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

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

Abstract

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

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

Introduction

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

In the 1990's, the JGOFS program substantially increased our understanding of the standing stock, vertical flux and fate of marine particles, with the focus largely on carbon and associated nutrient cycles (Fasham *et al.*, 2001). However, because of data scarcity and the large variability of particle fluxes in time and space, the full characterization of marine particle concentrations, flux, and composition was a difficult task, and remained far from being achieved. The JGOFS era also suffered from a lack of methodologies for determining TEIs, which are extremely helpful for quantifying specific particle processes in the water column. Although the analytical protocols for assessing some TEIs were available and applied during some JGOFS research projects, they did not yet represent the major research target. As a consequence, the global distribution of dissolved and particulate TEIs is poorly known today. Because some TEIs are powerful tracers of particle origin and processes (e.g. settling velocity, rates of dissolution, precipitation, adsorption, and desorption) and some are essential micronutrients whose speciation in the solid and dissolved phases is of prime importance for their bioavailability, there is an urgent need to understand the global distribution of TEIs in the oceanic environment. Filling this gap by investigating the sources, behavior and sinks of these TEIs is the main goal of the GEOTRACES program (www.geotraces.org), which was developed following the model of its "parent" program GEOSECS but with more emphasis on the collection, observation, speciation and analysis of marine particles. As we enter the early stages of the new GEOTRACES era, the present work reviews our understanding, informed by GEOSECS and JGOFS, of the distribution of suspended and sinking marine particles of both biogenic and abiogenic origin, as well as the role of these particles as regulators of the marine biogeochemical cycles of TEIs. In addition to Anderson and Hayes' introduction (this issue), this paper provides the historical context for this special issue that proposes to browse the state of the art of our present knowledge on optically characterizing (Boss *et al*., this issue), collecting (McDonnell *et al*., this issue), analyzing (Lam *et al*., this issue) and modelling (Dutay *et al*., this issue; Jackson and Burd, this issue) marine particles. The issue is concluded by Henderson and Marchal's comments and perspectives.

1- The origin of marine particles

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

Sources external to the marine system: By means of erosion, continents bring particles (natural or anthropogenic, mineral or organic) into the ocean. These are transported via the atmosphere (winds, rains), rivers (sedimentary discharge), or by lateral transport from continental margin sediments. Before extensive damming, the annual solid flux discharged by rivers to the oceans was of the order of 19×10^{15} g (Peucker-Ehrenbrink, 2009), which is 50 times the atmospheric flux (Jickells *et al.*, 2005). Other particles are extraterrestrial, such as micrometeorites (10 to 100 µm in size) or cosmic dust that would represent a flux of between 7 to 14 X 10^9 g/y to the oceans (Johnson, 2001). Nano-metric-sized (and highly magnetic) particles were also detected in the Greenland and Antarctic ice caps and are identified as originating from atmospheric ablation of meteorites and micrometeorites at high (~100 km) altitude (Lanci *et al.*, 2004; 2007). Finally, hydrothermal vents also are a significant "external" source of particles for the deep ocean. Particles from hydrothermal vents precipitate within the plume, forming fine grained sulfide and oxide minerals that may be distributed over large regions of the deep ocean (Mottl and McConachy, 1990; Feely *et al.*, 1996; Sherrell *et al.*, 2000; Tagliabue *et al.*, 2010).

Sources internal to the marine system: A huge quantity of marine particles is produced by biological activity. Photoautotrophic plankton assimilates dissolved species (C, N, P, Si, trace metals) and uses solar energy to synthesize organic matter, and specific groups, including microheterotrophs, also secrete skeletal parts consisting of calcite, aragonite, opal, or celestite. The annual flux of material so produced represents ~ 60×10^{15} g/y of organic carbon (Fasham *et al.*, 2001). The magnitude of marine primary production is similar to terrestrial primary production, but the standing stock of fixed organic carbon is far less in the ocean than in terrestrial systems, resulting in much higher turnover rate of carbon in the ocean. This high turnover rate has consequences for the cycling of TEIs associated with this biogenic material. Other autotrophic organisms, such as nitrifiers, use chemical energy, and are called chemolithotrophic (Griffith *et al.*, 2012; Honjo *et al.*, 2012). These thrive throughout the oceanic water column and produce new biomass in-situ. Autotrophic carbon fixation is the point of departure of the trophic chain whose life and death cycle generates particles throughout the water column. Among the heterotrophs, microzooplankton species, such as foraminifera, radiolarians, but also larger multi-cellullar organisms such as salps and pteropods, represent a significant portion of living biomass (Buitenhuis *et al.*, 2013). Although less abundant than phytoplanktonic organisms, they are important because of their role in packaging and remineralization and for their potential as recorders, once incorporated in ocean sediments, of past environmental conditions. In addition, diel vertical migration by mesozooplankton may represent a significant pathway of particle redistribution in the mesopelagic zone, between water depths of about 100 m and 1000 m (Steinberg *et al.*, 2008). Another source of particles in the upper water column is through spontaneous aggregation of Dissolved Organic Matter (DOM) into larger particles, from the molecular size up to a typical size of 4 µm, therefore becoming Particulate Organic Matter (POM). These particles have been termed microgels (Verdugo, 2012). Barium sulfate, and manganese and iron oxides and hydroxides are also known to precipitate within the water column, incorporating other elements in the process, or scavenging trace elements by adsorption or other particle surface phenomena (Krishnaswami *et al.*, 1976a,b; Bishop and Fleisher, 1987; Dehairs *et al.*, 1990; Sherrell and Boyle, 1992; Paytan *et al.*, 1993; van Beek *et al.*, 2007; van Beek *et al.*, 2009).

Marine particles are often divided in 2 different types: small (micron-size) and slowly sinking particles on one hand and large (> 50-100 micron-size) and rapidly sinking on the other hand. The cut off is both poorly defined and somewhat arbitrary. However, it corresponds to 2 modes of marine particle sampling: filtration on filters with (sub-) micron size porosity for the small particles and collection in sediment trap and/or filtration with large porosity for large particles. Hence, this operational definition is still used in the GEOTRACES program.

2- Small suspended particles and TEI behavior

2-1 Oceanic distribution of suspended particles

Small particles (0.2-53 µm) constitute the bulk of the particle standing stock in the ocean. In the upper 1000 m, particles < 53 µm represent on average ~80% of total suspended particle mass (Bishop *et al.*, 1977; Bishop *et al.*, 1978; Bishop *et al.*, 1980; Bishop *et al.*, 1985; Bishop *et al.*, 1986; Lam and Bishop, 2007; Bishop and Wood, 2008). Their amount and their large surface areas propel them as active players in the solution-solid exchanges that impact TEI distribution (Krishnaswami *et al.*, 1976; Anderson *et al.*, 1983a,b; Bishop and Fleisher, 1987; Sherrell and Boyle, 1992; Jeandel *et al.*, 1995; Roy-Barman *et al.*, 1996). The vertical distribution of particles is characterized by a surface maximum sustained by primary production, which decreases very quickly in the upper 200 m and exponentially at greater depth (Figure 2). Some regions are also characterized by strong intermediate (*e.g.,* Iberian margin) and/or bottom (*e.g.,* western boundary of the Atlantic basin) nepheloid layers, resulting in profiles with surface and near-bottom maxima and a clear-water minimum in the 2000 - 3000 m depth range as illustrated in Figures 2 and 3 (Brewer *et al.,* 1976; Biscaye and Eittreim, 1977).

Particles in the upper 1000 m, especially in open ocean areas, are produced internally in the marine system and are composed primarily of biogenic materials: particulate organic matter, $CaCO_3$, and biogenic silica. Particles in regions with high external inputs, such as the North Atlantic and the Mediterranean Sea with their high dust deposition and high sedimentary inputs, have a composition characterized by a higher fraction of lithogenic material –which could reach 70% of the total mass (Roy-Barman *et al.,* 2009)- particularly at depths where biogenic matter is being remineralized (POM) or is dissolving (biogenic silica or $CaCO_3$). A relatively high fraction of mineral particles is also found in benthic nepheloid layers, where surface sediment particles that are relatively poor in biogenic components are resuspended into bottom waters (Figure 3; Gardner *et al.,* 1983).

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

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

2-2 Role of suspended particles in oceanic processes

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

- Dissolved and particulate ^{230}Th and ^{231}Pa activity distributions provide an efficient tool for estimating apparent particle settling velocities and therefore residence times in the water column (Krishnaswami *et al.*, 1976; Bacon and Anderson, 1982; Bacon *et al.*, 1985; Roy-Barman *et al.*, 1996). The apparent settling velocity is the net effect of all processes that are described in Figure 4. We acknowledge that reducing the particle distribution in 2 categories only as represented in Figure 4 are simplified views of this distribution, driven partly by sampling and analytical logistics. However, a two particle class model captures two of the most important particle processes that are of interest here: scavenging and sinking (see also McDonnell *et al.*, this issue). The particle residence times deduced from these radionuclide distributions can therefore be applied to other poorly soluble TEIs.
- $\Delta^{14}C$, $\delta^{13}C$ and $\delta^{15}N$ distributions allow the identification of terrestrial versus marine origin of organic matter, episodes of re-suspension of shelf or slope organic matter, penetration of atmospheric nitrogen and carbon and/or oxidative processes (Williams *et al.*, 1992; Mollenhauer *et al.*, 2003; Mollenhauer *et al.*, 2005)
- Biologically driven barite precipitation in the surface or sub-surface waters provide a good tool for surface productivity reconstruction, in the modern as well as in the past ocean (Dehairs *et al.*, 1991; Dehairs *et al.*, 1992; Jeandel *et al.*, 2000; Cardinal *et al.*, 2001; Jacquet *et al.*, 2008; Sternberg *et al.*, 2008; Paytan *et al.*, 1993; van Beek and Reyss, 2001; van Beek *et al*., 2002).
- Rare earth elements (REEs) and Nd isotopes trace the origin of suspended material as well as dissolved-particle exchanges in the water column (Jeandel *et al*., 1995; Tachikawa *et al.*, 1999a; Kuss *et al.*, 2001).
- Manganese and iron are redox sensitive and less soluble when oxidized. In the surface waters, photochemistry can efficiently change the speciation of these tracers and

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

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

3- The role of sinking particles in TEI cycling

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

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

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

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

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

Several studies have noted correlations between the fluxes of POC and $CaCO_3$ in deep sediment traps (> 1000 m) and have sparked numerous other studies as to the processes behind this correlation. In contrast, the fraction of net primary production that is exported from the euphotic zone is often correlated with the abundance of large phytoplankton taxa, especially diatoms (Buesseler, 1991; Buesseler *et al.*, 2007a; Guidi *et al.*, 2009; Honda and Watanabe, 2010), illustrating that controls on shallow export flux may be decoupled from controls on deep POC flux (François *et al.*, 2002; Lomas *et al.*, 2010). Several time-series stations such as Bermuda Atlantic Time Series Study (http://bats.bios.edu), Hawaii Ocean Time series in the Pacific (http://hahana.soest.hawaii.edu/hot/hot-dogs/interface.html), DYFAMED time series in the Mediterranean Sea (http://www.eurosites.info/dyfamed.php; Miquel *et al.*, 2011) and ESTOC time series north of the Canary Islands (Neuer *et al.*, 1997; Patsch *et al.*, 2002), as well as dedicated programs such as EUMELI (Bory *et al.*, 2001), VERTIGO (Buesseler *et al.*, 2007a; Lamborg *et al.*, 2008), and MedFlux (Lee *et al.*, 2009) have also shown wide ranging temporal variability in particle flux and composition. Even though there are relatively few studies that have measured TEIs directly on sinking particles (Huang and Conte, 2009), the wide geographic and temporal variability in particle sinking flux implies that the sinks of particle-reactive TEIs will experience similar variability (Antia *et al.*, 2001; Scholten *et al.*, 2001).

At some of the sites listed above and elsewhere, TEIs were measured in the trapped material too. Most of these works used U-Th series to reconstruct or calibrate POC fluxes (Cochran *et al.*, 1993; Sarin *et al.*, 2000; Roy-Barman *et al.*, 2005; Stewart *et al.*, 2007; Trull *et al.*, 2008; Cochran *et al.*, 2009; Roy-Barman *et al.*, 2009). Others used stable ^{13}C and ^{15}N or barite to differentiate biogeochemical cycles (Jeandel *et al.*, 2000; Lourey *et al.*, 2004; Casciotti *et al.*, 2008), and a few have used REE and radiogenic isotope data to trace the origin of the particles (Jeandel *et al.*, 1995; Tachikawa *et al.*, 1997; Chavagnac *et al.*, 2008). The pioneer VERTEX program allowed investigations of the major and trace element composition of sinking particles from the Pacific (Knauer *et al.*, 1979; Fowler and Knauer, 1986) but the measurement of contamination-prone TEIs in sediment trap samples has only become more common recently (Kuss and Kremling, 1999; Frew *et al.*, 2006; Lamborg *et al.*, 2008; Bowie *et al.*, 2009; Ho *et al.*, 2010; Ho *et al.*, 2011). When studying fluxes of trace elements collected by sediment traps, one must be aware of the tendency for TEIs to dissolve into supernatant solutions (Kumar *et al*., 1996).

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

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

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

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

For elements having isotopes produced *in situ* such as Th and Pa, two methods have been used: 1) correlation of isotopes produced *in situ* with the main components of sinking marine particles collected by sediment traps, and 2) sorption experiments using natural or artificial seawater and particles. Sediment trap analyses have shown correlations between radioisotopes and inorganic phases, but fortuitous correlations between components have produced conflicting interpretations (Chase *et al.*, 2002; Luo and Ku, 2004; Roy-Barman *et al.*, 2005; Roy-Barman *et al.*, 2009). Some of these fortuitous correlations could be avoided by directly studying small filtered particles, because they dominate the solid surface area per volume and thus are more likely to adsorb tracers from seawater. This would require that the total mass and the major components of filtered particles be determined (Lam *et al.*, this issue). While focus has mainly been on the impact of major components on K_d (see references above), minor phases such as Mn oxides could play a significant role in the scavenging of Th (Roy-Barman *et al.*, 2009) and Pa (Anderson *et al.*, 1983a,b) in deep waters. Sorption experiments have shown a relatively low affinity of Th for inorganic phases and a high affinity for organic compounds (Santschi *et al.*, 2006). These results are consistent with ^{234}Th scavenging in shallow waters, but they fail to explain the correlations between ^{230}Th and inorganic phases (carbonate, lithogenic or Mn oxides, see previous paragraph) observed in sediment trap data.

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

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

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

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

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

5- Benthic and Intermediate Nepheloid Layers and their impacts on TEI distribution

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

5-1 Local re-suspension

The generation of a BNL and the distribution and size spectra of particles have been described by McCave (1984, 1986, 2001). Vertical mixing in bottom layers was studied during GEOSECS with ^{222}Rn (Sarmiento *et al.*, 1976). The vertical extent of BNLs is enhanced by the detachment of bottom mixed layers (Armi and D'Asaro, 1980). If surface sediments are in adsorption equilibrium with the bottom water, re-suspension need not change this equilibrium. However, there are cases in which interaction between re-suspension and bioturbation can change the distribution of dissolved components in the BNL relative to the water layer just above the BNL: 1) if the tracer decays within the bioturbated zone, or 2) if K_d changes as a result of diagenetic changes (e.g. MnO_2 enrichment) or particle dynamics like aggregation-disaggregation (Rutgers van der Loeff and Boudreau, 1997). There is no indication that the particle concentration has an effect on the K_d in the BNL (Honeyman *et al.*, 1988).
For short-lived radionuclides like ^{234}Th and ^{210}Pb, condition (1) above is clearly met. Profiles of dissolved ^{234}Th provide clear evidence for enhanced removal of dissolved TEIs from bottom waters in the presence of nepheloid layers (Bacon and Rutgers van der

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

5-2 Long-range transport in the BNL

Strong bottom currents occur along the western boundaries of the ocean basins (Warren, 1981), and deep wind and buoyancy-driven currents such as the Antarctic Circumpolar Current can reach abyssal depths (e.g. in the Drake Passage; Renault *et al.*, 2011). Through re-suspension or, rather, selective deposition, these currents can maintain high loads of suspended sediments. In the BNL, particles may be transported over large distances as shown for clay minerals (Griffin *et al*., 1968; Petschik *et al.*, 1996; Diekmann *et al.*, 2004). This means that particles are not only redistributed locally (winnowing and focusing) but also transported between areas with widely different local sediment compositions.

5-3 TEI fractionation

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

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

5-4 Intermediate Nepheloid Layers

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

6- "Historical" understanding of particle dynamics and perspectives

Despite its fundamental role in controlling the chemical composition of the ocean (Goldberg, 1954; Turekian, 1977) and the different cruises conducted in the 70s and 80s, the "oceanic microcosm of particles"-as christened by Lal (1977) – is far from being understood yet. In addition, sampling strategies and scientific focus differed between the GEOSECS and JGOFS programs. GEOSECS carried out large sections across the oceanic basins, where particles were collected by membrane filtration after bottle sampling, biasing the sampling towards the small particle pool. Analyses mostly informed us about the distribution of particle concentrations (mass/L), their major element compositions, as well as a few tracers and selected morphological and qualitative composition descriptions, thanks to the first Scanning Electron Microscopy (SEM) analyses. Subsequent box and one-dimensional (vertical) models described the different fluxes exchanged in and out the oceanic system as well as along the water column. These pioneering efforts led to the emergence of the fundamental notion of "reversible scavenging" (Brewer *et al.*, 1976; Krishnaswami *et al.*, 1976; Lal, 1980; Nozaki *et al.*, 1981; Bacon and Anderson, 1982; Anderson *et al.*, 1983a). They also highlighted the role of "particle-rich" continental margins on the distribution of ocean tracers (Anderson and Henderson, 2003; Jeandel *et al.*, 2011).

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

Conclusion

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

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

ii) What is the role of remineralization in the mesopelagic zone (Dehairs *et al.*, 1995; Dehairs *et al.*, 1997; Frew *et al.*, 2006; Boyd and Trull, 2007; Buesseler *et al.*, 2007b; Dehairs *et al.*, 2008)?

iii) What is the impact of sediment diagenesis on the composition of resuspended particles and on their ability to scavenge additional TEIs, despite having previously equilibrated with dissolved species in the water column (Kretschmer *et al.*, 2010; Kretschmer *et al.*, 2011)?

iv) What are the roles of the BNLs and INLs on the boundary scavenging and boundary exchange processes (Bacon *et al.*, 1988; Roy-Barman *et al.*, 2005; Roy-Barman *et al.*, 2009)?

v) What is the importance of other surface processes like chemoautotrophy as a source of particles in the deep ocean (Honjo *et al*., 2012)?

Answers to these questions can be provided by the GEOTRACES program with the implementation of a comprehensive sampling and analytical strategies (pumps, optics, observations and analysis of particles, see McDonnell *et al*., this issue; Boss *et al.*, this issue, Lam *et al*., this issue...), designed to elucidate the role of particles as agents of supply and removal of TEIs in the ocean. There is an urgent need for re-focusing on discrete particle composition, speciation and morphologies.

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Figure Captions

Figure 1

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

Figure 2

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

Figure 3

Longitudinal section of the dry weight of particulate matter along the western Atlantic Ocean from the GEOSECS program. *Reprinted from "Brewer, P. G., Spencer, D. W., Biscaye, P. E., Hanley, A., Sachs, P. L., Smith, C. L., Kadar, S., Fredericks, J., 1976. The distribution of particulate matter in the Atlantic Ocean. Earth and Planetary Science Letters 32, 393-402. Copyright (2014), with permission from Elsevier"*

Figure 4

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

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

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Figure 1

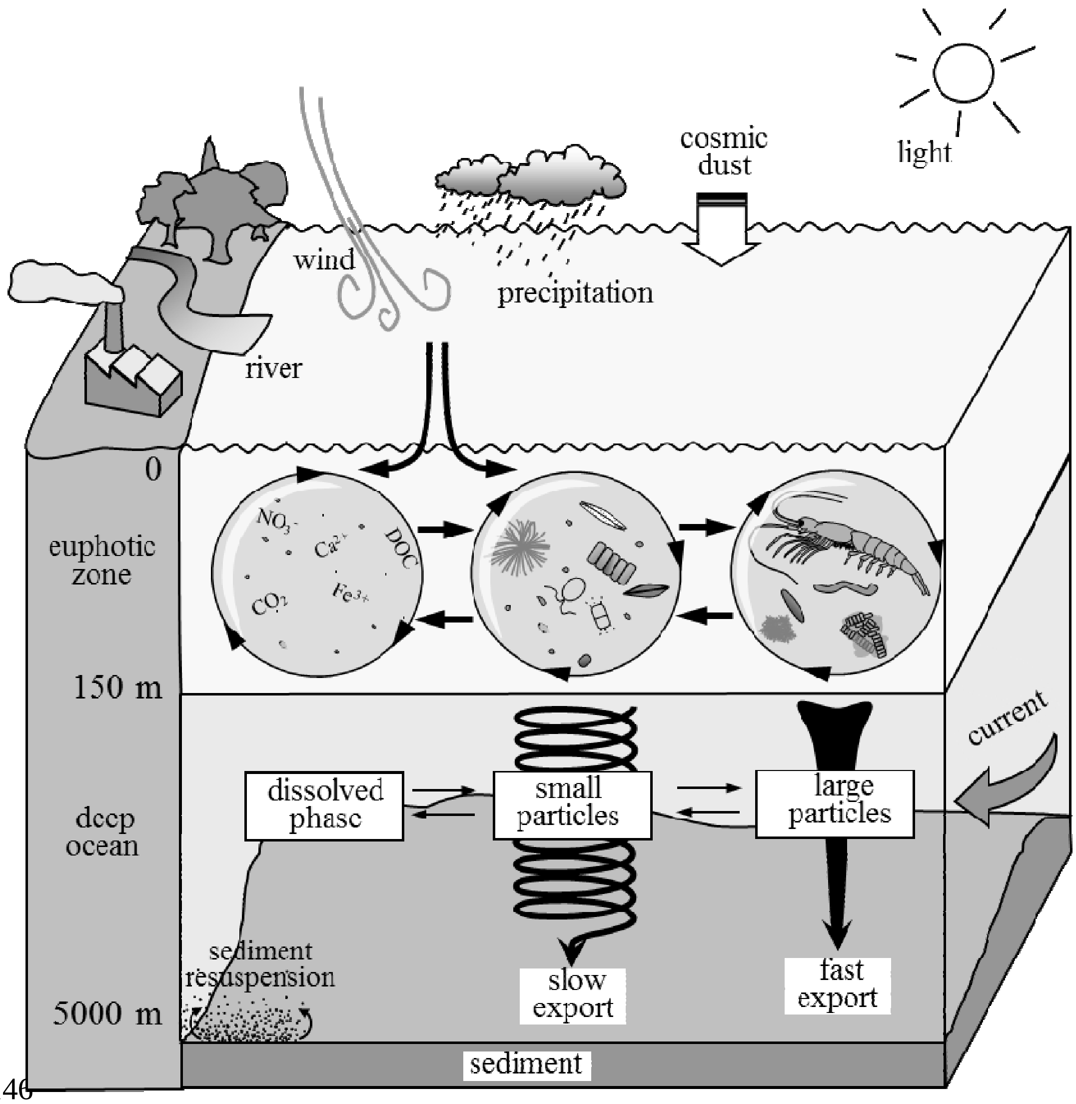
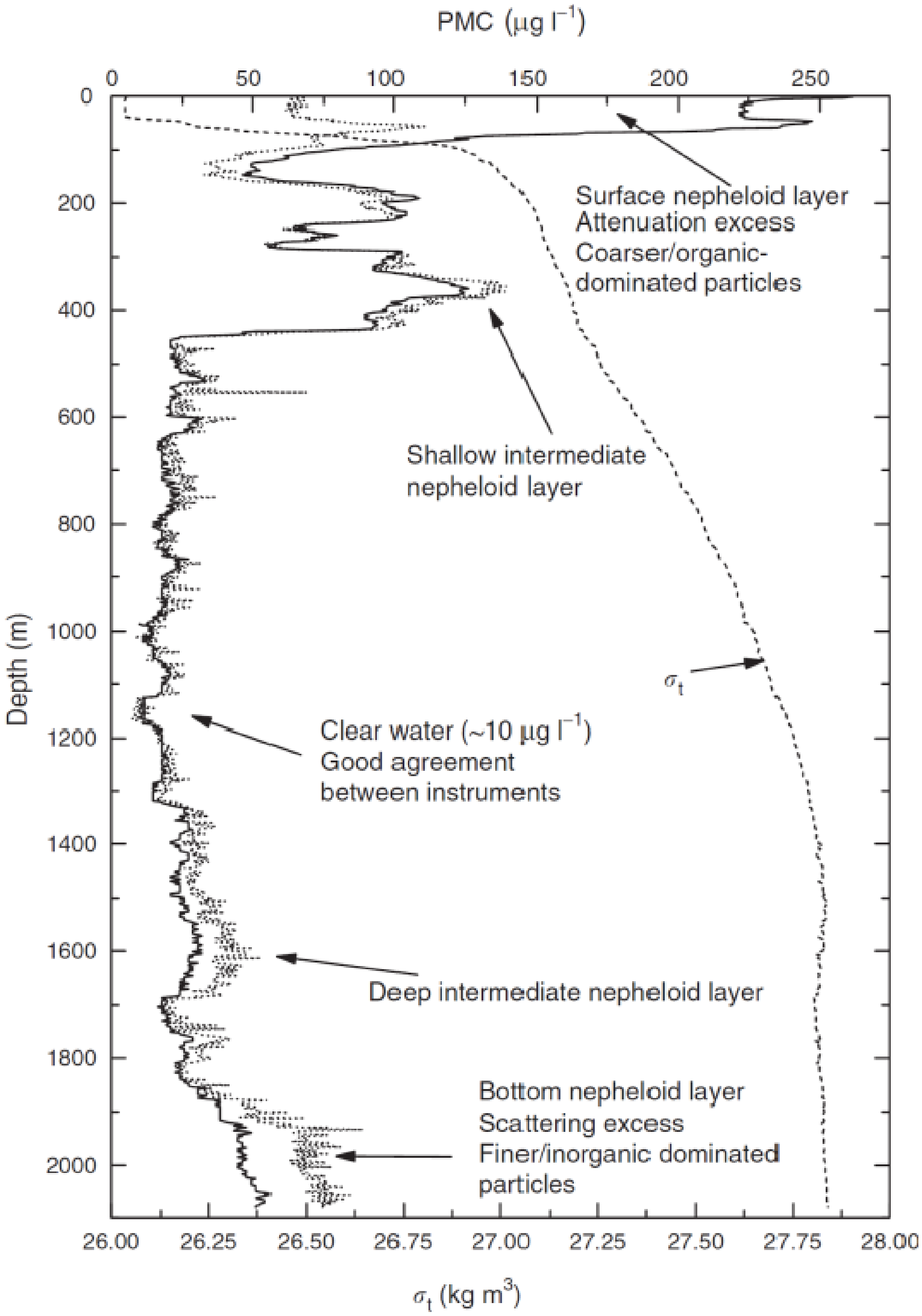


Figure 2



PMC (µg l⁻¹)
0
50
100
150
200
250
Depth (m)
0
200
400
600
800
1000
1200
1400
1600
1800
2000
Surface nepheloid layer
Attenuation excess
Coarser/organic-
dominated particles
Shallow intermediate
nepheloid layer
σt
Clear water (~10 µg l⁻¹)
Good agreement
between instruments
Deep intermediate nepheloid layer
Bottom nepheloid layer
Scattering excess
Finer/inorganic dominated
particles
26.00
26.25
26.50
26.75
27.00
27.25
27.50
27.75
28.00
σt (kg m³)

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Figure 3

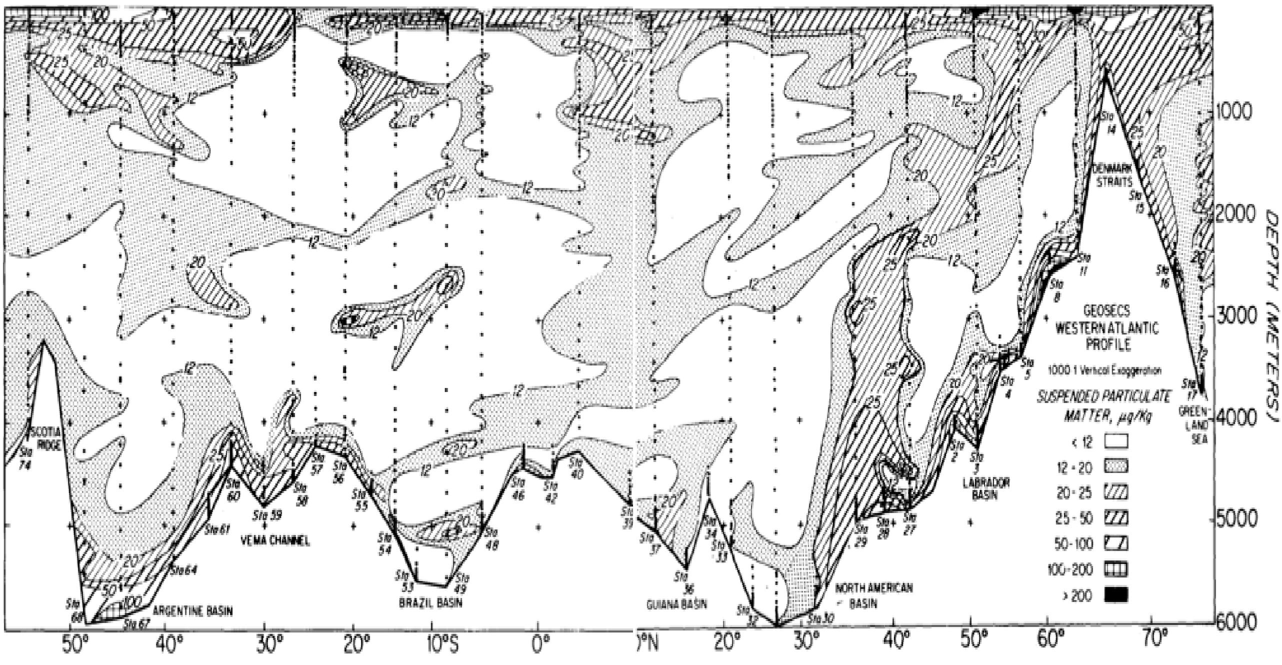
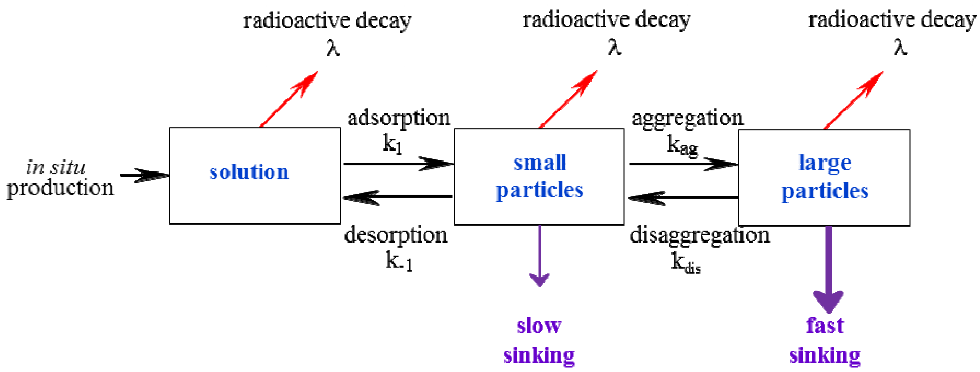


Fig. 2. Longitudinal section of the dry weight of particulate matter in the western Atlantic Ocean.

Figure 4


radioactive decay
λ
radioactive decay
λ
radioactive decay
λ
in situ
production
solution
adsorption
k_1
desorption
k_{-1}
small
particles
aggregation
k_{ag}
disaggregation
k_{dis}
large
particles
slow
sinking
fast
sinking