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The role of dense shelf water cascading in the transfer of organochlorine compounds to open marine waters

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30 **Abstract**

31 Settling particles were collected by an array of sediment trap moorings deployed along the
32 Cap de Creus (CCC) and Lacaze-Duthiers (LDC) submarine canyons and on the adjacent
33 southern open slope (SOS) between October 2005 and October 2006. This array collected
34 particles during common settling processes and particles transferred to deep waters by dense
35 shelf water cascading (DSWC). Polychlorobiphenyls (PCBs), dichlorodiphenyltrichloroethane
36 and its metabolites (DDTs), chlorobenzenes (CBzs) –pentachlorobenzene and
37 hexachlorobenzene- and hexachlorocyclohexanes were analyzed in all samples. The results
38 show much higher settling fluxes of these compounds during DSWC than during common
39 sedimentation processes. The area of highest deposition was located between 1000 and 1500
40 m depth and extended along the canyons and outside them showing their channelling effects
41 but also overflows of dense shelf water from these canyons. Higher fluxes were observed near
42 the bottom (30 m above bottom; mab) than at intermediate waters (500 mab) which is
43 consistent with the formation and sinking of dense water close to the continental shelf and
44 main displacement through the slope by the bottom. DSWC involved the highest settling
45 fluxes of these compounds ever described in marine continental slopes and pelagic areas, e.g.
46 peak values of PCBs ($960 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$), DDTs ($2900 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$), CBzs ($340 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and
47 lindane ($180 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$).

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65 **Introduction**

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67 Organochlorine compounds (OCs), such as polychlorobiphenyls (PCBs) and chlorinated
68 pesticides, constitute a group of persistent organic pollutants of worldwide concern due to
69 their toxic effects.^{1,2} Their high lipophilicity, hydrophobicity, chemical stability and resistance
70 to biological degradation have led to the accumulation of these compounds in biological
71 tissues and their biomagnification through to the food chain.^{3,4} These compounds are also
72 susceptible of long-range atmospheric transport to remote areas such as polar regions⁵,
73 mountain lakes^{6,7} and marine environments⁸.

74 OCs can enter into the marine environment by effluent discharges, atmospheric
75 deposition, runoff and other means.⁸ Once in the water column association to particulate
76 matter, transport and settling play an important role in their transfer from surface to deep
77 waters and sediments.⁹ Particle settling is also favoured by biological processes involving
78 incorporation into the food web, organism migration, vertical mixing, formation of large
79 particles such as fecal pellets, organic matter aggregation, and others.

80 Besides these mechanisms, other hydrodynamic processes induce lateral (cross-slope)
81 transport from continental shelves to the adjoining environments.¹⁰⁻¹² This is the case of the
82 Gulf of Lion (GoL) where a major mechanism controlling shelf-deep ocean exchange is dense
83 shelf water cascading (DSWC). The strongest exchange by this process takes place in the
84 westernmost end of the GoL through the Cap the Creus canyon (CCC)^{13,14}. Various studies
85 have described the rapid export of sediment from the shelf and upper slope to deep
86 environments by DSWC events and the role of submarine canyons in funneling the waters
87 generated in these episodes.¹⁵⁻¹⁹

88 To date no study has considered the influence of these cascading processes in the
89 transport of OCs from the continental shelf to the deep marine environment. For this purpose,
90 mooring lines with sediment traps were deployed in nine sites of the western GoL following
91 the physiography of the CCC and Lacaze-Duthiers canyon (LDC), the most active in the area
92 regarding sediment transfer. Traps were set between 300 m and 1900 m water depth and
93 settling particles were sampled between October 2005 and October 2006 at fortnightly
94 intervals. Two DSWC events occurred during this period and this experiment offered the
95 possibility to study transfer of OCs to the deep realm both during common settling and
96 intense events. The present study is aimed to analyze the spatial and temporal variability of

97 OCs deposition in order to assess the role of DSWC and the submarine canyons in the
98 transport of these compounds to the deep sea. Data are also compared with results from
99 previous sediment trap studies that have generally been performed in coastal areas, bays or
100 lakes.^{7,20-23}

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102 **Results and discussion**

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104 **Dense shelf water cascading.** Two major deep DSWC pulses occurred during early January
105 and late March 2006 (Fig. S1). Strongest temperature drops and current speed peaks occurred
106 at the onset of each cascading period and in late March 2006^{17,18}, e.g. 60-100 cm·s⁻¹ in
107 CCC1000. High current intensities were recorded at CCC300 and LDC300 (up to 95 cm·s⁻¹;
108 Fig. S1) during these episodes. At 1000 m depth the intensities reached up to 70 and 50 cm·s⁻¹
109 in CCC and LDC, respectively. In CCC1500 the current intensities were in the range of 10-35
110 cm·s⁻¹ during these events. These intensities were also recorded at the intersect of CCC and
111 LDC (CCC1900 m; Fig. S1) as well as outside the canyons (50 and 30 cm·s⁻¹ in SOS1000 and
112 SOS1900).

113 In the canyons, these episodes also involved temperature decreases down to 2°C at 300
114 m depth and 1°C at 1000 m depth (Fig. S1). In SOS1000 and SOS1900 the observed
115 temperatures decreased down to 0.8°C and 0.1°C, respectively. In CCC1500 temperatures
116 decreased down to 0.3°C and no significant change was observed at 1900 m depth. These
117 temperature measurements and the preferential directions along slope of the DSWC water
118 mass at CCC1900 and SOS1900 stations¹⁸ indicate that dense water reached equilibrium
119 (neutral density contrast) at about 1900 m depth and flowed along the SOS slope following
120 the cyclonic circulation once dense water is no longer constrained by the canyon walls.

121 DSWC entrained a strong increase of settling particle fluxes. The highest increase was
122 recorded in CCC1000 with total mass fluxes up to 90 g·m⁻²·d⁻¹ (Fig. 2). In the LDC and the
123 SOS the highest fluxes were also observed at 1000 m depth (40 g·m⁻²·d⁻¹). The mid-water
124 LDC trap (500 m above bottom; mab) also showed a total mass flux maximum during DSWC
125 (10 g·m⁻²·d⁻¹) in agreement with the near-bottom trap in the same line (Fig. 2). These values
126 strongly contrasted with those recorded during periods without deep DSWC for which fluxes
127 were lower than 5 g·m⁻²·d⁻¹.

128 **Concentrations and fluxes of organochlorine compounds. PCBs.** The PCB
129 concentrations (sum of 7 ICES congeners) in the settling particles ranged between 2.7 and
130 160 ng·g⁻¹ (Fig. S2). The highest concentrations were found in the deepest traps deployed in

131 the CCC and LDC (CCC1500, CCC1900 and LDC1500) and in the SOS (SOS1900). The
132 peak values were recorded during November-December 2005 and July-August 2006. Average
133 concentrations in the different sediment trap stations ranged between 16 (CCC1000) and 26
134 $\text{ng}\cdot\text{g}^{-1}$ (LDC1000 (500)) (Table 1). The observed concentrations did not increase during
135 DSWC (Fig. S2). These concentration values were lower than those reported in the LDC 20
136 years ago ($31\text{-}1500 \text{ ng}\cdot\text{g}^{-1}$)²⁴, although in that case they were reported as Aroclor 1254
137 equivalents.

138 The observed congener profiles are similar to those found in other marine settling
139 particles, such as the Mediterranean Sea²⁴⁻²⁷, the Sargasso Sea²⁸ and the Arabian Sea.²⁹ PCB-
140 101 was the highest congener in all samples (Fig. S3). Nevertheless, during DSWC (January-
141 March 2006) a relative enrichment of PCB-153 and PCB-138 was observed in all stations
142 (Fig. S3A) whereas enrichment in the less chlorinated isomers, especially PCB-52 and PCB-
143 101 (Fig. S3B), was found during common sedimentation. This contrast may reflect PCB
144 composition differences between shelf sediment and settling particles. The former, dragged
145 by DSWC, may be depleted in the less chlorinated congeners by long-term dissolution and
146 washing while the latter may better reflect atmospheric deposition³⁰.

147 All traps except one, LDC300, showed highest PCB fluxes in the period of DSWC
148 (Table 2; Fig. 2). During these DSWC episodes a maximum flux of $960 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ was
149 recorded at CCC1000 station (Table 1). Average PCB deposition fluxes at CCC1000,
150 CCC1500, LDC1000, LDC1500 and SOS1000 sites during DSWC ranged between 63.5 and
151 $570 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$. These moorings were all located in the 1000-1500 m water column depth
152 range. The trap situated close to the bottom (30 mab) at LDC1000 station exhibited higher
153 fluxes, $400 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, than the one at high position (500 mab), $135 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 2). Other
154 average fluxes during DSCW ranged between 24 and $31 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 2).

155 The average settling fluxes in the period without DSWC ranged between 12 and 160
156 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and decreased with increasing distance from the mud belt on the shelf (Table 2).
157 According to these results, while PCB settling without cascading occurred in higher intensity
158 closer to the mud belt, deposition during DSWC was dominant in an area between 1000 and
159 1500 m in agreement with the location of the CCC1000, LDC1000, LDC1500 and SOS1000
160 sites. This area extends inside and outside the canyons denoting both their channeling effect
161 on the sediments washed by DSWC but also overflows of dense shelf water from these
162 canyons.

163 Comparison of the average settling fluxes during DSWC and common sedimentation
164 (D/R ratios; Table 2) shows highest values for the moorings in the above mentioned 1000-

165 1500 m depth sites, involving between 5.3 and 12 times higher fluxes during DSWC. In all
166 the other mooring sites except LDC300 the PCB settling fluxes were also higher during
167 DSCW with D/R ratio values between 1.5 and 4.2. LDC300, the only exception, had an
168 average PCB settling flux during DSWC of $24 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ that was similar to that observed in
169 CCC1900 and SOS1900 in the same period. However, during common sedimentation the
170 highest average flux, $160 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, was observed in this site close to the mud belt.

171 Comparison of the settling fluxes during common sedimentation found in the western
172 GoL with those observed in other sites shows values in the same range as those in the Arabian
173 Sea (1988-91; 4017 m)²⁹, $16 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (13 PCB congeners), the Alboran Sea (1992; 1200
174 m)²⁶, $18.5 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (7 ICES PCB congeners), or in the Botnian Sea (1991-93; 115 m)³¹, 34
175 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (68 PCB congeners). However, in other marine areas the observed settling fluxes
176 are much lower, e.g. in the eastern Mediterranean Sea (2001; 186-1426 m)²⁷, $0.63 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
177 (54 PCB congeners), in the Sargasso Sea (1978-80; 3200 m)²⁸, $4.3 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (12 PCB
178 congeners), and in the Swedish western coast (1999-01; 125-155 m)³², $5.5 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (6 PCB
179 congeners). In this context, the observed PCB settling fluxes during DSWC (Fig. 2; Table 2)
180 are the highest ever described in marine continental slopes and open sea areas.

181 *DDTs*. The DDT concentrations in the settling particles (sum of 2,4'-DDE, 4,4'-DDE,
182 2,4'-DDD, 4,4'-DDD, 2,4'-DDT, 4,4'-DDT) ranged between 0.8 and $85 \text{ ng}\cdot\text{g}^{-1}$ (Table 1). The
183 highest concentrations, $85 \text{ ng}\cdot\text{g}^{-1}$, were found at SOS1000 in the first fortnight of January
184 2006. With the exception of this result the DDT concentrations followed rather closely the
185 changes observed for the PCBs. The average concentrations in the different sediment trap
186 stations ranged between 7.0 (CCC300) and $41 \text{ ng}\cdot\text{g}^{-1}$ (SOS1000) (Table 1). These
187 concentrations are lower than those observed in a sediment trap study performed in the LDC
188 ($3.1\text{-}64.5 \text{ ng}\cdot\text{g}^{-1}$) in 1985-86.²⁴ Nevertheless, the present GoL concentrations are higher than
189 those found in sediment traps deployed in the Alboran Sea ($1.9 \text{ ng}\cdot\text{g}^{-1}$)²⁶, in the Baltic Sea (8.1
190 $\text{ng}\cdot\text{g}^{-1}$)³¹ and in the Arabian Sea ($4.1 \text{ ng}\cdot\text{g}^{-1}$).²⁹

191 In all moorings except LDC300 the average total DDT fluxes during DSWC were
192 higher than in the period of common sedimentation (Fig. 2; Table 2). The highest average
193 total DDT fluxes during DSWC, between 60 and $530 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, were observed in the
194 moorings located at 1000 m, CCC1000, LDC1000 and SOS1000 (Table 2). In this context,
195 the trap located close to the bottom (30 mab, LDC1000) exhibited higher fluxes, e.g. 190
196 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ than the one at mid-water (500 mab, LDC1000 (500)), e.g. $60 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 2).
197 The moorings located at 1500 m, CCC1500 and LDC1500, defined a second group of high
198 average settling fluxes, $32\text{-}33 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 2). The geographic area of high average total

199 DDT settling during DSWC is about the same as the one observed from PCB deposition
200 during these episodes. The settling fluxes in the moorings at 1900 m, CCC1900 and SOS
201 1900, ranged between 11 and 17 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ during DSWC (Table 2). The average settling
202 fluxes during common sedimentation ranged between 3.6 and 73 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, and, as for PCBs,
203 decreased from increasing distance from the mud belt on the shelf (Table 2). However,
204 deposition during DSWC was accumulated in an area between 1000 and 1500 m (CCC1000,
205 LDC1000, LDC1500, SOS1000) both inside and outside the canyons.

206 The average settling fluxes in the moorings of the above mentioned 1000-1500 m
207 depth sites were between 6.8 and 79 times higher during DSWC than during common
208 sedimentation (Table 2). In all the other mooring sites except LDC300 the DDT settling
209 fluxes were also higher during DSCW than during common sedimentation with D/R ratio
210 values between 2.3 and 4.5. Only in LDC300 the average total DDT flux was lower during
211 DSWC than in the common sedimentation period, D/R ratio 0.16 (Table 2).

212 The observed DDT settling fluxes during common sedimentation in the GoL are in the
213 same order of magnitude than those found in the Botnian Sea (1991-93; 115 m)³¹, 16 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
214 ¹, and higher than those observed in the Arabian Sea (1988-91; 4017 m)²⁹, 0.6 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, and
215 in the Alboran Sea (1992; 1200 m)²⁶, 1.4 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$. According to these previous studies, the
216 observed DDT settling fluxes during DSWC (Fig. 2; Table 2) are the highest ever described in
217 marine continental slopes and pelagic areas.

218 The percent ratio $100\cdot(2,4'\text{-DDT} + 4,4'\text{-DDT})/(2,4'\text{-DDT} + 4,4'\text{-DDT} + 2,4'\text{-DDE} +$
219 $4,4'\text{-DDE} + 2,4'\text{-DDD} + 4,4'\text{-DDD})$ can be used to assess whether DDT emission occurred
220 recently or in the past. High ratios involve predominance of the original insecticide and
221 therefore recent applications of it whereas low ratios correspond to predominance of
222 metabolites and indicate older use. In most samples these ratios are below 50% (Fig. S4), in
223 consistency with predominance of older use. In some cases the ratio exceeded 50% which
224 may reflect some contributions of recent applications of the insecticide.

225 *Chlorobenzenes.* Total chlorobenzenes (CBzs) encompass the sum of
226 pentachlorobenzene (PeCB) and hexachlorobenzene (HCB). Even though the production of
227 PeCB and HCB was banned globally under the Stockholm Convention (2004), these
228 compounds are still released into the environment as byproducts of the synthesis of several
229 organochlorine solvents and as a result of backyard trash burning and municipal waste
230 incineration.³³ The concentrations of these compounds in the settling particles varied between
231 not detected and 21 $\text{ng}\cdot\text{g}^{-1}$ in CCC1000 and CCC1500, respectively (Fig. S2). The average
232 concentrations ranged between 3.8 and 6.6 $\text{ng}\cdot\text{g}^{-1}$ in LDC1000 and CCC1500, respectively

233 (Table 1). These values correspond to average percentages of 31% and 67 % for PeCB and
234 HCB, respectively, and both compounds showed the same temporal pattern in the sediment
235 trap stations (not shown). The temporal and geographic distribution of these CBzs is very
236 similar to that observed for the PCBs, with the highest concentrations found in the deepest
237 traps. In comparison to other studies, the HCB concentrations (nd-14.5 ng·g⁻¹; average 3.5
238 ng·g⁻¹) are similar to those found in the Baltic Sea (0.49 – 3.4 ng·g⁻¹; average 1.9 ng·g⁻¹)³¹ and
239 in the LDC in 1985-86 (0.3-3.7 ng·g⁻¹; average 1.7 ng·g⁻¹).²⁴

240 Consistently with the trends observed for DDTs and PCBs, all trap stations except
241 LDC300 showed higher deposition fluxes during DSWC (Fig. 2; Table 2). The average
242 deposition fluxes during DSWC ranged between 6.6 and 200 ng·m⁻²·d⁻¹ and the average fluxes
243 during common sedimentation were 1.3-37 ng·m⁻²·d⁻¹ (Table 2). This flux range during the
244 period without cascading was found in LDC and is consistent with previous flux
245 measurements, 9.2 ng·m⁻²·d⁻¹, in this canyon in 1985-86²⁴. These values are higher than those
246 observed in the Botnian Sea³¹, 3.7 ng·m⁻²·d⁻¹, as for DDTs and PCBs. The results observed in
247 the GoL show that the deposition fluxes during DSWC are the highest ever reported in the
248 literature.

249 The highest deposition CBzs fluxes during DSWC were observed at 1000 m depth in
250 CCC, LDC and SOS, 42-200 ng·m⁻²·d⁻¹ (Table 2). The deposition flux in LDC1000 (500 mab)
251 was also high, 35 ng·m⁻²·d⁻¹, but a bit lower than at 30 m depth, 95 ng·m⁻²·d⁻¹. The enhanced
252 settling during DSWC was also recorded at 1500 m depth which showed lower values than in
253 the mooring located at shallower positions, 20-24 ng·m⁻²·d⁻¹, but still higher than those in the
254 deep mooring sites, 6.6-8.9 ng·m⁻²·d⁻¹. The moorings at 1000 and 1500 m depth defined a
255 geographic area of highest DSWC CBzs accumulation that is coincident with the area of PCB
256 and DDT accumulation during these events (Fig. 1). Again, the CBzs results showed that the
257 canyons channel the supply of these compounds during DSWC but there was an overflow
258 which resulted into enhanced transfer in the SOS areas.

259 The DSWC involved higher settling fluxes of CBzs than during common
260 sedimentation. Thus, in the moorings at 1000 m the D/R ratios ranged between 7.9 and 20
261 (5.7 for the trap at 500 mab). In the moorings at 1500 m these ratios were 9.2 and 13
262 (CCC1500 and LDC1500, respectively) and in the moorings at 1900 m the ratios were 3.7
263 (CCC1900) and 6.8 (SOS1900). LDC300 was the only mooring in which the opposite trend
264 was observed and the D/R was 0.21. This differential behavior of this mooring is in
265 agreement with the observations in PCB and DDT sedimentation fluxes. Like in these
266 previous cases, the smaller distance of this mooring from the mud belt on the shelf suggests

267 that during common sedimentation in this area there is a regular deliver of pollutants stored in
268 the mud belt which is also consistent with the enhanced total mass flux recorded in this
269 mooring (Fig. 2).

270 *Lindane*. γ -HCH was the only hexachlorocyclohexane isomer detected in most of the
271 samples. The concentrations of lindane in the settling particles varied between not detected
272 (CCC1000, SOS1000) and $11 \text{ ng}\cdot\text{g}^{-1}$ (LDC1000 500mab, LDC1500) and the average trap
273 concentrations in each mooring ranged between 0.5 and $4.1 \text{ ng}\cdot\text{g}^{-1}$, the former in CCC300 and
274 the latest in CCC1900 (Fig. S2, Table 1). These concentrations were similar to those found in
275 the Baltic Sea,³¹ $3.6 \text{ ng}\cdot\text{g}^{-1}$ and in the LDC 20 years ago²⁴, $2.8 \text{ ng}\cdot\text{g}^{-1}$, but lower than those
276 found in the Alboran Sea²⁶, $24 \text{ ng}\cdot\text{g}^{-1}$.

277 The depositional fluxes of lindane were higher during DSWC, 5.1 - $98 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, than
278 during common sedimentation, 0.6 - $10 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, in all cases except LDC300 (Fig. 2; Table
279 2). The highest fluxes during DSWC were found in the moorings located at 1000 m depth
280 both in the traps at 30 m depth, 20 - $90 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, and in the trap at 500 m depth, $23.5 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
281 (Table 2). The trap at 1500 m from LDC also showed important fluxes during this event, 14
282 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$. In the case of CCC1500 the DSWC deposition flux was smaller and similar to
283 those at 1900 m depth or LDC300, 5.1 - $6.35 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 2). The deposition fluxes during
284 common sedimentation were much lower 0.6 - $3.5 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, except in LDC300, $10 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
285 (Table 2), the mooring situated closer to the mud belt. This pattern is well in agreement with
286 the flux differences observed for the other OCs.

287 The fluxes during common sedimentation were higher than the average fluxes
288 observed in the Alboran Sea²⁶, $4.2 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, but lower than those found in the Botnian Sea³¹,
289 $34 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, and in the LDC in 1985-86²⁴, $24 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$. However, the deposition fluxes
290 during DSWC were similar to those from these two last cases and much higher than those
291 recorded in the Alboran Sea²⁶.

292 The DSWC involved higher deposition fluxes of lindane than during common
293 sedimentation. Thus, in the moorings at 1000 m the D/R ratios ranged between 12 and 41 (6.7
294 for the trap at 500 mab). In LDC1500 the ratio was 22. The D/R ratios in the other moorings
295 with higher flux during the DSWC ranged between 3.4 and 9.0. LDC300 was the only
296 mooring in which the opposite trend was observed (D/R = 0.51). These ratios in each of the
297 moorings are well in agreement with the ratios found in PCB, DDT and CBzs.

298 **The role of DSWC in the transport of organochlorine compounds.** The two DSWC
299 pulses in early January and late March (Fig. S1) show the capacity of cascading waters to
300 transport OCs over several tens of kilometers offshore in a few days. The first pulse was more

301 intense at the upper reaches of the CCC (1000 m depth) involving peak fortnightly-averaged
302 settling fluxes of $960 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $630 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $340 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and $180 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for PCBs,
303 DDTs, CBzs and lindane, respectively (Fig. 2; Table 1). In the LDC, peak settling values were
304 also found at 1000 m depth, $280 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $110 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $77 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and $51 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$,
305 respectively. In this canyon, DDTs constituted the only exception and the highest settling flux
306 occurred at 300 m depth, not being related to DSWC (Fig. 2; Table 1). On the SOS, peak
307 settling flux also occurred at 1000 m depth and related to DSWC, showing fortnightly-
308 averaged deposition fluxes of $640 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $2900 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, $100 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ and $44 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$
309 for PCBs, DDTs, CBzs and lindane, respectively (Fig. 2; Table 1). The second pulse was less
310 intense but again involved a peak settling flux of OCs at 1000 m depth (Fig. 2).

311 The enhanced settling fluxes of OCs during DSWC were paralleled by increases of
312 total mass flux. Representation of the settling fluxes of OCs and total particulate organic
313 carbon from the fortnightly-averaged samples collected during DSWC and common
314 sedimentation show very strong correlation coefficients ($r^2 = 0.94, 0.89, 0.92$ and 0.79 for
315 PCBs, DDTs, CBzs and lindane, respectively; Fig. 3). The changes in deposition fluxes are
316 therefore related to the general dynamics of the settling particles in the GoL. Accordingly,
317 during common sedimentation, the observed increase in fluxes of PCBs, DDTs and CBzs in
318 CCC300 and LDC300 during September-October 2006 occurred in parallel to increases of
319 total mass flux. Such increase was caused by sporadic events of shelf and/or canyon-head
320 sediment resuspension¹⁰ due to the increase of waves and western river discharges during
321 these months.^{18,34,35}

322 In this respect, the above mentioned low OCs fluxes in LDC300 during DSWC that
323 were even lower than the fluxes at 1000 m in the same canyon during common sedimentation,
324 e.g. $24 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ vs $52\text{-}76 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for PCBs, or $12 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ vs $20\text{-}28 \text{ ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for DDTs
325 (Fig. 2; Table 2), may have been caused by low particle deposition due to very strong currents
326 at the canyon head during these DSWC episodes.

327 The overall significance of the enhanced flux deposition due to DSWC episodes can
328 be assessed by calculation of the inventories by integration of the average fluxes during the
329 time intervals in which DSWC and common sedimentation were predominant. The PCB
330 inventories show that deposition during DSWC was more important than during common
331 sedimentation in the mooring sites at 1000 and 1500 m depth, those with highest settling
332 fluxes during DSWC (Table 2). In these sites, the accumulated PCBs during the DSWC
333 episodes represented about 58-80% of the total annual input. In the moorings at 1900 m
334 depth, the settling PCBs during DSWC involved 33-46% of the whole annual budget and in

335 LDC300 they only involved 5% (Table 2). The lower rate in LDC1000 (500) than in
336 LDC1000, 41% vs 63%, respectively, is consistent with the physical characteristics of the
337 DSWC episodes involving formation and sinking of dense water close to the continental shelf
338 and main displacement through the slope by the bottom.

339 Similar figures were obtained for DDTs, in which sedimentation during DSWC at
340 1000 and 1500 m depth involved 60-96% of the annual input, at 1900 m depth 45-58% and at
341 300 m depth only 5% (Table 2). The inventories of CBzs showed that sedimentation during
342 DSWC was very significant at 1000 m and 1500 m depth, 72-87%, but also at 1900 m depth,
343 55-69%, and not in LDC300, 7% (Table 2). The same is the case for lindane, with 53-93% in
344 the mooring traps at 1000 and 1500 m depth, 56-75% at 1900 m depth and 14% at 300 m
345 depth (Table 2). In all these cases higher inventories in LDC1000 were found in the trap
346 located at 30 m depth than at 500 m depth, e.g. 69% vs 50%, 72% vs 64% and 91% vs
347 71% for DDTs, CBzs and lindane, respectively, which is consistent with the preferred
348 transport close to the bottom sea as consequence of the formation of dense waters.
349 Representation of the summed inventories of all measured compounds (Table 2) shows an
350 area of highest settling of OCs during DSWC that is related to the morphology of the canyons
351 but that also extends outside them, in the SOS region (Fig. 4).

352

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354

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362

363 **Supporting information available**

364

365 The study area, oceanographic setting, sample collection methods, materials and
366 analytical procedures are described in a Supporting Information section. This information is
367 available free of charge via the internet at <http://pubs.acs.org/>.

368

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- 478

479 **Table 1.** Settling fluxes ($\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and concentrations ($\text{ng}\cdot\text{g}^{-1}$) of PCBs, DDTs, CBzs and HCHs and total mass
 480 (TM; $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and organic carbon (TOC; %) in particles collected in sediment traps deployed along the Cap de
 481 Creus Canyon (CCC), Lacaze-Duthiers Canyon (LDC) and the Southern Open Slope (SOS). nd = not detected;
 482 nq = not quantified.

Station		TM ^c	TOC	PCBs ^d		DDTs		CBzs		Lindane	
		Flux	%	Flux ^e	Conc	Flux	Conc	Flux	Conc	Flux	Conc
CCC300 ^a	Max	12.5	2.9	320	30	92	7.5	62	5.1	6.1	0.7
	Min	3.8	1.4	99	21	28	5.5	5.2	1.2	2.0	0.4
	Mean ^b	7.0	1.8	180	25	50	7.0	30	4.2	3.4	0.5
CCC1000	Max	90	2.9	960	51	630	34	340	5.1	180	7.2
	Min	1.1	0.8	28	8.5	11	4.0	nd	nd	nd	nd
	Mean	12	1.3	190	16	100	8.6	57	5.0	26	2.4
CCC1500*	Max	6.3	9.5	120	95	58	36	53	21	18	5.9
	Min	<0.1	1.4	1.4	12	0.6	6.4	nq	nq	0.1	0.6
	Mean	1.3	1.9	28	20	14	10	8.8	6.6	2.7	2.0
CCC1900*	Max	3.2	6.5	82	81	35	28	20	14	19	7.5
	Min	<0.1	1.4	2.6	11	1.0	5.5	0.4	1.0	0.1	0.6
	Mean	0.7	2.1	18	25	6.2	9.3	3.1	5.0	2.5	4.1
LDC300	Max	18	3.5	460	33	350	25	120	9.7	52	6.5
	Min	0.3	1.4	6.4	9.1	3.9	4.4	2.4	2.5	1.7	0.3
	Mean	6.7	1.8	125	19	57	9.5	29	4.8	8.9	1.6
LDC1000 (500mab)	Max	11	4.5	280	46	130	19	77	12	51	11
	Min	0.6	1.5	15	14	5.7	7.2	1.4	1.5	1.0	0.6
	Mean	2.9	2.2	80	26	30	10	14	5.2	8.5	3.3
LDC1000*	Max	40	2.3	570	35	260	14	140	5.4	96	6.4
	Min	2.1	1.2	25	11	13	5.3	6.1	2.5	nq	nq
	Mean	6.7	1.5	105	18	42	8.0	20	3.8	12	2.4
LDC1500	Max	12	5.4	270	160	100	49	61	19	45	11
	Min	<0.1	1.4	0.8	2.7	0.3	0.8	0.1	0.3	<0.1	0.1
	Mean	1.4	1.9	30	21	11	8.2	6.3	4.8	3.9	3.1
SOS1000*	Max	34	4.2	640	59	2900	85	100	8.9	44	5.5
	Min	0.2	1.2	3.4	6.5	2.0	3.4	0.6	1.8	nd	nd
	Mean	3.4	1.5	70	20	145	41	13	3.9	6.6	1.9
SOS1900*	Max	5.7	6.5	64	140	41	44	20	17	13	7.4
	Min	<0.1	1.2	1.2	9.8	0.4	7.1	0.2	1.5	<0.1	0.8
	Mean	0.8	1.9	20	18	7.4	8.7	3.3	4.2	2.2	3.8

483 ^aSediment trap material was collected fortnightly from mid October 2005 to late October 2006. A maintenance
 484 and turnaround was performed in mid April 2006 and no samples were collected during this period. Failure of
 485 the sediment traps rotating motor resulted in the absence of samples during the first six months at the CCC300
 486 station and from February to April 2006 in LDC1000. Due to low amounts of collected particles during July 15,
 487 2006 to October 30, 2006 no samples were analyzed in CCC1500, CCC1900 and SOS1900.

488 ^bMean concentrations for each specific OC were calculated by as follows:

489 where i is the indicator of each cup, C_i is the concentration of the OC in cup i , T the total mass collected in
 490 cup i and n the total number of cups.

491 ^cTotal mass fluxes in each cup: $F = \frac{T}{d}$ where d is the number of days in which the cup was deployed.

492 ^d7 ICES PCB Congeners

493 ^eMean fluxes for each specific OC were calculated by as follows:

where

494 F_i is the flux of the OC in cup i and the other variables are defined as above.

495

Table 2. Mean settling fluxes ($\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and inventories ($\mu\text{g}\cdot\text{m}^{-2}$) of organochlorine compounds during the dense seawater cascading episode (D; January-March 2006) and during the period of common sedimentation (R).

498

		PCBs				DDTs				CBzs				Lindane				OCs ^d			
		Flux		Inventory ^a		Flux		Inventory		Flux		Inventory		Flux		Inventory		Flux		Inventory	
CCC1000	D	570	8.8 ^b	51	74 ^c	320	13 ^b	29	81 ^c	200	20 ^b	18	87 ^c	98	41 ^b	8.8	93 ^c	1200	12 ^b	110	80 ^c
	R	65		18		24		6.6		9.9		2.7		2.4		0.66		100		27.5	
CCC1500	D	63.5	4.2	5.7	58	32	4.5	2.9	60	24	9.2	2.2	76	5.5	3.4	0.495	53	125	4.8	11	61
	R	15		4.1		7.0		1.9		2.6		0.71		1.6		0.44		26		7.5	
CCC1900	D	24	1.5	2.2	33	11	2.3	0.99	45	6.6	3.7	0.59	55	5.5	3.9	0.495	56	47	1.9	4.2	39
	R	16		4.4		4.7		1.2		1.8		0.49		1.4		0.385		24		6.6	
LDC300	D	24	0.15	2.2	5	12	0.16	1.1	5	8.1	0.21	0.73	7	5.1	0.51	0.46	14	49	0.23	4.4	5
	R	160		44		73		20		37		10		10		2.75		290		80	
LDC1000 (500)	D	135	2.2	12	41	60	3.0	5.4	50	35	5.4	3.15	64	23.5	6.7	2.1	71	250	2.7	23	48
	R	62		17		20		5.5		6.5		1.8		3.5		0.85		93		25	
LDC1000	D	400	5.3	36	63	190	6.8	17	69	95	7.9	8.55	72	79	33	7.1	91	760	6.3	69	68
	R	76		21		28		7.7		12		3.3		2.4		0.66		120		33	
LDC1500	D	80	6.7	7.2	69	33	9.2	3.0	75	20	13	1.8	81	13	22	1.8	92	150	8.3	13	73
	R	12		3.3		3.6		0.99		1.5		0.41		0.60		0.165		18		4.9	
SOS1000	D	220	12	20	80	530	79	48	96	42	14	3.8	82	20	12	1.8	80	810	27	73	90
	R	18		4.9		6.7		1.8		3.0		0.82		1.7		0.46		30		8.2	
SOS1900	D	31	2.6	2.8	46	17	4.2	1.5	58	8.9	6.8	0.80	69	6.3	9.0	0.57	75	63	3.5	5.7	54
	R	12		3.3		4.0		1.1		1.3		0.36		0.70		0.19		18		4.9	

499 ^aInventories during DSWC and common sedimentation were calculated by multiplication of the average fluxes during these two periods by 90 and 275 days, respectively.

500 ^bD/R ratio of deposition fluxes.

501 ^c100·D/(R+D) ratio of inventories.

502 ^dOCs, sum of all measured organochlorine compounds.

503 **Figure captions**

504

505 **Figure 1.** Map of the Gulf of Lion (GoL) and location of the sediment trap mooring lines
506 along transects in Lacaze-Duthiers (LDC) and Cap de Creus (CCC) Canyons and in Southern
507 Open Slope (SOS) sites. The arrow highlights the dominant Northern Current (NC). Numbers
508 indicate the water column depths at each site. The traps were deployed at 30 m above sea
509 bottom. In LCD1000 another trap was installed at 500 m above sea bottom.

510

511 **Figure 2.** Deposition fluxes of organochlorine compounds (PCBs, DDTs, CBzs and Lindane;
512 $\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and total mass (TMF; $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) measured in sediment traps deployed in the Cap de
513 Creus (CCC) and Lacaze-Duthiers (LDC) canyons and in Southern Open Slope (SOS) sites
514 (Fig. 1). See Fig. 1 for details.

515

516 **Figure 3.** Correlations between fluxes of total organic carbon (TOC; $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and
517 organochlorine compounds ($\text{ng}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) in the particles collected with the sediment traps
518 deployed in the Cap de Creus and Lacaze-Duthiers canyons and in Southern Open Slope sites
519 (Fig. 1).

520

521 **Figure 4.** Inventories ($\mu\text{g}\cdot\text{m}^{-2}$) of settling organochlorine compounds (sum of PCBs, DDTs,
522 CBzs and lindane) during the DSWC episode (January-March 2003; in red) and during the
523 period of common sedimentation (in orange). The acronyms refer to Cap de Creus (CCC) and
524 Lacaze-Duthiers (LDC) canyons and Southern Open Slope (SOS) sites. Numbers besides the
525 acronyms refer to water column depths.

526