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1 **The influence of particle composition on Thorium scavenging**
2 **in the Mediterranean Sea**

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5 M. Roy-Barman (1)*, C. Lemaître (1,2), S. Ayrault (1), C. Jeandel (3), M. Souhaut (3), J.-C. Miquel
6 (4)

7
8 (1) LSCE/IPSL Laboratoire CNRS/CEA/UVSQ, 91198 Gif-sur-Yvette Cedex, France

9 (2) LATMOS (IPSL/ UVSQ / CNRS / Paris VI), 75252 Paris Cedex 05, France

10 (3) LEGOS (CNRS/CNES/IRD/UPS), Observatoire Midi-Pyrénées, 14, Av. E. Belin, 31400
11 Toulouse, France.

12 (4) IAEA Marine Environment Laboratories, 4 Quai Antoine 1^{er}, MC-98000, Monaco
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19 * corresponding author : Phone: (33)1-69-82-35-66; Fax: (33)1-69-82-35-68;

20 E-mail address: Matthieu.Roy-Barman@lsce.ipsl.fr

21 Abstract

22 Sediment trap data are crucial for the study of marine biochemical cycles but they need careful
23 validation by Thorium (Th) isotopes. In the present study, ^{230}Th , ^{232}Th , Uranium (U), Aluminum (Al),
24 Barium (Ba) and Manganese (Mn) were analyzed in sinking particles collected by moored sediment
25 traps in the Ligurian sea (DYFAMED site) in order to determine the collection efficiency of the traps
26 and to constrain which particulate phase(s) carries Th isotopes in a region with strong lithogenic
27 inputs. The $^{230}\text{Th}_{\text{xs}}$ evaluation was based on the U content of each sample rather than on the ^{232}Th
28 content of each sample and the average $^{238}\text{U}/^{232}\text{Th}$ ratio of the continental crust because the latter
29 method introduced too much uncertainty in the calculation. High trapping efficiencies (187 ± 85 % at
30 200 m and 87 ± 11 % at 1000 m) indicated no evidence of particle under-collection by the traps. The
31 lack of correlation between $^{230}\text{Th}_{\text{xs}}$ and the carbonate or the organic matter fraction suggests that these
32 phases are not major $^{230}\text{Th}_{\text{xs}}$ -carrying phases in the deep ocean. For most samples of the time series,
33 the $^{230}\text{Th}_{\text{xs}}$ concentration is correlated with the lithogenic fraction and the Mn concentration, but
34 pulses of particles with high lithogenic and Mn content and low $^{230}\text{Th}_{\text{xs}}$ concentration occur in winter.
35 Assuming that $^{230}\text{Th}_{\text{xs}}$ is only carried by lithogenic particles, we estimate $K_{\text{d_litho}}^{\text{Th}}$ ranges from $(0.42 \pm$
36 $0.04) \times 10^7$ ml/g to $(0.8 \pm 0.2) \times 10^7$ ml/g. Assuming that $^{230}\text{Th}_{\text{xs}}$ is only carried by authigenic Mn
37 oxide precipitates (MnO_2), we estimate that $K_{\text{d_MnO}_2}^{\text{Th}}$ ranges from $(0.6 \pm 0.1) \times 10^{10}$ ml/g to $(1.1 \pm$
38 $0.4) \times 10^{10}$ ml/g. The relative variation of $K_{\text{d_MnO}_2}^{\text{Th}}$ (a factor 7) between different oceanic sites is
39 significantly lower than the relative variation of $K_{\text{d_litho}}^{\text{Th}}$ (a factor 50), suggesting that $^{230}\text{Th}_{\text{xs}}$ has a
40 tighter link with MnO_2 rather than with lithogenic particles and hence that $^{230}\text{Th}_{\text{xs}}$ may be more likely
41 scavenged by MnO_2 than by lithogenic particles. The unambiguous determination of the particles
42 carrying $^{230}\text{Th}_{\text{xs}}$ remains to be done.

43

44
45 *Keywords: thorium, scavenging, sediment trap, trapping efficiency, marine particle, Mediterranean*
46 *Sea, DYFAMED*
47

48 **1. Introduction**

49 Understanding the oceanic carbon cycle requires reliable measurements of the particulate carbon
50 fluxes in the ocean. More generally, the quantification of particulate fluxes is a cornerstone of the
51 study of marine biogeochemical cycles. These fluxes are estimated with sediment traps. While
52 sediment trap data are very limited spatially and temporally for logistical and financial reasons, they
53 are critically important to tune the ocean biological pump in global biogeochemical models used to
54 evaluate the fate of the carbon cycle (Usbeck et al., 2003). However, turbulence around the trap
55 aperture can prevent a significant fraction of the sinking particles from being collected (Buesseler et
56 al., 2007). Therefore, it is critical to validate sediment trap data. Uranium (U) - Thorium (Th)
57 radioactive decay series are used to evaluate the efficiency of moored sediment traps (Bacon et al.,
58 1985). ^{230}Th is produced uniformly in the ocean by radioactive decay of ^{234}U . Thorium is a very
59 particle-reactive element, so that ^{230}Th is rapidly scavenged on sinking particles and transported
60 toward the bottom of the ocean (Bacon and Anderson, 1982). If a trap works correctly, it should
61 collect an average ^{230}Th sinking flux equal to the production of ^{230}Th by radioactive decay of ^{234}U
62 above the trap. As the ^{230}Th flux collected by a sediment trap varies seasonally with the particle flux,
63 annually averaged fluxes are considered in order to cancel seasonal variations of the ^{230}Th flux that
64 reflect short-term departures from the longer-term steady state between production and removal of
65 ^{230}Th (Bacon et al., 1985). Yearly averaged trapping efficiencies (the ratio between the trapped flux of
66 ^{230}Th and the radioactive production) as low as 10% have been obtained in surface waters and in the
67 mesopelagic zone (where horizontal currents are high) confirming that some traps largely undercollect
68 the particles (Guieu et al., 2005; Scholten et al., 2001). Very different evolutions of the particle flux
69 throughout the water column have been obtained when trapping efficiency corrections are used and
70 when they are ignored (Guieu et al., 2005; Scholten et al., 2001; Yu et al., 2001). Sediment traps may
71 also discriminate among different types of particles, with large and rapidly sinking particles expected

72 to be collected more efficiently into the traps than small and slowly sinking particles (Buesseler et al.,
73 1992). Therefore, a Th-based trapping efficiency may not be relevant for all particles and compounds
74 collected by the traps. Similar potential difficulties exist in paleo-oceanography with the
75 determination of sedimentary focussing/winning factors by the ^{230}Th -normalisation method
76 (Adkins et al., 2006; Francois et al., 2004). Thus, the determination of the phase(s) carrying Th
77 isotopes is a key issue.

78 One way to address this question is to consider the correlations of ^{230}Th with the major phases
79 (carbonate, biogenic silica, lithogenic particles, organic matter) constituting the particles compositions
80 (Chase and Anderson, 2004; Chase et al., 2002; Luo and Ku, 2004a; Luo and Ku, 2004b; Roy-Barman
81 et al., 2005; Scholten et al., 2005). However, this method suffers from multiple correlations between
82 the different components of the marine particles producing spurious correlations between ^{230}Th and
83 some phases unrelated with ^{230}Th scavenging, so that the result remains controversial. A recent work
84 has highlighted the possible role of minor phases such as Manganese (Mn) oxides on ^{230}Th scavenging
85 (Roy-Barman et al., 2005). However, it was based on a limited dataset that did not include samples
86 with very high concentration in lithogenic particles. Here, we present a set of sediment trap data
87 obtained at the DYFAMED site in the Ligurian Sea. This site receives sporadically a heavy load of
88 Saharan dust particles and the particle dynamics are particularly well documented (Migon et al.,
89 2002). Hence, this data set gives a good opportunity to test the Mn hypothesis in a lithogenic-rich
90 environment. It was also the opportunity to evaluate the collection efficiency of the traps at this site:
91 while the DYFAMED (DYnamique des Flux Atmosphériques en MEDiterranée) time series was
92 started in 1987, this evaluation has not been done yet.

93

94 **2. Sampling and analytical methods**

95 *2.1. Trap deployment*

96 The DYFAMED station (Fig. 1) is located in the western basin of the Mediterranean Sea, 30
97 nautical miles (54 km) off Nice (43°25 N - 07°54 E, water depth: 2330 m). Sinking particulate matter
98 was collected at 200 m and 1000 m from July 1999 to July 2000 as part of the DYFAMED sediment
99 trap time series. The traps were multisampling conical sediment-traps (Technicap PPS5) with a
100 collection surface of 1 m². Sampling cups were poisoned with formaldehyde prior to trap deployment
101 in order to prevent grazing in the traps. The sampling interval was of 7 days, but in order to reduce the
102 number of analyses, composite samples were prepared with sampling steps ranging from 7 to 35 days
103 (a higher temporal resolution was used during high flux periods).

104

105 *2.2. Analysis of particulate phases*

106 Sampling procedures followed the JGOFS protocols and can be found elsewhere (Miquel et
107 al., 1994). Back in the laboratory, swimmers were removed from the samples. The whole sample was
108 then rinsed with ultra pure (MilliQ) and freeze-dried. Concentrations of total (TC) and organic (POC)
109 carbon were measured in Monaco in triplicates with a Vario-El CHN elemental microanalyzer on
110 aliquots of the desiccated samples. Aliquots of approximately 20 mg of the desiccated samples were
111 digested in acid (HNO₃ + HF). Digestions were performed in a pressure-assisted microwave oven. For
112 all the acid-digested samples Barium (Ba) was analyzed by quadrupole ICP-MS (Perkin Elmer Elan
113 6000, LEGOS, Toulouse). Aluminum (Al), Mn and U concentrations were determined by quadrupole
114 ICP-MS (ThermoElectron XseriesII, LSCE, Gif sur Yvette).

115 Details on the material collected by the traps can be found at <http://www.obs-vlfr.fr/sodyf/>.
116 Trapped particles are mainly composed of marine snow, detritus, fecal pellets and lithogenic particles
117 in variable proportions. The fraction of aluminosilicate is given by $f_{\text{litho}} = (11.1 \pm 1.1) \times \text{Al}$ (in g/g). It

118 corresponds to an Al content (9 ± 1) % in the insoluble fraction of Saharan dusts of different origins
119 (Avila et al., 2007). The carbonate fraction was determined from Particulate Inorganic Carbon (PIC)
120 as follows: $f_{\text{CaCO}_3} = 8.3 \times \text{PIC}$ by assuming that all PIC is associated to CaCO_3 . The organic matter
121 content is assumed to be twice the POC content: $f_{\text{om}} = 2 \times \text{POC}$ (Klaas and Archer, 2002). Biogenic
122 silica is known for not being a major ^{230}Th scavenging phase in trapped material (Chase et al., 2002;
123 Roy-Barman et al., 2005). Therefore, it will not be considered in the following discussion. Excess Ba
124 (Ba_{ex}) was used to evaluate the abundance of biogenic Ba (Sternberg et al., 2007). It was obtained by
125 subtracting a Ba lithogenic fraction estimated with the Al content of the trapped material and a Ba/Al
126 ratio of $(6.7 \pm 1.0) \times 10^{-3}$ g/g that corresponds to the insoluble fraction of Saharan dusts (Avila et al.,
127 2007). Mn was measured to evaluate the abundance of Mn oxides. The authigenic Mn fraction
128 (Mn_{auth}) was obtained by subtracting a lithogenic fraction estimated with the Al content of the trapped
129 material and a Mn/Al ratio of $(8.5 \pm 0.5) \times 10^{-3}$ g/g that corresponds to the insoluble fraction of Saharan
130 dusts (Avila et al., 2007).

131

132 2.3. Analysis of ^{230}Th and ^{232}Th

133 ^{230}Th and ^{232}Th were analyzed on an aliquot of the same solution obtained for major and trace
134 elements (see above). ^{229}Th spike was added to this aliquot. After isotopic equilibration, the Th was
135 purified by ion exchange chemistry (Roy-Barman et al., 1996). Procedural blanks (around 20 pg of
136 ^{232}Th and 0.1 fg of ^{230}Th) represent typically less than 1-2 % of the Th mass in the samples. The
137 samples were analyzed in Toulouse by MC-ICP-MS on a Neptune (Finnigan) instrument following a
138 procedure described previously (Guieu et al., 2005; Roy-Barman et al., 2005). The agreement between
139 the measured $^{230}\text{Th}/^{232}\text{Th}$ ratio and the recommended value (Banner et al., 1990) of a Th standard
140 indicates that the accuracy of the MC-ICP-MS measurement is typically better than 2%.

141

142 3. Results

143 The ^{232}Th concentration ranges from 0.90 ± 0.01 ppm to 9.9 ± 0.2 ppm at 200 m and from 4.30
144 ± 0.02 ppm to 9.4 ± 0.1 ppm at 1000 m (Tab.1). The ^{230}Th concentration ranges from 4.9 ± 0.3 pg/g to
145 46.3 ± 0.7 pg/g at 200 m and from 28.8 ± 0.6 pg/g to 47.0 ± 0.6 pg/g at 1000 m (Tab.1).

146 The ^{230}Th produced by *in situ* decay of dissolved ^{234}U and that is scavenged on particles
147 ($^{230}\text{Th}_{\text{xs}}$) is often calculated by subtracting the lithogenic ^{230}Th component to the total ^{230}Th :

$$148 \quad ^{230}\text{Th}_{\text{xs}} = ^{230}\text{Th}_{\text{measured}} - ^{232}\text{Th}_{\text{measured}} \times (^{230}\text{Th}/^{232}\text{Th})_{\text{litho}} \quad (1)$$

149 In previous studies of Th isotopes in the Mediterranean sea (Arraes-Mescoff et al., 2001; Roy-Barman
150 et al., 2002), a ratio $(^{230}\text{Th}/^{232}\text{Th})_{\text{litho}} = 4.4 \times 10^{-6}$ mol/mol was used based on a mean composition of
151 the continental crust (Andersson et al., 1995). However, in the present study, we have analyzed
152 samples with $(^{230}\text{Th}/^{232}\text{Th})$ as low as $(4.0 \pm 0.1) \times 10^{-6}$ mol/mol at 200 m in winter (sample 7B). This
153 indicates that the value $(^{230}\text{Th}/^{232}\text{Th})_{\text{litho}} = 4.4 \times 10^{-6}$ mol/mol used in these earlier works is too high
154 because a negative $^{230}\text{Th}_{\text{xs}}$ obtained with equation 1 would have no physical meaning. The $^{230}\text{Th}/^{232}\text{Th}$
155 ratio of the lithogenic material ranges typically from 3.2×10^{-6} to 5.4×10^{-6} mol/mol (Anderson et al.,
156 1990). The $^{230}\text{Th}/^{232}\text{Th}$ ratio of aerosols collected in Monaco ranges from $(4.6 \pm 0.1) \times 10^{-6}$ to $(7.4 \pm$
157 $0.1) \times 10^{-6}$ mol/mol (Pham et al., 2005). Using these average values to correct for the lithogenic ^{230}Th
158 would also produce negative $^{230}\text{Th}_{\text{xs}}$ for numerous samples.

159 Alternatively, $^{230}\text{Th}_{\text{xs}}$ can be determined if the lithogenic U content ($^{238}\text{U}_{\text{litho}}$) of the sample is
160 known, assuming that in this lithogenic material ^{230}Th should be at secular equilibrium with ^{234}U and
161 ^{238}U (supported ^{230}Th):

$$162 \quad ^{230}\text{Th}_{\text{xs}} = ^{230}\text{Th}_{\text{measured}} - ^{238}\text{U}_{\text{litho}} \times (\lambda_{230\text{Th}} * m_{230\text{Th}}) / (\lambda_{238\text{U}} * m_{238\text{U}}) \quad (2)$$

163 where $\lambda_{230\text{Th}}$ ($= 9.24 \times 10^{-6} \text{ y}^{-1}$) and $\lambda_{238\text{U}}$ ($1.55 \times 10^{-10} \text{ y}^{-1}$) are the time constants for the radioactive
 164 decay of ^{230}Th and ^{238}U and $m_{230\text{Th}}$ and $m_{238\text{U}}$ their molar masses (this calculation neglects ^{235}U whose
 165 abundance is less than 1%). The hypothesis of the secular equilibrium between ^{230}Th and ^{238}U is
 166 necessary to use equation 1 and equation 2, although it is not always fulfilled in weathered lithogenic
 167 detritus (Pham et al., 2005). Further work is needed to precisely constrain the average $^{230}\text{Th}/^{238}\text{U}$ ratio
 168 of atmospheric aerosols particularly in areas dominated by these inputs.

169 The U content of the trapped sample ranges from 0.24 ± 0.25 ppm to 2.02 ± 0.06 ppm. The U
 170 content in crustal rocks ranges from 1 ppm (lower crust) to 2.8 ppm (upper crust) and hence
 171 atmospheric dust have variable U content (Taylor and McLennan, 1995). Organic matter is enriched
 172 in U at ppm level, whereas pelagic carbonates are depleted in U (< 0.1 ppm) (Anderson, 1982;
 173 Henderson and O'Nions, 1995). Therefore, a significant fraction of the U present in the sample could
 174 be authigenic U recently incorporated in organic matter. This authigenic U must not be taken into
 175 account to evaluate the supported ^{230}Th . Therefore, U_{litho} is bracketed with 2 extreme assumptions:

176 1) All the U in the trapped material is lithogenic and no correction of authigenic contribution has to be
 177 done ($U_{\text{auth}} = 0$):

$$178 \quad U_{\text{litho}} = U_{\text{measured}} \quad (3)$$

179 2) Some U is lithogenic and an authigenic U fraction is associated to organic matter (and to organic
 180 matter only). To estimate the authigenic U concentration in organic matter, we normalise the total U
 181 concentration and the lithogenic fraction by the fraction represented by the lithogenic and organic
 182 material ($f_{\text{litho}} + f_{\text{om}}$):

$$183 \quad U_{\text{norm}} = U / (f_{\text{litho}} + f_{\text{om}}) \quad (4)$$

$$184 \quad f_{\text{litho-norm}} = f_{\text{litho}} / (f_{\text{litho}} + f_{\text{om}}) \quad (5)$$

185 This normalisation removes the dilution effect of U-poor phases (carbonates). In a diagram U_{norm}
186 versus $f_{\text{litho-norm}}$, the U_{norm} value for $f_{\text{litho-norm}} = 0$ gives U_{om} , the authigenic U concentration in the
187 organic matter (Fig. 2). It appears that $U_{\text{om}} \approx 0.5\text{-}1.5$ ppm. To maximize the authigenic U correction,
188 we take $U_{\text{om}} = 1.5$ ppm. The authigenic U content of each sample is given by:

$$189 \quad U_{\text{auth}} = U_{\text{om}} \times f_{\text{om}} \quad (6)$$

190 With the 2 limiting cases, the mean U_{litho} and the range of uncertainty on the value are:

$$191 \quad U_{\text{litho}} = U_{\text{measured}} - U_{\text{auth}}/2 \pm (U_{\text{auth}}/2 + \text{error on } U_{\text{measured}}) \quad (7)$$

192 The U_{measured} value is the upper bound of this range. We note that the analytical uncertainty on U_{measured}
193 is small compared to the uncertainty introduced by the 2 limiting cases. In future studies, the analysis
194 of the $^{234}\text{U}/^{238}\text{U}$ ratio could contribute to further constrain the nature of U in marine particles and
195 sediments.

196 The $^{230}\text{Th}_{\text{xs}}$ is then given by equation 2. The $^{230}\text{Th}_{\text{xs}}$ concentration ranges from -0.6 ± 1.8 pg/g
197 to 19.1 ± 2.4 pg/g at 200 m and from 2.8 ± 2.1 pg/g to 21.3 ± 2.6 pg/g at 1000 m (Tab. 1). The highest
198 $^{230}\text{Th}_{\text{xs}}$ concentrations are generally found in the deepest traps in summer whereas the $^{230}\text{Th}_{\text{xs}}$
199 concentrations are low in winter. On average, $^{230}\text{Th}_{\text{xs}}$ represents 28 % of the total ^{230}Th at 200 m and
200 between 35% of the total ^{230}Th at 1000 m.

201

202 **4. Discussion**

203 *4.1. Particle dynamics at the DYFAMED site*

204 *4.1.1. Temporal evolution and control of the particle flux*

205 Despite its proximity to the continent, the DYFAMED site is considered as an “open ocean”
206 site because it is protected from direct inputs of coastal waters by the Ligurian current (Marty et al.,
207 2002). The Saharan dusts reaching DYFAMED have travelled at least 1000 or 2000 km since they

208 have left their desert sources. As a consequence, the lithogenic material found at the DYFAMED site
209 is characteristic of eolian material rather than of direct river inputs. Hence, the temporal variability of
210 the particle concentration and flux is due to local forcing rather than to advective inputs (Stemmann et
211 al., 2002). The vertical particule flux is influenced by the biological production (regulated by the
212 vertical inputs of nutrients), the atmospheric dust inputs of Saharan and European origin and winter
213 mixing (Miquel et al., 1994).

214
215 The fate of the atmospheric dusts deposited at DYFAMED deserves attention because it will
216 be used in the discussion in section 4.2.3. The deposition of Saharan dust over the Mediterranean Sea
217 occurs mostly in spring and summer although strong pulses may also occur in winter (Migon et al.,
218 2002). This general pattern can be compared with the total atmospheric deposition of Al recorded
219 from July 1999 to July 2000 at the Pirio site on the NW coast of Corsica (Loÿe-Pilot et al., 2001). This
220 site located 240 km SE of DYFAMED (Fig. 1) is free of local contamination and the atmospheric
221 fluxes recorded at Pirio are consistent with those collected on the French Riviera (Loÿe-Pilot et al.,
222 2001). Therefore, it represents a good recording of the Saharan dust deposition events that are
223 susceptible to have reached DYFAMED. At Pirio, strong Al deposition from July to October 1999
224 ($1.25\text{-}9.24\text{ mg/m}^2/\text{d}$) and from Mid-April to July 2000 ($1.20\text{-}3.67\text{ mg/m}^2/\text{d}$), whereas very low fluxes
225 are recorded from November 1999 to Mid-April 2000 ($0.03\text{-}0.16\text{ mg/m}^2/\text{d}$) (Fig. 3). This lack of
226 Saharan events during this period was also observed over the same period at the La Castanya
227 Biological Station (NE Spain)), where no red rain event was recorded from November 15th 1999 to
228 March 25th 2000 (Avila et al., 2007).

229 The vertical Al flux recorded at DYFAMED at 200 m are clearly not synchronized with the
230 atmospheric deposition described above (Fig. 3). From July 15th 1999 to December 5th 1999, the Al

231 flux trapped at 200 m is always lower than 1.5 mg/m²/d, whereas from December 9th 1999 to June 25th
232 2000, it ranges from 4.9 mg/m²/d to 16.9 mg/m²/d.

233 In previous studies, the vertical transfer of marine particles at the DYFAMED site was
234 described as a three step seasonal transfer scenario (Migon et al., 2002; Sarthou and Jeandel, 2001;
235 Sicre et al., 1999; Ternois et al., 1996) that accounts for the observations presented above:

236 (i) In summer and autumn, during the period of water stratification, biological fluxes are low and do
237 not allow the transfer of lithogenic material down to 200 m depth. Atmospheric material accumulates
238 along the seasonal thermocline, while below the thermocline a series of physico-chemical processes
239 lead to the formation of small non-biogenic aggregates. During that period, the residence time of the
240 suspended particles in the surface water can exceed 200 days (Schmidt et al., 2002).

241 (ii) In winter, the disappearance of the thermocline (and the sinking of dense water when it occurs) is
242 responsible for a rapid downward transfer of the lithogenic matter accumulated in the surface layer.

243 (iii) In spring, the availability of nutrients brought to surface waters by the winter vertical mixing
244 allows phytoplankton blooms. The sinking of particulate matter (including atmospheric dust) is then
245 governed by biological activity and can be very fast (Buat-Ménard et al., 1989; Fowler et al., 1987;
246 Schmidt et al., 1992).

247 Due to the lack of sedimentation between the end of the summer bloom and the winter mixing,
248 the lithogenic rich particles collected at 200 m or 1000 m in winter have spent several (3-5) months
249 above and at the seasonal thermocline. This is less than the residence time estimated for the suspended
250 particles between the surface and 200 m or 1000 m. At DYFAMED, the mean settling speed of the
251 small (> 0.2 µm) particle estimated by the ²³⁰Th_{xs} method is $S = 250$ m/y (Roy-Barman et al., 2002).
252 This settling speed of small particles is in the lower end of the range determined in the ocean. In the
253 oligotrophic Central North Pacific, the small particle settling speed is of 330 m/y (Roy-Barman et al.,

254 1996) whereas in coastal areas the settling speed is of the order of 1000 m/y (Huh and Beasley, 1987).
255 The estimate of the settling speed at DYFAMED (Roy-Barman et al., 2002) may not be very accurate
256 because it is based on a budget that depends more on the ventilation rate of the deep waters than on
257 the thorium scavenging rate. Therefore, we suspect that this value underestimates the true settling
258 speed. Hence, the mean settling time of the suspended particles is expected to be less than 1 year
259 between 0 and 200 m and of the order of 3 years between 200 m and 1000 m.

260 The particles collected at 200 m or 1000 m are not simply surface-derived particles. Their
261 chemical composition is affected by the mineralization of organic compounds (Sempéré et al., 2000),
262 by the dissolution of inorganic elements (Arraes-Mescoff et al., 2001) and aggregation of suspended
263 particles (Roy-Barman et al., 2002, Coppola et al., 2006). Aggregation is best visible by the increase
264 of the $^{230}\text{Th}_{\text{xs}}$ flux between 200 m ($0.24 \pm 0.11 \text{ pg/m}^2/\text{y}$) and 1000 m ($0.56 \pm 0.07 \text{ pg/m}^2/\text{y}$). It is likely
265 that this increase of $^{230}\text{Th}_{\text{xs}}$ flux between 200 m and 1000 m is due to aggregation of suspended
266 particles that are enriched in *in situ* produced ^{230}Th due to their high specific surface and long
267 residence time in the water column rather than to direct adsorption of *in situ* produced ^{230}Th on
268 trapped particles (Roy-Barman et al., 2002).

269

270 4.1.2. Sediment trap efficiency

271 It is worth evaluating the collection efficiency of the trap because a low efficiency can modify
272 the chemical composition of the trapped material by, as an example, an under-sampling of the finest
273 particles (Roy-Barman et al., 2005). To cancel the short-term seasonal variations of the $^{230}\text{Th}_{\text{xs}}$ flux
274 induced by the seasonal variations of the particle flux, the annually averaged $^{230}\text{Th}_{\text{xs}}$ flux is considered
275 (Bacon et al., 1985). Assuming that $^{230}\text{Th}_{\text{xs}}$ is transported vertically by the settling particles only, the
276 collection efficiency (E) is just the ratio between the flux of $^{230}\text{Th}_{\text{xs}}$ collected by the trap during a full

277 year (F) and the quantity of $^{230}\text{Th}_{\text{xs}}$ produced over the trap by radioactive decay of ^{234}U (Guieu et al.,
278 2005):

$$279 \quad E = \frac{F}{P \times h} \quad (8)$$

280
281 where F is the year-averaged $^{230}\text{Th}_{\text{xs}}$ flux collected by the trap, P is the ^{230}Th production rate (P = 0.65
282 fg/l/y in the salty Mediterranean sea) and h is the trap depth. Under these assumptions, we obtain E =
283 $187 \pm 85 \%$ at 200 m and $E = 87 \pm 11 \%$ at 1000 m.

284 Overall, the trapping efficiency is close to 100 % considering the uncertainties and there is no
285 evidence of strong under-trapping. This high trapping efficiency can be related to the generally low
286 speed of the currents at 200 m and 1000 m at DYFAMED (Miquel and La Rosa, 2001; Roy-Barman et
287 al., 2002). This is the first published determination of the PPS5 trap efficiency at DYFAMED and it
288 validates the numerous sediment trap based studies at DYFAMED.

289

290 4.2. Testing the potential carriers of $^{230}\text{Th}_{\text{xs}}$

291 In this section, we look at the correlations between the concentration of $^{230}\text{Th}_{\text{xs}}$ and of different
292 phases constituting the particulate material. With this approach of comparing concentrations rather
293 than vertical fluxes, we try to determine the phases on which $^{230}\text{Th}_{\text{xs}}$ is adsorbed but not necessarily
294 the ones that create the vertical flux through aggregation and ballasting. The parameters controlling
295 the partition coefficient $K_{\text{d_bulk}}^{\text{Th}}$ between the bulk particulate and the dissolved phases are
296 controversial. Field-based estimate of $K_{\text{d_bulk}}^{\text{Th}}$ is obtained by dividing the amount of nuclides per g of
297 bulk trapped particles by the total amount of nuclides (dissolved+particulate) per ml of seawater in the
298 water through which the particle sunk.

$$299 \quad K_{\text{d_bulk}}^{\text{Th}} = \frac{[^{230}\text{Th}_{\text{xs}}]_{\text{particulate matter}}}{[^{230}\text{Th}_{\text{xs}}]_{\text{seawater}}} \quad (9)$$

300 And for the fractionation between a given phase of the particulate matter and seawater:

301
$$K_{d_phase}^{Th} = K_{d_bulk}^{Th} / [phase]_{particulate\ matter} \quad (10)$$

302 In the southern and Pacific oceans, the correlation between $K_{d_bulk}^{Th}$ and the carbonate content
303 of trapped particles suggests that ^{230}Th scavenging is controlled by carbonates (Chase and Anderson,
304 2004; Chase et al., 2002; Chase et al., 2003). In regions with higher lithogenic content in the trapped
305 material, both carbonate and lithogenic material should scavenge ^{230}Th (Chase et al., 2002; Narita et
306 al., 2003). Revisiting the same data set, Luo and Ku (2004) noted a strong correlation between
307 $K_{d_bulk}^{Th}$ and the lithogenic content of the trapped particles and proposed that ^{230}Th is scavenged
308 mainly by the lithogenic phase (Luo and Ku, 2004a; Luo and Ku, 2004b). However, the $K_{d_litho}^{Th}$
309 (2.3×10^8 ml/g) obtained at these sites is more than one order of magnitude larger than those obtained
310 in regions with strong lithogenic inputs (1×10^7 ml/g) (Chase et al., 2002). In the Arctic Ocean where
311 biological productivity is very low, ice-rafted lithogenic particles are proposed to scavenge ^{230}Th
312 (Edmonds et al., 1998; Trimble et al., 2004). In the Arabian Sea, the lack of consistency between
313 $K_{d_bulk}^{Th}$ and the major components of the sinking particles suggested that Th scavenging is not
314 controlled by major phases (Scholten et al., 2005). More recently, it was proposed that Mn oxides
315 rather than lithogenic particles or carbonates control ^{230}Th scavenging on marine particles (Roy-
316 Barman et al., 2005). This was based on (1) the strong correlation between Mn and ^{230}Th content of
317 marine particles collected in the NE Atlantic Ocean and (2) the consistency of the $K_{d_MnO2}^{Th}$ estimates
318 over a large range of oceanic environment. However, only a limited number of samples from area with
319 strong lithogenic inputs were available in this study and we now re-explore the possible relationships
320 between $^{230}Th_{xs}$ and the different components of the DYFAMED trapped particles.

321

322 4.2.1. *CaCO₃*

323 The lack of correlation between CaCO_3 and $^{230}\text{Th}_{\text{xs}}$ (Fig. 4a) is consistent with the results
324 obtained in the NE Atlantic and in the Arabian Sea (Roy-Barman et al., 2005; Scholten et al., 2005). It
325 confirms that the linear correlation observed between CaCO_3 and $^{230}\text{Th}_{\text{xs}}$ in the Southern Ocean and in
326 the Equatorial Pacific (Chase et al., 2002) is not a general feature. This correlation may be due to the
327 fortuitous correlation of CaCO_3 and $^{230}\text{Th}_{\text{xs}}$ bearing phases, possibly MnO_2 or lithogenic particles
328 (Roy-Barman et al., 2005).

329

330 4.2.2. *Particulate Organic Matter*

331 The lack of correlation between Particulate Organic matter (estimated through POC
332 concentration) and $^{230}\text{Th}_{\text{xs}}$ (Fig. 4b) is consistent with the results obtained in the NE Atlantic, in the
333 Southern Ocean, in the Equatorial Pacific and in the Arabian Sea (Chase et al., 2002; Roy-Barman et
334 al., 2005; Scholten et al., 2005). It suggests that in the deep ocean ^{230}Th is not scavenged significantly
335 by organic matter (Roy-Barman et al., 2005) contrary to what is probably the case in surface waters
336 (Coppola et al., 2002, Quigley et al., 2002).

337

338 4.2.3. *Lithogenic particles*

339 There is not a single correlation between $^{230}\text{Th}_{\text{xs}}$ and ^{232}Th (Fig. 4c). This is not surprising
340 because $^{230}\text{Th}_{\text{xs}}$ is scavenged from seawater, whereas ^{232}Th is mostly introduced in the ocean
341 imbedded in lithogenic particles (Adkins et al., 2006; Roy-Barman et al., 2002). This is reflected by
342 the high correlation between the ^{232}Th concentration and the Al concentration in the trapped material
343 (not shown, $R^2 = 0.98$). Most samples define a gross linear relationship between $^{230}\text{Th}_{\text{xs}}$ and ^{232}Th (200

344 m: $R^2 = 0.87$ and 1000 m: $R^2 = 0.75$). Lithogenic-rich samples collected during winter (and early
345 spring) do not fall on the main trend (circles with a cross inside in Fig. 4).

346 A double correlation between $^{230}\text{Th}_{\text{xs}}$ and ^{232}Th was also obtained during the POMME
347 experiment in the North Atlantic (Roy-Barman et al., 2005). The main correlation was explained by a
348 mixture between small suspensions (enriched in ^{230}Th) and lithogenic particles, whereas winter
349 samples with high ^{232}Th content and low $^{230}\text{Th}_{\text{xs}}$ were due to a rapid transfer at depth of atmospheric
350 dust deposited at the sea surface (within 2 weeks at 400 m and the week after at 1000 m). This rapid
351 transfer may have been due to incorporation of the dusts in faecal pellets or in marine snow.
352 Therefore, it was not possible to determine if these samples were not enriched in $^{230}\text{Th}_{\text{xs}}$ because the
353 $K_{\text{d_litho}}^{\text{Th}}$ is low or because $^{230}\text{Th}_{\text{xs}}$ had not enough time to equilibrate with the newly arrived lithogenic
354 particles and seawater.

355 The DYFAMED site offers the opportunity to tackle this question. As explained in section
356 4.1.1., the lithogenic particles deposited in summer and autumn are not transferred directly through
357 the thermocline but rather stored at the top of the thermocline (during 3-5 months) until winter
358 convection allow them to cross the thermocline. Then, it takes 2 additional months for the lithogenic
359 particles to go from 200 m to 1000 m as indicated by the time lag of the Al flux between 200 m and
360 1000 m (Fig. 3). Despite this longer equilibration time between the lithogenic particles and seawater
361 compared to the POMME data, the lithogenic rich samples plot below the main $^{230}\text{Th}_{\text{xs}}$ – lithogenic
362 fraction correlation at both 200 m and 1000 m. The transit time of these particles is not as long as the
363 average residence time of the bulk suspended particles in the water column (see section 4.1.1.). So it is
364 possible that there was not enough time for $^{230}\text{Th}_{\text{xs}}$ to reach complete equilibration between the winter
365 lithogenic particles and seawater. However, in the North Atlantic, the particles transporting a Saharan
366 dust pulse at 1000 m within 3 weeks have a larger $K_{\text{d_litho}}^{\text{Th}}$ than particles transporting a Saharan dust
367 pulse in several months at 1000 m at DYFAMED. It suggests that the kinetic of ^{230}Th adsorption on

368 lithogenic particles is not the limiting factor of the ^{230}Th uptake by sinking particles. This is also
369 consistent with in vitro experiments indicating a fast sorption of ^{230}Th on lithogenic particles (Geibert
370 and Usbeck, 2004). Nevertheless, deposition of larger dust particles at DYFAMED compared to the
371 North Atlantic could explain the lower affinity of ^{230}Th for DYFAMED particles due to their lower
372 surface to volume ratio. Alternatively, it was proposed that the $^{230}\text{Th}_{\text{xs}}$ content of trapped particles is
373 controlled by the incorporation of $^{230}\text{Th}_{\text{xs}}$ -rich, Ba-rich and Mn-rich suspended particles to the rapidly
374 sinking particles (Huang and Conte, 2009; Roy-Barman et al., 2005). This mixing would then control
375 the apparent $K_{\text{d_litho}}^{\text{Th}}$ of the trapped particles. The low Ba content of lithogenic-rich winter samples
376 indicate a low contribution of the small suspended particle pool (Barite tend to accumulate in this
377 small particle pool (Sternberg et al., 2007)) despite high lithogenic content. In any case, the
378 DYFAMED results confirm that arrival of atmospheric pulses of Saharan material decreases the
379 degree of correlation between the $^{230}\text{Th}_{\text{xs}}$ and f_{litho} for the whole dataset.

380 Assuming that all the $^{230}\text{Th}_{\text{xs}}$ is adsorbed on the lithogenic material and that there is a chemical
381 equilibration of $^{230}\text{Th}_{\text{xs}}$ between lithogenic particles and seawater, we can estimate $K_{\text{d_litho}}^{\text{Th}}$ with the
382 trapped particles analyzed in the present work and seawater data (Roy-Barman et al., 2002). We focus
383 on the 1000 m samples, because their $^{230}\text{Th}_{\text{xs}}$ is less sensitive to uncertainties on lithogenic correction
384 and because there is some concern that at shallower depth the equilibration between seawater and
385 particulate matter is not achieved (Chase et al., 2002). We obtain $K_{\text{d_litho}}^{\text{Th}} = (0.7 \pm 0.2) \times 10^7$ for the
386 main trend samples and $K_{\text{d_litho}}^{\text{Th}} = (0.36 \pm 0.04) \times 10^7$ for the winter samples (Tab. 2). These values are
387 consistent with the result of $K_{\text{d_litho}}^{\text{Th}} \approx 10^7$ obtained by (Chase et al., 2002) based on Atlantic samples
388 and much lower than the value $K_{\text{d_litho}}^{\text{Th}} \approx 23 \times 10^7$ estimated by (Luo and Ku, 2004a) based on sample
389 collected in area experiencing very low eolian inputs.

390
391 4.2.4. *Mn oxides*
392 The relationship between $^{230}\text{Th}_{\text{xs}}$ and Mn is quite similar to the relationship between $^{230}\text{Th}_{\text{xs}}$
393 and the lithogenic fraction (Fig. 4c,e): excluding the winter samples, good correlations are obtained
394 between $^{230}\text{Th}_{\text{xs}}$ and Mn (200 m: $R^2 = 0.83$ and 1000 m: $R^2 = 0.86$). In the NE Atlantic, the lithogenic
395 content of the trapped particle was low enough (<10%) so that the lithogenic contribution to the total
396 Mn was generally less than 50 % and a good correlation was found between $^{230}\text{Th}_{\text{xs}}$ and Mn (Roy-
397 Barman et al., 2005). The winter samples are still not on the main trend on a $^{230}\text{Th}_{\text{xs}}$ and Mn_{auth}
398 diagram (fig 4f) and excluding the winter samples, the goodness of fit of the $^{230}\text{Th}_{\text{xs}}$ versus Mn_{auth} data
399 (200 m : $R^2 = 0.56$ and 1000 m: $R^2 = 0.69$) is not as good as the $^{230}\text{Th}_{\text{xs}}$ and Mn correlation. However,
400 correcting the DYFAMED samples for the lithogenic Mn contribution requires caution because the
401 Mn/Al ratio of the insoluble fraction of Saharan dusts $(8.5 \pm 0.5) \times 10^{-3}$ g/g is variable (Avila et al.,
402 2007). In addition, the Mn/Al ratio of the “Saharan aerosols” collected in “clean” sites of Corsica or of
403 the French Riviera ($\text{Mn/Al} = 16 \times 10^{-3} - 38 \times 10^{-3}$) can be 3-4 times higher indicating the contribution of
404 an anthropogenic Mn source (possibly European aerosols) (Ridame et al., 1999). Finally, Mn
405 dissolution can occur when it enters seawater (Guieu et al., 1994) or during particulate organic matter
406 mineralization (Arraes-Mescoff et al., 2001).

407 The Mn/Al ratio of the DYFAMED samples range from 5×10^{-3} to 16×10^{-3} (average: 12.0×10^{-3})
408 at 200 m and from 11×10^{-3} to 18×10^{-3} (average: 13.4×10^{-3}) at 1000 m. These values are within the
409 range of the crustal rock average and at the low end of the atmospheric inputs range so that it is
410 difficult to correct for the “non authigenic” Mn fraction. The maximum of anthropogenic inputs
411 occurs at the end of summer (before winter mixing) and the end of winter (during winter mixing)
412 (Migon et al., 2008). Therefore, we cannot rule out that the Mn-rich and $^{230}\text{Th}_{\text{xs}}$ -poor winter samples

413 fall out of the main correlation because the Mn/Al ratio of their dusts is larger than the value used to
414 calculate Mn_{auth} .

415 In surface waters, photoreduction limits the formation of Mn oxides, so that particulate Mn is
416 associated with organic matter (Johnson et al., 1996). Below the photic zone, Mn oxides are
417 precipitated on marine particles throughout the water column (Huang and Conte, 2009; Johnson et al.,
418 1996). Assuming that all the $^{230}Th_{xs}$ is adsorbed on these authigenic Mn oxides and that there is a
419 chemical equilibration of $^{230}Th_{xs}$ between these oxides and seawater, we can estimate $K_{d_MnO_2}^{Th}$. Still
420 focussing on the 1000 m samples, we obtain $K_{d_MnO_2}^{Th} = (1.1 \pm 0.4) \times 10^{10}$ for the main trend samples
421 and $K_{d_MnO_2}^{Th} = (0.6 \pm 0.1) \times 10^{10}$ for the winter samples (Tab. 2). These values are lower but grossly
422 consistent with the result of $K_{d_MnO_2}^{Th} \approx 2 \times 10^{10}$ obtained for the Atlantic Ocean (Roy-Barman et al.,
423 2005) and $K_{d_MnO_2}^{Th} \approx 3.5 \times 10^{10}$ estimated from sample collected in area experiencing very low eolian
424 inputs (Luo and Ku, 1999; Roy-Barman et al., 2002).

425 In the previous section and in the present one, we successively assumed that all the $^{230}Th_{xs}$ is
426 carried by the lithogenic particles or by the authigenic Mn oxides. We now try to choose between
427 these 2 hypotheses. It is well established that the slope of the correlation between the $^{230}Th_{xs}$
428 concentration and the lithogenic fraction of trapped particles is site-dependent (Chase and Anderson,
429 2004; Chase et al., 2002; Luo and Ku, 2004a; Luo and Ku, 2004b; Roy-Barman et al., 2005). Much of
430 the controversy on affinity of $^{230}Th_{xs}$ for lithogenic particles resides in the interpretation of this site-
431 dependent slope that may reflect (1) true variations of $K_{d_litho}^{Th}$ from one site to the other depending on
432 the chemical and size characteristics of the lithogenic particles (Luo and Ku, 2004a; Luo and Ku,
433 2004b) or (2) more or less fortuitous correlation between the lithogenic fraction and another phase by
434 which the $^{230}Th_{xs}$ is effectively taken up such as carbonates (Chase and Anderson, 2004; Chase et al.,
435 2002) or Mn oxides (Roy-Barman et al., 2005). The present study does not allow choosing clearly

436 between the 2 hypotheses because the Mn concentration and the lithogenic fraction are strongly
437 correlated ($R^2=0.92$). However, we note that if neither $K_{d_litho}^{Th}$ nor $K_{d_MnO_2}^{Th}$ are constant between
438 high dust and low dust sites, $K_{d_MnO_2}^{Th}$ estimated from sediment trap data varies by at most a factor 7,
439 whereas $K_{d_litho}^{Th}$ varies by a factor 50 (Tab. 2). Thus the affinity of $^{230}Th_{xs}$ for Mn oxides appears to
440 be much less “site-dependent” than its affinity for lithogenic particles, suggesting a stronger link
441 between $^{230}Th_{xs}$ scavenging and authigenic MnO_2 precipitation than with lithogenic particles. In that
442 case lithogenic matter may serve as support for Mn oxide precipitation and then as ballast for its
443 transport.

444 These results combined with the lack of correlation found between the $^{230}Th_{xs}$ concentration
445 and the $CaCO_3$ content of trapped particles at DYFAMED as well as in previous studies (Roy-Barman
446 et al., 2005; Scholten et al., 2005), suggests that the high $K_{d_CaCO_3}^{Th}$ used in various modelling studies
447 (Dutay et al., 2009; Siddall et al., 2005) should be re-evaluated and that Mn coatings have to be taken
448 into account in these models. The correlation between $^{230}Th_{xs}$ and carbonates at some sites could be
449 explained if carbonates are substrates for Mn oxide precipitation (Martin and Knauer, 1983).

450
451
452

453 **5. Conclusion**

454 The present study brings a new insight on thorium scavenging in regions with strong lithogenic
455 inputs. The lack of correlation between $^{230}Th_{xs}$ and the carbonate and organic matter fractions suggests
456 that carbonates and organic matter are not the main phases carrying the in situ ^{230}Th in the
457 Mediterranean Sea. The high temporal resolution of DYFAMED sediment trap time series indicates
458 that different correlations exist between $^{230}Th_{xs}$ concentration and the lithogenic fraction of the
459 trapped particles. Most samples define a correlation between $^{230}Th_{xs}$ and the lithogenic fraction but
460 lithogenic-rich and $^{230}Th_{xs}$ – poor particle pulses occur in winter. Assuming that $^{230}Th_{xs}$ is only carried

461 by lithogenic particles, the estimated value of $K_{d_litho}^{Th}$ range from $(0.42 \pm 0.04) \times 10^7$ ml/g to $(0.8 \pm$
462 $0.2) \times 10^7$ ml/g. Similarly, a correlation between the $^{230}Th_{xs}$ concentration and the Mn concentrations
463 occurs with also Mn-rich and $^{230}Th_{xs}$ – poor particle pulses in winter. Assuming that $^{230}Th_{xs}$ is only
464 carried by authigenic MnO_2 precipitates, the estimated value of $K_{d_MnO_2}^{Th}$ ranges from $(0.6 \pm 0.1) \times$
465 10^{10} ml/g to $(1.1 \pm 0.4) \times 10^{10}$ ml/g. Comparing K_d^{Th} obtained at different sites indicates that the range
466 of variation of $K_{d_MnO_2}^{Th}$ is significantly lower than the range of variation $K_{d_litho}^{Th}$, suggesting that
467 $^{230}Th_{xs}$ has a tighter link with MnO_2 rather than with lithogenic particles and that hence MnO_2 is a
468 more likely carrier of $^{230}Th_{xs}$ than lithogenic particles. The unambiguous determination of the particles
469 carrying thorium isotopes remains to be done.

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488 **Figure Captions**

489

490 Figure 1: The DYFAMED station, in the western Mediterranean (from Ocean Data View)

491

492 Figure 2: U_{norm} versus $f_{\text{litho_norm}}$. See text for definitions. The straight lines represent mixing lines

493 between hypothetical (lithogenic free) organic matter end members and lithogenic end members. It

494 suggests that the organic matter end-member contains at most 1.5 ppm of authigenic U. Crustal end-

495 member from (Taylor and McLennan, 1995).

496

497 Figure 3: Temporal evolution of the trapped material at 200 m and 1000 m and atmospheric deposit at

498 Pario in Corsica. A: Mass flux. B: Al flux.

499

500 Figure 4: $^{230}\text{Th}_{\text{xs}}$ as a function of the particle composition. a: $^{230}\text{Th}_{\text{xs}}$ versus CaCO_3 . b: $^{230}\text{Th}_{\text{xs}}$ versus

501 organic matter. c: $^{230}\text{Th}_{\text{xs}}$ versus lithogenic matter d: $^{230}\text{Th}_{\text{xs}}$ versus Ba_{xs} . e: $^{230}\text{Th}_{\text{xs}}$ versus Mn_{total} . f:

502 $^{230}\text{Th}_{\text{xs}}$ versus Mn_{auth} . Large error bars on the lithogenic fraction, Ba_{xs} and Mn_{auth} are due to the

503 variability of the composition of the insoluble fraction of Saharan aerosols (Avila et al., 2007).

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Table 1: sediment trap data

sample	Sampling dates	mass flux (mg/m ² /d)	²³² Th (ppm)	²³⁰ Th (ppt)	²³⁰ Th/ ²³² Th (mol/mol)	U (ppm)	U _{lith} (ppm)	²³⁰ Th _{xs} (ppt)	Al (w%)	Mn (ppm)	Mn _{auth} (ppm)	Ba (ppm)	Ba _{ex} (ppm)	POC (w%)	CaCO ₃ (w%)
200 m															
1B	15.07.99-08.08.99	5.8	1.87 ± 0.01	11.6 ± 0.8	6.2 ± 0.4	0.49	0.24 ± 0.25	7.6 ± 5.0	1.18	157	58 ± 12	347	268 ± 20	14	47
2B	09.08.99-05.09.99	25.8	7.95 ± 0.05	46.3 ± 0.7	5.8 ± 0.1	1.76	1.66 ± 0.12	19.1 ± 2.6	5.58	787	315 ± 58	673	300 ± 76	13	14
3B	06.09.99-03.10.99	4.0	4.66 ± 0.02	24.7 ± 0.9	5.3 ± 0.2	1.10	0.80 ± 0.31	11.7 ± 6.0	3.45	287	-5 ± 32	454	223 ± 48	9	40
4B	04.10.99-31.10.99	16.5	5.61 ± 0.02	30 ± 1.4	5.4 ± 0.2	1.28	1.08 ± 0.22	12.6 ± 5.0	3.91	332	1 ± 36	617	356 ± 56	11	27
5B	01.11.99-05.12.99	9.0	7.10 ± 0.02	33.2 ± 0.9	4.7 ± 0.1	1.38	1.18 ± 0.21	13.8 ± 4.4	4.66	512	117 ± 46	625	313 ± 65	16	27
6B	13.12.99-09.01.00	100	8.07 ± 0.03	34.2 ± 0.6	4.2 ± 0.1	1.65	1.55 ± 0.14	8.8 ± 3.0	5.92	852	351 ± 62	548	153 ± 77	11	13
7B	10.01.00-30.01.00	97	9.85 ± 0.07	39.7 ± 0.5	4.0 ± 0.1	1.86	1.75 ± 0.12	11.0 ± 2.5	6.99	850	258 ± 70	506	39 ± 89	14	15
8B	31.01.00-20.02.00	223	9.89 ± 0.07	40 ± 1.2	4.1 ± 0.1	2.05	1.98 ± 0.08	7.7 ± 2.5	6.89	872	288 ± 70	333	-127 ± 84	12	9
9B	21.02.00-12.03.00	222	9.34 ± 0.06	39.3 ± 0.5	4.2 ± 0.1	1.98	1.89 ± 0.12	8.4 ± 2.5	6.45	779	233 ± 65	462	31 ± 82	13	12
10B	13.03.00-26.03.00	315	6.38 ± 0.03	27.8 ± 0.6	4.4 ± 0.1	1.35	1.30 ± 0.09	6.5 ± 2.0	5.37	640	185 ± 54	399	40 ± 68	8	7
11B	03.04.00-09.04.00	813	0.90 ± 0.01	4.9 ± 0.3	5.4 ± 0.3	0.43	0.34 ± 0.10	-0.6 ± 1.8	0.93	48	-31 ± 8	321	259 ± 17	4	12
12B	10.04.00-01.05.00	434	2.13 ± 0.01	11.0 ± 0.3	5.2 ± 0.1	0.75	0.60 ± 0.16	1.2 ± 2.8	1.77	114	-36 ± 16	264	146 ± 25	14	20
13B	01.05.00-21.05.00	267	1.87 ± 0.01	10.2 ± 0.4	5.4 ± 0.2	0.65	0.49 ± 0.17	2.2 ± 3.2	1.79	172	20 ± 17	415	295 ± 28	18	22
14B	22.05.00-11.06.00	134	2.00 ± 0.01	11.6 ± 0.5	5.8 ± 0.2	0.64	0.50 ± 0.15	3.5 ± 2.9	1.62	259	121 ± 18	567	459 ± 30	17	19
15B	12.06.00-25.06.00	537	1.32 ± 0.01	8.0 ± 0.3	6.0 ± 0.3	0.47	0.37 ± 0.11	1.9 ± 2.2	0.92	133	55 ± 10	343	281 ± 17	31	14
16B	26.06.00-09.07.00	78	1.28 ± 0.01	8.2 ± 0.3	6.4 ± 0.3	0.41	0.30 ± 0.12	3.4 ± 2.3	0.76	124	60 ± 8	276	225 ± 14	29	16
17B	10.07.00-16.07.00	536	1.80 ± 0.01	10.7 ± 0.4	6.0 ± 0.2	0.65	0.53 ± 0.12	2.0 ± 2.4	1.23	127	23 ± 12	377	295 ± 21	29	15
1000 m															
1A	15.07.99-08.08.99	68	6.80 ± 0.13	45 ± 1.2	6.7 ± 0.2	1.55	1.47 ± 0.09	21.3 ± 2.6	5.00	871	448 ± 55	823	489 ± 73	12	11
2A	09.08.99-05.09.99	123	8.10 ± 0.13	45 ± 2.6	5.6 ± 0.3	1.73	1.63 ± 0.11	18.7 ± 4.3	5.65	716	237 ± 57	674	296 ± 77	12	13
3A	06.09.99-03.10.99	61	6.76 ± 0.06	40.3 ± 0.7	6.0 ± 0.1	1.59	1.51 ± 0.09	15.5 ± 2.1	4.95	638	220 ± 50	643	313 ± 68	12	10
4A	04.10.99-31.10.99	42	7.51 ± 0.04	47 ± 0.6	6.3 ± 0.1	1.79	1.69 ± 0.11	19.3 ± 2.3	5.09	671	240 ± 52	797	457 ± 73	13	12
5A	01.11.99-05.12.99	9.8	6.82 ± 0.04	42 ± 1.0	6.2 ± 0.2	1.59	1.50 ± 0.10	17.7 ± 2.6	4.67	610	215 ± 48	679	367 ± 66	10	11
6A	13.12.99-09.01.00	8.3	4.89 ± 0.02	27 ± 1.1	5.5 ± 0.2	1.08	0.96 ± 0.14	11.0 ± 3.4	3.52	449	151 ± 36	471	236 ± 49	17	16
7A	10.01.00-30.01.00	20	9.21 ± 0.03	41 ± 1	4.4 ± 0.1	1.78	1.71 ± 0.08	12.7 ± 2.3	6.45	821	275 ± 66	575	144 ± 84	10	9
8A	31.01.00-20.02.00	165	4.80 ± 0.01	32.4 ± 0.5	6.8 ± 0.1	1.40	1.36 ± 0.05	10.3 ± 1.4	3.91	509	179 ± 40	784	523 ± 60	9	5
9A	21.02.00-12.03.00	129	9.39 ± 0.10	43.9 ± 0.7	4.7 ± 0.1	2.06	2.02 ± 0.06	10.9 ± 1.7	6.61	857	297 ± 68	502	60 ± 84	8	6
10A	13.03.00-26.03.00	283	8.81 ± 0.06	42.4 ± 0.7	4.8 ± 0.1	1.90	1.86 ± 0.06	12.1 ± 1.8	6.02	801	291 ± 62	479	76 ± 77	9	6
11A	03.04.00-09.04.00	298	2.74 ± 0.02	17.5 ± 0.5	6.4 ± 0.2	1.00	0.90 ± 0.12	2.8 ± 2.5	2.24	244	54 ± 22	491	341 ± 35	5	13
12A	10.04.00-01.05.00	311	3.55 ± 0.03	22.9 ± 0.4	6.4 ± 0.1	1.18	1.10 ± 0.08	4.8 ± 1.8	3.04	333	76 ± 30	507	304 ± 44	9	10
13A	01.05.00-21.05.00	227	9.32 ± 0.03	42.1 ± 0.5	4.5 ± 0.1	1.96	1.88 ± 0.10	11.2 ± 2.2	6.76	921	349 ± 70	438	-14 ± 85	12	10
14A	22.05.00-11.06.00	289	3.06 ± 0.01	22.5 ± 0.4	7.4 ± 0.1	0.80	0.70 ± 0.11	11.1 ± 2.2	2.60	304	84 ± 26	587	413 ± 41	16	13
15A	12.06.00-25.06.00	528	2.81 ± 0.03	19.5 ± 0.5	6.9 ± 0.2	0.85	0.77 ± 0.09	6.9 ± 2.0	2.11	355	176 ± 23	576	435 ± 35	25	11
16A	26.06.00-09.07.00	277	2.79 ± 0.01	19.3 ± 0.4	6.9 ± 0.2	0.70	0.60 ± 0.10	9.4 ± 2.0	2.07	389	213 ± 24	721	582 ± 38	27	12
17A	10.07.00-16.07.00	422	4.30 ± 0.02	28.8 ± 0.6	6.7 ± 0.1	1.07	0.95 ± 0.13	13.2 ± 2.8	3.27	450	174 ± 34	846	628 ± 54	21	16

Table 2: Partition coefficient of Th between lithogenic particles, MnO₂ and seawater

	$K_{d-litho}^{Th} (10^7 \text{ ml/g})$	$K_{d-MnO_2}^{Th} (10^{10} \text{ ml/g})$
DYFAMED 1000 m, main correlation (this work)	0.8 ± 0.2	1.1 ± 0.4
DYFAMED 1000 m, winter samples (this work)	0.42 ± 0.04	0.6 ± 0.1
Eastern North Atlantic ¹	5	2
Eastern North Atlantic ²	0.5-10	0.7-4.2
North Atlantic ³	1	
Equatorial Pacific and Southern Ocean ⁴	23	
Equatorial Pacific and Southern Ocean ⁵	20	3.5
Panama Basin ⁶	2-4	0.6-3.7

¹ Roy-Barman et al., 2005. ² Scholten et al., 2001; Kuss and Kremling, 1999. ³ Chase et al., 2002. ⁴ Luo and Ku, 2004a; Luo and Ku, 2004b. ⁵ Luo and Ku, 1999, (unpublished data, 1993, <http://usjgofs.whoi.edu/jg/dir/jgofs/eqpac/tt013/>). ⁶ Anderson et al., 1983 (restricted to the samples with a sufficient Mn_{auth} content).

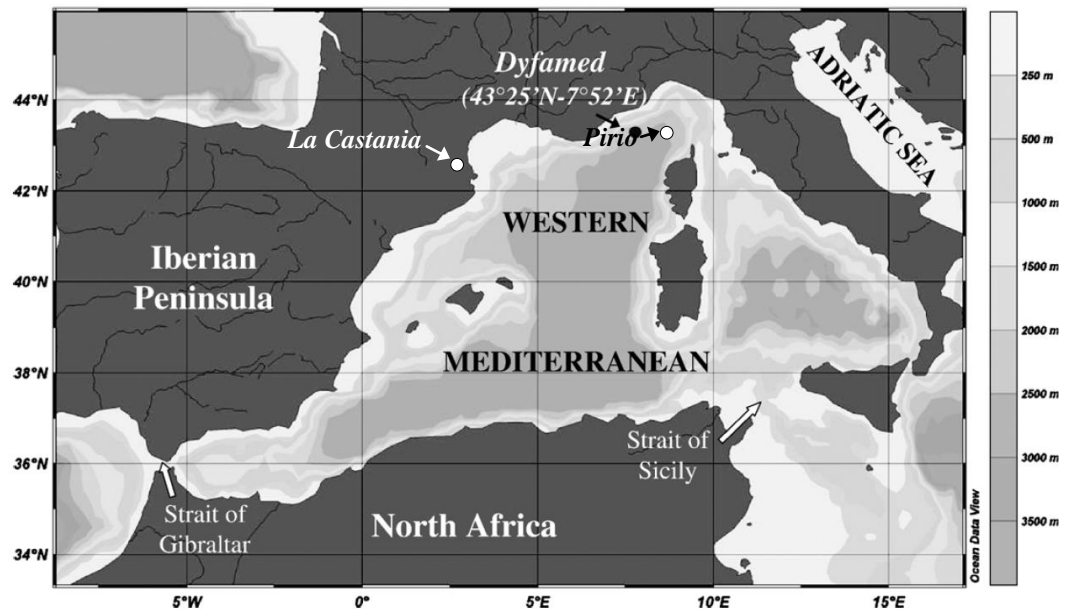


Figure 1

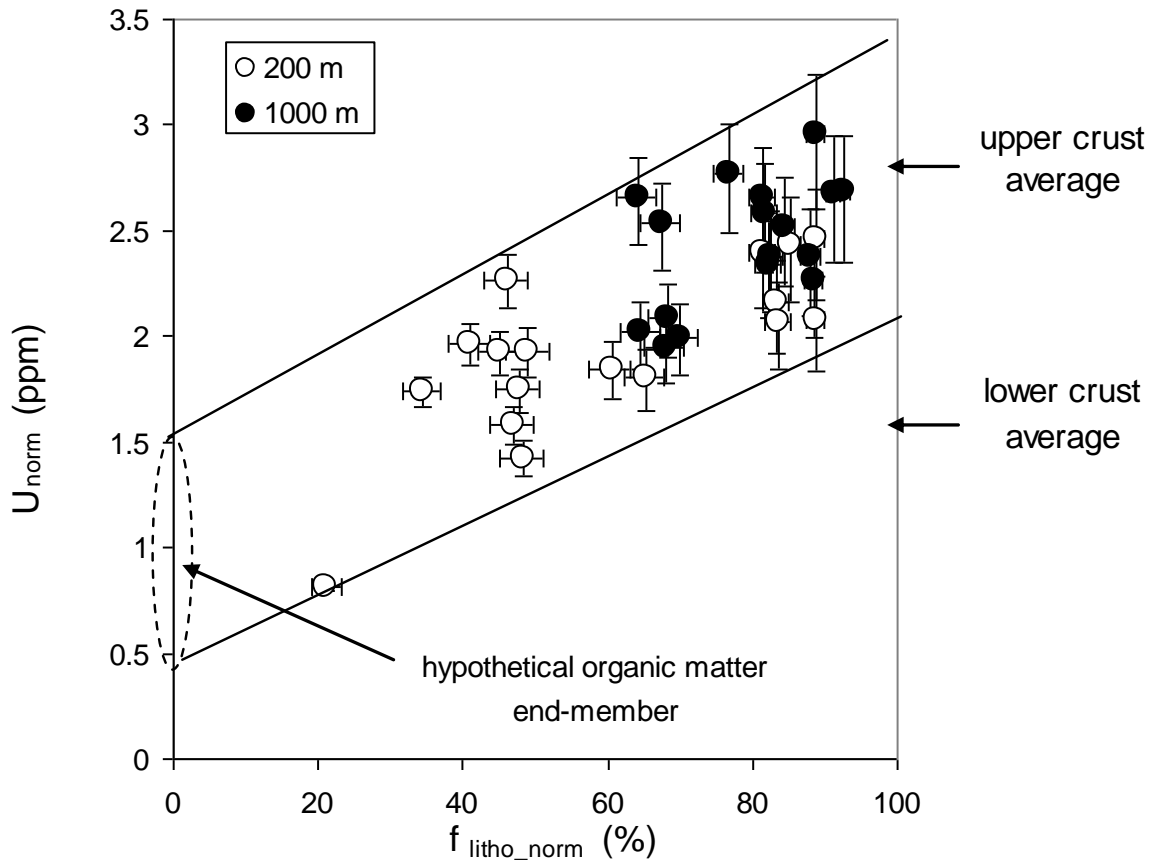


Figure 2

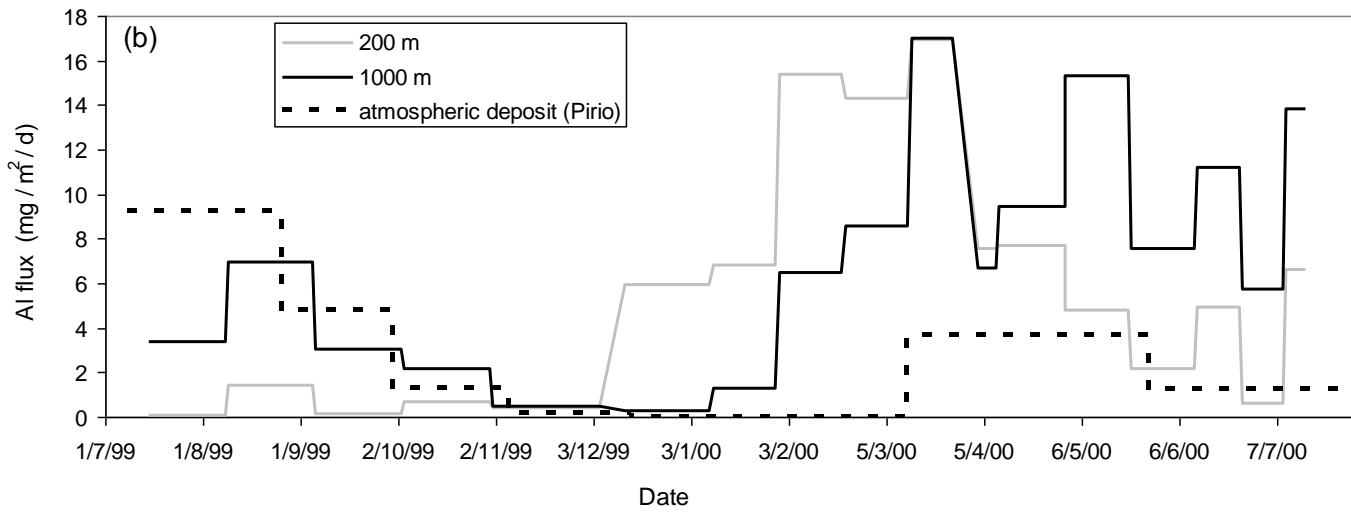
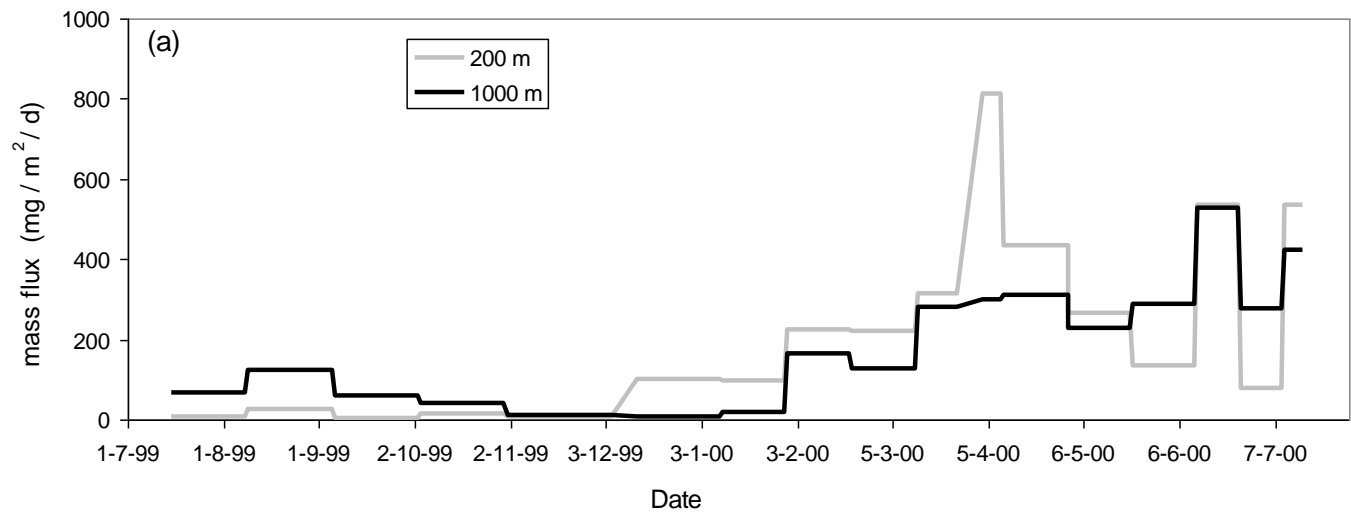


Figure 3

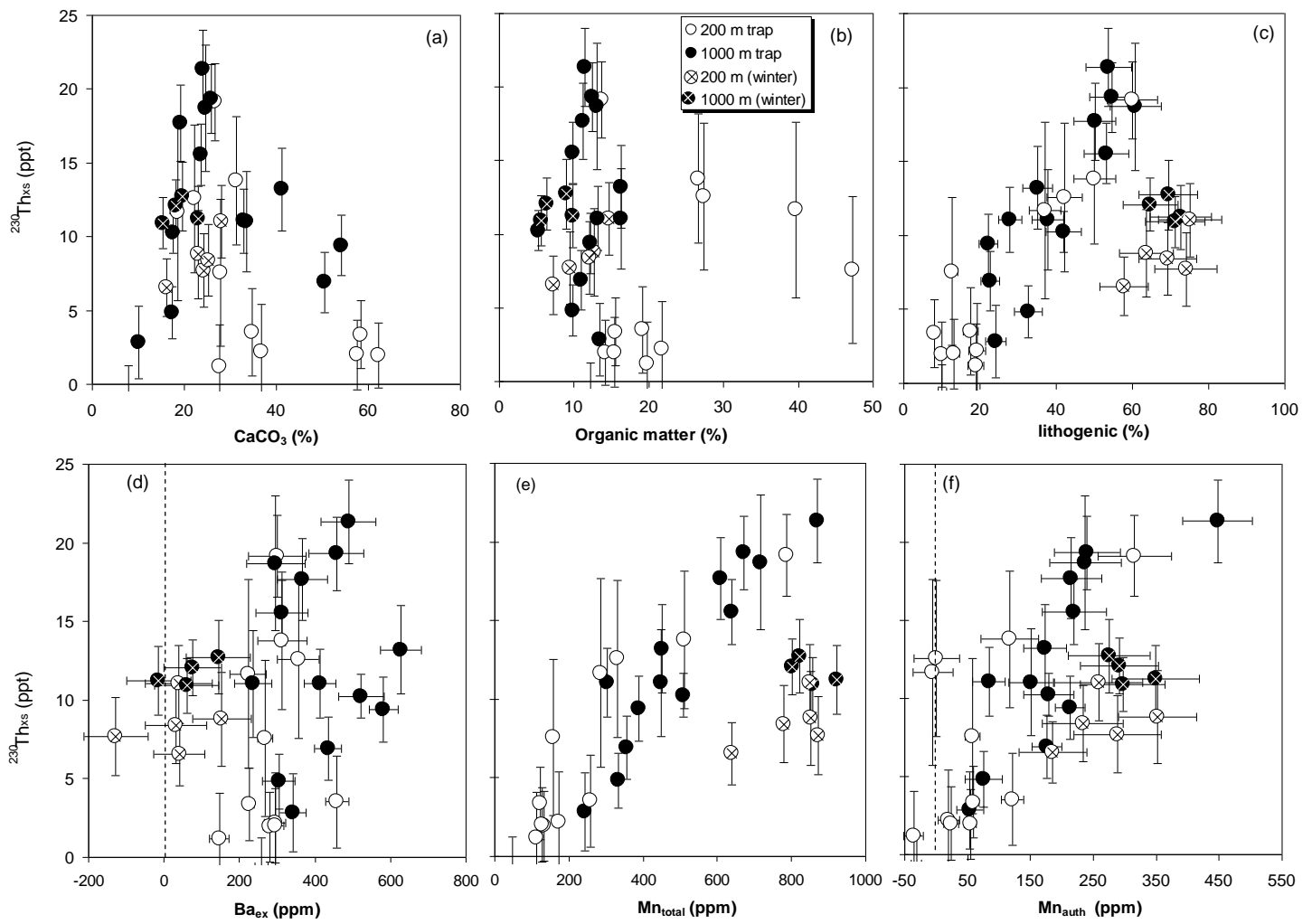


Figure 4