

1 **What did we learn about ocean particle dynamics in the GEOSECS-JGOFS era?**

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3 Catherine Jeandel¹, Michiel Rutgers van der Loeff², Phoebe J. Lam³, Matthieu Roy-
4 Barman⁴, Robert M. Sherrell⁵, Sven Kretschmer², Chris German³ and Frank Dehairs⁶

5 1- Observatoire Midi-Pyrénées-14, avenue Edouard Belin-31400-Toulouse-France
6 *Catherine.jeandel@legos.obs-mip.fr*

7 2- Alfred-Wegener Institute for Polar and Marine Research, am Handelshafen 12,
8 D-27570, Bremerhaven, Germany.

9 3- Woods Hole Oceanographic Institution, Woods Hole, MA 02543, USA

10 4- LSCE/IPSL Laboratoire CNRS/CEA/UVSQ, Domaine du CNRS, Bat 12
11 Avenue de la Terrasse, 91198 Gif-sur-Yvette Cedex, France

12 5-Institute of Marine and Coastal Sciences and Department of Earth and Planetary
13 Sciences, Rutgers University, 71 Dudley Road New Brunswick, NJ 08901-8521

14 6- Vrije Universiteit Brussel, ESSC Research Group, Pleinlaan 2, 1050 Brussels,
15 Belgium

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17 **Key words: Historical review. Oceanic Particle distribution sources and sinks;**
18 **GEOSECS; JGOFS; GEOTRACES.**

19
20 ** This article is dedicated to the memory of Devendra Lal (1929-2012) who wrote a*
21 *seminal contribution to the study of "the oceanic microcosm of particles".*

22
23 **Abstract**

24 Particles determine the residence time of many dissolved elements in seawater. Although
25 a substantial number of field studies were conducted in the framework of major
26 oceanographic programs as GEOSECS and JGOFS, knowledge about particle dynamics
27 is still scarce. Moreover, the particulate trace metal behavior remains largely unknown.
28 The GEOSECS sampling strategy during the 1970's focused on large sections across
29 oceanic basins, where particles were collected by membrane filtration after Niskin bottle
30 sampling, biasing the sampling towards the small particle pool. Late in this period, the
31 first in situ pumps allowing large volume sampling were also developed. During the
32 1990's, JGOFS focused on the quantification of the "exported carbon flux" and its
33 seasonal variability in representative biogeochemical provinces of the ocean, mostly
34 using sediment trap deployments. Although scarce and discrete in time and space, these
35 pioneering studies allowed an understanding of the basic fate of marine particles. This
36 understanding improved considerably, especially when the analysis of oceanic tracers
37 such as natural radionuclides allowed the first quantification of processes such as
38 dissolved-particle exchange and particle settling velocities. Because the GEOTRACES

39 program emphasizes the importance of collecting, characterizing and analyzing marine
40 particles, this paper reflects our present understanding of the sources, fate and sinks of
41 oceanic particles at the early stages of the program.

42

43 **Introduction**

44

45 The ocean contains 1.4×10^{18} cubic meters of water and holds approximately 10^{10} metric
46 tons of solid material in the form of suspended particles that are present at an average
47 concentration in the deep sea ranging from 5 to 20 μg per liter (Brewer *et al.*, 1976;
48 Bishop and Fleisher, 1987; Sherrell and Boyle, 1992). Although not abundant, particles
49 act as an essential regulator of ocean chemistry because they determine the residence
50 time of many dissolved elements in seawater (Lal, 1977; Turekian, 1977). Vertical and
51 horizontal distributions of many trace elements and their isotopes (TEIs) are clearly
52 influenced by particle formation, remineralization, and transport. Because of their
53 importance, numerous studies during the past 50 years have focused on characterizing
54 these marine particles. In the 1970's, the Geochemical Ocean Sections study (GEOSECS;
55 Craig and Turekian, 1976) allowed a first description of the particle distribution in the
56 ocean, and mostly focused on suspended particles collected by filtration from Niskin-type
57 bottles. During those times, only a few pioneering studies attempting to characterize and
58 quantify particle fluxes were conducted (McCave, 1975; Honjo, 1976; Shanks and Trent,
59 1980). Nevertheless, these first results were invaluable in that i) they were the first
60 suggesting that vertical flux is dominated by rare large particles (McCave, 1975) and ii)
61 they guided the strategy of the Joint Global Ocean Flux Study (JGOFS) program (Fowler
62 and Knauer, 1986). However, laboratory and field technologies at that time were such
63 that measurements of TEIs in these particles with a good precision and resolution (spatial
64 as well as temporal) were difficult.

65 In the 1990's, the JGOFS program substantially increased our understanding of the
66 standing stock, vertical flux and fate of marine particles, with the focus largely on carbon
67 and associated nutrient cycles (Fasham *et al.*, 2001). However, because of data scarcity
68 and the large variability of particle fluxes in time and space, the full characterization of
69 marine particle concentrations, flux, and composition was a difficult task, and remained
70 far from being achieved. The JGOFS era also suffered from a lack of methodologies for

71 determining TEIs, which are extremely helpful for quantifying specific particle processes
72 in the water column. Although the analytical protocols for assessing some TEIs were
73 available and applied during some JGOFS research projects, they did not yet represent
74 the major research target. As a consequence, the global distribution of dissolved and
75 particulate TEIs is poorly known today. Because some TEIs are powerful tracers of
76 particle origin and processes (e.g. settling velocity, rates of dissolution, precipitation,
77 adsorption, and desorption) and some are essential micronutrients whose speciation in the
78 solid and dissolved phases is of prime importance for their bioavailability, there is an
79 urgent need to understand the global distribution of TEIs in the oceanic environment.
80 Filling this gap by investigating the sources, behavior and sinks of these TEIs is the main
81 goal of the GEOTRACES program (www.geotraces.org), which was developed following
82 the model of its “parent” program GEOSECS but with more emphasis on the collection,
83 observation, speciation and analysis of marine particles. As we enter the early stages of
84 the new GEOTRACES era, the present work reviews our understanding, informed by
85 GEOSECS and JGOFS, of the distribution of suspended and sinking marine particles of
86 both biogenic and abiogenic origin, as well as the role of these particles as regulators of
87 the marine biogeochemical cycles of TEIs. In addition to Anderson and Hayes’
88 introduction (this issue), this paper provides the historical context for this special issue
89 that proposes to browse the state of the art of our present knowledge on optically
90 characterizing (Boss *et al.*, this issue), collecting (McDonnell *et al.*, this issue), analyzing
91 (Lam *et al.*, this issue) and modelling (Dutay *et al.*, this issue; Jackson and Burd, this
92 issue) marine particles. The issue is concluded by Henderson and Marchal’s comments
93 and perspectives.

94

95 **1- The origin of marine particles**

96

97 Marine particles have two main origins, as illustrated in Figure 1.

98 *Sources external to the marine system:* By means of erosion, continents bring particles
99 (natural or anthropogenic, mineral or organic) into the ocean. These are transported via
100 the atmosphere (winds, rains), rivers (sedimentary discharge), or by lateral transport from
101 continental margin sediments. Before extensive damming, the annual solid flux
102 discharged by rivers to the oceans was of the order of 19×10^{15} g (Peucker-Ehrenbrink,

103 2009), which is 50 times the atmospheric flux (Jickells *et al.*, 2005). Other particles are
104 extraterrestrial, such as micrometeorites (10 to 100 μm in size) or cosmic dust that would
105 represent a flux of between 7 to 14 $\times 10^9$ g/y to the oceans (Johnson, 2001). Nano-
106 metric-sized (and highly magnetic) particles were also detected in the Greenland and
107 Antarctic ice caps and are identified as originating from atmospheric ablation of
108 meteorites and micrometeorites at high (~ 100 km) altitude (Lanci *et al.*, 2004; 2007).
109 Finally, hydrothermal vents also are a significant “external” source of particles for the
110 deep ocean. Particles from hydrothermal vents precipitate within the plume, forming fine
111 grained sulfide and oxide minerals that may be distributed over large regions of the deep
112 ocean (Mottl and McConachy, 1990; Feely *et al.*, 1996; Sherrell *et al.*, 2000; Tagliabue *et*
113 *al.*, 2010).

114 *Sources internal to the marine system:* A huge quantity of marine particles is produced by
115 biological activity. Photoautotrophic plankton assimilates dissolved species (C, N, P, Si,
116 trace metals) and uses solar energy to synthesize organic matter, and specific groups,
117 including microheterotrophs, also secrete skeletal parts consisting of calcite, aragonite,
118 opal, or celestite. The annual flux of material so produced represents $\sim 60 \times 10^{15}$ g/y of
119 organic carbon (Fasham *et al.*, 2001). The magnitude of marine primary production is
120 similar to terrestrial primary production, but the standing stock of fixed organic carbon is
121 far less in the ocean than in terrestrial systems, resulting in much higher turnover rate of
122 carbon in the ocean. This high turnover rate has consequences for the cycling of TEIs
123 associated with this biogenic material. Other autotrophic organisms, such as nitrifiers, use
124 chemical energy, and are called chemolithotrophic (Griffith *et al.*, 2012; Honjo *et al.*,
125 2012). These thrive throughout the oceanic water column and produce new biomass in-
126 situ. Autotrophic carbon fixation is the point of departure of the trophic chain whose life
127 and death cycle generates particles throughout the water column. Among the
128 heterotrophs, microzooplankton species, such as foraminifera, radiolarians, but also
129 larger multi-cellular organisms such as salps and pteropods, represent a significant
130 portion of living biomass (Buitenhuis *et al.*, 2013). Although less abundant than
131 phytoplanktonic organisms, they are important because of their role in packaging and
132 remineralization and for their potential as recorders, once incorporated in ocean
133 sediments, of past environmental conditions. In addition, diel vertical migration by

134 mesozooplankton may represent a significant pathway of particle redistribution in the
135 mesopelagic zone, between water depths of about 100 m and 1000 m (Steinberg *et al.*,
136 2008). Another source of particles in the upper water column is through spontaneous
137 aggregation of Dissolved Organic Matter (DOM) into larger particles, from the molecular
138 size up to a typical size of 4 μm , therefore becoming Particulate Organic Matter (POM).
139 These particles have been termed microgels (Verdugo, 2012). Barium sulfate, and
140 manganese and iron oxides and hydroxides are also known to precipitate within the water
141 column, incorporating other elements in the process, or scavenging trace elements by
142 adsorption or other particle surface phenomena (Krishnaswami *et al.*, 1976a,b; Bishop
143 and Fleisher, 1987; Dehairs *et al.*, 1990; Sherrell and Boyle, 1992; Paytan *et al.*, 1993;
144 van Beek *et al.*, 2007; van Beek *et al.*, 2009).
145 Marine particles are often divided in 2 different types: small (micron-size) and slowly
146 sinking particles on one hand and large (> 50-100 micron-size) and rapidly sinking on the
147 other hand. The cut off is both poorly defined and somewhat arbitrary. However, it
148 corresponds to 2 modes of marine particle sampling: filtration on filters with (sub-)
149 micron size porosity for the small particles and collection in sediment trap and/or
150 filtration with large porosity for large particles. Hence, this operational definition is still
151 used in the GEOTRACES program.

152

153 **2- Small suspended particles and TEI behavior**

154

155 **2-1 Oceanic distribution of suspended particles**

156

157 Small particles (0.2-53 μm) constitute the bulk of the particle standing stock in the ocean.
158 In the upper 1000 m, particles < 53 μm represent on average ~80% of total suspended
159 particle mass (Bishop *et al.*, 1977; Bishop *et al.*, 1978; Bishop *et al.*, 1980; Bishop *et al.*,
160 1985; Bishop *et al.*, 1986; Lam and Bishop, 2007; Bishop and Wood, 2008). Their
161 amount and their large surface areas propel them as active players in the solution-solid
162 exchanges that impact TEI distribution (Krishnaswami *et al.*, 1976; Anderson *et al.*,
163 1983a,b; Bishop and Fleisher, 1987; Sherrell and Boyle, 1992; Jeandel *et al.*, 1995; Roy-
164 Barman *et al.*, 1996). The vertical distribution of particles is characterized by a surface

165 maximum sustained by primary production, which decreases very quickly in the upper
166 200 m and exponentially at greater depth (Figure 2). Some regions are also characterized
167 by strong intermediate (*e.g.*, Iberian margin) and/or bottom (*e.g.*, western boundary of the
168 Atlantic basin) nepheloid layers, resulting in profiles with surface and near-bottom
169 maxima and a clear-water minimum in the 2000 - 3000 m depth range as illustrated in
170 Figures 2 and 3 (Brewer *et al.*, 1976; Biscaye and Eittrheim, 1977).

171 Particles in the upper 1000 m, especially in open ocean areas, are produced internally in
172 the marine system and are composed primarily of biogenic materials: particulate organic
173 matter, CaCO₃, and biogenic silica. Particles in regions with high external inputs, such as
174 the North Atlantic and the Mediterranean Sea with their high dust deposition and high
175 sedimentary inputs, have a composition characterized by a higher fraction of lithogenic
176 material –which could reach 70% of the total mass (Roy-Barman *et al.*, 2009)-
177 particularly at depths where biogenic matter is being remineralized (POM) or is
178 dissolving (biogenic silica or CaCO₃). A relatively high fraction of mineral particles is
179 also found in benthic nepheloid layers, where surface sediment particles that are
180 relatively poor in biogenic components are resuspended into bottom waters (Figure 3;
181 Gardner *et al.*, 1983).

182 Before the advent of the GEOTRACES program, full water column profiles of trace
183 metal and isotopic composition of suspended particles were measured in only a few
184 locations. The trace element composition (including Al, Mn, Fe, Co, Ni, Cu, Zn, Cd and
185 Pb) of suspended particles was measured at BATS in the Sargasso Sea (Sherrell and
186 Boyle, 1992), in the North Pacific subtropical gyre (Bruland *et al.*, 1994), and off Point
187 Conception (CA) in the Northeast Pacific (Sherrell *et al.*, 1998). The acetic acid leachable
188 and refractory fractions of particulate iron, manganese, and aluminum have been
189 measured in the North Pacific (Orians and Bruland, 1986; Landing and Bruland, 1987).

190 During the GEOSECS Atlantic cruises in the seventies, full water column data for a
191 whole suite of trace and minor elements were obtained by neutron activation of total
192 suspended matter (Ba, Ti, Sr, Mn, Mg, Cu, V, Al, Ca, La, Au, Hg, Cr, Sb, Sc, Fe, Zn, Co;
193 Peter Brewer, unpublished results). These data are currently being compared to those
194 obtained as part of the early GEOTRACES cruises. Such quality controlled data will be
195 further stored in the GEOTRACES Data Center, under the label “historical data”.

196

197 **2-2 Role of suspended particles in oceanic processes**

198

199 Once we understand what drives the dissolved/colloidal/particulate partitioning of a
200 tracer, this information can then be used in turn to trace oceanic processes. Non-
201 exhaustively, we can cite:

202 - Dissolved and particulate ^{230}Th and ^{231}Pa activity distributions provide an efficient tool
203 for estimating apparent particle settling velocities and therefore residence times in the
204 water column (Krishnaswami *et al.*, 1976; Bacon and Anderson, 1982; Bacon *et al.*,
205 1985; Roy-Barman *et al.*, 1996). The apparent settling velocity is the net effect of all
206 processes that are described in Figure 4. We acknowledge that reducing the particle
207 distribution in 2 categories only as represented in Figure 4 are simplified views of this
208 distribution, driven partly by sampling and analytical logistics. However, a two particle
209 class model captures two of the most important particle processes that are of interest here:
210 scavenging and sinking (see also McDonnell *et al.*, this issue). The particle residence
211 times deduced from these radionuclide distributions can therefore be applied to other
212 poorly soluble TEIs.

213 - $\Delta^{14}\text{C}$, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ distributions allow the identification of terrestrial versus marine
214 origin of organic matter, episodes of re-suspension of shelf or slope organic matter,
215 penetration of atmospheric nitrogen and carbon and/or oxidative processes (Williams *et*
216 *al.*, 1992; Mollenhauer *et al.*, 2003; Mollenhauer *et al.*, 2005)

217 - Biologically driven barite precipitation in the surface or sub-surface waters provide a
218 good tool for surface productivity reconstruction, in the modern as well as in the past
219 ocean (Dehairs *et al.*, 1991; Dehairs *et al.*, 1992; Jeandel *et al.*, 2000; Cardinal *et al.*,
220 2001; Jacquet *et al.*, 2008; Sternberg *et al.*, 2008; Paytan *et al.*, 1993; van Beek and
221 Reyss, 2001; van Beek *et al.*, 2002).

222 - Rare earth elements (REEs) and Nd isotopes trace the origin of suspended material as
223 well as dissolved-particle exchanges in the water column (Jeandel *et al.*, 1995;
224 Tachikawa *et al.*, 1999a; Kuss *et al.*, 2001).

225 - Manganese and iron are redox sensitive and less soluble when oxidized. In the surface
226 waters, photochemistry can efficiently change the speciation of these tracers and

227 therefore their distributions. In the water column, co-precipitation and/or adsorption of
228 TEIs on Mn and Fe oxyhydroxides result in removal of elements such as Co, Cu, Ni, Zn,
229 Th but also REEs (Anderson *et al.*, 1983a, b; Landing and Bruland, 1987; Sherrell and
230 Boyle, 1992; Moffett and Ho, 1996; Cardinal *et al.*, 2001; Roy-Barman *et al.*, 2009; Kuss
231 *et al.*, 1999; Tachikawa *et al.*, 1999b).

232 - Particle formation above submarine hydrothermal vents plays an important role in
233 modifying the gross flux from hydrothermal systems to the oceans. Approximately 50%
234 of the dissolved Fe released from a high-temperature vent is predicted to be precipitated
235 in the form of polymetallic sulfides in buoyant hydrothermal plumes within minutes of
236 emission from the seafloor (Rudnicki and Elderfield, 1993). The remaining Fe
237 precipitates more slowly, in the form of Fe-oxyhydroxides (Sherrell *et al.*, 2000) which
238 can significantly impact the scavenging of trace elements and isotopes (oxyanions, Be, Y,
239 REE, Th, Pa) from the water column (Michard *et al.*, 1983; Lilley *et al.*, 1993). This
240 hydrothermal scavenging can be so pronounced as to match the boundary scavenging
241 effects seen at high productivity ocean margins (German *et al.*, 1997). Most prior works
242 assumed that hydrothermal plume particle formation is an inorganic process, but recent
243 studies have shown that significant concentrations of organic carbon are incorporated into
244 hydrothermal particles (Bennett *et al.*, 2011; Toner *et al.*, 2009) and further that the
245 formation of these particulate phases may be microbially mediated (Sylvan *et al.*, 2012).

246

247 **3- The role of sinking particles in TEI cycling**

248

249 Large particles (> 53 μm , under typical methodological size fractionation) make up most
250 of the vertical flux and therefore contribute to the sequestration of most elements in the
251 deep ocean. The size criterion for separating suspended and sinking particles is more an
252 operational definition than a biogeochemical one: small and dense particles (as fecal
253 pellets for example) can sink faster than large fluffy aggregates (McCave, 1975). Indeed,
254 in the ocean, particle distribution follows a continuous spectrum whose sinking rates do
255 not necessarily increase monotonically with size (McDonnell and Buesseler, 2010;
256 McDonnell *et al.*, this issue).

257 There are two approaches to sampling sinking particles to study TEI cycling: 1) size-
258 fractionated filtration, which separates the particle pool into an operationally defined
259 “suspended” size class (e.g. $< 53 \mu\text{m}$) and a “sinking” size class (e.g. $> 53 \mu\text{m}$), and 2)
260 direct collection of sinking particles of various sizes in sediment traps (Honjo, 1978;
261 McDonnell *et al.*, this issue). In the first approach, geochemists analyze the TEI contents
262 of the different fractions, providing “state variables” of the systems to the modelers (Lam
263 *et al.*, this issue; Dutay *et al.*, 2009; Dutay *et al.*, this issue). Despite this crude and
264 operational separation of the particle pool, thorium isotope distribution studies have
265 nonetheless shown that small and large particles exchange with each other throughout the
266 water column, as well as with the dissolved phases. This has yielded the conceptual
267 model for particle dynamics first proposed by Bacon *et al.* (1985) and represented in
268 Figure 4.

269 In addition to thorium isotopes, measurements of the size-fractionated concentrations of
270 other TEIs such as manganese, neodymium and barium have also yielded insights into
271 particle aggregation and disaggregation processes (Bishop and Fleisher, 1987; Jeandel *et al.*,
272 1995; Bishop and Wood, 2008).

273 In the second approach, sinking particles collected from sediment traps are analyzed
274 directly. The majority of sediment trap studies have had as their goal a better
275 understanding of the biological pump. As such, sediment trap studies most frequently
276 report measurements of particulate organic carbon (POC) and particle mass, and often
277 also major particle phases such as CaCO_3 , biogenic silica, and lithogenic material, but
278 TEI measurements are much more rare (Brewer *et al.*, 1980).

279 Compilations of the major phase composition (POM, CaCO_3 , biogenic Si, lithogenics) of
280 sinking particles from bottom-tethered sediment traps during the JGOFS era have been
281 published (Antia *et al.*, 2001; Armstrong *et al.*, 2002; François *et al.*, 2002; Klaas and
282 Archer, 2002; Lutz *et al.*, 2007; Honjo *et al.*, 2008; Honjo *et al.*, 2010) and show a wide
283 geographic range in the magnitude and efficiency of POC flux to depth. Analysis of a
284 compilation of $>53\mu\text{m}$ POC, CaCO_3 and biogenic Si concentrations also show wide
285 geographic and temporal range in the transfer of POC to depth (Lam *et al.*, 2011).

286 Several studies have noted correlations between the fluxes of POC and CaCO_3 in deep
287 sediment traps ($> 1000 \text{ m}$) and have sparked numerous other studies as to the processes

288 behind this correlation. In contrast, the fraction of net primary production that is
289 exported from the euphotic zone is often correlated with the abundance of large
290 phytoplankton taxa, especially diatoms (Buesseler, 1991; Buesseler *et al.*, 2007a; Guidi *et al.*,
291 *et al.*, 2009; Honda and Watanabe, 2010), illustrating that controls on shallow export flux
292 may be decoupled from controls on deep POC flux (François *et al.*, 2002; Lomas *et al.*,
293 2010). Several time-series stations such as Bermuda Atlantic Time Series Study
294 (<http://bats.bios.edu>), Hawaii Ocean Time series in the Pacific
295 (<http://hahana.soest.hawaii.edu/hot/hot-dogs/interface.html>), DYFAMED time series in
296 the Mediterranean Sea (<http://www.eurosites.info/dyfamed.php>; Miquel *et al.*, 2011) and
297 ESTOC time series north of the Canary Islands (Neuer *et al.*, 1997; Patsch *et al.*, 2002),
298 as well as dedicated programs such as EUMELI (Bory *et al.*, 2001), VERTIGO
299 (Buesseler *et al.*, 2007a; Lamborg *et al.*, 2008), and MedFlux (Lee *et al.*, 2009) have also
300 shown wide ranging temporal variability in particle flux and composition. Even though
301 there are relatively few studies that have measured TEIs directly on sinking particles
302 (Huang and Conte, 2009), the wide geographic and temporal variability in particle
303 sinking flux implies that the sinks of particle-reactive TEIs will experience similar
304 variability (Antia *et al.*, 2001; Scholten *et al.*, 2001).

305 At some of the sites listed above and elsewhere, TEIs were measured in the trapped
306 material too. Most of these works used U-Th series to reconstruct or calibrate POC
307 fluxes (Cochran *et al.*, 1993; Sarin *et al.*, 2000; Roy-Barman *et al.*, 2005; Stewart *et al.*,
308 2007; Trull *et al.*, 2008; Cochran *et al.*, 2009; Roy-Barman *et al.*, 2009). Others used
309 stable ^{13}C and ^{15}N or barite to differentiate biogeochemical cycles (Jeandel *et al.*, 2000;
310 Lourey *et al.*, 2004; Casciotti *et al.*, 2008), and a few have used REE and radiogenic
311 isotope data to trace the origin of the particles (Jeandel *et al.*, 1995; Tachikawa *et al.*,
312 1997; Chavagnac *et al.*, 2008). The pioneer VERTEX program allowed investigations of
313 the major and trace element composition of sinking particles from the Pacific (Knauer *et al.*,
314 *et al.*, 1979; Fowler and Knauer, 1986) but the measurement of contamination-prone TEIs
315 in sediment trap samples has only become more common recently (Kuss and Kremling,
316 1999; Frew *et al.*, 2006; Lamborg *et al.*, 2008; Bowie *et al.*, 2009; Ho *et al.*, 2010; Ho *et al.*,
317 *et al.*, 2011). When studying fluxes of trace elements collected by sediment traps, one must

318 be aware of the tendency for TEIs to dissolve into supernatant solutions (Kumar *et al.*,
319 1996).

320

321 **4- Partition coefficients of trace elements: from the ocean to the models**

322

323 The chemical behavior of particle-reactive metals such as Th, Pa, Nd and other REE is
324 often characterized by a partition coefficient K_d between seawater and marine particles
325 defined as:

$$326 K_d = \frac{\text{mass of particulate tracer per mass of particles}}{\text{mass of dissolved tracer per volume of seawater}}$$

327

328 To first order, K_d for a given element is expected to depend of the chemical bulk
329 composition of the marine particles. Several approaches have been used to determine the
330 relationship between K_d and particle composition.

331 For elements having isotopes produced *in situ* such as Th and Pa, two methods have been
332 used: 1) correlation of isotopes produced *in situ* with the main components of sinking
333 marine particles collected by sediment traps, and 2) sorption experiments using natural or
334 artificial seawater and particles. Sediment trap analyses have shown correlations between
335 radioisotopes and inorganic phases, but fortuitous correlations between components have
336 produced conflicting interpretations (Chase *et al.*, 2002; Luo and Ku, 2004; Roy-Barman
337 *et al.*, 2005; Roy-Barman *et al.*, 2009). Some of these fortuitous correlations could be
338 avoided by directly studying small filtered particles, because they dominate the solid
339 surface area per volume and thus are more likely to adsorb tracers from seawater. This
340 would require that the total mass and the major components of filtered particles be
341 determined (Lam *et al.*, this issue). While focus has mainly been on the impact of major
342 components on K_d (see references above), minor phases such as Mn oxides could play a
343 significant role in the scavenging of Th (Roy-Barman *et al.*, 2009) and Pa (Anderson *et*
344 *al.*, 1983a,b) in deep waters. Sorption experiments have shown a relatively low affinity of
345 Th for inorganic phases and a high affinity for organic compounds (Santschi *et al.*, 2006).
346 These results are consistent with ^{234}Th scavenging in shallow waters, but they fail to

347 explain the correlations between ^{230}Th and inorganic phases (carbonate, lithogenic or Mn
348 oxides, see previous paragraph) observed in sediment trap data.

349 For elements derived from continental erosion, such as Neodymium (Nd) or Hafnium
350 (Hf), with no in situ sources of isotopes, the authigenic fraction of the elements in
351 particles can be determined by subtraction of the lithogenic fraction (Kuss *et al.*, 2001,
352 Garcia-Solsona *et al.*, 2014), chemical leaching or isotopic balance (Tachikawa *et al.*,
353 1999b; Tachikawa *et al.*, 2004). These methods do not necessarily give consistent results.
354 In the case of leaching, the selective dissolution of authigenic phases, without
355 contamination from other phases, remains to be demonstrated. More importantly, re-
356 adsorption of leached TEIs to refractory phases, and the incomplete removal of colloidal
357 materials mobilized during leaching procedures, can confound the interpretation of the
358 original carrier of TEIs (Lam *et al.*, this issue). Consequently, an approach based on
359 isotopic mass balance or on the statistical correlation among end member particulate
360 phases is preferred.

361 Recently, physical separations have brought new insights by partially isolating and
362 enriching some carriers (Kretschmer *et al.*, 2010; 2011). The development of the analysis
363 of individual particles allows the unambiguous determination of some carriers (Roy-
364 Barman, pers. comm.). Particle observation should be systematically coupled to particle
365 analysis (Lam *et al.*, this issue). Besides methodological aspects, fundamental aspects of
366 the tracer's behavior must be addressed:

- 367 - Possible disequilibrium between particles and seawater (Coppola *et al.*, 2006;
368 Venchiarutti *et al.*, 2011).
- 369 - The role of the colloidal phase for both organic and inorganic compounds.
- 370 - The impact of mineralization on the particle composition and K_d .

371 The present uncertainties on the K_d of Pa, Th and Nd have direct impacts on our
372 understanding of the distribution of these tracers in the ocean. For example, several
373 models "successfully" represent the Nd concentration and isotopic composition in the
374 ocean but in fact use different particle models (particle mineralization or boundary
375 exchange) and K_d (equilibrium versus adsorption-desorption) that are adjusted to
376 eventually match the data (synthesis in Rempfer *et al.*, 2011, 2012; Arsouze *et al.*, 2009).

377 More dissolved and particle data from representative oceanic regimes are required to
378 constrain models, one of the main missions of GEOTRACES.

379

380

381 **5- Benthic and Intermediate Nepheloid Layers and their impacts on TEI**

382 **distribution**

383

384 Benthic Nepheloid Layers (BNLs) occur wherever bottom currents interact with the
385 (deep) sea floor (Biscaye and Eittrheim, 1977, McCave *et al.*, 2001). In the discussion of
386 the effect of a BNL on the distribution of TEIs, we can distinguish the effects at two
387 spatial scales: (i) the effects on a local scale, like those related to currents characterized
388 by high level of eddy kinetic energy and to currents over seamounts (Turnewitsch *et al.*,
389 2008) and (ii) the effects on a larger scale related to large-scale abyssal circulation.

390

391 **5-1 Local re-suspension**

392

393 The generation of a BNL and the distribution and size spectra of particles have been
394 described by McCave (1984, 1986, 2001). Vertical mixing in bottom layers was studied
395 during GEOSECS with ^{222}Rn (Sarmiento *et al.*, 1976). The vertical extent of BNLs is
396 enhanced by the detachment of bottom mixed layers (Armi and D'Asaro, 1980). If surface
397 sediments are in adsorption equilibrium with the bottom water, re-suspension need not
398 change this equilibrium. However, there are cases in which interaction between re-
399 suspension and bioturbation can change the distribution of dissolved components in the
400 BNL relative to the water layer just above the BNL: 1) if the tracer decays within the
401 bioturbated zone, or 2) if K_d changes as a result of diagenetic changes (e.g. MnO_2
402 enrichment) or particle dynamics like aggregation-disaggregation (Rutgers van der Loeff
403 and Boudreau, 1997). There is no indication that the particle concentration has an effect
404 on the K_d in the BNL (Honeyman *et al.*, 1988).

405 For short-lived radionuclides like ^{234}Th and ^{210}Pb , condition (1) above is clearly met.

406 Profiles of dissolved ^{234}Th provide clear evidence for enhanced removal of dissolved

407 TEIs from bottom waters in the presence of nepheloid layers (Bacon and Rutgers van der

408 Loeff, 1989). Enhanced removal of particle-reactive TEIs near the sea bed has been
409 evident since GEOSECS-era studies of ^{210}Pb (Craig *et al.*, 1973), and the concept of
410 bottom scavenging has been reintroduced recently through the study of ^{230}Th (Okubo *et*
411 *al.*, 2012). However, developing a direct link between sediment re-suspension and
412 enhanced removal of TEIs near the sea bed will require joint research on particles as well
413 as on the distribution of dissolved TEIs.

414

415 **5-2 Long-range transport in the BNL**

416

417 Strong bottom currents occur along the western boundaries of the ocean basins (Warren,
418 1981), and deep wind and buoyancy-driven currents such as the Antarctic Circumpolar
419 Current can reach abyssal depths (e.g. in the Drake Passage; Renault *et al.*, 2011).

420 Through re-suspension or, rather, selective deposition, these currents can maintain high
421 loads of suspended sediments. In the BNL, particles may be transported over large
422 distances as shown for clay minerals (Griffin *et al.*, 1968; Petschik *et al.*, 1996;
423 Diekmann *et al.*, 2004). This means that particles are not only redistributed locally
424 (winnowing and focusing) but also transported between areas with widely different local
425 sediment compositions.

426

427 **5-3 TEI fractionation**

428

429 The composition of material suspended in the BNL is different from that in the clear
430 water above it. Grain size fractionation has been described in detail by the studies of I.
431 McCave (McCave, 2001). The possible effect of grain size fractionation on the isotopic
432 composition of deposited sediments was studied by Kretschmer *et al.* (2010; 2011) who
433 found that :

- 434 • ^{230}Th , ^{231}Pa and ^{10}Be adsorb preferentially onto the smallest grain sizes
- 435 • $^{231}\text{Pa}/^{230}\text{Th}$ and $^{10}\text{Be}/^{230}\text{Th}$ ratios are enhanced in a slowly settling pure opal
436 fraction
- 437 • Settling rate fractionation during sediment focusing causes an increase in the bulk
438 ^{230}Th concentration and in the $^{231}\text{Pa}/^{230}\text{Th}$ ratio.

439

440 5-4 Intermediate Nepheloid Layers

441

442 There are many examples of Intermediate Nepheloid Layers (INLs) caused by the
443 detachment of a BNL at the shelf break and other breaks in slope where internal tidal
444 energy is focused, followed by offshore advection (McCave *et al.*, 2001). It would be
445 important to study the link between the dispersal of particulate (INLs) and dissolved
446 tracer signals from the shelf (e.g. Fe and Mn releases, ²¹⁰Pb removal, Nd isotope
447 exchange (Sherrell *et al.*, 1998; Lacan and Jeandel, 2005; Lam and Bishop, 2008). The
448 particulate signal disappears by sinking and aggregate formation (Clegg and Whitfield,
449 1990, 1991; Karakas *et al.*, 2006; 2009). The time scale of distribution of dissolved shelf
450 inputs can be studied with short lived Ra isotopes and ²²⁸Th.

451

452 6- “Historical” understanding of particle dynamics and perspectives

453

454 Despite its fundamental role in controlling the chemical composition of the ocean
455 (Goldberg, 1954; Turekian, 1977) and the different cruises conducted in the 70s and 80s,
456 the “oceanic microcosm of particles”-as christened by Lal (1977) – is far from being
457 understood yet. In addition, sampling strategies and scientific focus differed between the
458 GEOSECS and JGOFS programs. GEOSECS carried out large sections across the
459 oceanic basins, where particles were collected by membrane filtration after bottle
460 sampling, biasing the sampling towards the small particle pool. Analyses mostly
461 informed us about the distribution of particle concentrations (mass/L), their major
462 element compositions, as well as a few tracers and selected morphological and qualitative
463 composition descriptions, thanks to the first Scanning Electron Microscopy (SEM)
464 analyses. Subsequent box and one-dimensional (vertical) models described the different
465 fluxes exchanged in and out the oceanic system as well as along the water column. These
466 pioneering efforts led to the emergence of the fundamental notion of “reversible
467 scavenging” (Brewer *et al.*, 1976; Krishnaswami *et al.*, 1976; Lal, 1980; Nozaki *et al.*,
468 1981; Bacon and Anderson, 1982; Anderson *et al.*, 1983a). They also highlighted the role

469 of "particle-rich" continental margins on the distribution of ocean tracers (Anderson and
470 Henderson, 2003; Jeandel *et al.*, 2011).
471 JGOFS identified representative biogeochemical provinces of the ocean, where most of
472 the work was dedicated to the quantification of the "exported carbon flux" and its
473 seasonal variability (Fasham *et al.*, 2001). Except for rare studies just prior to JGOFS
474 that conducted small particle sampling and deployed the first in situ pumps allowing
475 large volume filtration (Krishnaswami *et al.*, 1976; Bacon and Anderson, 1982; Bishop *et al.*,
476 1985; Rutgers van der Loeff and Berger, 1993; Jeandel *et al.*, 1995; Tachikawa *et al.*,
477 1999b), most of the field work conducted during JGOFS deployed moored and (or)
478 drifting sediment traps. TEIs were barely measured, except perhaps ^{234}Th and ^{230}Th
479 isotopes, which were recognized as useful for POC flux calibration and quantification.
480 Resulting models describe the exported carbon flux as it was related to the surface
481 nutrient distribution using 1D and 3D models coupling physics and biology (Bopp *et al.*,
482 2002). Most of the particle models developed in the late 80s and in the 90s are
483 mechanistic and abiotic (Dutay *et al.*, this issue; Burd and Jackson, 2009; Jackson and
484 Burd, this issue). Early models coupled particle dynamics to ocean circulation in an
485 OGCM, although processes describing the particle behavior in such 3D dynamical
486 models remained one-dimensional (Henderson and Maier-Reimer, 2002; Gehlen *et al.*,
487 2003; Gehlen *et al.*, 2006; Arsouze *et al.*, 2009; Dutay *et al.*, 2009; Rempfer *et al.*, 2011).

488

489 **Conclusion**

490

491 At the beginning of the GEOTRACES program, we have to admit that our collective
492 understanding of the processes governing the solution-particle exchange has made little
493 progress in the preceding two decades. Key questions remain:

494 i) What are the affinities of the various TEIs for the different particulate phases
495 (Rutgers van der Loeff and Berger, 1993; Chase *et al.*, 2002; Anderson and Henderson,
496 2003; Geibert and Usbeck, 2004; Luo and Ku, 2004; Roy-Barman *et al.*, 2005; Santschi
497 *et al.*, 2006; Roy-Barman *et al.*, 2009)?

- 498 ii) What is the role of remineralization in the mesopelagic zone (Dehairs *et al.*, 1995;
499 Dehairs *et al.*, 1997; Frew *et al.*, 2006; Boyd and Trull, 2007; Buesseler *et al.*, 2007b;
500 Dehairs *et al.*, 2008)?
- 501 iii) What is the impact of sediment diagenesis on the composition of resuspended
502 particles and on their ability to scavenge additional TEIs, despite having previously
503 equilibrated with dissolved species in the water column (Kretschmer *et al.*, 2010;
504 Kretschmer *et al.*, 2011)?
- 505 iv) What are the roles of the BNLs and INLs on the boundary scavenging and boundary
506 exchange processes (Bacon *et al.*, 1988; Roy-Barman *et al.*, 2005; Roy-Barman *et al.*,
507 2009)?
- 508 v) What is the importance of other surface processes like chemoautotrophy as a source of
509 particles in the deep ocean (Honjo *et al.*, 2012)?
- 510 Answers to these questions can be provided by the GEOTRACES program with the
511 implementation of a comprehensive sampling and analytical strategies (pumps, optics,
512 observations and analysis of particles, see McDonnell *et al.*, this issue; Boss *et al.*, this
513 issue, Lam *et al.*, this issue...), designed to elucidate the role of particles as agents of
514 supply and removal of TEIs in the ocean. There is an urgent need for re-focusing on
515 discrete particle composition, speciation and morphologies.

516

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524

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1100 **Figure Captions**

1101

1102 **Figure 1**

1103 Illustration of the different sources, internal cycling and sinks of oceanic particles. Reproduced
1104 from Roy-Barman and Jeandel (2011).

1105

1106 **Figure 2**

1107 Profiles of particulate matter concentration (PMC) at the Northern Iberian Margin (43°N, June
1108 1997) calculated from beam attenuation (solid line) and light scattering (dotted line) against
1109 depth, together with the density structure (σ_t) of the water column (dashed line). *Reprinted from*
1110 *“Hall, I. R., Schmidt, S., McCave, I. N., Reyss, J.-L., 2000. Deep Sea Research Part I:*
1111 *Oceanographic Research Papers 47, 557-582. Copyright (2014), with permission from Elsevier”*

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1113 **Figure 3**

1114 Longitudinal section of the dry weight of particulate matter along the western Atlantic Ocean
1115 from the GEOSECS program. *Reprinted from “Brewer, P. G., Spencer, D. W., Biscaye, P.*
1116 *E., Hanley, A., Sachs, P. L., Smith, C. L., Kadar, S., Fredericks, J., 1976. The*
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1118 *Letters 32, 393-402. Copyright (2014), with permission from Elsevier”*

1119

1120 **Figure 4**

1121 Particle dynamics as depicted by thorium (Th) isotopes in the mid-80s. Th isotopes are produced
1122 in solution by radioactive decay of the soluble U or Ra isotopes. Due to their very low solubility,
1123 Th isotopes are rapidly adsorbed on small particles and colloids that represent most of the
1124 available solid surface. Th isotopes then follow the dynamics of particles. Th isotopes differ by
1125 their radioactive decay constants (λ) and input functions. Combining the different isotopes
1126 allows determining the other time constants: k_{ads} for adsorption, k_{des} for desorption, k_{aggr} for
1127 aggregation, k_{dis} for disaggregation, as well as the sinking speeds of the different types of
1128 particles. Remineralization of large particles was neglected due to the low solubility of thorium.
1129 Colloids were not included either because their impact on Th isotopes was highlighted later
1130 (Honeyman and Santschi, 1989). The main 1-D scheme is certainly dramatically oversimplified
1131 compared to the ecosystem-driven real processes. Small particles are aggregated into large
1132 particles either by zooplankton grazing (producing fecal pellets) or by abiotic aggregation of
1133 organic and inorganic material in fluff, due to sticking exudates produced at the end of the bloom.

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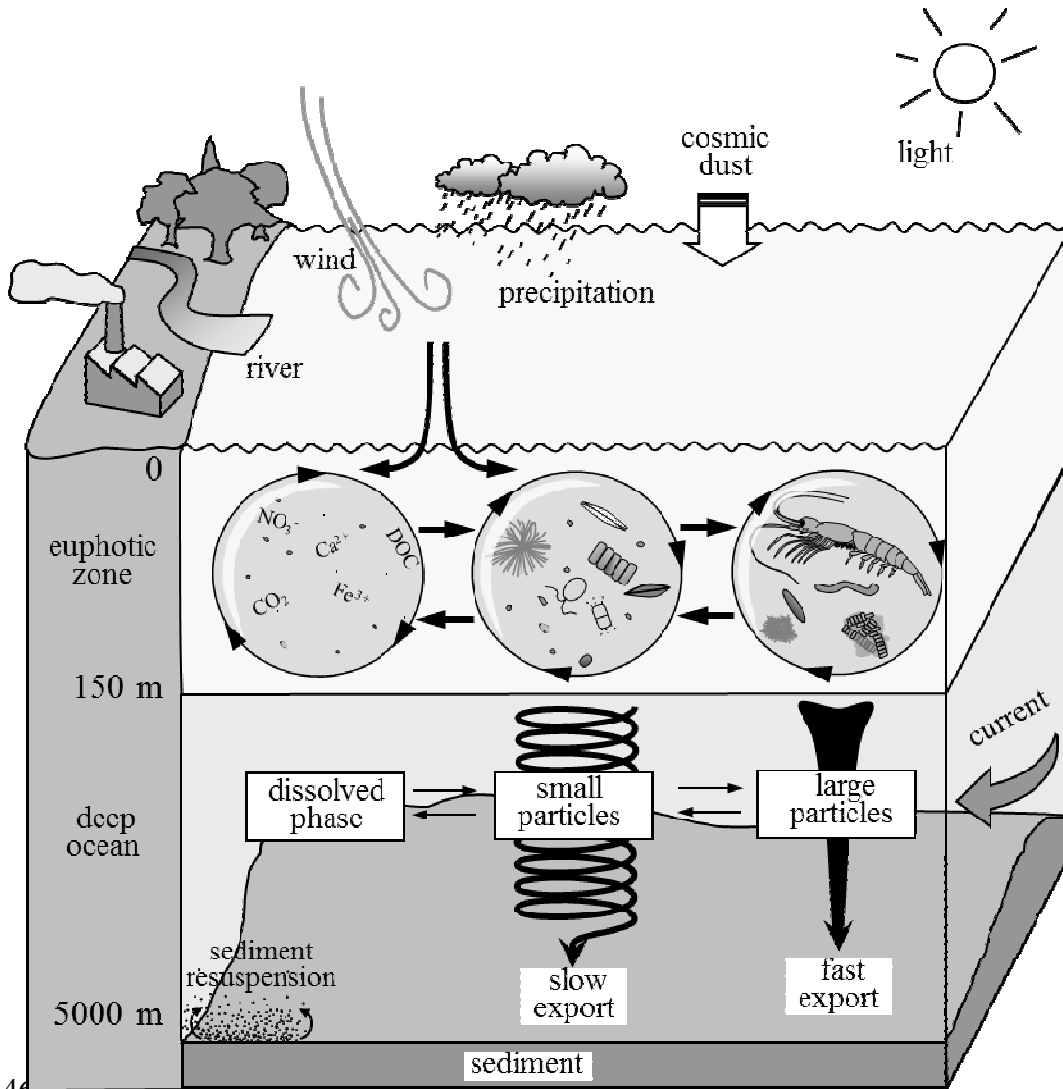
1134 Large particles can scavenge and drag small ones in a sort of "oceanic piggy-back" process (Lal,
1135 1980). Large particles can also disaggregate into small particles when sinking. Indeed, the most
1136 fragile large particles, such as marine snow, can be broken by the turbulence of the current. Fecal
1137 pellets can also be destroyed by bacterial activity. The apparent settling velocity is the net effect
1138 of all these processes. The deduced particle residence time can therefore be applied to other
1139 poorly soluble TEIs. From Roy-Barman and Jeandel (2011) and redrawn from Bacon *et al.*
1140 (1985).
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1142 **Figure 1**

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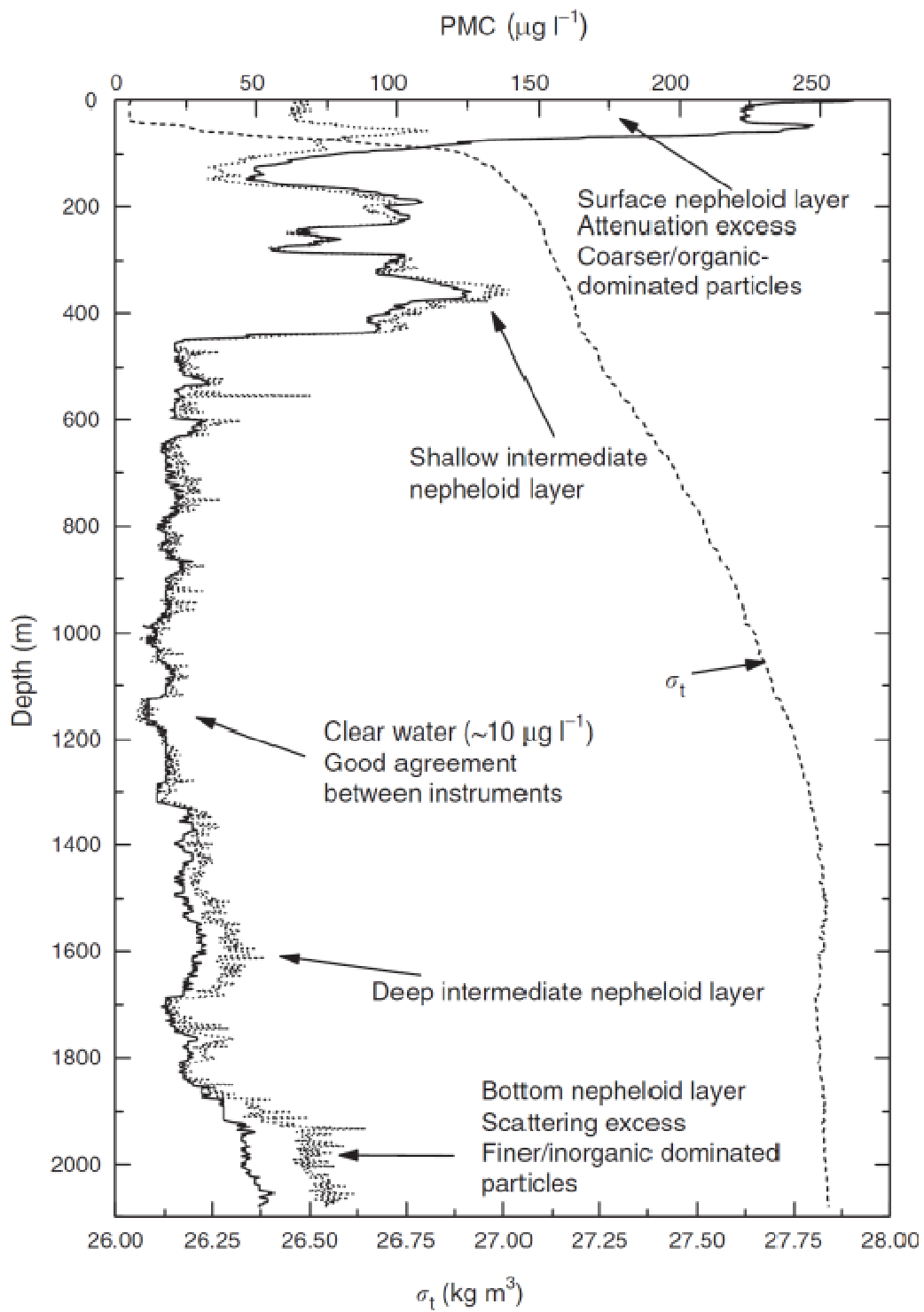
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Figure 2



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1153 **Figure 3**
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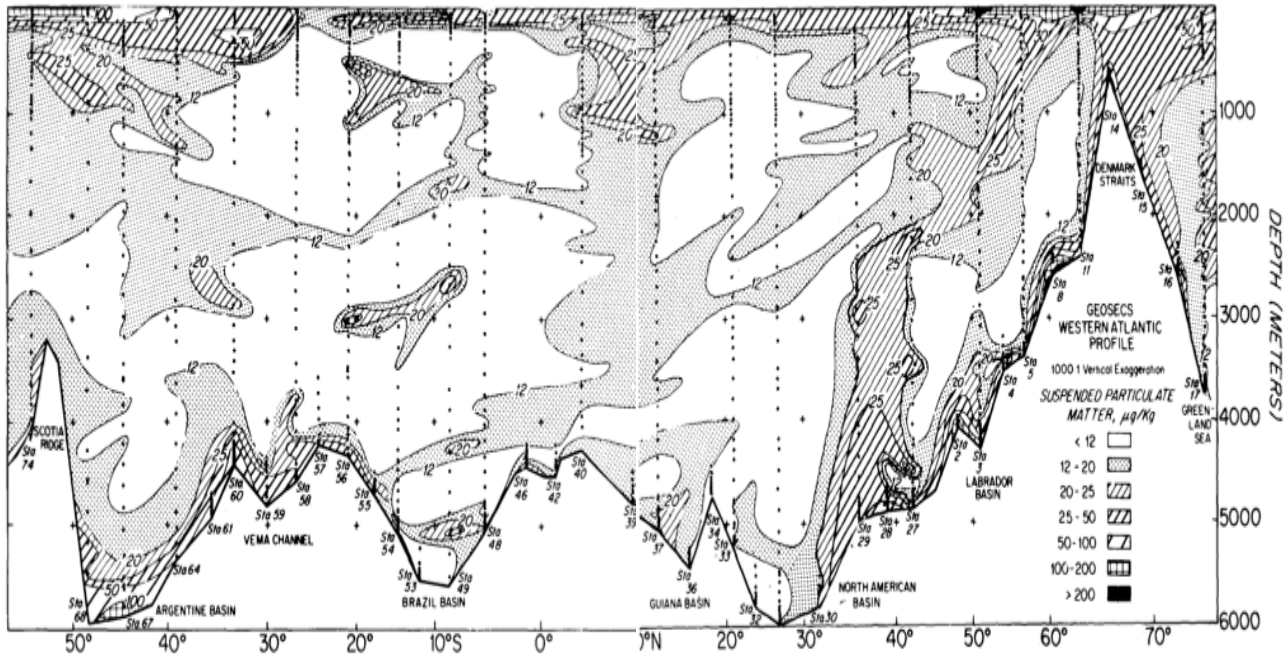
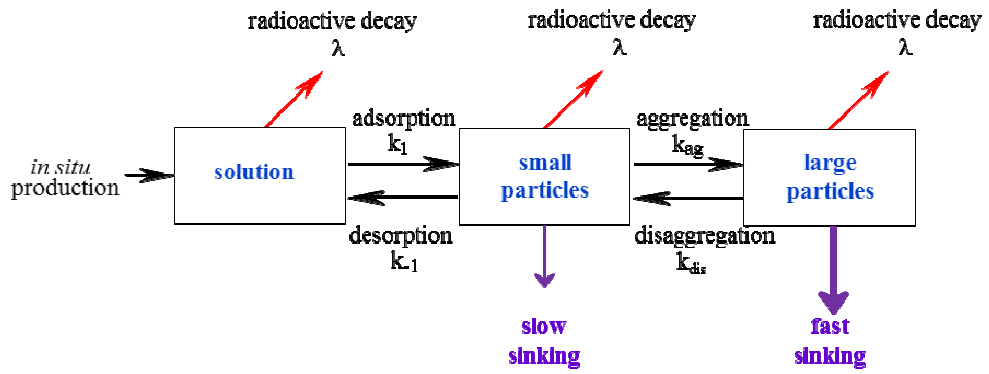


Fig. 2. Longitudinal section of the dry weight of particulate matter in the western Atlantic Ocean.

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Figure 4



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