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). Lauerman 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 $^{234}$Th/$^{238}$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 scav-

enging.). 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 $^{234}$Th/$^{137}$Cs and $^{234}$Th/$^{239,\ 240}$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 $^{7}$Be ($t_{1/2}$ = 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 $^{7}$Be 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 $^{7}$Be/$^{234}$Th in sediments increases substantially over that measured from the water column, then boundary scavenging has occurred (Fig. 1f). For example, in Casco Bay in the Gulf of Maine, Gustafsson et al. (1998) measured water column and sediment activities of $^{234}$Th, $^{7}$Be, 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 $^{7}$Be/$^{234}$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 $^{7}$Be inputs over the same area remain relatively constant, measurements of $^{234}$Th and $^{7}$Be 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 $^{7}$Be and $^{234}$Th in the Hudson River estuary and determined high $^{234}$Th/$^{7}$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 10–20 km, comparable to the tidal excursion. In contrast, during high discharge, $^{234}$Th/$^{7}$Be

activity ratios were found to decrease, indicating either down-river transport of particles or enhanced resuspension. Furthermore, similarities between the $^{234}$Th/$^{7}$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, $^{7}$Be 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 $^{234}$Th/$^{230}$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 $^{234}$Th/$^{238}$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, Fig. 1d). The short half-life of $^{234}$Th (i.e., 24.1 days) implies that activity in excess of that supplied by $^{238}$U within 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 off-shore 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 cm$^2$ year$^{-1}$ in estuarine and slope sediments (Aller and Cochran, 1976; Aller et al., 1980; Sun et al., 1994; Green et al., 2002) to >10 cm$^2$ year$^{-1}$ 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) (Fig. 1c). 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 et al., 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 $^7$Be 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, however, is no easy task. Therefore, 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., Druffel 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. (this volume) in this issue).

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, $^{234}$Th may also provide insight into processes that occur deeper in the water column (i.e., in the mesopelagic "twilight zone" at ~100 to 1000 m 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. $^{234}$Th may be used to help answer some of these pressing research questions. Although studies are limited, $^{234}$Th has been shown to be an effective tracer of horizontal sediment transport, particularly in combination with other radionuclides (e.g., $^7$Be 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 inves-

tigate 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 $^{234}$Th/$^{238}$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, 2003) 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., this volume, in this issue). 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 $^{210}$Pb/$^{137}$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 Corbett, 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 hyperpycnal 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., this volume 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.

# References

Alexander, C.R., Venherm, C., 2003. Modern sedimentary processes in the Santa Monica, California continental margin: sediment accumulation, mixing and budget. Marine Environmental Research 56 (1–2), 177–204.

Alldredge, A.L., Jackson, G.A., 1995. Aggregation in marine systems. Deep-Sea Research 42, 1–8.

Aller, R.C., 1978. Experimental studies of changes produced by deposit feeders on pore water, sediment, and overlying water chemistry. American Journal of Science 278, 1185–1234.

Aller, R.C., Cochran, J.K., 1976. $^{234}$Th/$^{238}$U disequilibrium in nearshore sediment: particle reworking and diagenetic timescales. Earth and Planetary Science Letters 29, 37–50.

Aller, R.C., DeMaster, D.J., 1984. Estimates of particle flux and reworking at the deep-sea floor using $^{234}$Th/$^{238}$U disequilibrium. Earth and Planetary Science Letters 67, 308–318.

Aller, R.C., et al., 1980. Tracking particle-associated processes in nearshore environments by use of $^{234}$Th/$^{238}$U disequilibrium. Earth and Planetary Science Letters 47, 161–175.

Aller, R.C., Heilbrun, C., Panzeca, C., Zhu, Z.B., Baltzer, F., 2004. Coupling between sedimentary dynamics, early diagenetic processes, and biogeochemical cycling in the Amazon-Guianas mobile mud belt: coastal French Guiana. Marine Geology 208 (2–4), 331–360.

Allison, M.A., Nittrouer, C.A., Kineke, G.C., 1995. Seasonal sediment storage on mudflats adjacent to the Amazon river. Marine Geology 125 (3–4), 303–328.

Amiel, D., Cochran, J.K., Hirschberg, D.J., 2002. Th-234/U-238 disequilibrium as an indicator of the seasonal export flux of particulate organic carbon in the North Water. Deep-Sea Research II 49 (22–23), 5191–5209.

Amin, B.S., Krishnaswamy, S., Somayajulu, B.L.K., 1974. $^{234}$Th/$^{238}$U activity ratios in Pacific Ocean bottom waters. Earth and Planetary Science Letters 21, 342–344.

Anschutz, P., Jorissen, F.J., Chaillou, G., Abu-Zied, R., Fontanier, C., 2002. Recent turbidite deposition in the eastern Atlantic: early diagenesis and biotic recovery. Journal of Marine Research 60 (6), 835–854.

Assinder, D.J., Mudge, S.M., Bourne, G.S., 1997. Radiological assessment of the Ribble Estuary: 1. Distribution of radionuclides in surface sediments. Journal of Environmental Radioactivity 36 (1), 1–19.

Bacon, M.P., Anderson, R.F., 1982. Distribution of thorium isotopes between dissolved and particulate forms in the deep sea. Journal of Geophysical Research 87, 2045–2056.

Bacon, M.P., Rutgers van der Loeff, M.M., 1989. Removal of Th-234 by Scavenging in the Bottom Nepheloid Layer of the Ocean. Earth and Planetary Science Letters 92 (2), 157–164.

Bacon, M.P., Huh, C.A., Fleer, A.P., Deuser, W.G., 1985. Seasonality in the flux of natural radionuclides and plutonium in the deep Sargasso Sea. Deep-Sea Research 32, 273–286.

Bacon, M.P., Huh, C.A., Moore, R.M., 1989. Vertical profiles of some natural radionuclides over the Alpha Ridge, Arctic Ocean. Earth and Planetary Science Letters 95 (1), 15–22.

Bacon, M.P., Belastock, R.A., Bothner, M.H., 1994. 210Pb balance and implications for particle transport on the continental shelf, U.S. Middle Atlantic Bight. Deep-Sea Research. Part II 41 (2–3), 511–535.

Bacon, M.P., Cochran, J.K., Hirschberg, D., Hammar, T.R., Fleer, A.P., 1996. Export flux of carbon at the equator during the EqPac time-series cruises estimated from Th-234 measurements. Deep-Sea Research II 43 (4–6), 1133–1153.

Barbeau, K., Kujawinski, E.B., Moffett, J.W., 2001. Remineralization and recycling of iron, thorium and organic carbon by heterotrophic marine protists in culture. Aquatic Microbial Ecology 24 (1), 69–81.

Baskaran, M., 2001. Scavenging of thorium isotopes in the Arctic regions: implications for the fate of particle-reactive pollutants. Marine Pollution Bulletin 42 (1), 16–22.

Baskaran, M., Santschi, P.H., 1993. The role of particles and colloids in the transport of radionuclides in coastal environments of Texas. Marine Chemistry 43 (1–4), 95–114.

Baskaran, M., Krishnaswami, S., Bhandari, N., 1986. The geochemistry of uranium and thorium isotopes in the salt lakes and adjacent groundwaters of Rajasthan. Journal of the Geological Society of India 27, 90–101.

Baskaran, M., Santschi, P.H., Benoit, G., Honeyman, B.D., 1992. Scavenging of thorium isotopes by colloids in seawater of the Gulf of Mexico. Geochimica et Cosmochimica Acta 56 (9), 3375–3388.

Baskaran, M., Santschi, P.H., Guo, L.D., Bianchi, T.S., Lambert, C., 1996. Th-234:U-238 disequilibria in the Gulf of Mexico: the importance of organic matter and particle concentration. Continental Shelf Research 16 (3), 353–380.

Baskaran, M., Swarzenski, P.W., Porcelli, D., 2003. Role of colloidal material in the removal of Th-234 in the Canada basin of the Arctic Ocean. Deep-Sea Research I 50 (10–11), 1353–1373.

Benitez-Nelson, C.R., Buesseler, K.O., Crossin, G., 2000. Upper ocean carbon export, horizontal transport, and vertical eddy diffusivity in the southwestern Gulf of Maine. Continental Shelf Research 20 (6), 707–736.

Benitez-Nelson, C., Buesseler, K.O., Karl, D.M., Andrews, J., 2001. A time-series study of particulate matter export in the North Pacific Subtropical Gyre based on Th-234 : U-238 disequilibrium. Deep-Sea Research I 48 (12), 2595–2611.

Benner, R., 2002. Chemical composition and reactivity. In: Hansell, D., Carlson, C. (Eds.), Biogeochemistry of Marine Dissolved Organic Matter. Academic Press, New York, pp. 59–90.

Bentley, S.J., Nittrouer, C.A., 1997. Environmental influences on the formation of sedimentary fabric in a fine-grained carbonate-shelf environment: Dry Tortugas, Florida Keys. Geo-Marine Letters 17 (4), 268–275.

Bentley, S.J., Nittrouer, C.A., 1999. Physical and biological influences on the formation of sedimentary fabric in an oxygen-restricted depositional environment: Eckernforde Bay, southwestern Baltic Sea. Palaios 14 (6), 585–600.

Bentley, S.J., Nittrouer, C.A., Sommerfield, C.K., 1996. Development of sedimentary strata in Eckernforde Bay, southwestern Baltic Sea. Geo-Marine Letters 16 (3), 148–154.

Bhat, S.G., Krishnaswamy, S., Lal, D., Rama and Moore, W.S., 1969. $^{234}$Th/$^{238}$U ratios in the ocean. Earth and Planetary Science Letters 5, 483–491.

Biddanda, B., Cotner, J., 2002. Love handles in aquatic ecosystems: the role of dissolved organic carbon drawdown, resuspended sediments, and terrigenous inputs in the carbon balance of Lake Michigan. Ecosystems 5, 431–445.

Bidigare, R.R., et al., 2003. Influence of a cyclonic eddy on microheterotroph biomass and carbon export in the lee of Hawaii. Geophysical Research Letters 30 (6).

Bollhofer, A., et al., 1994. High-resolution Pb-210 dating of Lake Constance sediments-stable lead in Lake Constance. Environmental Geology 24 (4), 267–274.

Bou-Rabee, F., Bem, H., 1997. Natural radionuclides and cesium-137 content in Arabian Gulf bottom sediments. Journal of Radioanalytical and Nuclear Chemistry 222 (1–2), 219–221.

Bowles, M.C., Livingston, H., 1997. Assessing the ocean's role in the global carbon cycle: the joint global ocean flux study. McGraw-Hill's Yearbook of Science and Technology.

Brewer, P.G., Nozaki, Y., Spencer, D., Fleer, A.P., 1980. Sediment trap experiments in the deep North Atlantic: isotopic and elemental fluxes. Journal of Marine Research 38 (4), 703–728.

Broecker, W.S., Peng, T.-H., 1982. Tracers in the Sea. Eldigio Press, Palisades, NY.

Broecker, W.S., Kaufman, A., Trier, R.M., 1973. The residence time of thorium in surface sea water and its implication regarding the rate of reactive pollutants. Earth and Planetary Science Letters 20, 35–44.

Buesseler, K.O., 1991. Do upper-ocean sediment traps provide an accurate record of particle-flux. Nature 353 (6343), 420–423.

Buesseler, K.O., 1998. The decoupling of production and particulate export in the surface ocean. Global Biogeochemical Cycles 12 (2), 297–310.

Buesseler, K.O., Bacon, M.P., Cochran, J.K., Livingston, H.D., 1992. Carbon and nitrogen export during the Jgofs North-Atlantic Bloom Experiment estimated from Th-234u-238 Disequilibria. Deep-Sea Research I 39 (7–8A), 1115–1137.

Buesseler, K.O., Michaels, A.F., Siegel, D.A., Knap, A.H., 1994. A 3-dimensional time-dependent approach to calibrating sediment trap fluxes. Global Biogeochemical Cycles 8 (2), 179–193.

Buesseler, K.O., Andrews, J.A., Hartman, M.C., Belastock, R., Chai, F., 1995. Regional estimates of the export flux of particulate organic-carbon derived from Th-234 during the Jgofs eqpac program. Deep-Sea Research II 42 (2–3), 777.

Buesseler, K.O., et al., 1996. An intercomparison of cross-flow filtration techniques used for sampling marine colloids: overview and organic carbon results. Marine Chemistry 55, 1–31.

Buesseler, K., et al., 1998. Upper ocean export of particulate organic carbon in the Arabian Sea derived from thorium-234. Deep-Sea Research II 45 (10–11), 2461–2487.

Buesseler, K.O., et al., 2000. A comparison of the quantity and composition of material caught in a neutrally buoyant versus surface-tethered sediment trap. Deep-Sea Research I 47 (2), 277–294.

Buesseler, K.O., et al., 2001. Upper ocean export of particulate organic carbon and biogenic silica in the Southern Ocean along 170 degrees W. Deep-Sea Research II 48 (19–20), 4275–4297.

Buesseler, K.O., Andrews, J.E., Pike, S.M., Charette, M.A., 2004. The effects of iron fertilization on carbon sequestration in the Southern Ocean. Science 304 (5669), 414–417.

Buesseler, K.O. et al., this volume. An assessment of particulate organic carbon to thorium-234 ratios in the ocean and their impact on the application of $^{234}$Th as a POC flux proxy. Marine Chemistry.

Buesseler, K.O. Antia A.N., Chen, M., Fowler, S.W., Gardner, W.D., Gustaffson, Ö., Harada, K., Michaels, A.F., Rutgers van der Loeff, M., Sarin, M., Steinberg, D.K., Trull, T., submitted for publication. Estimating upper ocean particle fluxes with sediment traps: a progress report. Progress in Oceanography.

Burd, A.B., Jackson, G.A., Moran, S.B., 2000. A coupled adsorption-aggregation model for the POC/$^{234}$Th ratio of marine particles. Deep-Sea Research I 47, 103–120.

Cai, P.H., Huang, Y.P., Chen, M., Liu, G.S., Qiu, Y.S., 2001. Export of particulate organic carbon estimated from Th-234-U-238 disequilibria and its temporal variation in the South China Sea. Chinese Science Bulletin 46 (20), 1722–1726.

Cai, P.H., et al., 2002. New production based on Ra-228-derived nutrient budgets and thorium-estimated POC export at the intercalibration station in the South China Sea. Deep-Sea Research I 49 (1), 53–66.

Charette, M.A., Moran, S.B., 1999. Rates of particle scavenging and particulate organic carbon export estimated using Th-234 as a tracer in the subtropical and equatorial Atlantic Ocean. Deep-Sea Research II 46 (5), 885–906.

Charette, M.A., Moran, S.B., Bishop, J.K.B., 1999. Th-234 as a tracer of particulate organic carbon export in the subarctic northeast Pacific Ocean. Deep-Sea Research II 46 (11–12), 2833–2861.

Charette, M.A., Moran, S.B., Pike, S.M., Smith, J.N., 2001. Investigating the carbon cycle in the Gulf of Maine using the natural tracer thorium 234. Journal of Geophysical Research, [Oceans] 106 (C6), 11553–11579.

Chen, M., et al., 2002. Biological productivity and carbon cycling in the Arctic Ocean. Chinese Science Bulletin 47 (12), 1037–1040.

Clegg, S.L., Whitfield, M., 1991. A generalized-model for the scavenging of trace-metals in the open ocean: 2. Thorium scavenging. Deep-Sea Research I 38 (1), 91–120.

Clegg, S.L., Whitfield, M., 1993. Application of a generalized scavenging model to time-series Th-234 and particle data obtained during the Jgofs North-Atlantic bloom experiment. Deep-Sea Research I 40 (8), 1529–1545.

Clegg, S.L., Bacon, M.P., Whitfield, M., 1991. Application of a generalized scavenging model to thorium isotope and particle data at equatorial and high-latitude sites in the Pacific-Ocean. Journal of Geophysical Research, [Oceans] 96 (C11), 20655–20670.

Chen, M., Huang, Y.P., Cai, P.G., Guo, L.D., 2003. Particulate organic carbon export fluxes in the Canada Basin and Bering Sea as derived from Th-234/U-238 disequilibria. Arctic 56 (1), 32–44.

Coale, K.H., Bruland, K.W., 1985. $^{234}$Th:$^{238}$U disequilibria within the California current. Limnology and Oceanography 30, 22–33.

Coale, K.H., Bruland, K.W., 1987. Oceanic stratied euphotic zone as elucidated by $^{234}$Th:$^{238}$U disequilibria. Limnology and Oceanography 32, 189–200.

Cochran, J.K., Aller, R.C., 1979. Particle reworking in sediments from the New York Bight Apex: evidence from. 234 Th/238 U disequilibrium. Estuarine and Coastal Marine Science 9, 739–747.

Cochran, J.K., Masqué, P., 2003. Short-lived U/Th series radionuclides in the ocean: tracers for scavenging rates, export fluxes and particle dynamics. In: Bourdon, B., Henderson, G.M., Lundstrom, C.C., Turner, S.P. (Eds.), Uranium-Series Geochemistry, Reviews in Mineralogy and Geochemistry, vol. 52, pp. 461–492.

Cochran, J.K., Buesseler, K.O., Bacon, M.P., Livingston, H.D., 1993. Thorium isotopes as indicators of particle dynamics in the upper ocean- results from the Jgofs North-Atlantic Bloom Experiment. Deep-Sea Research I 40 (8), 1569–1595.

Cochran, J.K., Barnes, C., Achman, D., Hirschberg, D.J., 1995a. Thorium-234/Uranium-238 disequilibrium as an indicator of scavenging rates and particulate organic-carbon fluxes in the Northeast Water Polynya, Greenland. Journal of Geophysical Research, [Oceans] 100 (C3), 4399–4410.

Cochran, J.K., Frignani, M., Hirschberg, D.J., Barnes, C., 1995b. Thorium isotopes as indicators of scavenging rates in the Venice Lagoon. Marine and Freshwater Research 46 (1), 215–221.

Cochran, J.K., et al., 2000. Short-lived thorium isotopes (Th-234, Th-228) as indicators of POC export and particle cycling in the Ross Sea, Southern Ocean. Deep-Sea Research II 47 (15–16), 3451–3490.

Cole, J.J., 1999. Aquatic microbiology for ecosystem scientists: new and recycled paradigms in ecological microbiology. Ecosystems 2, 215–225.

Coppola, L., Roy-Barman, M., Wassmann, P., Mulsow, S., Jeandel, C., 2002. Calibration of sediment traps and particulate organic carbon export using Th-234 in the Barents Sea. Marine Chemistry 80 (1), 11–26.

Corbett, D.R., McKee, B., Duncan, D., 2004. An evaluation of mobile mud dynamics in the Mississippi River deltaic region. Marine Geology 209 (1–4), 91–112.

Crookes, W., 1900. Radio-activity of uranium. Proceedings of the Royal Society of London 66 (1899–1900), 409–423.

Dai, M.H., Benitez-Nelson, C.R., 2001. Colloidal organic carbon and Th-234 in the Gulf of Maine. Marine Chemistry 74 (2–3), 181–196.

Del Giorgio, P.A., Cole, J.J., Peters, R.H., 1999. Linking planktonic biomass and metabolism to net gas fluxes in northern temperate lakes. Ecology 80, 1422.

DeMaster, D.J., McKee, C.A., Nittrouer, D.C., Brewster, D.C., Biscaye, P.E., 1985. Rates of sediment reworking at the Hebble site based on measurement of $^{234}$Th, $^{137}$Cs and $^{210}$Pb. Marine Geology 66, 133–148.

DeMaster, D.J., Kuehl, S., Nittrouer, C.A., 1986. Effects of suspended sediments on geochemical processes near the mouth of the Amazon River: examination of biological silica uptake and the fate of particle-reactive elements. Continental Shelf Research 6 (1/2), 107–125.

DeMaster, D.J., Brewster, D.C., McKee, B.A., Nittrouer, C.A., 1991. Rates of particle scavenging, sediment reworking, and longitudinal ripple formation at the hebble site based on measurements of Th-234 and Pb-210. Marine Geology 99 (3–4), 423–444.

DeMaster, D.J., Pope, R.H., Levin, L.A., Blair, N.E., 1994. Biological mixing intensity and rates of organic-carbon accumulation in North-Carolina slope sediments. Deep-Sea Research, Part II: Topical Studies in Oceanography 41 (4–6), 735–753.

Demopoulos, A.W.J., Smith, C.R., DeMaster, D.J., Fornes, W.L., 2003. Evaluation of excess Th-234 activity in sediments as an indicator of food quality for deep-sea deposit feeders. Journal of Marine Research 61 (2), 267–284.

Dominik, J., Schuler, C., Santschi, P.H., 1989. Residence times of $^{234}$Th and $^{7}$Be in Lake Geneva. Earth and Planetary Science Letters 93 (3–4), 345–358.

Druffel, E.R.M., Williams, P.M., Bauer, J.E., Ertel, J.R., 1992. Cycling of dissolved and particulate organic matter in the open ocean. Journal of Geophysical Research 97, 15639–15659.

Dukat, D.A., Kuehl, S.A., 1995. Non-steady-state Pb-210 flux and the use of Ra-228/Ra-226 as a geochronometer on the Amazon Continental-Shelf. Marine Geology 125 (3–4), 329–350.

Dunne, J.P., Murray, J.W., Young, J., Balistrieri, L.S., Bishop, J., 1997. Th-234 and particle cycling in the central equatorial Pacific. Deep-Sea Research II 44 (9–10), 2049–2083.

Dunne, J.P., Murray, J.W., Rodier, M., Hansell, D.A., 2000. Export flux in the western and central equatorial Pacific: zonal and temporal variability. Deep-Sea Research I 47 (5), 901–936.

Eckman, J.E., et al., 2001. Performance of cages as large animal-exclusion devices in the deep sea. Journal of Marine Research 59 (1), 79–95.

Edwards, R.L., Gallup, C.D., Cheng, H., 2003. Uranium-series dating of marine and lacustrine carbonates. Reviews in Mineralogy and Geochemistry 52, 363–405.

Fahnenstiel, G.L., Lang, G.A., Nalepa, T.F., Johengen, T.H., 1995. Effects of zebra mussel (*Dreissena polymorpha*) colonization on water quality parameters in Saginaw Bay, Lake Huron. Journal of Great Lakes Research 211, 435–448.

Feng, H., Cochran, J.K., Hirschberg, D.J., Wilson, R.E., 1998. Small-scale spatial variations of natural radionuclide and trace metal distributions in sediments from the Hudson River estuary. Estuaries 21 (2), 263–280.

Feng, H., Cochran, J.K., Hirschberg, D.J., 1999a. Th-234 and Be-7 as tracers for the sources of particles to the turbidity maximum of the Hudson River estuary. Estuarine, Coastal and Shelf Science 49 (5), 629–645.

Feng, H., Cochran, J.K., Hirschberg, D.J., 1999b. Th-234 and Be-7 as tracers for the transport and dynamics of suspended particles in a partially mixed estuary. Geochimica et Cosmochimica Acta 63 (17), 2487–2505.

Feng, H., Cochran, J.K., Hirschberg, D.J., 1999c. Th-234 and Be-7 as tracers for transport and sources of particle-associated contaminants in the Hudson River Estuary. Science of the Total Environment 238, 401–418.

Fisher, N.S., Teyssie, J.L., Krishnaswami, S., Baskaran, M., 1987. Accumulation of Th, Pb, U, and Ra in marine-phytoplankton and its geochemical significance. Limnology and Oceanography 32 (1), 131–142.

Fisher, N.S., Cochran, J.K., Krishnaswami, S., Livingston, H.D., 1988. Predicting the oceanic flux of radionuclides on sinking biogenic debris. Nature 335, 622–625.

Fornes, W.L., DeMaster, D.J., Levin, L.A., Blair, N.E., 1999. Bioturbation and particle transport in Carolina slope sediments: a radiochemical approach. Journal of Marine Research 57 (2), 335–355.

Fornes, W.L., DeMaster, D.J., Smith, C.R., 2001. A particle introduction experiment in Santa Catalina Basin sediments: testing the age-dependent mixing hypothesis. Journal of Marine Research 59 (1), 97–112.

Foster, J.M., Shimmield, G.B., 2002. Th-234 as a tracer of particle flux and POC export in the northern North Sea during a coccolithophore bloom. Deep-Sea Research II 49 (15), 2965–2977.

Friedrich, J., Rutgers van der Loeff, M.M., 2002. A two-tracer (Po-210–Th-234) approach to distinguish organic carbon and biogenic silica export flux in the Antarctic Circumpolar Current. Deep-Sea Research I 49 (1), 101–120.

Frignani, M., Courp, T., Cochran, J.K., Hirschberg, D., Codina, L.V.I., 2002. Scavenging rates and particle characteristics in and near the Lacaze-Duthiers submarine canyon, northwest Mediterranean. Continental Shelf Research 22 (15), 2175–2190.

Fuller, C.C., van Geen, A., Baskaran, M., Anima, R., 1999. Sediment chronology in San Francisco Bay, California, defined by Pb-210, Th-234, Cs-137, and Pu-239,Pu-240. Marine Chemistry 64 (1–2), 7–27.

Gardner, W.D., 2000. Sediment trap sampling in surface waters. In: Hanson, R.B., Ducklow, H.W., Field, J.G. (Eds.), The Changing Ocean Carbon Cycle: A Midterm Synthesis of the Joint Global Ocean Flux Study. Cambridge University Press, pp. 240–281.

Gerino, M., et al., 1998. Comparison of different tracers and methods used to quantify bioturbation during a spring bloom: 234-thorium, luminophores and chlorophyll a. Estuarine, Coastal and Shelf Science 46 (4), 531–547.

German, C.R., Sparks, R.S.J., 1993. Particle recycling in the tag hydrothermal plume. Earth and Planetary Science Letters 116 (1–4), 129–134.

German, C.R., Fleer, A.P., Bacon, M.P., Edmond, J.M., 1991. Hydrothermal scavenging at the Mid-Atlantic Ridge-radionuclide distributions. Earth and Planetary Science Letters 105 (1–3), 170–181.

Giffin, D., Corbett, D.R., 2003. Evaluation of sediment dynamics in coastal systems via short-lived radioisotopes. Journal of Marine Systems 42 (3–4), 83–96.

Goldberg, E.D., 1954. Marine chemistry: 1. Chemical scavengers of the sea. Journal of Geology 62, 249–265.

Green, M.O., et al., 1995. Storm sediment transport: observations from the British North Sea shelf. Continental Shelf Research 15, 889–912.

Green, M.A., Aller, R.C., Cochran, J.K., Lee, C., Aller, J.Y., 2002. Bioturbation in shelf/slope sediments off Cape Hatteras, North Carolina: the use of Th-234, Chl-a, and Br to evaluate rates of particle and solute transport. Deep-Sea Research II 49 (20), 4627–4644.

Greenamoyer, J.M., Moran, S.B., 1997. Investigation of Cd, Cu, Ni and Th-234 in the colloidal size range in the Gulf of Maine. Marine Chemistry 57 (3–4), 217–226.

Gulin, S.B., 2000. Seasonal changes of Th-234 scavenging in surface water across the western Black Sea: an implication of the cyclonic circulation patterns. Journal of Environmental Radioactivity 51 (3), 335–347.

Gulin, S.B., et al., 1995. Study of seasonal dynamics of the suspended matter, nutrients and pollutants scavenging out of the Black-Sea surface from 1992 to 1994. Geokhimiya 6, 863–873.

Guo, L.D., Santschi, P.H., Baskaran, M., Zindler, A., 1995. Distribution of dissolved and particulate Th-230 and Th-232 in seawater from the Gulf-of-Mexico and Off Cape-Hatteras as measured by sims. Earth and Planetary Science Letters 133 (1–2), 117–128.

Guo, L.D., Santschi, P.H., Baskaran, M., 1997. Interactions of thorium isotopes with colloidal organic matter in oceanic environments. Colloids and Surfaces. A, Physicochemical and Engineering Aspects 120 (1–3), 255–271.

Guo, L.D., Hung, C.C., Santschi, P.H., Walsh, I.D., 2002. Th-234 scavenging and its relationship to acid polysaccharide abundance in the Gulf of Mexico. Marine Chemistry 78 (2–3), 103–119.

Gust, G., Bowles, W., Giordano, S., Huettel, M., 1996. Particle accumulation in a cylindrical sediment trap under laminar and turbulent steady flow: an experimental approach. Aquatic Sciences 58, 297–326.

Gustafsson, Ö., Gschwend, P.M., Buesseler, K.O., 1997a. Using Th-234 disequilibria to estimate the vertical removal rates of polycyclic aromatic hydrocarbons from the surface ocean. Marine Chemistry 57 (1–2), 11–23.

Gustafsson, Ö., Gschwend, P.M., Buesseler, K.O., 1997b. Settling removal rates of PCBs into the Northwestern Atlantic derived from $^{238}$U–$^{234}$Th disequilibria. Environmental Science and Technology 31 (12), 3544–3550.

Gustafsson, Ö., Buesseler, K.O., Geyer, W.R., Moran, S.B., Gschwend, P.M., 1998. An assessment of the relative importance of horizontal and vertical transport of particle-reactive chemicals in the coastal ocean. Continental Shelf Research 18 (7), 805–829.

Gustafsson, Ö., et al., 2000. Colloid dynamics and transport of major elements through a boreal river- brackish bay mixing zone. Marine Chemistry 71 (1–2), 1–21.

Gustafsson, Ö., Long, C.M., Macfarlane, J., Gschwend, P.M., 2001. Fate of linear alkylbenzenes released to the coastal environment near Boston Harbor. Environmental Science and Technology 35 (10), 2040–2048.

Gustafsson, Ö., et al., 2004. Evaluation of the collection efficiency of upper ocean sub-photic-layer sediment traps: a 24-month in situ calibration in the open Baltic Sea using $^{234}$Th. Limnology and Oceanography Methods 2, 62–74.

Hall, I.R., Schmidt, S., McCave, I.N., Reyss, J.L., 2000. Particulate matter distribution and Th-234/U-238 disequilibrium along the Northern Iberian Margin: implications for particulate organic carbon export. Deep-Sea Research I 47 (4), 557–582.

Hernes, P.J., et al., 2001. Particulate carbon and nitrogen fluxes and compositions in the central equatorial Pacific. Deep-Sea Research I 48 (9), 1999–2023.

Hirschberg, D.J., Chin, P., Feng, H., Cochran, J.K., 1996. Dynamics of sediment and contaminant transport in the Hudson River Estuary: evidence from sediment distributions of naturally occurring radionuclides. Estuaries 19 (4), 931–949.

Honeyman, B.D., Santschi, P.H., 1989. A Brownian pumping model for oceanic trace-metal scavenging-evidence from Th isotopes. Journal of Marine Research 47, 951–992.

Honeyman, B.D., Balistrieri, L.S., Murray, J.W., 1988. Oceanic trace metal scavenging: the importance of particle concentration. Deep-Sea Research 35, 227–246.

Huettel, M., Ziebis, W., Forster, S., 1996. Flow-induced uptake of particulate matter in permeable sediments. Limnology and Oceanography 41 (2), 309–322.

Huettel, M., Ziebis, W., Forster, S., Luther, G.W., 1998. Advective transport affecting metal and nutrient distributions and interfacial fluxes in permeable sediments. Geochimica et Cosmochimica Acta 62 (4), 613–631.

Huettel, M., Røy, H., Precht, E., Ehrenhauss, S., 2003. Hydrodynamical impact on biogeochemical processes in aquatic sediments. Hydrobiologia 494, 231–236.

Hughes, D.J., Ansell, A.D., Atkinson, R.J.A., 1996. Sediment bioturbation by the echiuran worm *Maxmuelleria lankesteri* (Herdman) and its consequences for radionuclide dispersal in Irish Sea sediments. Journal of Experimental Marine Biology and Ecology 195 (2), 203–220.

Huh, C.A., Beasley, T.M., 1987. Profiles of dissolved and particulate thorium isotopes in the water column of coastal Southern-California. Earth and Planetary Science Letters 85 (1–3), 1–10.

Huh, C.A., Prahl, I.G., 1995. Role of colloids in upper ocean biogeochemistry in the Northeast Pacific-Ocean elucidated from U-238 Th-234 disequilibria. Limnology and Oceanography 40 (3), 528–532.

Huh, C.A., Zahnle, D.L., Small, L.F., Noshkin, V.E., 1987. Budgets and behaviors of uranium and thorium series isotopes in Santa Monica Basin sediments. Geochimica et Cosmochimica Acta 51, 1743–1754.

Huh, C.A., Ku, T.L., Luo, S.D., Landry, M.R., Williams, P.M., 1993. Fluxes of Th isotopes in the Santa-Monica basin, offshore California. Earth and Planetary Science Letters 116 (1–4), 155–164.

Hussain, N., Krishnaswami, S., 1980. U-238 series radioactive disequilibrium in groundwaters: implications to the origin of excess U-234 and fate of reactive pollutants. Geochimica et Cosmochimica Acta 44, 1287–1291.

Ishikawa, Y., Kagaya, H., Saga, K., 2004. Biomagnification of Be-7, Th-234, and Ra-228 in marine organisms near the northern Pacific coast of Japan. Journal of Environmental Radioactivity 76 (1–2), 103–112.

Jaeger, J.M., Nittrouer, C.A., 1999a. Marine record of surge-induced outburst floods from the Bering Glacier, Alaska. Geology 27 (9), 847–850.

Jaeger, J.M., Nittrouer, C.A., 1999b. Sediment deposition in an Alaskan Fjord: controls on the formation and preservation of sedimentary structures in Icy Bay. Journal of Sedimentary Research 69 (5), 1011–1026.

Jahnke, R.A., Jahnke, D.B., 2004. Calcium carbonate dissolution in deep sea sediments: reconciling microelectrode, pore water and benthic flux chamber results. Geochimica et Cosmochimica Acta 68 (1), 47–59.

Johnson, T.C., 2003. A Report of the Workshop on the Science of Freshwater Inland Seas, Contributions of the SOFIS Working Group (2003) Large Lakes Observatory Technical Report 2003-1 University of Minnesota, Duluth, Minnesota. 40 pp.

Kadko, D., Feely, R., Massoth, G., 1994. Scavenging of Th-234 and phosphorus removal from the hydrothermal effluent plume over the north cleft segment of the Juan-De-Fuca Ridge. Journal of Geophysical Research, [Solid Earth] 99 (B3), 5017–5024.

Kaufman, A., Li, Y.-H., Turekian, K.K., 1981. The removal rates of $^{234}$Th and $^{228}$Th from waters of the New York Bight. Earth and Planetary Science Letters 54 (3), 385–392.

Keafer, B.A., Buesseler, K.O., Anderson, D.M., 1992. Burial of living dinoflagellate cysts in estuarine and nearshore sediments. Marine Micropaleontology 20 (2), 147–161.

Kershaw, P., Young, A., 1988. Scavenging of 234Th in the eastern Irish sea. Journal of Environmental Radioactivity 6 (1), 1–23.

Kersten, M., Thomsen, S., Priebsch, W., Garbe-Schonberg, C.D., 1998. Scavenging and particle residence times determined from Th-234/U-238 disequilibria in the coastal waters of Mecklenburg Bay. Applied Geochemistry 13 (3), 339–347.

Kersten, M., Leipe, T., Tauber, F., 2005. Storm disturbance of sediment contaminants at a Hot-Spot in the Baltic Sea assessed by $^{234}$Th radionuclide tracer profiles. Environmental Science and Technology 39, 984–990.

Kigoshi, K., 1971. Alpha-recoil Thorium-234: dissolution into water and the Uranium-234/Uranium-238 disequilibrium in nature. Science 173, 47–48.

Kim, G., Church, T.M., 2001. Seasonal biogeochemical fluxes of Th-234 and Po-210 in the upper Sargasso Sea: influence from atmospheric iron deposition. Global Biogeochemical Cycles 15 (3), 651–661.

Klump, J.V., et al., 2003. Sampling methods and approaches using radionuclide tracers in the study of sediment resuspension and cross margin transport in the nearshore of the Laurentian Great Lakes. International Journal of Sediment Research 18 (2), 266–277.

Knauss, K.G., Ku, T.L., Moore, W.S., 1978. Radium and thorium isotopes in the surface waters of the East Pacific and coastal Southern California. Earth and Planetary Science Letters 39 (2), 235–249.

Krishnaswami, S., Lal, D., Somayajulu, B.L.K., 1976. Large-volume in-situ filtration of deep Pacific waters: mineralogical and radioisotope studies. Earth and Planetary Science Letters 32 (2), 420–429.

Krishnaswami, S., Graustein, W.C., Turekian, K.K., 1982. Radium, thorium, and radioactive lead isotopes in groundwaters: applications to the in situ determination of adsorption–desorption rate constants and retardation factors. Water Resources Research 18, 1633–1675.

Krishnaswami, S., Monaghan, M.C., Westrich, J.T., Bennett, J.T., Turekian, K.K., 1984. Chronologies of sedimentary processess in sediments of the FOAM site, Long Island Sound, Connecticut. American Journal of Science 284, 706–733.

Krishnaswami, S., Baskaran, M., Fowler, S.W., Heyraud, M., 1985. Comparative role of salps and other zooplankton in the cycling and transport of selected elements and natural radionuclides in Mediterranean waters. Biogeochemistry 1 (4), 353–360.

Kryger, J., Riisgard, H.U., 1988. Filtration rate capacities in six species of European freshwater bivalves. Oecologia 77, 34–38.

Kuptsov, V.M., 1994. Th-234 in the Karskoye sea-water. Geokhimiya 8–9, 1346–1353.

Kuptsov, V.M., Lisitzin, A.P., Shevchenko, V.P., 1994. Th-234 as an indicator of particulate fluxes in the Kara Sea. Okeanologiya 34 (5), 759–765.

Kuptzov, V.M., Lisitzin, A.P., Shevchenko, V.P., Burenkov, V.I., 1999. Suspended matter fluxes in the bottom sediments of the Laptev Sea. Okeanologiya 39 (4), 597–604.

Langone, L., Frignani, M., Cochran, J.K., Ravaioli, M., 1997. Scavenging processes and export fluxes close to a retreating seasonal ice margin (Ross Sea, Antarctica). Water, Air and Soil Pollution 99 (1–4), 705–715.

Lauerman, L.M.L., Smoak, J.M., Shaw, T.J., Moore, W.S., Smith, K.L., 1997. Th-234 and Pb-210 evidence for rapid ingestion of settling particles by mobile epibenthic megafauna in the abyssal NE Pacific. Limnology and Oceanography 42 (3), 589–595.

Lavelle, J.W., Cudaback, C.N., Paulson, A.J., Murray, J.W., 1991. A rate for the scavenging of fine particles by macroaggregates in a deep estuary. Journal of Geophysical Research, [Oceans] 96 (C1), 783–790.

Lee, T., Barg, E., Lal, D., 1991. Studies of vertical mixing in the Southern California Bight with cosmogenic radionuclides P-32 and Be-7. Limnology and Oceanography 36 (5), 1044–1053.

Lee, T., Barg, E., Lal, D., Azam, F., 1993. Bacterial scavenging of Th-234 in surface ocean waters. Marine Ecology. Progress Series 96 (2), 109–116.

Legeleux, F., Reyss, J.L., Schmidt, S., 1994. Particle mixing rates in sediments of the Northeast Tropical Atlantic—evidence from Pb-210(Xs), Cs-137, Th-228(Xs) and Th-234(Xs) downcore distributions. Earth and Planetary Science Letters 128 (3–4), 545–562.

Levin, L.A., Childers, S.E., Smith, C.R., 1991. Epibenthic, agglutinating foraminiferans in the Santa-Catalina basin and their response to disturbance. Deep-Sea Research I 38 (4), 465–483.

Levin, L., et al., 2002. Benthic processes on the Peru margin: a transect across the oxygen minimum zone during the 1997–98 El Nino. Progress in Oceanography 53 (1), 1–27.

Luo, S.D., Ku, T.L., Roback, R., Murrell, M., McLing, T.L., 2000. In-situ radionuclide transport and preferential groundwater flows at INEEL (Idaho): decay-series disequilibrium studies. Geochimica et Cosmochimica Acta 64 (5), 867–881.

Martin, W.R., Sayles, F.L., 1987. Seasonal cycles of particle and solute transport processes in nearshore sediments: $^{222}$Rn/$^{226}$Ra and $^{234}$Th/$^{238}$U disequilibrium at a site in Buzzards Bay, MA. Geochimica et Cosmochimica Acta 51 (4), 927–943.

Matsumoto, E., 1975. $^{234}$Th–$^{238}$U radioactive disequilibrium in the surface layer of the ocean. Geochimica et Cosmochimica Acta 39 (2), 205–212.

Mayer, L.M., 1994. Surface area control of organic carbon accumulation in continental shelf sediments. Geochimica et Cosmochimica Acta 58, 1271–1284.

McCartney, M., Kershaw, P.J., Allington, D.J., Young, A.K., Turner, D., 1992. Industrial sources of naturally-occurring radionuclides in the Eastern Irish Sea. Radiation Protection Dosimetry 45 (1–4), 711–714.

McKee, B.A., DeMaster, D.J., Nittrouer, C.A., 1984. The use of 234T/h238 disequilibrium to examine the fate of particle-reactive species on the Yangtze continental shelf. Earth and Planetary Science Letters 68 (3), 431–442.

McKee, B.A., Demaster, D.J., Nittrouer, C.A., 1986. Temporal variability in the partitioning of thorium between dissolved and particulate phases on the Amazon shelf- implications for the

scavenging of particle-reactive species. Continental Shelf Research 6 (1–2), 87–106.

Merritt, E., 1903. Recent developments in the study of radioactive substances. Science 18 (445), 41–47.

Michaels, A.F., Bates, N.R., Buesseler, K.O., Carlson, C.A., Knap, A.H., 1994. Carbon-cycle imbalances in the Sargasso sea. Nature 372 (6506), 537–540.

Miller, L.A., Svaeren, I., 2003. Th-234 distributions in coastal and open ocean waters by non-destructive beta-counting. Journal of Radioanalytical and Nuclear Chemistry 256 (3), 431–444.

Miller, R.J., Smith, C.R., DeMaster, D.J., Fornes, W.L., 2000. Feeding selectivity and rapid particle processing by deep-sea megafaunal deposit feeders: a Th-234 tracer approach. Journal of Marine Research 58 (4), 653–673.

Minagawa, M., Tsunogai, S., 1980. Removal of $^{234}$Th from a coastal sea: Funka Bay, Japan. Earth and Planetary Science Letters 47 (1), 51–64.

Moore, R.M., Hunter, K.A., 1985. Thorium adsorption in the ocean: reversibility and distribution amongst particle sizes. Geochimica et Cosmochimica Acta 49 (11), 2253–2257.

Moore, R.M., Millward, G.E., 1988. The kinetics of reversible Th reactions with marine particles. Geochimica et Cosmochimica Acta 52 (1), 113–118.

Moore, W.S., Wilson, A., 2005. Advective flow through the upper continental shelf driven by storms, buoyancy, and submarine groundwater discharge. Earth and Planetary Science Letters 235 (3–4), 564–576.

Moore, W.S., DeMaster, D.J., Smoak, J.M., McKee, B.A., Swarzenski, P.W., 1996. Radionuclide tracers of sediment–water interactions on the Amazon shelf. Continental Shelf Research 16 (5–6), 645–665.

Moore, W.S., et al., 2002. Thermal evidence of water exchange through a coastal aquifer: implications for nutrient fluxes. Geophysical Research Letters 29.

Moran, S.B., Buesseler, K.O., 1992. Short residence time of colloids in the upper ocean estimated from U-238 Th-234 disequilibria. Nature 359 (6392), 221–223.

Moran, S.B., Buesseler, K.O., 1993. Size-fractionated Th-234 in continental-shelf waters off New-England—implications for the role of colloids in oceanic trace-metal scavenging. Journal of Marine Research 51 (4), 893–922.

Moran, S.B., Moore, R.M., 1989. The distribution of colloidal aluminium and organic carbon in coastal and open ocean waters off Nova Scotia. Geochimica et Cosmochimica Acta 53, 2519–2527.

Moran, S.B., Moore, R.M., 1992. Kinetics of the removal of dissolved aluminium by diatoms in seawater—a comparison with thorium. Geochimica et Cosmochimica Acta 56 (9), 3365–3374.

Moran, S.B., Smith, J.N., 2000. Th-234 as a tracer of scavenging and particle export in the Beaufort Sea. Continental Shelf Research 20 (2), 153–167.

Moran, S.B., Ellis, K.M., Smith, J.N., 1997. Th-234/U-238 disequilibrium in the central Arctic Ocean: implications for particulate organic carbon export. Deep-Sea Research II 44 (8), 1593–1606.

Moran, S.B., et al., 2003. Does Th-234/U-238 disequilibrium provide an accurate record of the export flux of particulate organic carbon from the upper ocean? Limnology and Oceanography 48 (3), 1018–1029.

Morel, F.M.M., Price, N.M., 2003. The biogeochemical cycles of trace metals in the oceans. Science 300, 944–947.

Mudge, S.M., Assinder, D.J., Bourne, G.S., 1997. Radiological assessment of the ribble estuary: 3. Redistribution of radionuclides. Journal of Environmental Radioactivity 36 (1), 43–67.

Mulder, T., Weber, O., Anschutz, P., Jorissen, F.J., Jouanneau, J.M., 2001. A few months-old storm-generated turbidite deposited in the Capbreton Canyon (Bay of Biscay, SW France). Geo-Marine Letters 21 (3), 149–156.

Muller, F.L.L., Gulin, S.B., Kalvoy, A., 2001. Chemical speciation of copper and zinc in surface waters of the western Black Sea. Marine Chemistry 76 (4), 233–251.

Murnane, R.J., Cochran, J.K., Buesseler, K.O., Bacon, M.P., 1996. Least-squares estimates of thorium, particle, and nutrient cycling rate constants from the JGOFS north Atlantic bloom experiment. Deep-Sea Research I 43 (2), 239–258.

Murray, J.W., Downs, J.N., Strom, S., Wei, C., Jannash, H.W., 1989. Nutrient assimilation, export production and $^{234}$Th scavenging in the eastern equatorial Pacific. Deep-Sea Research 36, 1471–1489.

Murray, R.W., Leinen, M., Isern, A.R., 1993. Biogenic flux of A1 to sediment in the central equatorial Pacific-Ocean—evidence for increased productivity during glacial periods. Paleoceanography 8 (5), 651–670.

Murray, J.W., et al., 1996. Export flux of particulate organic carbon from the central equatorial Pacific determined using a combined drifting trap Th-234 approach. Deep-Sea Research II 43 (4–6), 1095–1132.

Nittrouer, C.A., DeMaster, D.J., McKee, B.A., Cutshall, N.H., Larsen, I.L., 1984. The effect of sediment mixing on Pb-210 accumulation rates for the Washington continental shelf. Marine Geology 54, 201–221.

Niven, S.E.H., Moore, R.M., 1993. Thorium sorption in seawater suspensions of aluminum-oxide particles. Geochimica et Cosmochimica Acta 57 (10), 2169–2179.

Niven, S.E.H., Kepkay, P.E., Boraie, A., 1995. Colloidal organic carbon and colloidal $^{234}$Th dynamics during a coastal phytoplankton bloom. Deep-Sea Research II 42, 257–273.

Nodder, S.D., et al., 2001. Particle transformations and export flux during an in situ iron-stimulated algal bloom in the Southern Ocean. Geophysical Research Letters 28 (12), 2409–2412.

Nozaki, Y., Horibe, Y., Tsubota, H., 1981. The water column distributions of thorium isotopes in the western North Pacific. Earth and Planetary Science Letters 54 (2), 203–216.

Palm, A., et al., 2004. Evaluation of sequentially-coupled POP fluxes estimated from simultaneous measurements in multiple compartments of an air–water–sediment system. Environmental Pollution 128 (1–2), 85–97.

Pope, R.H., Demaster, D.J., Smith, C.R., Seltmann, H., 1996. Rapid bioturbation in equatorial Pacific sediments: evidence from excess Th-234 measurements. Deep-Sea Research II 43 (4–6), 1339–1364.

Porcelli, D., Andersson, P.S., Baskaran, M., Wasserburg, G.J., 2001. Transport of U-and Th-series nuclides in a Baltic Shield watershed and the Baltic Sea. Geochimica et Cosmochimica Acta 65 (15), 2439–2459.

Quigley, M.S., Santschi, P.H., Guo, L., Honeyman, B.D., 2001. Sorption irreversibility and coagulation behavior of $^{234}$Th with surface-active marine organic matter. Marine Chemistry 76, 27–45.

Quigley, M.S., Santschi, P.H., Hung, C.-C., Guo, L., Honeyman, B.D., 2002. Importance of polysaccharides for $^{234}$Th complexation to marine organic matter. Limnology and Oceanography 47, 367–377.

Radakovitch, O., Frignani, M., Giuliani, S., Montanari, R., 2003. Temporal variations of dissolved and particulate Th-234 at a coastal station of the northern Adriatic Sea. Estuarine, Coastal and Shelf Science 58 (4), 813–824.

Ralph, E., 2002. Scales and structures of large lake eddies. Geophysical Research Letters 29, 2177.

Reimers, C., Friedrichs, C., Bebout, B., Howd, P., Huettel, M., Jahnke, R., MacCready, P., Ruttenberg, K., Sanford, L., Trowbridge, J., 2004. Coastal Benthic Exchange Dynamics, COOP Report #10. 97 pp.

Riedl, R.J., Huang, N., Machan, R., 1972. The subtidal pump: a mechanism of interstitial water exchange by wave action. Marine Biology 13, 210–221.

Roberts, K.A., Santschi, P.H., 2004. Inverse dependency of particle residence times in ponds to the concentration of phosphate, the limiting nutrient. Journal of Environmental Radioactivity 76 (3), 311–318.

Rose, C.L., McKay, W.A., Toole, J., 1994. The use of the Th-234/U-238 disequilibrium in studying surf-zone sediment processes in the Eastern Irish Sea. Marine Environmental Research 37 (4), 393–416.

Rudnicki, M.D., Elderfield, H., 1993. A chemical-model of the buoyant and neutrally buoyant plume above the tag vent field, 26 degrees-N, Mid-Atlantic Ridge. Geochimica et Cosmochimica Acta 57 (13), 2939–2957.

Rutgers van der Loeff, M.M., Berger, G.W., 1991. Scavenging and particle-flux- seasonal and regional variations in the Southern-Ocean (Atlantic Sector). Marine Chemistry 35 (1–4), 553–567.

Rutgers van der Loeff, M.M., Boudreau, B.P., 1997. The effect of resuspension on chemical exchanges at the sediment–water interface in the deep sea—A modelling and natural radiotracer approach. Journal of Marine Systems 11 (3–4), 305–342.

Rutgers van der Loeff, M.M., Friedrich, J., Bathmann, U.V., 1997. Carbon export during the spring bloom at the antarctic polar front, determined with the natural tracer Th-234. Deep-Sea Research II 44 (1–2), 457–478.

Rutgers van der Loeff, M.M., Buesseler, K., Bathmann, U., Hense, I., Andrews, J., 2002a. Comparison of carbon and opal export rates between summer and spring bloom periods in the region of the Antarctic Polar Front, SE Atlantic. Deep-Sea Research II 49 (18), 3849–3869.

Rutgers van der Loeff, M.M., Meyer, R., Rudels, B., Rachor, E., 2002b. Resuspension and particle transport in the benthic nepheloid layer in and near Fram Strait in relation to faunal abundances and Th-234 depletion. Deep-Sea Research I 49 (11), 1941–1958.

Rutgers van der Loeff, M. et al., this volume. A review of present techniques and methodological advances in analyzing [234]Th in aquatic systems. Marine Chemistry.

Rutherford, E., Soddy, F., 1902. The cause and nature of radioactivity. Philosophical Magazine 4, 569–585.

Sambrotto, R.N., 2001. Nitrogen production in the northern Arabian Sea during the Spring Intermonsoon and Southwest Monsoon seasons. Deep-Sea Research II 48 (6–7), 1173–1198.

Santschi, P.H., 1984. Particle flux and trace metal residence time in natural waters. Limnology and Oceanography 29 (5), 1100–1108.

Santschi, P.H., Li, Y.-H., Bell, J., 1979. Natural radionuclides in the water of Narragansett Bay. Earth and Planetary Science Letters 45 (1), 201–213.

Santschi, P.H., et al., 1995. Isotopic evidence for the contemporary origin of high-molecular-weight organic-matter in oceanic environments. Geochimica et Cosmochimica Acta 59 (3), 625–631.

Santschi, P.H., Guo, L., Walsh, I.D., Quigley, M.S., Baskaran, M., 1999. Boundary exchange and scavenging of radionuclides in continental margin waters of the Middle Atlantic Bight: implications for organic carbon fluxes. Continental Shelf Research 19 (5), 609–636.

Santschi, P.H., et al., 2001. Accumulation rates and sources of sediments and organic carbon on the Palos Verdes shelf based on radioisotopic tracers (Cs-137, Pu-239,Pu-240, Pb-210, Th-234, U-238 and C-14). Marine Chemistry 73 (2), 125–152.

Santschi, P.H., et al., 2003. Control of acid polysaccharide production and Th-234 and POC export fluxes by marine organisms. Geophysical Research Letters 30 (2).

Santschi, P.H. et al., this volume. Thorium speciation in seawater. Marine Chemistry.

Sarin, M.M., Bhushan, R., Rengarajan, R., Yadav, D.N., 1992. Simultaneous determination of U-238 series nuclides in waters of Arabian sea and Bay of Bengal. Indian Journal of Marine Sciences 21 (2), 121–127.

Sarin, M.M., Krishnaswami, S., Ramesh, R., Somayajulu, B.L.K., 1994a. U-238 decay series nuclides in the Northeastern Arabian Sea— scavenging rates and cycling processes. Continental Shelf Research 14 (2–3), 251–265.

Sarin, M.M., Rengarajan, R., Somayajulu, B.L.K., 1994b. Natural radionuclides in the Arabian Sea and Bay of Bengal— distribution and evaluation of particle scavenging processes. Proceedings of the Indian Academy of Sciences. Earth and Planetary Sciences 103 (2), 211–235.

Sarin, M.M., Rengarajan, R., Ramaswamy, V., 1996. Th-234 scavenging and particle export fluxes from the upper 100 m of the Arabian Sea. Current Science 71 (11), 888–893.

Savoye, N., Buesseler, K.O., Cardinal, D., Dehairs, F., 2004. [234]Th deficit and excess in the Southern Ocean during spring 2001: particle export and remineralization. Geophysical Research Letters 31 (12).

Savoye, N. et al., this volume. [234]Th sorption and export models in the water column: A review. Marine Chemistry.

Schaff, T., et al., 1992. Spatial heterogeneity of benthos on the carolina continental-slope- large (100-Km)-scale variation. Marine Ecology. Progress Series 88 (2–3), 143–160.

Schmidt, S., Reyss, J.L., Nguyen, H.V., Buatmenard, P., 1990. Th-234 cycling in the upper water column of the Northwestern Mediterranean Sea. Global and Planetary Change 3 (1–2), 25–33.

Schmidt, S., Nival, P., Reyss, J.L., Baker, M., Buatmenard, P., 1992. Relation between Th-234 scavenging and zooplankton biomass in Mediterranean surface waters. Oceanologica Acta 15 (2), 227–231.

Schmidt, S., de Stigter, H.C., van Weering, T.C.E., 2001. Enhanced short-term sediment deposition within the Nazare Canyon, North-East Atlantic. Marine Geology 173 (1–4), 55–67.

Schmidt, S., Chou, L., Hall, I.R., 2002a. Particle residence times in surface waters over the north-western Iberian Margin: comparison of pre-upwelling and winter periods. Journal of Marine Systems 32 (1–3), 3–11.

Schmidt, S., Andersen, V., Belviso, S., Marty, J.C., 2002b. Strong seasonality in particle dynamics of north-western Mediterranean surface waters as revealed by Th-234/U-238. Deep-Sea Research I 49 (8), 1507–1518.

Schmidt, S., van Weering, T.C.E., Reyss, J.L., van Beek, P., 2002c. Seasonal deposition and reworking at the sediment–water interface on the northwestern Iberian margin. Progress in Oceanography 52 (2–4), 331–348.

Shimmield, G.B., Ritchie, G.D., Fileman, T.W., 1995. The impact of marginal ice-zone processes on the distribution of Pb-210, Po-210 and Th-234 and implications for new production in the Bellingshausen Sea, Antarctica. Deep-Sea Research II 42 (4–5), 1313–1335.

Shull, D.H., 2001. Transition-matrix model of bioturbation and radionuclide diagenesis. Limnology and Oceanography 46 (4), 905–916.

Shull, D.H., Mayer, L.M., 2002. Dissolution of particle-reactive radionuclides in deposit-feeder digestive fluids. Limnology and Oceanography 47 (5), 1530–1536.

Shum, K.T., Sundby, B., 1996. Organic matter processing in continental shelf sediments – the subtidal pump revisited. Marine Chemistry 53 (12), 81–87.

Silverberg, N., 1986. Radionuclide profiles, sedimentation rates, and bioturbation in modern sediments of the Laurentian Trough, Gulf of St. Lawrence. Oceanology Acta 9 (3), 285–290.

Smith, C.R., Pope, R.H., Demaster, D.J., Magaard, L., 1993. Age-dependent mixing of deep-sea sediments. Geochimica et Cosmochimica Acta 57 (7), 1473–1488.

Smith, C.R., et al., 1996. Phytodetritus at the abyssal seafloor across 10 degrees of latitude in the central equatorial Pacific. Deep-Sea Research II 43 (4–6), 1309–1338.

Smith, C.R., et al., 1998. Sediment community structure around a whale skeleton in the deep Northeast Pacific: macrofaunal, microbial and bioturbation effects. Deep-Sea Research II 45 (1–3), 335–364.

Smith, K.J., Vintro, L.L., Mitchell, P.I., de Bois, P.B., Boust, D., 2004. Uranium-thorium disequilibrium in north-east Atlantic waters. Journal of Environmental Radioactivity 74 (1–3), 199–210.

Smoak, J.M., DeMaster, D.J., Kuehl, S.A., Pope, R.H., McKee, B.A., 1996. The behavior of particle-reactive tracers in a high turbidity environment: Th-234 and Pb-210 on the Amazon continental shelf. Geochimica et Cosmochimica Acta 60 (12), 2123–2137.

Smoak, J.M., Moore, W.S., Thunell, R.C., Shaw, T.J., 1999. Comparison of Th-234, Th-228, and Pb-210 fluxes with fluxes of major sediment components in the Guaymas Basin, Gulf of California. Marine Chemistry 65 (3–4), 177–194.

Smoak, J.M., Moore, W.S., Thunell, R.C., 2000. Influence of boundary scavenging and sediment focusing on Th-234, Th-228 and Pb-210 fluxes in the Santa Barbara Basin. Estuarine, Coastal and Shelf Science 51 (3), 373–384.

Smoak, J.M., et al., 2004. Radionuclide fluxes and particle scavenging in Cariaco Basin. Continental Shelf Research 24 (13–14), 1451–1463.

Somayajulu, B.L.K., Rengarajan, R., Jani, R.A., 2002. Geochemical cycling in the Hooghly estuary, India. Marine Chemistry 79 (3–4), 171–183.

Sommerfield, C.K., Nittrouer, C.A., Alexander, C.R., 1999. Be-7 as a tracer of flood sedimentation on the northern California continental margin. Continental Shelf Research 19 (3), 335–361.

Sommerfield, C.K., Aller, R.C., Nittrouer, C.A., 2001. Sedimentary carbon, sulfur, and iron relationships in modern and ancient diagenetic environments of the Eel River Basin (USA). Journal of Sedimentary Research 71 (3), 335–345.

Spencer, D.W., et al., 1978. Chemical fluxes from a sediment trap experiment in the deep Sargasso Sea. Journal of Marine Research 36 (3), 493–523.

Sun, M.Y., Aller, R.C., Lee, C., 1991. Early diagenesis of chlorophyll-a in long-island sound sediments— a measure of carbon flux and particle reworking. Journal of Marine Research 49 (2), 379–401.

Sun, M.Y., Aller, R.C., Lee, C., 1994. Spatial and temporal distributions of sedimentary chloropigments as indicators of benthic processes in long-island sound. Journal of Marine Research 52 (1), 149–176.

Swarzenski, P.W., Porcelli, D., Andersson, P.S., Smoak, J.M., 2003. The behavior of U-and Th-series nuclides in the estuarine environment. Review of Mineral Geochemistry 52, 577–606.

Sweeney, E.N., McGillicuddy, D.J. Jr., Buesseler, K.O., 2004. Biogeochemical impacts due to mesoscale eddy activity in the Sargasso Sea as measured at the Bermuda Atlantic Time-series Study (BATS). Deep-Sea Research, Part II: Topical Studies in Oceanography 50, 3017–3039.

Tanaka, N., Takeda, Y., Tsunogai, S., 1983. Biological effect on removal of Th-234, Po-210 and Pb-210 from surface-water in Funka Bay, Japan. Geochimica et Cosmochimica Acta 47 (10), 1783–1790.

Thunell, R.C., Moore, W.S., Dymond, J., Pilskaln, C.H., 1994. Elemental and isotopic fluxes in the Southern California Bight— a time-series sediment trap study in the San-Pedro Basin. Journal of Geophysical Research, [Oceans] 99 (C1), 875–889.

Tricca, A., Wasserburg, G.J., Porcelli, D., Baskaran, M., 2001. The transport of U-and Th-series nuclides in a sandy unconfined aquifer. Geochimica et Cosmochimica Acta 65 (8), 1187–1210.

Trimble, S.M., Baskaran, M., Porcelli, D., 2004. Scavenging of thorium isotopes in the Canada basin of the Arctic Ocean. Earth and Planetary Science Letters 222 (3–4), 915–932.

Tsunogai, S., Taguchi, K., Harada, K., 1986. Seasonal variation in the difference between observed and calculated particulate fluxes of Th-234 in Funka Bay, Japan. Journal of the Oceanographical Society of Japan 42, 91–98.

Tsunogai, S., Noriki, S., Harada, K., Tate, K., 1990. Vertical-change index for the particulate transport of chemical and isotopic components in the ocean. Geochemical Journal 24 (4), 229–243.

Turekian, K.K., Benninger, L.K., Dion, E.P., 1983. 7Be and 210Pb total deposition fluxes at New Haven, Connecticut, and at Bermuda. Journal of Geophysical Research 88, 5411–5415.

Turnewitsch, R., Witte, U., Graf, G., 2000. Bioturbation in the abyssal Arabian Sea: influence of fauna and food supply. Deep-Sea Research II 47 (14), 2877–2911.

Turnewitsch, R., Springer, B.M., 2001. Do bottom mixed layers influence Th-234 dynamics in the abyssal near-bottom water column? Deep-Sea Research I 48 (5), 1279–1307.

Usbeck, R., Rutgers van der Loeff, M., Hoppema, M., Schlitzer, R., 2002. Shallow remineralization in the Weddell Gyre. Geochemistry Geophysics Geosystems, 3.

Vanderploeg, H.A., et al., 2002. Dispersal and emerging ecological impacts of Ponto-Caspian species in the Laurentian Great Lakes. Canadian Journal of Fisheries and Aquatic Sciences 59, 1209–1228.

Vogler, S., Jung, M., Mangini, A., 1996. Scavenging of Th-234 and Be-7 in Lake Constance. Limnology and Oceanography 41 (7), 1384–1393.

Walsh, J.P., Nittrouer, C.A., 2003. Contrasting styles of off-shelf sediment accumulation in New Guinea. Marine Geology 196 (3–4), 105–125.

Waples, J.T., Orlandini, K.A., Edgington, D.N., Klump, J.V., 2004. Seasonal and spatial dynamics of Th-234/U-238 disequilibria in southern Lake Michigan. Journal of Geophysical Research, [Oceans] 109 (C10).

Wei, C.L., Hung, C.C., 1998. Particle scavenging in the upper water column off Mindoro Island, Philippine: Th-234/U-238 disequilibria. Estuarine, Coastal and Shelf Science 46 (3), 351–358.

Wei, C.L., Murray, J.W., 1991. Th-234/U-238 disequilibria in the Black-Sea. Deep-Sea Research I 38, S855–S873.

Wei, C.L., Murray, J.W., 1992. Temporal variations of Th-234 activity in the water column of Dabob Bay—particle scavenging. Limnology and Oceanography 37 (2), 296–314.

Weinstein, S.E., Moran, S.B., 2005. Vertical flux of particulate Al, Fe, Pb, and Ba from the upper ocean estimated from $^{234}$Th/$^{238}$U disequilibria. Deep-Sea Research I 52, 1477–1488.

Wheatcroft, R.A., Martin, W.R., 1996. Spatial variation in short-term (Th-234) sediment bioturbation intensity along an organic-carbon gradient. Journal of Marine Research 54 (4), 763–792.

Wurtsbaugh, W., et al., 2002. Report of the Workshop on Emerging Research Questions for Limnology: The Study of Inland Waters, Boulder, Colorado.

Yamada, M., Aono, T., 2003. Pb-210 and Th-234 in settling particles collected by time-series sediment traps in the Okinawa Trough. Deep-Sea Research II 50 (2), 487–501.

Yang, Y.L., Han, X., Kusakabe, M., 2004. POC fluxes from euphotic zone estimated from Th-234 deficiency in winter in the northwestern North Pacific Ocean. Acta Oceanologica Sinica 23 (1), 135–147.