Biogeosciences Discuss., 6, 10737–10773, 2009
www.biogeosciences-discuss.net/6/10737/2009/

Biogeosciences Discussions

This discussion paper is/has been under review for the journal Biogeosciences (BG).
Please refer to the corresponding final paper in BG if available.

The impact of Saharan dust on the particulate export in the water column of the North Western Mediterranean Sea

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Received: 27 October 2009 – Accepted: 9 November 2009 – Published: 17 November 2009

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Simultaneous measurements of atmospheric deposition and of sinking particles at 200 m depth, were performed in the Ligurian Sea (North-Western Mediterranean) between 2003 and 2007 along with the historical time records of phytoplanktonic activity from satellite images. Atmospheric deposition of Saharan dust particles was very irregular and confirmed the importance of sporadic high magnitude events over the annual average (11.4 $gm^{-2}yr^{-1}$ for the 4 yr). The average marine total mass flux was 31 $gm^{-2}yr^{-1}$, the larger fraction being the lithogenic one (~37%). The marine total mass flux displayed a seasonal pattern with a maximum in winter occurring before the onset of the spring bloom. The highest POC fluxes did not occur during the spring bloom nor could they be related to any noticeable increase in the surface phytoplanktonic activity. Over the 4 yr of the study, the strongest POC fluxes were concomitant with large increases of the lithogenic marine flux, which had originated from either recent Saharan fallout events (February 2004, August 2005), or from "old" Saharan dust "stored" in the upper water column layer (March 2003, February 2005), or alternatively from lithogenic material originating from Ligurian riverine flooding (December 2003, Arno, Roya and Var rivers). These "lithogenic events" are believed to result from a combination of physical processes (winter mixing), the aggregation of faecal pellets resulting from zooplankton activity, and also organic-mineral aggregation inducing a ballast effect. However, such an event also occurred in August 2005 without any physical mixing, and was attributed to Saharan dust-induced biological enhancement. POC export provoked by the simultaneous occurrence of winter mixing and an extreme dust event (February 2004, 22 gm^{-2} deposition) was shown to represent 50% of the total annual POC export at 200 m in the water column that year, as compared to only ~25% for the bloom period. This indicates the importance of atmospheric deposition for POC fluxes in the Mediterranean Sea, which is an area strongly influenced by Saharan dust inputs.

BGD

6, 10737–10773, 2009

1 Introduction

The Mediterranean Sea is a semi-enclosed basin receiving one of the highest rates of aeolian material deposition in the world (Guerzoni et al., 1999). It receives mineral dust from the Saharan desert in the form of strong pulses, and also continuous anthropogenic aerosol inputs from industrial and domestic activities on both sides of the basin. It also receives lithogenic material from coastal margins and rivers which contribute to the overall pool of lithogenic particles of the water column.

Recent studies have shown the importance of lithogenic particles in the export of the organic matter through the aggregation process (Armstrong et al., 2002; François et al., 2002; Hamm, 2002). Nevertheless, the physical role of large sporadic inputs of mineral dust during extreme Saharan dust events still remains poorly documented.

The Mediterranean Sea is characterized by strong stratification of the upper water column during at least five months of the year during the summer period, during which time the atmosphere then becomes the main external source of nutrients for the mixed layer (see Guieu et al., 2009). By influencing the marine nutrient cycle and the nutrient budgets of nitrogen, phosphorus, iron (Loÿe-Pilot et al., 1990; Markaki et al., 2003; Krom et al., 2004; Bonnet and Guieu, 2006), these atmospheric inputs can impact on the heterotrophic (Thingstad et al., 1998; Pulido-Villena et al., 2008) and autotrophic production (Klein et al., 1997; Kouvarakis et al., 2001; Bonnet et al., 2005; Guieu et al., 2009) of the Mediterranean Sea. The biological production depends in part on the atmospheric inputs, and so therefore the marine particulate flux should also be indirectly linked to atmospheric deposition via its dependence on the biological production, this is in addition to its more direct link via the sedimentation of insoluble atmospheric particles.

The proximity and the diversity of these aerosol sources, as well as the biogeochemistry of the surface layer, make the Mediterranean Sea an excellent natural laboratory to study the transfer of atmospheric lithogenic material and the potential role in the export of carbon. This 4-yr time-series of both atmospheric and marine fluxes allows

BGD
6, 10737–10773, 2009

us to investigate (i) the effect of the atmospheric lithogenic deposition on the intensity and composition of the marine flux, (ii) the role of lithogenic particles in the transfer of organic matter from the surface to the deeper layers, and (iii) the response of the biota to atmospheric inputs, and the associated marine flux occurring during the stratified period.

2 Materials and methods

2.1 Atmospheric sampling

Atmospheric deposition samples were collected at coastal sampling sites located on both sides of the Ligurian Sea. Data for insoluble particulate atmospheric deposition were obtained in two different ways: (i) between 2004 and 2006 the data published in respectively Bonnet and Guieu (2006) and Pulido-Villena et al. (2008) for bulk deposition at Cap Ferrat were used (see details in Table 1 and Fig. 1); (ii) for 2003 and 2005, bulk atmospheric deposition samples were collected in three Corsican sites: Ostriconi, Ile Rousse, and Ponte Leccia, on a weekly basis (see details in Table 1). The insoluble fraction of atmospheric deposition was collected on 0.4 μm pore size and 47 mm diameter polycarbonate filters and determined by weighing; the contribution of Saharan dust was estimated according to Loÿe-Pilot and Martin, 1996. The Saharan dust fallout is very irregular (Loÿe-Pilot and Martin, 1996), and varies from 50 mg m^{-2} to 22 g m^{-2} from different events during the study period. Atmospheric sampling sites were considered as representative of the atmospheric deposition actually depositing in the central Ligurian Sea (Fig. 1).

2.2 Marine sampling

Since 1988, marine settling particulates have been collected at the DYFAMED site (43°25′N 07°52′E) with 2 multisampling cylindrical sediment traps (Technicap

PPS 5, height of 2.3 m-collection surface of 1 m^2) moored at 200 m and 1000 m. This time-series site (http://www.obs-vlfr.fr/sodyf/) is located in an open ocean area (depth=2330 m), 50 km offshore from the French coast and 130 km from Corsica. Only the data from the upper trap (200 m) was considered for the observation of the export of the particulate material from the surface layer.

The 24 trap collector cups were filled with a solution of 2% buffered formaldehyde in filtered seawater in order to prevent in situ microbial degradation and grazing by swimmers. Traps were set to sample sinking particles for consecutive periods ranging between 7 and 15 days. After retrieval of the traps (operated by IAEA up to November 2006, and then by LOV laboratory (Service National "Cellule Piège" INSU), the samples were stored in the dark at 4 °C, until processed. Swimmers were identified and carefully removed, first by sieving through 1500 and 160 μm mesh and then by handpicking the remainder under a binocular microscope. Any visible material attached to the swimmers was carefully removed as best as possible, to avoid biasing the particulate flux result. The remaining sample was desalted with ultrapure water and freeze-dried for future analyses. Mass flux was measured by weighing the freeze-dried sample five times with the accuracy of the weighing being about 1% over the whole data series (Miquel et al., 1994).

2.2.1 Particulate matter analysis

Carbon and nitrogen: For the samples treated by IAEA, total carbon and nitrogen were analysed in duplicates using a VarioEL microanalyser (CHN) elemental analyser on 2–4 mg aliquots of the desiccated samples (Miquel et al., 1994). For samples treated at LOV, the concentration of total carbon and nitrogen was analysed in triplicate with a Perkin Elmer 2400 elemental analyser on 3–4 mg sub-samples of the desiccated samples, all according to Guieu et al., 2005.

Aluminium and calcium were analyzed by ICP-AES Jobin Yvon (JY 138 "Ultrace"), on acid digested samples. The detection limit of the apparatus, defined as 3 times the

standard deviation of 10 measurements of the blank (9 ppb for aluminium and 4 ppb for calcium), was well below the lowest concentration of the digested aliquot. The acid digestion was performed in 7 ml Teflon flasks inside an oven (150°C) and suprapur acids were added to 20 mg of sample, following two steps (1 mL 65% HNO_3, followed by 500 μL 65% HNO_3+500 μL of 40% HF). After each treatment, samples were oven heated at 150 °C for 5 h (see Journel, 1998). Eight aliquots of both blanks and certified reference material (GBW07313: marine sediment from National Research Centre for Certified Reference Materials of China) were digested and analyzed in the same conditions. Results were consistent for the blanks (9 ppb for aluminium and 4 ppb for calcium), and also for the reference material (recovery being 95±1% for aluminium and 96±2% for calcium, n=8).

2.2.2 The composition of the marine particulate matter

The elemental analyses were used to calculate the 4 main fractions of the collected material: carbonates, organic matter, lithogenic material and opal.

Carbonates. For samples treated by IAEA, the organic carbon was measured after removal of carbonates (1 M phosphoric acid), by CHN analysis as described above. The inorganic carbon was then estimated by subtraction ($C_{inorg}=C_{tot}-C_{org}$) and the carbonate fraction was calculated as $CaCO_3$=PIC×8.33. For samples treated at the LOV laboratory the carbonate fraction was determined from particulate Calcium concentration measured by ICP-AES ($CaCO_3$=2.5×Ca). Particulate inorganic carbon was then deduced from the carbonate fraction (PIC=0.12×$CaCO_3$).

Organic matter. For samples treated at the IAEA, the particulate organic carbon (POC) was analysed by CHN elemental analyser, as mentioned above. Whereas for those treated at the LOV, the POC was calculated by subtracting the inorganic particulate carbon from the total carbon. The organic matter fraction (OM) was calculated as 2×(%POC).

The lithogenic fraction was estimated from the particulate aluminium concentrations assuming that lithogenic particles have, on average, Aluminium concentrations typical

BGD
6, 10737–10773, 2009

of the upper continental crust (Al=7.74, Wedepohl 1995), but also they can be more influenced by their Saharan origins (Al=7.1%, Guieu et al., 2002), or can have Al concentrations typical of riverine particles (Al=9.4%, Martin and Windom, 1991).

The opal fraction was calculated as the difference between the total mass, and the sum of the other components ($CaCO_3$+OM+lithogenic material). Here again, a range is given, resulting from the range of the lithogenic fraction.

2.3 Additional in situ and satellite data

Daily surface chlorophyll concentrations estimated from the NASA sensor MODIS, using reprocessing #4, (carried by AQUA), were spatially averaged by considering all the data from a square of 10 km^2 (around 9 pixels) surrounding the DYFAMED site. Despite the missing data due to clouds, the time series, from March 2003 to end of 2007, reach a temporal cover of up to 60% during that period.

Salinity and water temperature, measured routinely by CTD casts between 0 and 200 m depth during the monthly DYFAMED surveys (43°25′N, 7°52′E, http://www.obs-vlfr.fr/sodyf/), and BOUSSOLE cruises (43°22′N, 7°54′E, http://www.obs-vlfr.fr/Boussole/), were used to estimate the mixed layer depth (data not shown). During these cruises, biological measurements such as pigment concentrations were also determined and the data were used to help interpret our results. Physical data such as air and sea surface temperature, sea surface salinity, as well as wind speed and direction were continuously recorded by the Météo France Buoy (43°38′N, 7°83′E, http://www.meteo.shom.fr/real-time/html/dyfamed.html). As a high temporal resolution of the biological activity was required to examine the potential response of biota to atmospheric inputs, ocean colour data from satellite derived sources was also used.

French riverine data (Var and Roya) were obtained from the free access website of the French governmental hydrological office (http://www.hydro.eaufrance.fr/). The Italian river data (Arno) were obtained from the Servizio Idrologico Regionale – Centro Funzionale – of the Regione Toscana (Fiorini, personal communication).

BGD
6, 10737–10773, 2009

3 Results and discussion

3.1 Atmospheric flux

During the 4 years of this study, Saharan dust events covered a huge range of frequency and intensity. Very low (0.05 $g m^{-2}$) dust flux events occurred relatively often (27 events in 4 years) but there was only one extreme event (22 $g m^{-2}$) (Fig. 2). That particular Saharan event represented almost 90% of the Saharan input reported for the whole of 2004 at the Cap Ferrat site. The importance of a single event as part of the annual flux is consistent with previous observations by Loÿe-Pilot and Martin, 1996. The monthly dust deposition during the study period shows a strong inter-annual variability (Fig. 3). The average annual dust flux over the 4 yr (11.4 $g m^{-2} y^{-1}$) is very similar to the one observed in Corsica for the period 1984–1994 (12.5 $g m^{-2} y^{-1}$, Loÿe-Pilot and Martin, 1996). Most of the dust deposition occurred as wet deposition (Bonnet and Guieu, 2006; Pulido-Villena et al., 2008). This is consistent with Loÿe-Pilot and Martin, (1996) who showed that more than 95% of the Saharan events in Corsica are associated to wet deposition.

3.2 Hydrological and biological features of the study site

The DYFAMED site which has been monitored since 1991 is well described, and is characterized by important seasonal variations in both hydrological and biological features (Marty et al., 2002). The hydrological seasonality is characterized by a vertical mixing period that generally occurs from late December until late March (defined as winter mixing), and a period of strong thermal stratification usually from June to October, which is defined as summer stratification. The maximum winter mixing generally occurs in February and is to about 150–200 m deep. The biological activity within the water column is driven by the hydrology, where the winter mixing brings nutrients into the surface layers, with a spring bloom generally initiated in early March, a peak in April and ending in late May (Marty et al., 2002; Bosc et al., 2004). The beginning of the

BGD
6, 10737–10773, 2009

bloom is dominated by large and opportunistic phytoplanktonic cells (diatoms), followed later by smaller cell species (nanoflagellates; prymnesiophytes). Later in the season, during the stratification period, the biomass declines and is dominated by smaller picoplankton. It is also then found at greater depth (30–50 m), and is in fact following the establishment of a deeper nutricline layer (Marty et al., 2002).

The evolution of the hydrology during this study was generally consistent with the pattern described in Marty et al. (2002). However, the years 2004, 2005 and especially 2006 were characterized by strong mixing events with mixing depths reaching 400, 700 and 2300 m, respectively. Contrary to these, the years 2003 and 2007 were characterized by weak mixing (~100 to 150 m depth) that began in late January. As shown on Fig. 4a, the evolution of the biological features over this 4-yr study is consistent with the pattern described by Marty et al. (2002), Bonnet and Guieu (2006) and Bosc et al. (2004). According to pigment data the succession of phytoplankton species was the same as previously described by these authors. Nevertheless, over the period, the intra-annual pattern and maximum intensity of the surface chlorophyll a concentration showed an important inter-annual variability (Fig. 4a).

3.3 Marine flux

3.3.1 Marine total mass flux

From 2003 to 2007, the total mass flux (TMF) was 86 $mg m^{-2} d^{-1}$ on average, and presented a strong temporal variability over the 4 years (5–1228 $mg m^{-2} d^{-1}$, Fig. 4b). If the DYFAMED site is oligotrophic most of the year, the total mass flux is higher than those reported for other open-ocean oligotrophic sites located far from continental influence, for example 66 $mg m^{-2} d^{-1}$ at Station ALOHA (Karl et al., 1996), and 26 $mg m^{-2} d^{-1}$ at BATS (Conte et al., 2001). Table 2 compares the data obtained at DYFAMED during this study with data from other Mediterranean sites and also from oceanic oligotrophic sites subject to noticeable atmospheric inputs (EUMELIE, BATS). However, the comparison between all these studies remains difficult as the sampling depths and du-

BGD
6, 10737–10773, 2009

ration were highly variable, with some studies not covering the key periods such as the spring bloom, the stratification period or the winter mixing. As compared to studies covering at least one year, our data are consistent with the values reported for similar depths, at different Mediterranean sites: 111 $mgm^{-2}d^{-1}$ for the same site during the year 1987 (Miquel et al., 1994), 106 $mgm^{-2}d^{-1}$ for the Adriatic (Boldrin et al., 2002) and 96 $mgm^{-2}d^{-1}$, for the Algero-Provencal Basin (Zuniga et al., 2007). Some other Mediterranean sites characterized by different hydrological and biological features, present either higher or lower total mass flux values: 300 and 647 $mgm^{-2}d^{-1}$ for respectively the Gulf of Lions and the Alboran Sea (Monaco et al., 1999; Sanchez-Vidal et al., 2005) and 35 $mgm^{-2}d^{-1}$ for the Ionian Sea (Boldrin et al., 2002).

Lee et al. (2009) measured the total mass flux in the same area and depth during the spring bloom, from March to May in 2003 and 2005, but with a different sampling device (sampling surface of 0.0184 m^2 vs. 1 m^2 in this study) and a different sampling scheme (5 d vs. 14 d in this study). In the situation of the low fluxes (most of the time), the total mass flux was similar in both studies. However, in the situation with high fluxes (March 2003), the total mass flux measured by Lee et al. (2009) was twice that of this study (~800 and 430 $mgm^{-2}d^{-1}$ respectively). However, this difference could be due to the different sampling methodologies; Lee et al. (2009) mentioned high analytical errors (50–75%) due to the very low sample size.

Over the 4 years of this study, the marine total mass flux showed a distinct seasonal pattern (Fig. 4b) with the highest fluxes in winter (303±426 $mgm^{-2}d^{-1}$, February) and lowest fluxes in summer (16±10 $mgm^{-2}d^{-1}$, July). Such a pattern is consistent with the one previously described for the same site by Miquel et al. (1994). Fluxes are also high during the spring bloom period (72±121 $mgm^{-2}d^{-1}$, April). It is noteworthy that the maximum of the total mass flux occurs before the peak of the bloom. This is still true if we do not take into account the particular event of February 2004. The high values of the standard deviation for some months denote significant inter-annual variations of this pattern (i.e. February with a standard deviation >100%, Fig. 4b). However, the fluxes recorded during the year 2006 seem abnormal with very low fluxes in late winter and

BGD
6, 10737–10773, 2009

moderate fluxes during the spring bloom. The year 2006 was characterized by a very strong deep winter mixing (over the whole water column to 2300 m), and also coupled with strong currents at 200 m depth, which were >15 cm/s, and up to 30 cm/s at times (Service d'Observation, Observatoire Océanologique de Villefranche-sur-Mer).

3.3.2 Composition of the marine flux

Average Composition. The relative average contributions of the major constituents (carbonates, organic matter, lithogenic material and opal) to the marine particulate flux are shown in Table 2. During the study, the marine particulate matter was composed of 27±8% of $CaCO_3$, 25±12% of OM, 37±19% of lithogenic material, and 13±12% of opal. A similar composition has already been reported for the Ligurian Sea at the same depth for the years 1986–1987 (Quétel et al., 1993; Miquel et al., 1994). The carbonate fraction could include a fraction of Saharan origin, as Saharan dusts may contain up to 20% $CaCO_3$ (Guieu et al., 2002). A high lithogenic contribution to the annual total mass flux has also been reported for other Mediterranean sites: 39% for both the Adriatic and Ionian seas (Boldrin et al., 2002) and 51% for the Algero-Balearic basin (Zuniga et al., 2007). The remaining part of the flux in our study (~60%) represented by the sum of the biogenic fluxes (carbonates+OM+opal) is quite similar to values reported throughout the Mediterranean basin: 53% (Boldrin et al., 2002), 52% (Zuniga et al., 2007), 55% (Stavrakakis et al., 2000).

Variability of the marine flux composition. The seasonal pattern observed for the total mass flux is also valid for both lithogenic and biogenic fractions. The highest lithogenic fluxes occurred during the winter mixing period (265±312 $mg m^{-2} d^{-1}$ in February over the 4-yr period) and the lowest during summer (5±5 $mg m^{-2} d^{-1}$ in July).

For high mass fluxes (winter mixing), the lithogenic fraction is the dominant component of the total mass flux, whereas for low mass fluxes (stratification period), the biogenic fraction dominates, as already mentioned by Migon et al. (2002). This indicates that in winter the biological activity is not the main parameter controlling the total mass flux composition and intensity.

BGD
6, 10737–10773, 2009

Reflecting the species succession during the spring bloom, the biogenic flux was mostly formed by opal at the beginning of the bloom (diatoms bloom in February–March) and then by carbonates (prymnesiophytes bloom in April–May). The dominant group of phytoplankton is the prymnesiophytes in the North Western Mediterranean Sea and this predominance is independent of the season (Marty et al., 2002). Thus, the carbonate fraction dominates the biogenic flux most of the time, and its contribution presents generally very little variation throughout the year. The highest fluxes of $CaCO_3$ and OM were reached in February 2004 with respective values of 295 and 71 $mg m^{-2} d^{-1}$. In contrast, lowest values for carbonates were observed in summer 2004 (0.8 $mg m^{-2} d^{-1}$) and for OM in summer 2006 (2 $mg m^{-2} d^{-1}$). On average, the highest biogenic fluxes occurred before the peak of the bloom (Fig. 5), at the end of the winter mixing (122±144 $mg m^{-2} d^{-1}$ in February) and the lowest during the stratification period (11±6 $mg m^{-2} d^{-1}$ in July). Only the year 2006 exhibited its highest biogenic fluxes during the spring bloom. The mismatch between the bloom occurrence and the vertical transport of biogenic particles, already observed at the DYFAMED site (Miquel et al., 1994; Migon et al., 2002), implies that the spring bloom is not the only factor driving the biogenic flux.

3.4 Comparison of the atmospheric and marine lithogenic fluxes

It has been suggested that atmospheric deposition of mineral particles from the Saharan source may be the major source of terrigenous sediments in the offshore Mediterranean Sea (Loÿe-Pilot et al., 1986; Bergametti et al., 1989; Tomadin and Lenaz 1989). The contribution of Saharan dust to lithogenic particles trapped in the water column at the DYFAMED site has already been documented (Buat-Menard et al., 1989; Quétel et al., 1993; Journel, 1998; Migon et al., 2002; Lee et al., 2009).

The comparison of atmospheric and marine lithogenic fluxes from 2003 to 2007 shows that they are in general not synchronous (Fig. 2), that the export of lithogenic material from the upper water column displays a strong seasonal signal with the maximum occurring in late winter/early spring (Fig. 2) and that also a "*t-test*" performed to com-

BGD
6, 10737–10773, 2009

pare atmospheric (31 mgm^{-2}d^{-1}) and marine (40–53 mgm^{-2}d^{-1}) annual lithogenic fluxes showed that the difference between both of these fluxes is not significantly different (P>0.05).

In order to better understand how the Saharan dust does impact the water column particulate export, three contrasting cases will be described in the following sections: (i) three strong marine lithogenic fluxes which were not related to any Saharan events (March and December 2003, and February 2005), (ii) two Saharan dust events (February 2004 and August 2005) with rapid transfer of material to the 200 m depth, and (iii) one Saharan event (June 2006) where no mineral dust was found in the traps.

3.4.1 Winter high marine lithogenic events

February–March 2003 and February 2005: late winter events (Fig. 2). Winter mixing constitutes a period of high marine mass fluxes in this area (Miquel et al., 1994; Quétel et al., 1993; Migon et al., 2002), and is confirmed by mass fluxes from February–March 2003 (430 mgm^{-2}d^{-1}) and February 2005 (451 mgm^{-2}d^{-1}). Mass fluxes were mainly formed by lithogenic material (respectively 40±5% and 80±11%) which was not from an identified Saharan event (Fig. 2). Atmospheric mineral particles deposited during the previous months are reported to remain stored in the surface layers of the water column during the stratification period (Migon et al., 2002), along with dissolved organic carbon and small biogenic particles. These could then be rapidly exported when the stratification breaks down and winter mixing starts. The reasons why these mineral particles are exported during the winter mixing is still not fully understood. However, three mechanisms favouring the quick export of the particles from the surface layer to the deeper layers can be proposed: i) the vertical convection processes during winter which homogenise the superficial part of the water column (0–250 m; Marty et al., 2002), ii) the incorporation of mineral particles into faecal pellets (as shown by Fowler et al., 1987; Buat-Menard et al., 1989 – biological aggregation; Burd and Jackson, 2009) which were shown to greatly contribute to the mass flux in that period (Miquel

BGD
6, 10737–10773, 2009

et al., 1994), and iii) the incorporation of mineral particles acting as ballast in organic aggregates formed by either "old" organic matter stored in the water column (Marty et al., 1994) or organic colloids such as TEP (transparent exocellular polysaccharides; Alldredge et al., 1993) generated by the of emerging diatom blooms (physical aggregation; Burd and Jackson, 2009).

It is also possible that dissolved aluminium be scavenged by siliceous organisms (Mackenzie et al., 1978; Hydes, 1979), increasing the particulate aluminium flux during the period of early diatoms blooms, as a result, the lithogenic flux derived from particulate aluminium would have also included any dissolved aluminium (also from eolian origin – Measures, 1995) stored, like Fe (Bonnet and Guieu, 2006), in the mixed layer during the stratification period.

The strong export of lithogenic particulate matter during the first part of the year (February–March) is attributed to the presence of "old" Saharan dust stored in the surface layer and the simultaneous occurrence of a physical process (winter mixing), the grazing of phytoplankton producing faecal pellets, and the presence of organic matter.

December 2003-January 2004: early winter lithogenic event. In December 2003, a strong export of lithogenic particles ($478 \pm 67\,mg\,m^{-2}\,d^{-1}$; Fig. 2) occurred at the very beginning of the winter mixing period (mixed layer between 20 and 90 m deep) whereas no Saharan dust deposition event was recorded at that time. The DYFAMED site is believed to be most of the time a 1-D site, presenting thus a weak advection (Andersen and Prieur, 2000). However, in the case of very strong floods from the Ligurian rivers, the DYFAMED site may be influenced by their inputs (Béthoux and Prieur, 1983; Stemmann, 1998; Stemmann et al., 2002), and this has also been observed on the ocean floor (Guidi-Guilvard, 2002).

Data from the three major Ligurian rivers show indeed severe floods on the 1 November and the 3 December 2003, while the high marine lithogenic flux was measured from the 21st of December until the end of January 2004 (Fig. 6). If the high lithogenic flux recorded at DYFAMED is linked to inputs from those rivers, it means that the transit

BGD
6, 10737–10773, 2009

time necessary for a riverine particle to reach the DYFAMED site is at least 3 weeks. This time is consistent with the observation made by Béthoux and Prieur (1983) who showed that strong freshwater inputs in Italy can be transported by the Ligurian current within 3 weeks to as far as 30 nautical miles away from the French coast. Those findings were confirmed by Stemmann et al., (2002), who showed that according to the Ligurian current structure, part of these riverine inputs could reach the DYFAMED area. This means that in winter 2003 a significant proportion of marine lithogenic matter at the DYFAMED site originated from riverine inputs and that the advection of particulate matter from the coastal margins could explain that, in some years, the marine lithogenic flux can be higher than Saharan inputs alone.

3.4.2 The cases of marine lithogenic fluxes following Saharan dust events

During the 4 years time series, 2 specific events illustrate well how a rapid transfer of Saharan particles down to a depth of 200m can occur, firstly during the mixing period in February 2004 and secondly during the stratification period in August 2005 (Fig. 2).

An extreme Saharan event in February 2004. While the winter mixing process was high (data not shown), an extreme Saharan dust input (22.2 $g m^{-2}$) occurred in the early hours of 20 February 2004. The strong and rapid marine lithogenic export that followed the event was attributed to both the winter mixing and the Saharan dust fallout. Taking into account that the atmospheric deposition was not precisely measured at the DYFAMED site, the amount of lithogenic particles retrieved in the traps within the following month (23 g) was of the same order of magnitude as the Saharan deposition (22.2 $g m^{-2}$). The marine lithogenic flux in February 2004 was the highest flux ever reported at the DYFAMED site (Buat-Menard et al., 1989; Quétel et al., 1993; Migon et al., 2002; Lee et al., 2009, this study), and the event may have favoured the removal of the older lithogenic particles accumulated in the surface layer during the stratification period as evidenced in late winter 2003 and 2005 (see Sect. 3.4.1 above), and in 1998 (Migon et al., 2002). Because of the extremely high flux due to this atmospheric input, the part due to "older" lithogenic material is not quantifiable.

BGD
6, 10737–10773, 2009

Several studies performed at the same site (Fowler et al., 1987; Buat-Menard et al., 1989) showed that aluminosilicate minerals are easily incorporated into faecal pellets or trapped in organic aggregates, thus enabling the possibility of a rapid export of lithogenic and organic material to the deeper layers and implying that lithogenic particles do not sink following Stokesian settling calculations. Indeed, Saharan particles were also directly transferred to 1000 m (with a minimal settling velocity estimated for 35% of this lithogenic material at 100 $m d^{-1}$, (data not shown)), confirming very high sedimentation rates during those sporadic large dust events. At the same site, Armstrong et al., (2009) measured a settling velocity with an average of 353±76 $m d^{-1}$ for the fast sinking particles. Thus, it is not surprising that more than half of the amount of the Saharan dust input was retrieved within less than 10 d at 200 m depth.

Again, a fraction of this lithogenic flux, attributed to the scavenging of dissolved aluminium by diatoms, should not be neglected.

Evidence of carbon and lithogenic material export linked to Saharan inputs during the stratification period. A series of Saharan events in July (18th) and in August (10th to 12th, and 18th) brought up to 2 $g m^{-2}$ of mineral particles into the water column. An increase of the marine lithogenic fluxes occurred during the following month (14 August–11 September) which was quite surprising as generally the marine summer fluxes are low, with the well established stratification limiting the downward transport of atmospheric material.

During that period, in the Mediterranean Sea, the atmosphere represents a significant forcing for the biogeochemical cycles in the surface mixed layer, and providing the nutrients able to sustain new production (see for example Guieu et al., 2009). Phosphorus is thought to be the limiting element in summer for both bacterioplankton and phytoplankton communities (Moutin et al., 2002; Bonnet et al., 2005; Pulido-Villena et al., 2008). Therefore, an input of "new" phosphorus released by Saharan dust to the P-depleted mixed layer would enhance the bacterial and the phytoplankton activity. These Saharan dust fallouts were accompanied by moderate winds (13 knots), but these were not sufficiently strong to disrupt the stratification and induce any in-

BGD
6, 10737–10773, 2009

jection of new nutrients from below the thermocline. The dust fallout of the 11–12 August was also accompanied by rain, and the end of the event was typically a "mixed" one. In "mixed" rains (Loÿe-Pilot and Morelli, 1988; Loÿe-Pilot et al., 1990), anthropogenic aerosols are scavenged with Saharan dust, and their high inorganic nitrogen content and acidic/complexing components increase the dissolution of the inorganic phosphate of the Saharan particles to provide a notable input of bioavailable nutrients. Within 14 d after the dust event, an increase of biogenic flux was recorded, (POC: 140 $mg m^{-2}$ (Fig. 5) and carbonate: 570 $mg m^{-2}$). The exported POC could have resulted from the surface new primary production induced by the Saharan dust event. This increase by a factor of ~3 can be compared to the 2-fold increase of POC export after an Asian dust input in the North Pacific (Bishop et al., 2002). The authors linked the observed POC flux increase to a dust-induced fertilisation of the surface layer of this HNLC zone.

Estimating that all the POC exported was formed by surface new primary production, the POC export and chlorophyll a concentration would thus respectively be 0.14 $g C m^{-2}$ and 0.2 $\mu g L^{-1}$ (based on a C/chl-*a* ratio of 50; Guieu et al., 2009). The calculated chlorophyll-*a* concentration would then represent 100% of the surface chlorophyll-*a* concentration observed at that time (0.2 $\mu g L^{-1}$ – MODIS data, Fig. 5). Although no increase in chlorophyll a concentration was observed by satellites, opal flux increased, after the event (data not shown), indicating a diatom growth. By using the N:C and the P:C ratios of Redfield (16:106 and 1:106), the calculated new primary production would have been supplied by an atmospheric input of 294 $\mu M m^{-2}$ of nitrogen and an input of 18 $\mu M m^{-2}$ of phosphorus, with these values being consistent with those reported for "mixed rain" samples in other Mediterranean studies (Loÿe-Pilot et al., 1990; Herut et al., 1999; Migon and Sandroni, 1999).

This mixed event could have supplied enough phosphorus and nitrogen to the depleted surface layer to enhance new primary production and thus lead to the export of particulate organic carbon. The fact that this dust-induced biological enhancement was not visible by satellite could be due to the inefficiency of satellites in detecting

small chlorophyll a variations, like those induced by Saharan events, but in contrast with Volpe et al. (2009) concerning the lack of marine biological response to dust fall-out in the Mediterranean.

3.4.3 The case of significant Saharan dust event with no induced lithogenic marine flux: June 2006

In June 2006, during the stratification period (mixed layer ~6 m) a Saharan event brought 2.5 g m^{-2} of dust into the area. Despite evidence that this dust deposition event played a fertilising role to the bacterial community in the surface mixed layer (Pulido-Villena et al., 2008), neither any increase of lithogenic (see Fig. 2), nor biogenic fluxes were noticed at 200 m during the following weeks. In order to explain this "non-export", three hypothesis are proposed: (i) that due to the strong stratification of the water column and their small size, mineral particles are suspected to remain in the surface layer during the summer months when there is little perturbation of the system (Migon et al., 2002), (ii) the low biological activity may also prevent any flux transfer via faecal pellet and/or export of organic aggregates (Buat-Ménard et al., 1989), or finally (iii) it is possible that on occasions the Ligurian Current could flow through the DYFAMED area (evidence from SST data: L. Prieur, personal communication) with the consequence that the particles reaching the surface waters may actually be exported away from the study site by lateral advection before reaching the 200 m sampling depth.

3.5 Lithogenic particles and organic carbon export

Data from this study shows that for the 4-yr time series, stronger POC fluxes were related to "lithogenic events": in February–March 2003, December 2003, February 2004, February 2005 and August 2005 (Fig. 7). These "lithogenic events" were observed in contrasted hydrological situations (winter mixing and in the stratification period, see Sects. 3.4.1 and 2), meaning that hydrodynamic factors are not the only factor responsible for such export. Biological and physical aggregation processes and mineral

BGD
6, 10737–10773, 2009

ballasting also play a major role and can occur at any period of the year with different hydrological configurations.

Recently, Armstrong et al. (2002), Francois et al. (2002), Klaas and Archer (2002) and Hamm (2002), proposed that ballast minerals (opal, carbonates and dust) were able to scavenge organic matter, affecting the POC flux sinking velocity and its remineralisation rate. The mineral matrix would first provide a protection for organic matter, and this would then act as a "glue" to bind particles together. A recent assumption of Passow (2004) suggested that rather than lithogenic particles, organic matter would be driving the aggregation process, where sinking POC would scavenge small lithogenic particles whilst they are in suspension in the water column. From those studies, it appears clear that whatever the aggregation catalyst is, the simultaneous presence (with a certain balance: Passow and de La Rocha, 2006) of organic matter and mineral particles is required to form aggregates. Additionally to those physical aggregations, several studies at the DYFAMED site have shown that zooplankton grazing and subsequent faecal pellet production may be the most efficient removal process of mineral particles from the surface waters (Fowler et al., 1987; Buat-Ménard et al., 1989).

"Lithogenic events" in this study suggest that Saharan dust and to a larger extent lithogenic particles from all origins, are able to form aggregates with organic matter in the surface layer, and thus participate to its large export flux, as already assumed by Lee et al. (2009). Those "lithogenic events" that occurred during our 4 yr study have been promoted by the simultaneous presence of organic matter ("old"– in winter (Marty et al., 1994) or freshly produced and induced by dust nutrient inputs in summer), and also by lithogenic material from "old" Saharan dust, that has been "stored" in the upper water column or originating from recent Saharan dust event or from coastal riverine flood events. Unfortunately, no zooplankton data are available for that period and we don't know to what extend zooplankton have mediated those events.

Two specific periods can be pointed out that can put the emphasis on the necessary simultaneous occurrence of both presence of organic matter and lithogenic particles with zooplankton activity to enable these "lithogenic events". Following the June

BGD
6, 10737–10773, 2009

2006 Saharan dust deposition there was no "lithogenic event" even though its intensity (2.5 mg m^{-2}) was of the same order of magnitude as the August 2005 Saharan event that was followed by a full export of lithogenic material to the sediment traps. Organic matter quality and low zooplankton activity may be responsible for that non-export. Indeed at the DYFAMED, the seasonal variation of the organic matter quality site was shown to impact the sinking flux composition and velocity (Bourguet et al., 2009; Wakeham et al., 2009) and low zooplanctonic abundance was measured at that time (Stemmann and Berline, personal communication). The second period where no "lithogenic event" occurred was during the winter mixing in 2006. A possible explanation could be the lack of "old stored" lithogenic particles in the water column as the late August 2005 Saharan event provoked an important export, effectively removing all the particulate material from the upper layer of the water column.

The particular extreme Saharan dust event of February 2004 occurred during the winter mixing period and so does not enable us to discriminate the processes driving the POC export, which could be from winter physical convection or provoked by the introduction of Saharan lithogenic particles. Usually, POC exported during winter mixing represents on average about 30% of the annual POC flux. This 2004 event produced an intense POC export (0.8 g C m^{-2}) at 200 m that represented 50% of the annual POC flux (2 g C m^{-2}) at 200 m, indicating that the Saharan event enhanced the fraction of POC exported.

It is noteworthy that the organic matter brought by a Saharan event would not significantly participate to the POC flux neither to the aggregates formation. Indeed, assuming that organic carbon in Saharan dust only represents 0.43%±0.02% of the particle mass (Ridame, 2001), an event of 22 g m^{-2} could only provide 94 mg m^{-2} of POC which is far below the marine POC fluxes registered by the traps after the event of February 2004 which was 807 mg m^{-2} within a month.

Saharan dust events play a significant role by providing lithogenic particles to the Mediterranean open-ocean area. Those lithogenic particles can be either simply transferred, or stored in the surface layer depending on the in situ hydrology, the water

column organic matter content (quantity and quality), and the zooplanktonic activity.

4 Conclusions

During our 4-year study, the Saharan dust inputs over the Ligurian Sea displayed a temporal pattern, and an annual mean flux was observed similar to previous findings in the same area (Loÿe-Pilot and Martin, 1996), highlighted by short events (like in February 2004) of high magnitude fluxes driving the inter- and intra-annual variability. The seasonal pattern of total marine mass flux also fitted the previous description at the same site (Miquel et al., 1994) with the highest fluxes in winter and the lowest in summer.

For the 4-yr averages, the lithogenic fraction represented ~37% of the total marine mass flux with the same seasonal trend as the total mass flux. Atmospheric deposition and lithogenic export were most of the time not synchronous, hence indicating that lithogenic export is not just controlled by direct atmospheric deposition.

Significant POC fluxes were associated to "lithogenic events" and occurred in several seasonal situations (winter mixing and stratification). From the data presented here, it appears clear that whatever the season, the simultaneous presence of lithogenic particles, organic matter and zooplanktonic activity were required for the occurrence of such "lithogenic events". However, if all parameters required are present and available, then the winter convection would enhance the export, generating the highest mass fluxes.

Thanks to programs such as MedFlux, recent advances have been made about the knowledge of the processes driving the POC export, and the relationship between mineral ballast and organic matter in the Mediterranean Sea. However, this study shows that the implication of parameters such as organic matter quality and zooplanktonic activity, in the control of the marine particulate lithogenic and organic carbon export, needs to be further studied.

Acknowledgement. The work presented here is part of the doctoral dissertation of E.T. (grant of the French Ministère de l'Education Nationale, de l'Enseignement Supérieur et de la

Recherche). We thank J-C Marty and L Coppola in charge of the DYFAMED program (funded by CNRS/INSU in the course of the French-JGOFS (now PROOF) program. The IAEA is grateful for the support provided to its Marine Environment Laboratories by the Government of the Principality of Monaco. We also thank the captain and crew of the Tethys II for their work at sea. NASA data acknowledgments: The authors thank the Ocean Biology Processing Group (Code 614.2) at the GSFC, Greenbelt, MD 20771, for the production and distribution of the ocean color data. We warmly thank M. Woodward who kindly checked the manuscript for scientific English.

The publication of this article is financed by CNRS-INSU.

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Table 1. Atmospheric sampling sites and sampling methodology.

Site	Latitude	Longitude	altitude (m)	Date	Sampling interval	Ground level	Type of Collector	Reference
Cap Ferrat	43°41′N	7°19′E	138	2004 2006	biweekly	3 m	4-liters polyethylen bottle+funnel 113 cm^2	Bonnet and Guieu, 2006 Pulido-Villena et al., 2008
Ile Rousse	42°38′N	8°55′E	135	2003 2005	weekly basis	1.2 m	Cylindric polyethylene collector	This study
Ostriconi	42°40′N	9°04′E	60	2003 2005	weekly basis	1.7 m	Bulk plastic collector (Météo France standard rain gauge)	This study
Ponte Leccia	42°29′N	9°12′E	200	2003 2005	event basis	1.7 m	Bulk plastic collector (Météo France standard rain gauge)	This study

BGD
6, 10737–10773, 2009

Table 2. Total mass fluxes, POC, $CaCO_3$, lithogenic and opal fluxes ($mg\,m^{-2}\,d^{-1}$) and composition of the total mass flux (percentage) for this study and data from the literature. Data which can be compared to our study are in bold (same depth and at least a one-year sampling).

Site name or campaign	Zone	depth (m) Water	Trap	Latitude	Longitude	Date Start	End	mass	Fluxes $mg\,m^{-2}\,d^{-1}$ POC	$CaCO_3$	Lithogenic	Opal	References
DYFAMED	Ligurian	2330	200	43°24′ N	7°52′ E	Mar 03	Feb 07	86 (5–1228)	11±12	22±42	50±128	8±20	This study
							% (std)	100	25 (12)	27 (8)	37 (19)	13 (12)	
DYFAMED	Ligurian	2330	200	43°24′ N	7°52′ E	**1986**	**1987**	**111**	**14**	**5**	**28***	**–**	**Miquel et al., 1994; Quétel et al., 1993***
						Oct 97	Apr 98	77	–	–	–	–	Migon et al., 2002
DYNAPROC	Ligurian		200			May 95		–	34	–	–	–	Goutx et al., 2000
						Jun 95		–	11	–	–	–	
MedFlux	Ligurian	2330	238	43°24′ N	7°52′ E	Mar 03	May 03	346	65	77	32	43	Lee et al., 2009
			117			May 03	Jun 03	46	25	27	4	5	
			313			Mar 05	Apr 05	426	35	104	94	107	
Other Mediterranean Sites													
ECORHONE	Lion Gulf	950	200	42°50′ N	4°49′ E	Jan 88	Jan 89	300	35	84	165	26	Monaco et al., 1999
MTP II-MATTER	Adriatic	1030	530	41°48′ N	17°19′ E	Nov 94	Oct 95	140	8	25	–	16	Estimated from Miserocchi et al., 1999
CINCS	**Cretean Sea**	**1550**	**200**	**35°44′ N**	**25°05′ E**	**Nov 94**	**Nov 95**	**50**	**3 (6%)**	**15 (31%)**	**23 (45%)**	**6 (13%)**	**Stavrakakis et al., 2000**
	Adriatic	**1200**	**150**	**41°49′ N**	**17°46′ E**	**Mar 97**	**Mar 99**	**106**	**10 (9%)**	**23 (22%)**	**41 (39%)**	**23 (22%)**	**Boldrin et al., 2002**
	Ionian	**2400**	**150**	**38°29′ N**	**17°59′ E**			**35**	**4 (11%)**	**10 (29%)**	**14 (40%)**	**3 (9%)**	
MTP II-MATTER	Alboran Sea	1337	396	36°01′ N	4°18′ W	Jul 97	May 98	802	47 (6%)	66 (8%)	613 (76%)	30 (4%)	Fabres et al., 2002
ALB 3-U	Alboran Sea	2260	680	36°40′ N	1°30′ W	Jul 97	May 98	323	14 (4%)	18 (6%)	204 (63%)	13 (4%)	Sanchez-Vidal et al., 2005
ALB 4-U		2240	645	36°13′ N	1° 33′ W			483	26 (5%)	18 (4%)	290 (60%)	31 (6%)	
ALB 5-U		2100	510	35°55′ N	1°30′ W			647	32 (5%)	18 (3%)	398 (62%)	37 (6%)	
	Algero Baleares	**2850**	**250**	**39°25′ N**	**06°04′ E**	**Apr 01**	**May 02**	**96**	**13 (13%)**	**24 (25%)**	**49 (43%)**	**6 (7%)**	**Zuniga et al., 2007**
Other Oligotrophic Sites													
ALOHA	**Tropical Pacific**	**4800**	**150**	**22°45′ N**	**158°′ W**	**Oct 88**	**Dec 93**	**66**	**–**	**–**	**–**	**–**	**Karl et al., 1996**
EUMELIE	**Tropical Atlantic**	**4600**	**1000**	**21°00′ N**	**31°00′ W**	**Feb 91**	**Nov 92**	**35**	**2**	**2**	**10**	**–**	**Bory et al., 2001**
BATS	**Sargasso Sea**	**4500**	**500**	**31°50′ N**	**64°10′ W**	**1989**	**1998**	**36**	**4**	**18**	**–**	**–**	**Conte et al., 2001**

*: reference corresponding to the lithogenic fraction data

BGD
6, 10737–10773, 2009

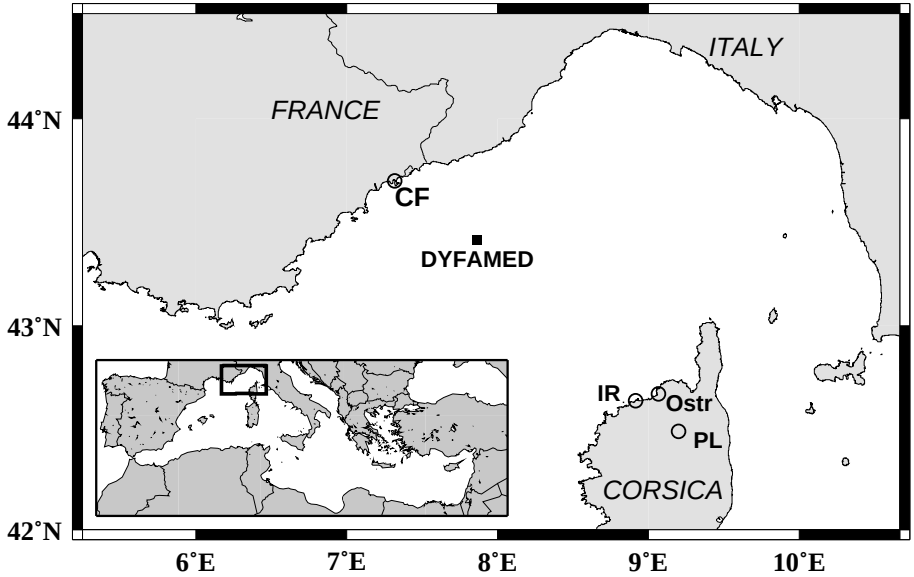

Fig. 1. Atmospheric and marine sampling sites. (Details on each site are available in Table 1; 0str.=Ostriconi; IR=Ile Rousse; PL=Ponte Leccia; CF=Cap Ferrat).

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6, 10737–10773, 2009

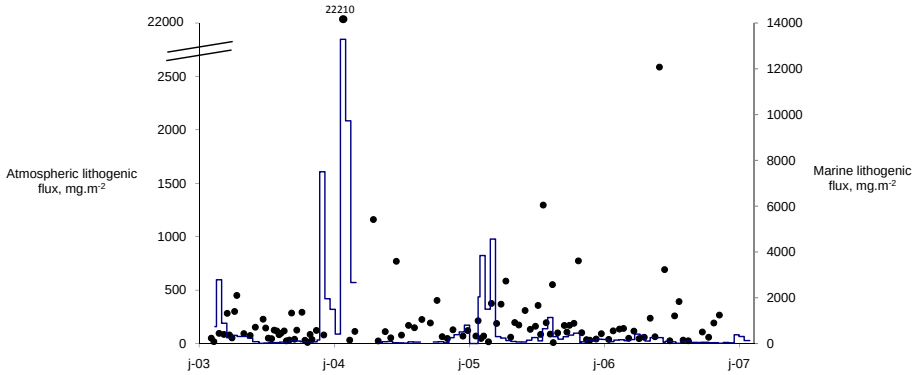

Fig. 2. 4 yr time series of simultaneous marine and atmospheric (Cap Ferrat and Corsica) lithogenic fluxes (200 m depth, Ligurian Sea: DYFAMED site).

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6, 10737–10773, 2009

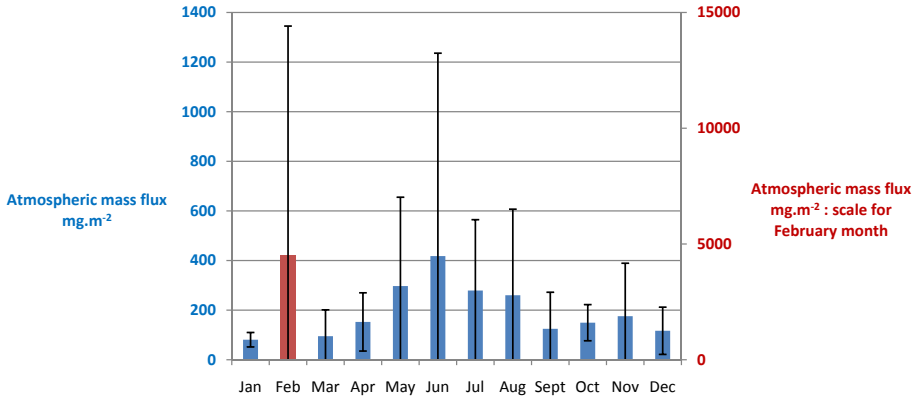

Fig. 3. Climatology of the atmospheric deposition of insoluble particles. (The scale for February 2004, on the right *Y* axis, is different in order to take into account the extreme Saharan event. The error bars represent the standard deviation of the mean monthly values).

BGD
6, 10737–10773, 2009

The impact of Saharan dust on the particulate export

E. Ternon et al.

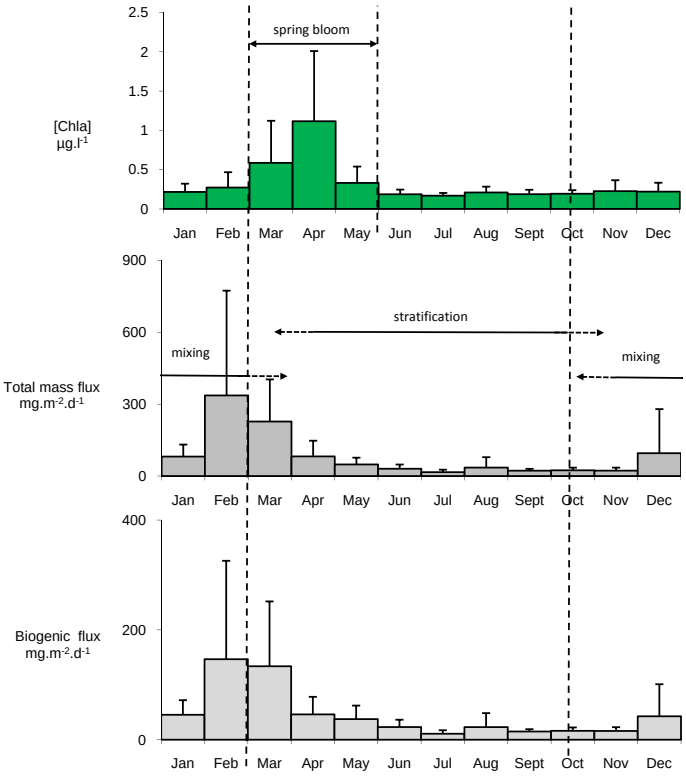

Fig. 4. Climatology of (i) above, the chlorophyll-*a* surface concentration measured by the MODIS satellite (upper figure), (ii) on the middle, the marine total mass flux at 200 m and (iii) last, the marine biogenic flux at 200 m.

BGD
6, 10737–10773, 2009

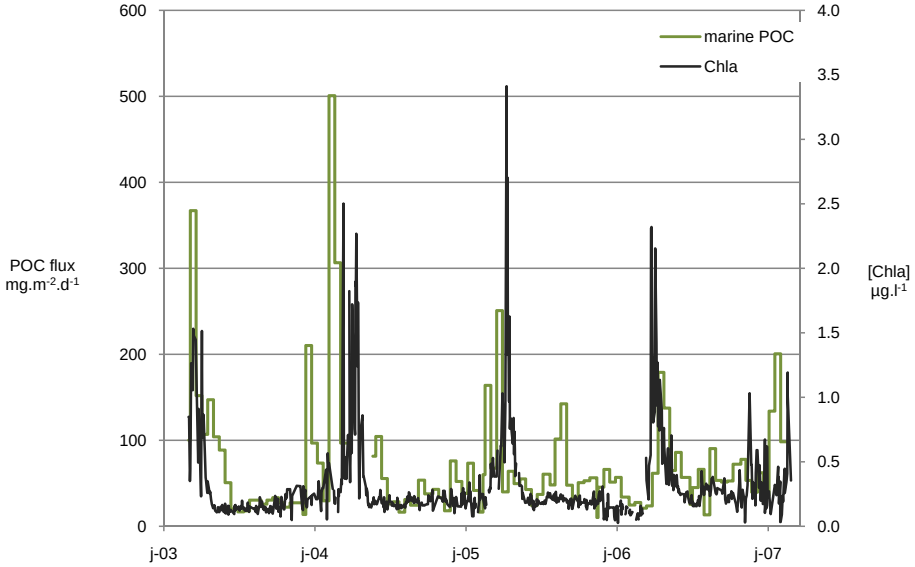

Fig. 5. Surface chlorophyll a concentration (from satellite observations) and POC export (in situ measurements) at 200 m depth (DYFAMED site).

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6, 10737–10773, 2009

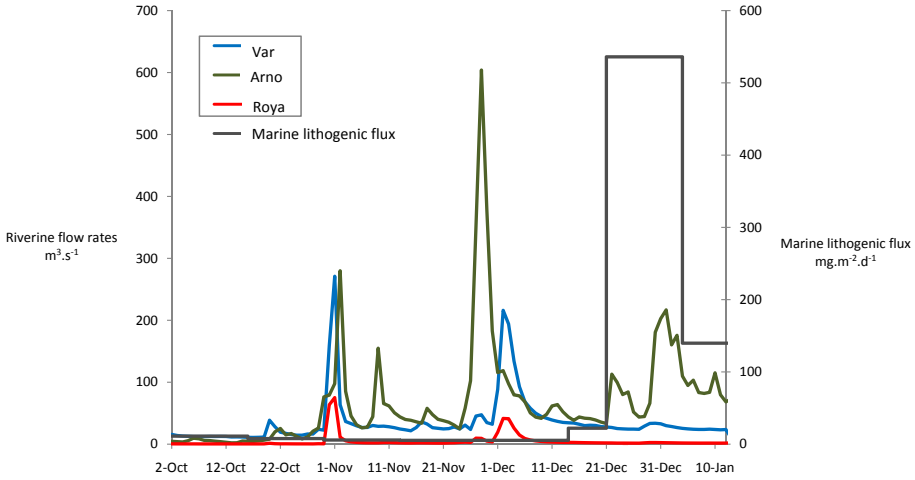

Fig. 6. Evolution of the marine lithogenic flux in the Ligurian Sea at the DYFAMED site and of the Ligurian rivers flow rates at the end of 2003.

BGD
6, 10737–10773, 2009

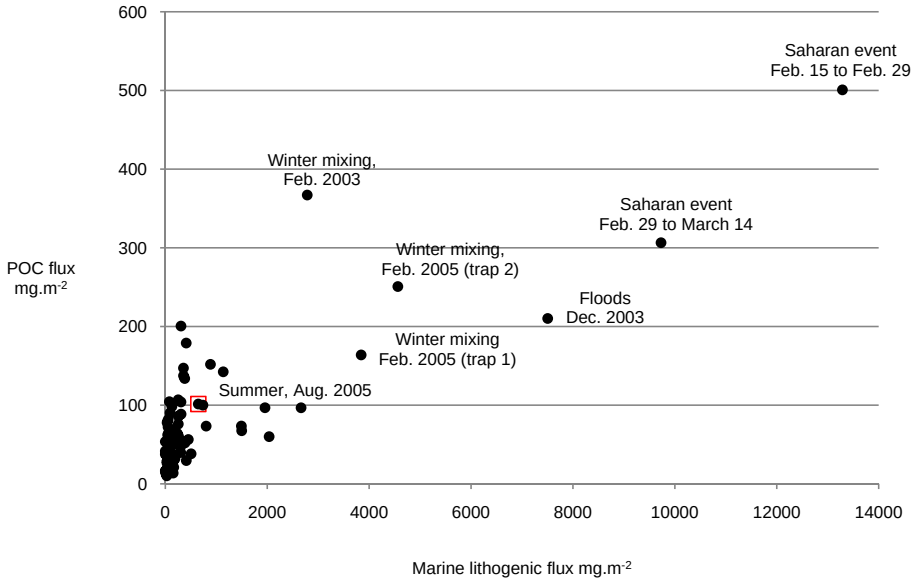

Fig. 7. Marine POC flux and marine lithogenic flux at the DYFAMED site, 200 m depth.

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