

## Atmospherically-promoted photosynthetic activity in a well-mixed ecosystem: Significance of wet deposition events of nitrogen compounds

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### Abstract

Wet atmospheric deposition of dissolved N, P and Si species is studied in well-mixed coastal ecosystem to evaluate its potential to stimulate photosynthetic activities in nutrient-depleted conditions. Our results show that, during spring, seawater is greatly depleted in major nutrients: Dissolved Inorganic Nitrogen (DIN), Dissolved Inorganic Phosphorus (DIP) and Silicic acid (Si), in parallel with an increase of phytoplanktonic biomass. In spring (March–May) and summer (June–September), wet atmospheric deposition is the predominant source (>60%, relative to riverine contribution) for nitrates and ammonium inputs to this N-limited coastal ecosystem. During winter (October–February), riverine inputs of DIN predominate (>80%) and are annually the most important source of DIP (>90%). This situation allows us to calculate the possibility for a significant contribution to primary production in May 2003, from atmospheric deposition (total input for DIN  $\approx 300 \text{ kg km}^{-2} \text{ month}^{-1}$ ). Based on usual Redfield ratios and assuming that all of the atmospheric-derived N (AD-N) in rainwater is bioavailable for phytoplankton growth, we can estimate new production due to AD-N of  $950 \text{ mg C m}^{-2} \text{ month}^{-1}$ , during this period of depletion in the water column. During the same episode (May 2003), photosynthetic activity rate, considered as gross primary production, was estimated to approximately  $30\,300 \text{ mg C m}^{-2} \text{ month}^{-1}$ . Calculation indicates that new photosynthetic activity due to wet atmospheric inputs of nitrogen could be up to 3%.

**Keywords:** primary production; coastal zone; atmospheric deposition; river plumes; nutrient cycles; Strait of Dover

## 1 **1. Introduction**

2 Coastal seas represent one of the most valuable resources on the planet, but they are under  
3 strong anthropogenic pressure such as fisheries, industry or tourism. As a case study, the  
4 Eastern English Channel and the Southern Bight of the North Sea constitute an area under  
5 direct influence of anthropogenic nitrogen (N) sources (De Leeuw *et al.*, 2003).

6 For many years, coastal ecosystems have been the subject of various studies on nutrient  
7 inputs as a possible cause for harmful algal blooms (e.g., Paerl, 1997; De Leeuw *et al.*, 2001,  
8 2002) and nuisance blooms of *Phaeocystis globosa* in Eastern English Channel waters  
9 (Gentilhomme and Lizon, 1998).

10 Nitrogen is generally viewed as the limiting factor for primary production (PP - Dugdale  
11 and Goering, 1967), especially in the North Sea basin (Kronvang *et al.*, 1993; De Leeuw *et*  
12 *al.*, 2003). The riverine supply of N to the sea has increased considerably in the last century  
13 (Paerl *et al.*, 2000; Carstensen *et al.*, 2005), but atmospheric inputs may constitute an  
14 important direct pathway for nutrients to the sea (Migon *et al.*, 1989; Bergametti *et al.*, 1992;  
15 Van Jaarsveld, 1995; De Leeuw *et al.*, 2003), especially for open ocean environments (Owens  
16 *et al.*, 1992; Galloway *et al.*, 1993; Jickells, 1995). Atmospheric deposition of nitrogen  
17 compounds has been shown to be significant in coastal seas, especially in summer and fall  
18 when riverine inputs are low (Paerl *et al.*, 2000; De Leeuw *et al.*, 2002). For the total North  
19 Sea, where there is a relatively high riverine input, direct atmospheric contribution to the total  
20 nitrogen load has been reported to be between 30 and 40% (Rendell *et al.*, 1993; De Leeuw *et*  
21 *al.*, 2002). The atmospheric input is highly episodic with, generally, little seasonal variations  
22 compared with continental inputs (Michaels *et al.*, 1993; Spokes *et al.*, 1993).

23 Previous studies in the Eastern English Channel (Bentley *et al.*, 1993; Laane *et al.*, 1993;  
24 Brunet *et al.*, 1996; Gentilhomme and Lizon, 1998) have shown typical seasonal cycle of  
25 nutrients and chlorophyll. In early spring, the rise of temperatures and the winter stock of

1 nutrients in the water body lead to a phytoplankton bloom that generally ends in May or June.  
2 This bloom is characterized by the succession of two groups of phytoplankton (Quisthoudt,  
3 1987; Lancelot, 1989): diatoms and Prymnesiophyceae such as *Phaeocystis globosa*. Some  
4 recent observations in the Eastern English Channel and other coastal ecosystems, have shown  
5 further phytoplankton blooms after late summer storms, suggesting an episodic direct  
6 atmospheric input of nutrients and/or a substantial riverine input after runoff of rain waters  
7 (Dupuy *et al.*, 2000; D. Vincent – pers. comm.; M. Schapira - pers. comm.). The present study  
8 aims:

- 9 - To quantify the direct atmospheric contribution to the N loading of coastal waters, by  
10 wet deposition,
- 11 - To estimate the significance of this input in comparison to riverine inputs, which  
12 integrate an indirect atmospheric contribution from land-based sources (Paerl *et al.*, 2000),
- 13 - And, in a case of clear N limitation, to determinate the gross primary production  
14 (GPP) rate supported by atmospheric input of nitrogen.

15 This study is not the first dealing with budget assessments of N-loading to estuaries  
16 and coastal water bodies (OSPAR, 2000; Valigura, 2000) and studies on the direct influence  
17 of atmospheric deposition on PP are not new (Paerl, 1985; Paerl and Whitall, 1999). This  
18 work represents one of the few studies on the effect of episodic impacts of AD-N on  
19 photosynthetic activity, in well-mixed water bodies.

20

## 21 **2. Study site and methods**

22 Eastern English Channel (Figure 1) is an epicontinental sea exhibiting high tidal flows  
23 with a transport of water masses from west to east (Pingree and Maddock, 1977). This results  
24 in an advection of Atlantic water masses to the North Sea, through the Strait of Dover. Near  
25 the French coasts, the structure is more complex due to the presence of rivers Seine, Somme,

1 Authie and Canche. These rivers generate a coastal water mass which drifts along the shore,  
2 named “Coastal Flow” (Brylinski *et al.*, 1991), separated from the open sea by a tidally-  
3 maintained frontal area (Brylinski and Lagadeuc, 1990). The inshore water mass is  
4 characterised by a lower salinity (32-33 PSU - Dupont *et al.*, 1991), when compared to the  
5 oceanic offshore waters (35 PSU). This coastal structure, under the direct influence of  
6 freshwaters, is used here to define a coastal box (CB), to calculate and compare atmospheric  
7 and riverine inputs in the Strait of Dover area (see Fig. 1). This CB “Strait of Dover” is  
8 geographically limited to 111 km<sup>2</sup> in order to ensure spatial representative to our comparisons,  
9 according to the location of the wet-only collector (Fig. 1). The western limit of the CB  
10 corresponds to the mean position of the Coastal Front. Taking into account the bathymetry,  
11 the volume of the water body is about 3 km<sup>3</sup>.

12 It is recognised that rainfall is two times less intense over the open sea than on the coast  
13 (Elliott and Reed, 1973; Bethoux, 1977), but the number of rain events is more or less equal.  
14 As indicated by Migon *et al.*, 1991, the number of rain events has a greater influence on flux  
15 than the intensity of the rain. This is confirmed in our survey in which, after long dry periods  
16 (typically several days), N concentrations in rainwaters are systematically higher than  
17 monthly mean concentrations, regardless the intensity of the rain event.

18 The Eastern English Channel is characterised by an oceanic temperate climate.  
19 Predominant winds ( $\approx 50\%$ ) are oriented south and west. This main direction corresponds to  
20 low pressure systems over the North Atlantic and leads to major rain events. Northeasterlies  
21 constitute the second main wind direction ( $\approx 25\%$ ) and corresponds to anticyclonic situations  
22 and dry periods. On an annual basis, the mean wind speed is about 20 km.h<sup>-1</sup> (Flament *et al.*,  
23 1996). Annual precipitations range between 800 and 1000 mm (40 to 60 mm per month  
24 between February and August, and 70 to 90 mm from September to January). In spring 2003,

1 precipitations were 20 to 60 % lower than the seasonal mean (Meteo-France Data,  
2 [www.meteofrance.com/FR/climat/](http://www.meteofrance.com/FR/climat/)).  
3 Two years of sea surveys (R.V. “Côtes de la Manche”, INSU/CNRS) were carried out in  
4 Eastern English Channel in springs 2003 (March, April, May and June) and 2004 (March,  
5 April and May). In each of the surveys, CTD hydrocasts (SBE 25 Sealogger CTD) were made  
6 to determine temperature and salinity down the water column at several stations (Figure 1),  
7 distributed regularly to give a good spatial representation. For each station, seawater was  
8 collected at 1 m depth using a NISKIN™ bottle.

9 The Eastern English Channel is well known to show homogeneous physical and chemical  
10 structure, through the water column (Lagadeuc *et al.*, 1997; Lizon and Lagadeuc, 1998). A  
11 small stratification can sometimes occur between coastal and offshore waters in neap tide  
12 conditions. The duration of this stratification is very short, since destratification occurs at low  
13 waters, due to high current speeds in the shallow waters.

14 After collection, an aliquot of the water sample was frozen (-18°C) on board, for nutrient  
15 analysis immediately following the cruise. Ammonium concentrations were measured directly  
16 on board, according to the Koroleff method (1969). Between 250 mL and 1L of seawater  
17 were filtered through 47 mm WHATMAN™ GF/C filters for chlorophyll *a*, pigments and  
18 suspended particulate matter. Filters were frozen immediately after (-18°C). Full details on  
19 the procedure are given in Gentilhomme and Lizon (1998). Riverine concentrations of DIN  
20 and DIP were measured at salinity 0 for Liane, Wimereux and Slack (See Fig. 1), using  
21 similar procedures for sampling and analysis.

22 Nutrients in seawater, rain and freshwater were photometrically analyzed by an  
23 “AutoAnalyzer Alliance Futura” spectrophotometer, using established methodologies  
24 (Gentilhomme and Lizon, 1998). In total, samples were analyzed for Dissolved Inorganic  
25 Nitrogen (DIN, representing the sum of  $\text{NO}_3^- + \text{NO}_2^- + \text{NH}_4^+$ ), Dissolved Inorganic

1 Phosphorus (DIP, representing  $\text{PO}_4^{3-}$ ) and Si (dissolved silicate concentrations). Chlorophyll *a*  
2 filters were extracted in 90% acetone in the dark for 4 hours. Concentration was determined  
3 using a multi-wave length spectrophotometer and calculated according with Gentilhomme and  
4 Lizon (1998). These data have been completed by data from the French Coastal Observation  
5 Network SOMLIT from June 2003 to January 2004 ([www.domino.u-](http://www.domino.u-bordeaux.fr/somlit_national)  
6 [bordeaux.fr/somlit\\_national](http://www.domino.u-bordeaux.fr/somlit_national)). These network data were acquired in the same experimental  
7 conditions as this study, but samples were only taken off Wimereux.

8        Additionally, PP rate measurements were performed by twice daily sampling of seawater  
9 ( $50^{\circ}48'N$ ,  $1^{\circ}34'E$ ), each week during May 2003. Light intensity measurements were carried  
10 out off Wimereux, where the rainwater collector is located. PP rates were computed from  
11 photosynthetic parameters, estimated with the  $^{14}\text{C}$  technique and short incubation times  
12 (Babin *et al.*, 1994), and from light intensity measured each hour, during the studied month.  
13 This method approximates the carbon fixed by photosynthesis, called “Gross Primary  
14 Production” (GPP). The GPP is defined as the sum of the net primary production plus the  
15 autotrophic respiration. This photosynthetic activity was computed using averaged light  
16 extinction values and interpolated photosynthetic parameters to obtain daily GPP rates. Before  
17 summing daily GPP rates for the month of comparison, they were vertically integrated  
18 (through a 20-m depth water column). Full details on calculations are given in Lizon *et al.*,  
19 1995.

20        Rainwater was collected at Wimereux ( $50^{\circ}75' N - 1^{\circ}65' E$ ), a semi-remote site located on  
21 the French coast of the Straits of Dover, from April 2003 to April 2004. The sampler is  
22 situated at the south of the town (8500 inhabitants), 50 m from the coastline and 10 m above  
23 sea level, relatively isolated and protected from local perturbations. Large agricultural fields  
24 and some buildings surround it. However, Wimereux is situated 10 km from Boulogne-sur-  
25 Mer (in the South) and 30 km from Calais (in the North), medium-sized cities along French

1 coast (100 000 inhabitants). Rainwater samples were collected on an event basis, using an  
2 automatic homemade wet-only collector. This collector only opens when it rains and not  
3 under the influence of the fog or dew. In addition, this device prevents from the  
4 contamination by sea spray, containing nutrients recycled from the sea surface microlayer.  
5 Rainwater is collected by a plastic funnel (area: 452 cm<sup>2</sup>), into pre-cleaned  
6 FluoroEthylenePolymer (FEP) bottles. After the rain event, an automatic cleaning system of  
7 the funnel is activated to rinse funnel and collecting network with ultrapure water ( $\rho = 18.2$   
8 M $\Omega$ .cm). A series of 8 bottles is placed on a carousel, which turns automatically every 24  
9 hours.

10 Funnel and bottles are changed every week when it does not rain or after each rain event.  
11 Sampling equipment was cleaned by soaking with 1M Nitric acid (Normapur<sup>®</sup> grade) 2 to 3  
12 days and then meticulously rinsed with ultra pure water. Funnel and bottles were soaked in  
13 0.1M Nitric acid (Normapur<sup>®</sup> grade), in ultrasonic bath for 1 hour, followed by 1 hour in  
14 ultrasonic bath of ultrapure water and dried under a laminar flow hood (Class 100, U.S.  
15 standard 209a).

16 To evaluate contamination, “field-blanks” were produced by simulating a rain event with  
17 ultrapure water (125 mL). Ultrapure water is then treated as a rain sample. Detection limits of  
18 this method are given in Table 1. 92 samples were collected between April 2003 and April  
19 2004 (13 months). Rainwater was filtered through Nuclepore<sup>™</sup> filters (diameter: 47 mm, cut-  
20 off diameter: 0.45  $\mu$ m, approximately the same as WHATMAN<sup>™</sup> GF/C filters), under a  
21 laminar flow hood. Nutrients concentrations have been determined for 76 events (82 %).  
22 Samples for ammonium were directly analyzed in glass bottle, whereas those for NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>,  
23 DIP and Si measurements were frozen (-18°C) in plastic tubes. Analytical methods were  
24 validated by analyzing certified rainwater (AES 02, NRC Canada). Results are given in  
25 Figure 2.

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**3. Results**

*3.1. Hydrological environment*

Vertically averaged salinity did not exhibit any temporal pattern over the two surveys, but a rather stationary behaviour fluctuating between  $33.1 \pm 0.4$  and  $34.9 \pm 0.5$  in March 2003 and between  $33.6 \pm 0.6$  (May) and  $35.2 \pm 0.0$  (March) in 2004. In contrast, vertically averaged temperature exhibited a clear seasonal cycle throughout the two years, increasing gradually from  $7.5 \pm 0.0^\circ\text{C}$  to  $18.7 \pm 0.2^\circ\text{C}$  between March and June 2003, and from  $8.7 \pm 0.1^\circ\text{C}$  to  $11.5 \pm 0.0^\circ\text{C}$  between March and May 2004. These temperature and salinity values were fully consistent with previous measurements done at the seasonal scale in this area (Brunet *et al.*, 1992; Brylinski *et al.*, 1996; F. Lizon - pers. comm., Seuront *et al.*, In Press). In addition, associated low standard deviation values ( $<0.1$  to  $0.6^\circ\text{C}$ ) indicated that water column was well-mixed over the both studied periods, as previously shown in the whole studied area (Lizon *et al.*, 1995; Gentilhomme and Lizon, 1998; Seuront and Schmitt, 1996; Seuront and Lagadeuc, 1998; Seuront *et al.*, 1999, 2002, In Press).

*3.2. Nutrients and chlorophyll a in seawater: measured concentrations and calculated stocks*

Arithmetic means and ranges of concentrations for DIN, DIP, Si and chlorophyll *a* are summarised in Table 2. DIN concentrations measured here range between 5 and 52  $\mu\text{M}$  in March, followed by a fall to between 0.3 and 6.6  $\mu\text{M}$  in early spring (April and May). This behaviour is not seen for DIP. Chlorophyll *a* concentrations show a decrease in spring 2003 (not in 2004). This decrease seems to be associated with the depletion of DIN and, to a lesser extent, Si. Stocks of DIN, DIP and Si for the CB area are presented in Figure 3. Nutrient stocks are calculated for each survey from the mean nutrient concentration. The DIN inventory ranged from around 1 100  $\text{T.km}^{-3}$  in winter to less than 33  $\text{T.km}^{-3}$  in spring. DIP



1 inventory ranged from 73 T.km<sup>-3</sup> in winter to around 10 T.km<sup>-3</sup> in summer. For Si, the  
2 inventory ranged from 670 T.km<sup>-3</sup> in winter, to less than 10 T.km<sup>-3</sup> in spring.

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### 4 3.3. Nutrient concentrations in rainwater and wet atmospheric inputs to the CB “Strait of 5 Dover”

6 Rainfall and nutrient concentrations in rainwater are given in Table 3. Percentages  
7 indicate the ratio of analyzed rain events. Clearly, major DIN species are nitrate and  
8 ammonium salts, nitrites are negligible in the DIN budget. The predominance of nitrates and  
9 ammonium salts in the DIN mass balance has been already established for other coastal  
10 ecosystems (Migon *et al.*, 1989; Scudlark *et al.*, 2005). For each rain event, the wet input of a  
11 nutrient ( $F$  in  $\mu\text{mol.m}^{-2}$ ) is calculated as:

$$12 \quad F = \frac{C \times V}{S} \quad (1)$$

13 where  $C$  is the nutrient concentration ( $\mu\text{mol.L}^{-1}$ ) in the rain sample,  $V$  is the volume (L) of the  
14 rainwater measured by a rain-gauge and  $S$  is the area of the funnel ( $0.0452 \text{ m}^2$ ). Rainfalls less  
15 than 2 mm are not taken into account due to lack of sample. Wet inputs are cumulated for  
16 each month and are presented in Figure 4. Atmospheric deposition of DIN was estimated to  
17 be  $\approx 415 \text{ T}$  for the CB, from April 2003 to April 2004. Seasonally, these inputs ranged  
18 approximately from 5 to 100 T.month<sup>-1</sup>. DIP inputs to the CB were estimated to be 12 T, from  
19 April 2003 to April 2004 and they ranged from 0.1 to about 6 T.month<sup>-1</sup>. Si inputs were  
20 estimated to be 10 T for the same period and ranged between 0.1 and 4 T.month<sup>-1</sup>.

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### 22 3.4. Riverine inputs

23 Riverine nutrient fluxes entering the coastal zone are modified within estuaries (Jickells,  
24 1998), due to trapping by particles, resuspension of both marine and freshwater sediments, as  
25 well as estuarine PP. The quantification of these processes is still poorly known. Sanders *et al.*

1 (1997) have assessed a net export of DIN and DIP, at the tidal limits of the Humber River of,  
2 respectively, 96 and 15%, relative to quantities entering the estuary. Loquet *et al.* (2000) have  
3 shown, for the Somme River, that during winter (December to March) the net export of DIN  
4 and DIP corresponded respectively to 89 and 85 % of the continental inputs to the Somme  
5 estuary. During the April to September period, inputs to coastal sea from the Somme estuary  
6 were limited due to depletion by phytoplankton and microphytobenthos. In summer (June to  
7 August), almost 100 % of DIN of continental input sinks in the Somme estuary. Morris et al  
8 (1995) show clearly that river-derived nutrients are conservatively exported to coastal sea in  
9 winter and that nutrients are consumed into plume estuaries faster than they are supplied by  
10 the river/estuary system, in spring. In summary, estuaries could be considered as a source of  
11 nutrients in winter and a sink in spring and summer. To evaluate the net export of riverine  
12 DIN and DIP to the CB “Strait of Dover” (Figure 5), we have adopted the quantitative  
13 estimations available for the closest estuary, i.e. the Bay of Somme (Loquet *et al.*, 2000).  
14 From April 2003 to April 2004, a retention factor of 15% has been applied to DIP fluxes.  
15 According to the same author, retention of 10% was applied to DIN fluxes during winter and  
16 90% during spring and summer.

17

## 18 **4. Discussion**

### 19 *4.1. Evolution of stocks and concentrations of nutrients and chlorophyll a*

20 There is evidence for seasonal cycles of nutrients and chlorophyll *a*, which is typical of  
21 temperate seas (OSPAR, 2000). These seasonal patterns of nutrient stocks and chlorophyll *a*  
22 concentrations have been already described in the Eastern English Channel (Bentley *et al.*,  
23 1993; Laane *et al.*, 1993; Gentilhomme and Lizon, 1998). During the winter period,  
24 phytoplankton growth is limited by light and temperature (Gentilhomme and Lizon, 1998)  
25 and the stocks of DIN, DIP and Si increase. These winter stocks (Figure 3) are strongly and

1 quickly depleted during spring (by a factor of 7 for DIP, 33 for DIN and 67 for Si), due to  
2 phytoplankton growth. In addition, the corresponding increase of mean chlorophyll *a*  
3 concentrations between spring 2003 and spring 2004 (Table 2) is consistent with previous  
4 observations of Gentilhomme and Lizon (1998). These authors illustrate the consequence of  
5 phytoplankton growth limitation, depending on nutrient availability, which is dependant on  
6 water column dynamics, meteorological patterns and external inputs. Whereas the bloom and  
7 its relationship with nutrient occur every years, the timing and strength of this event is very  
8 variable (Tungaraza *et al.*, 2003). In 2003, an important bloom of diatoms occurred from  
9 March. In 2004, two successive different blooms occurred in early March and in May (Table  
10 2 and Figure 3). It is generally accepted that there are two blooms in the North Sea and the  
11 Eastern English Channel (Lancelot, 1989). Large diatoms dominate the first bloom, which  
12 occurs in early spring, and the second is characterized by the Prymnesiophyceae *Phaeocystis*  
13 *globosa*. In summer, smaller diatoms are predominant but concentrations of chlorophyll *a* are  
14 relatively low (Quisthoudt, 1987). For 2003, the exceptional meteorological conditions  
15 (Meteo-France Data), in spring (relatively high light and temperature) could have results in  
16 both blooms occurring in a very short time (less than one month) and are not distinguished in  
17 our survey. Accordingly, as indicated by Figure 3, May 2003 is characterised by the lowest  
18 DIN stock recorded in this study. The same effect is observed for Si, but this nutrient is only  
19 limiting for diatoms. There are always measurable DIP concentrations in our study area,  
20 indicating that it is not limiting. From our study, the Eastern English Channel and the North  
21 Sea are clearly limited by N, in agreement with previous results from the ANICE project (de  
22 Leeuw *et al.*, 2003). In our coastal area, we are clearly, in May 2003, in N-limiting conditions  
23 for phytoplankton growth.

24

#### 25 4.2. Atmospherically-derived nitrogen deposition

1 The extreme variability of nutrients concentrations in rainwater is not surprising: many  
2 studies have shown at least one order of magnitude between minimum and maximum values  
3 for nutrient concentrations in rainwater (e.g. Beverland *et al.*, 1998; Scudlark *et al.*, 2005).  
4 For atmospherically-derived nitrogen (AD-N), wet deposition rates are influenced by urban  
5 and industrial activities (e.g. nitrate from NO<sub>x</sub> conversion). Agricultural activities and  
6 chicken/pig farming, emitting ammonium from NH<sub>3</sub> conversion, are mentioned in addition to  
7 explain this variability (Paerl, 1997 ; Paerl *et al.*, 2000). Maximum AD-N wet depositions are  
8 encountered in April, May and October 2003 (from 60 to 100 T.month<sup>-1</sup>; Fig.4). These three  
9 months are not particularly wet, but correspond to rain events occurring after long dry periods  
10 (typically several days). As indicated in Section 2, in this situation, N concentrations in  
11 rainwaters are systematically higher than monthly mean concentrations. This highlights the  
12 occurrence of wash-out processes and, indirectly, suggests the accumulation of nitrogen  
13 compounds in the low troposphere during long dry periods. During these episodes, typically  
14 anticyclonic, wind speeds are low and local winds oriented to the continental sector (the  
15 countryside around Boulogne sur Mer, Calais and the industrial zone of Dunkirk harbour).  
16 This sector can be view as a source of nutrients for the atmosphere (Paerl *et al.*, 2000), due to  
17 extensive agricultural and industrial activities. The Dunkirk harbour industrial area (about 80  
18 km from the Strait of Dover, in the Northeast) emits each year more than 10<sup>4</sup> T (NO + NO<sub>2</sub>).

19 Considering that maximum inputs of AD-N (DIN) occur between April and October 2003,  
20 we note that they correspond with the minimum stock of DIN in seawater (Figure 3).  
21 Approximately 60% of AD-N deposition occurs within 3 months (April, May and October  
22 2003). In May 2003, period corresponding to photosynthetic-activity measurements, the AD-  
23 N wet input is approximately 66 Tons and may represent a significant contribution of “new”  
24 N for PP, in this oligotrophic area (N-depleted waters), as has been proposed in other studies  
25 (Benitez-Nelson, 2000; de Leeuw *et al.*, 2003; Cartensen *et al.*, 2005; Bartoli *et al.*, 2005).

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### 2 4.3 Comparison with riverine inputs

3 Comparison of atmospheric versus riverine nutrient contributions to the CB is presented in  
4 Figure 6. Through the 13 months survey, riverine inputs are the most important source for  
5 DIP (> 90%), with the exception of the May to August 2003 period. In spring and summer,  
6 wet deposition becomes the major source ( $\geq 60\%$ ) for DIN, except in April 2004 ( $\approx 40\%$ ).  
7 During winter, DIN riverine inputs predominate (> 80%). This comparison is calculated from  
8 our data, with the caveat that we take into account wet atmospheric deposition. It is calculated  
9 that dry deposition represents 20 to 25 % of wet deposition in the North Sea basin (De Leeuw  
10 *et al.*, 2002). Even though this dry deposition is not considered here, our measurements fall  
11 into the range of ACDEP results (De Leeuw *et al.*, 2003). This indicates that not considering  
12 dry deposition does not preclude a rough comparison of our results with those from ACDEP.  
13 The efficiency of wash-out after long dry periods and the related deposition may explain the  
14 absence of discrepancy between this study and the more extensive ANICE Project, in the  
15 evaluation of N deposition rates.

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### 17 4.4. Estimation of primary production triggered by AD-N

18 An evaluation of new production theoretically triggered by an atmospheric event ( $NP_{\text{atmo}}$ ),  
19 can be calculated using Redfield ratios:

$$20 \quad NP_{\text{atmo}} = [X] \frac{C}{X} \times 12 \quad (2)$$

21 where  $[X]$  is the nutrient atmospheric input, expressed in  $\mu\text{mol m}^{-2}$ , with  $X = \text{N, P or Si}$ ,

22  $\frac{C}{X}$  is the carbon / nutrient Redfield ratio (Redfield *et al.*, 1963), and 12 is the molar mass of

23 carbon, expressed in  $\text{g mol}^{-1}$ .  $NP_{\text{atmo}}$  is viewed as the quantity of carbon available for

24 photosynthetic activity and, as such, can be compared to the activity calculated from

1 photosynthetic parameters, into the water column (see section 2). Nutrients cannot accumulate  
2 in depleted surface waters and are rapidly consumed immediately after their incorporation in  
3 seawater (Migon *et al.*, 1989). In case of stratified water masses, the atmospheric input of  
4 nutrients is often evaluated by comparison with PP within a thin layer (Migon and Sandroni,  
5 1999 ; Ridame and Guieu, 2002; Bartoli *et al.*, 2005). In this work, AD-N contribution is  
6 compared with photosynthetic activity measured for the entire water column, due to the  
7 absence of stratification. In May 2003, DIN concentrations in seawater were very low (0.3 to  
8 0.6  $\mu\text{M}$ ) and wet atmospheric inputs were estimated to 12  $\text{mmol}\cdot\text{m}^{-2}$ . Based on Redfield ratios  
9 and assuming that total DIN in rainwater is bioavailable for phytoplankton, we can estimate  
10 the  $\text{NP}_{\text{atmo}}$  to be 79.5  $\text{mmolC}\cdot\text{m}^{-2}$  (950  $\text{mgC}\cdot\text{m}^{-2}$ ). During the same period (May 2003), the  
11 photosynthetic activity, considered as a GPP, can be estimated to 30300  $\text{mgC}\cdot\text{m}^{-2}$ .  
12 Consequently, the atmospherically-promoted photosynthetic activity could be equivalent to  
13 3.1 % of the total monthly GPP of phytoplankton. Such a result is consistent with other  
14 studies in the south of the North Sea, where De Leeuw *et al.* (2003) have shown that total  
15 atmospheric inputs of DIN could support 5.5 % of the total primary production during  
16 summer. In the Western Mediterranean sea, Guerzoni *et al.* (1999) reported that new  
17 production triggered by rain source of nitrogen could be of 3 to 6 % of the total annual  
18 primary production rates.

19

## 20 **5. Conclusion**

21 In this work, our goal was to quantify the direct atmospheric contribution to the N  
22 loading of coastal waters, to estimate the significance of this input by comparison with  
23 riverine inputs, which integrate an indirect N atmospheric contribution from land-based  
24 sources. In a case of clear N limitation of the PP, the capacity of wet atmospheric deposition  
25 to support GPP, viewed as an atmospherically-promoted photosynthetic activity, has been

1 estimated. For the Strait of Dover area, the seasonal cycles of nutrients and chlorophyll *a*,  
2 typical of temperate coastal seas, is established. During spring, seawater is strongly depleted  
3 in major nutrients (DIN, DIP and Si), in parallel with an increase of chlorophyll *a*  
4 concentrations. The atmospheric input of nutrients occurring during this period may be use for  
5 new PP. These inputs are characterized by their highly episodic nature. 60% of AD-N  
6 deposition occurs within 3 months and more than 80% for DIP. Compared with riverine  
7 nutrient inputs, atmospheric deposition becomes the predominant source ( $\geq 60\%$ ) for DIN in  
8 spring and summer. That leads to the conclusion that atmospheric deposition can have a  
9 significant contribution to new production, particularly in late spring (April and May). Based  
10 on Redfield ratios and assuming that entire nitrogen in rainwater is bioavailable for  
11 phytoplankton, we can estimate the atmospherically-promoted photosynthetic activity  
12 equivalent to  $950 \text{ mgC.m}^{-2}$  during May 2003. During the same period, GPP rates (estimated  
13 by photosynthetic parameters) of coastal waters can be estimated to  $30300 \text{ mgC.m}^{-2}$ .  
14 Consequently, the theoretical new production due to atmospheric inputs of N compounds  
15 could represent about 3 % of total PP of phytoplankton. Numerous studies have been  
16 concerned with long-term assessments and comprehensive budgets of N-loading to estuaries  
17 and coastal water bodies (e.g. Valigura, 2000). Because the atmospheric source is  
18 quantitatively important and capable of stimulating PP in coastal ecosystems, the emphasis  
19 has been put on meaningful airsheds assessment studies and on comparisons of direct and  
20 indirect (i.e. through watersheds and estuarine waters) atmospheric contributions. Studies on  
21 the direct influence of atmospheric deposition on PP are not new. However, this work  
22 represents one of the sparse studies on episodic impacts of AD-N on GPP, computed from *In*  
23 *Situ* photosynthetic parameters, during a period of demonstrated N depletion.  
24

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11

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**Figures Captions:**

**Figure 1:** Study Site. Solid lines delimit the coastal box (CB) for stock calculations and comparison of atmospheric versus riverine inputs. Adapted from Seuront *et al.*, 2002.

**Figure 2:** Precision and accuracy of the rainwater sampling and analysis method (all results in  $\mu\text{M}$ ).

**Figure 3:** Evolution of DIN, DIP and Si concentrations for the CB “Strait of Dover”, from March 2003 to March 2004 ( $\text{T.km}^{-3}$ ).

**Figure 4:** Evolution of wet atmospheric inputs of nutrients to the CB “Strait of Dover”, from April 2003 to April 2004 ( $\text{Tons.month}^{-1}$ ).

**Figure 5:** Net export of riverine nutrients to the CB “Strait of Dover” for DIN [a] and DIP [b], from April 2003 to April 2004 ( $\text{Tons.month}^{-1}$ ).

**Figure 6:** Comparison of nutrients inputs to the CB “Strait of Dover” for DIN [a] and DIP [b], from April 2003 to April 2004 (Black: Net riverine inputs; Grey: Wet atmospheric inputs).



**Table 1:** Detection limits of the rainwater sampling and analysis method (Detection limits corresponds to 3 time the standard deviation)

Nutrients	Detection limits ( $\mu\text{M}$ )
$\text{NO}_3^-$	4.4
$\text{NO}_2^-$	0.1
$\text{NH}_4^+$	7.1
$\text{PO}_4^{3-}$	0.4
$\text{Si(OH)}_4$	0.4

**Table 2:** Arithmetic means ( $\mu_A$ ) and ranges of DIN, DIP, Si, chlorophyll *a* concentrations and temperatures, during sea-cruises. LD for Detection Limit

Date	DIN ( $\mu\text{M}$ )		DIP ( $\mu\text{M}$ )		Si ( $\mu\text{M}$ )		Chl <i>a</i> ( $\mu\text{g.L}^{-1}$ )		T ( $^{\circ}\text{C}$ )	
	$\mu_A$	range	$\mu_A$	range	$\mu_A$	range	$\mu_A$	range	$\mu_A$	range
march-03	13.2	0.8-52.1	0.4	0.3-0.8	1.6	0.8-5.2	35.2	2.1-96.4	7.6	7.1-7.8
april-03	0.6	0.1-1.9	0.2	0-4.2	0.5	0-3.1	11.2	2.4-53.5	9.9	9.1-11.3
may-03	0.3	0.1-0.6	0.2	0.05-0.7	0.01	< LD -0.07	3.1	< LD -17.4	11.8	10.8-13.3
june-03	2.2	0.1-18.9	0.3	0.1-3.7	1.0	0.07-2.4	5.2	< LD -25.7	16.8	15.3-19.0
march-04	9.7	4.8-15.9	0.25	0.1-0.3	2.2	0.1-3.1	9.8	5.2-12.7	7.1	6.8-7.3
april-04	2.6	0.7-6.6	0.2	0.1-0.5	1.4	0.8-6.3	7.6	1.4-13.9	9.0	8.7-9.3
may-04	1.2	0.5-2.2	0.2	0.05-0.8	1.2	0.8-1.8	15.5	1.6-33.3	11.1	10.7-11.8

**Table 3:** Pluviometry (mm) and mean concentrations of nutrients in rainwater, measured at Wimereux (See Fig. 1), from April 2003 to April 2004.  $\mu_A$  and  $\mu_G$  are respectively the arithmetic and geometric means, SD the standard deviation. LD for Detection Limit.

	mm	DIP ( $\mu\text{M}$ )	Si ( $\mu\text{M}$ )	$\text{NO}_2^-$ ( $\mu\text{M}$ )	$\text{NO}_3^-$ (mM)	$\text{NH}_4^+$ ( $\mu\text{M}$ )
<b>%</b>	100	82	82	82	82	66
<b><math>\mu_A</math></b>	5.1	3.5	3.5	0.8	0.14	85
<b><math>\mu_G</math></b>	2.9	0.65	0.67	0.3	0.05	52
<b>SD</b>	5.9	10.4	14.6	1.8	0.41	77
<b>Median</b>	2.6	0.4	0.5	0.2	0.04	58
<b>Max</b>	32	59	117	9.8	3.3	346
<b>Min</b>	0.3	<LD	<LD	<LD	<LD	<LD

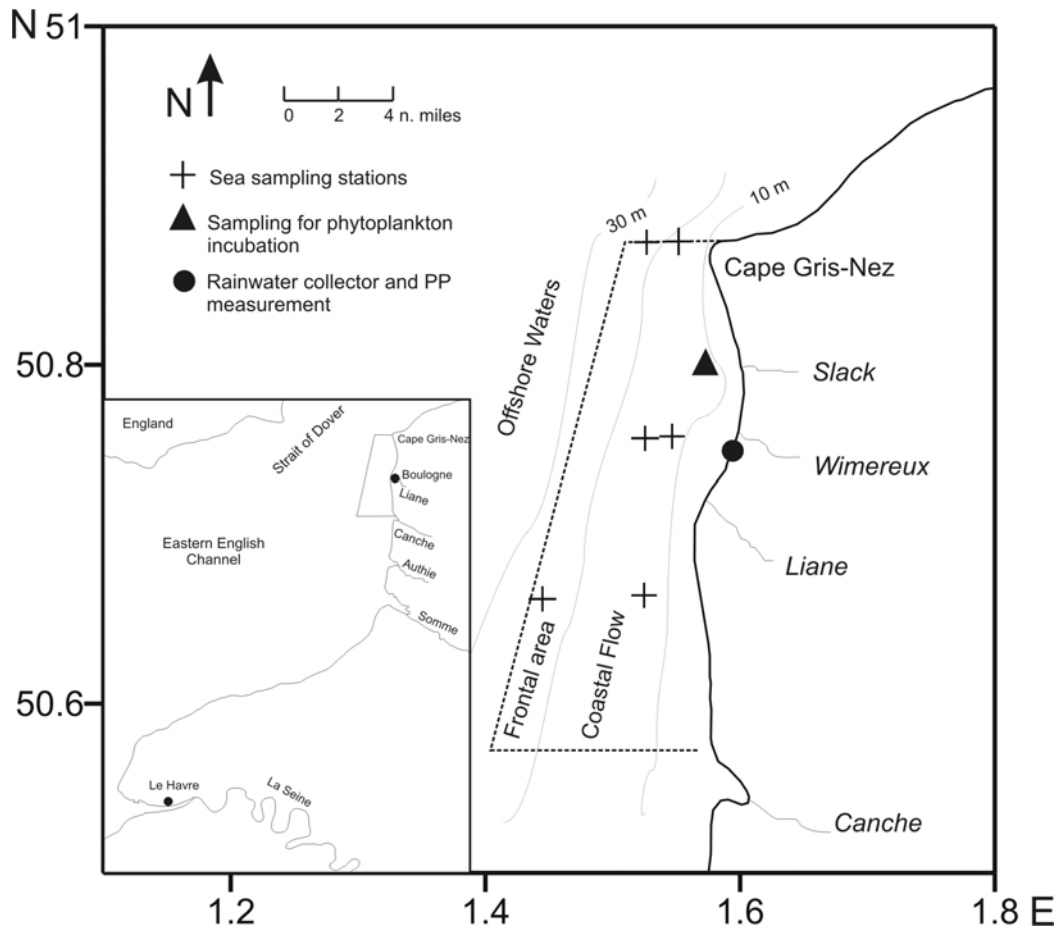


Figure 1: Study Site. Solid lines delimit the coastal box (CB) for stock calculations and comparison of atmospheric versus riverine inputs. Adapted from Seuront et al., 2002.

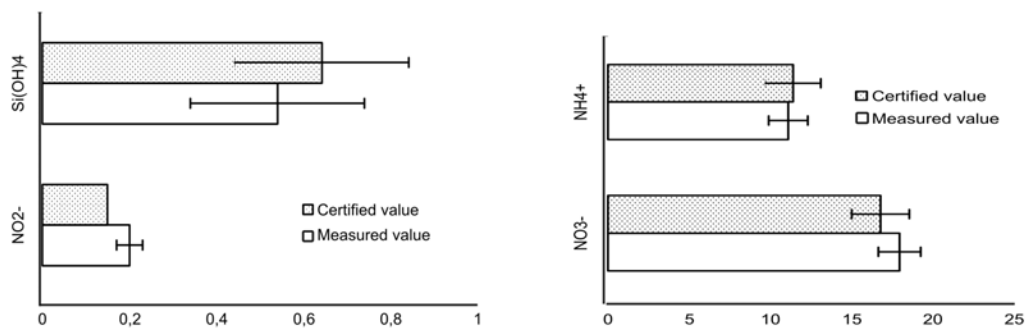
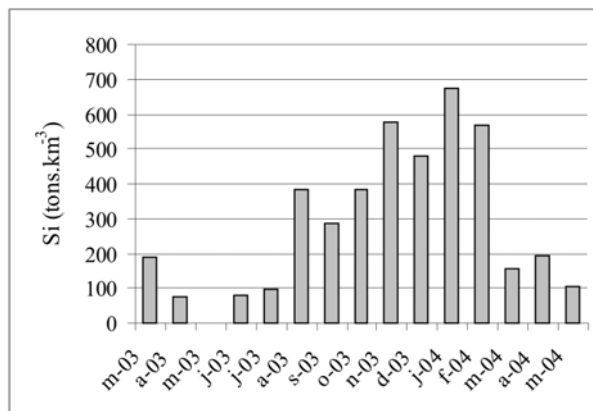
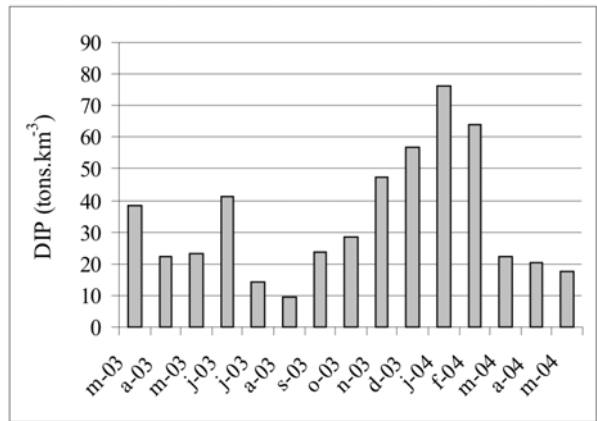
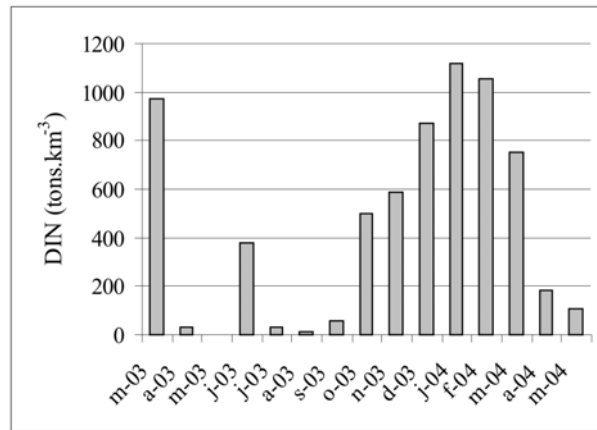


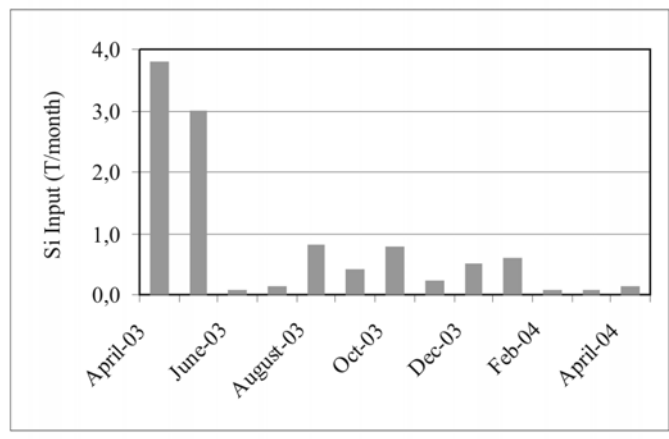
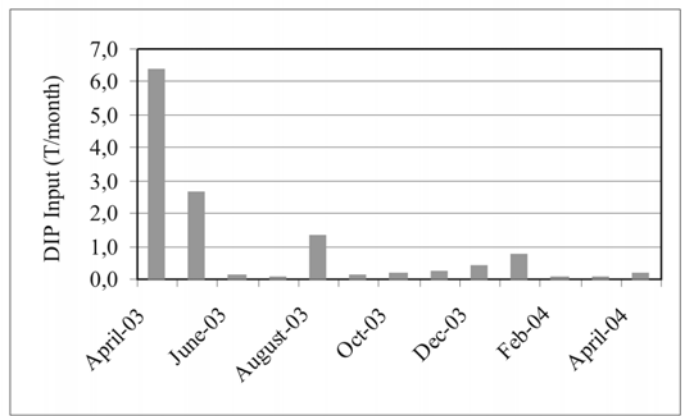
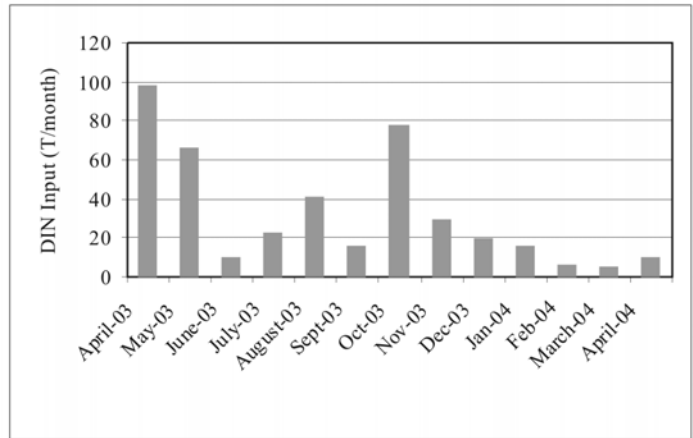
FIGURE 2

Figure 2: Precision and accuracy of the rainwater sampling and analysis method (all results in  $\mu\text{M}$ ).



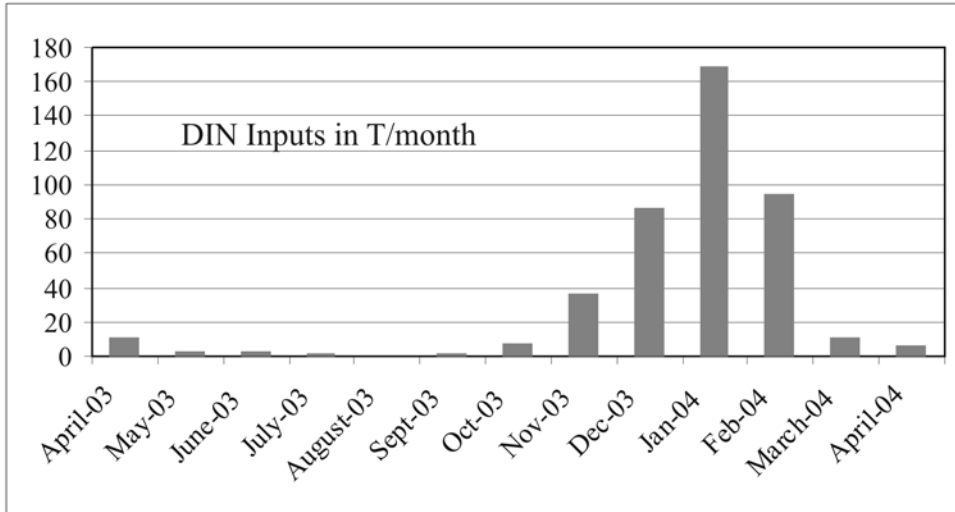
**FIGURE 3**

Figure 3: Evolution of DIN, DIP and Si concentrations for the CB "Strait of Dover", from March 2003 to March 2004 (T.km<sup>-3</sup>).

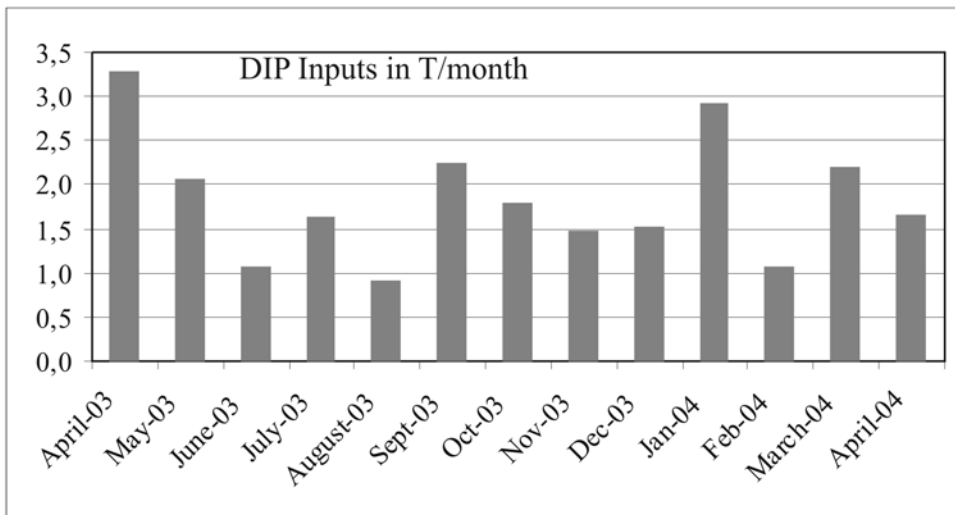


**FIGURE 4**

Figure 4: Evolution of wet atmospheric inputs of nutrients to the CB "Strait of Dover", from April 2003 to April 2004 (Tons.month<sup>-1</sup>).



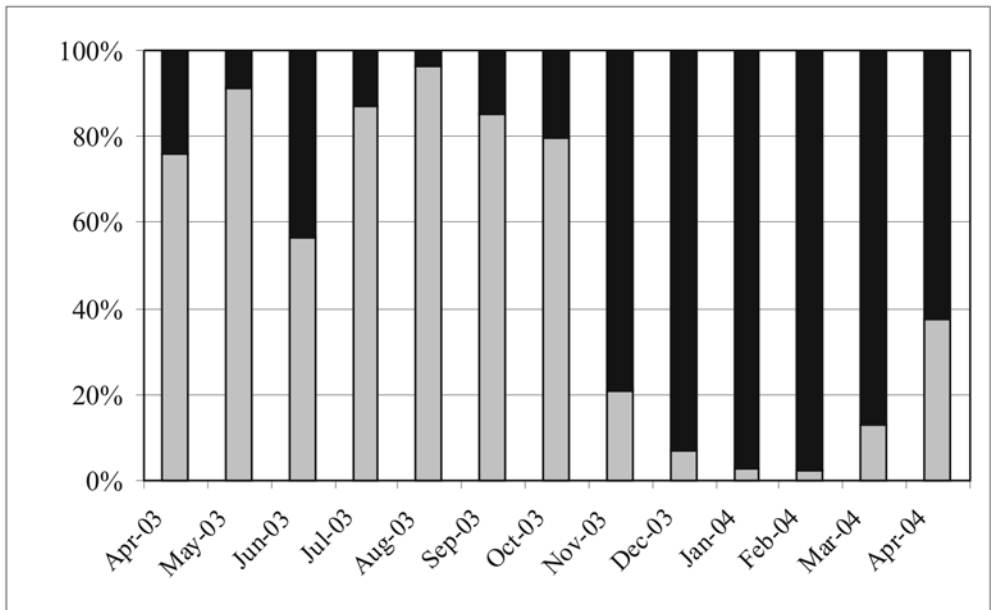
[a]



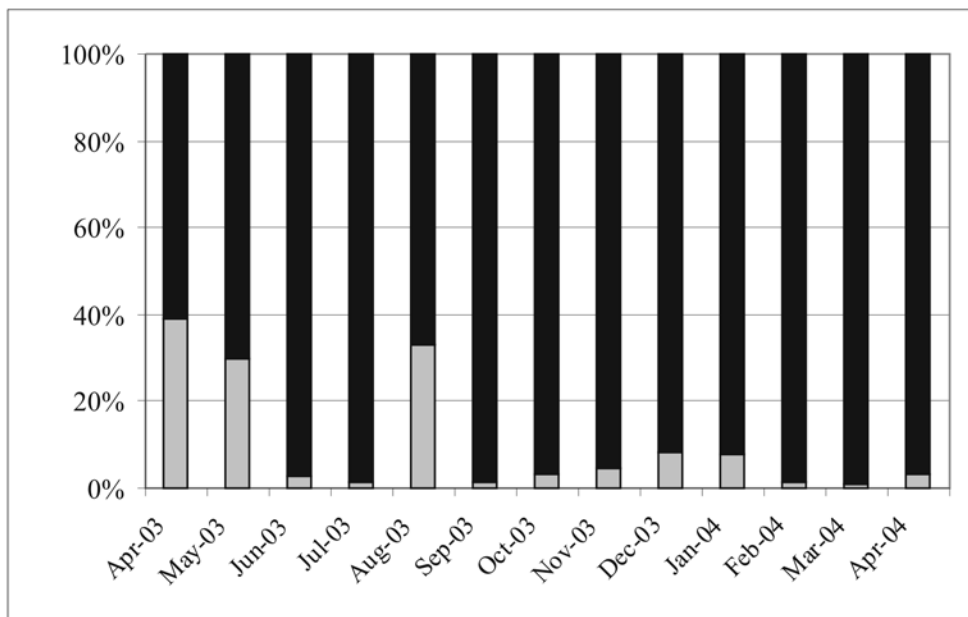
[b]

## FIGURE 5

Figure 5: Net export of riverine nutrients to the CB “Strait of Dover” for DIN [a] and DIP [b], from April 2003 to April 2004 (Tons.month<sup>-1</sup>).



[a]



[b]

## FIGURE 6

Figure 6: Comparison of nutrients inputs to the CB "Strait of Dover" for DIN [a] and DIP [b], from April 2003 to April 2004 (Black: Net riverine inputs; Grey: Wet atmospheric inputs).