

# Seawater $p\text{CO}_2$ Distribution and Air-Sea $\text{CO}_2$ Exchanges on the Atlantic European Shelf

Michel FRANKIGNOULLE<sup>1</sup>, Marc ELSKENS<sup>2</sup>,  
Michèle LOIJENS<sup>3</sup> and Patrick DAUBY<sup>1</sup>

<sup>1</sup> Laboratoire d'Océanologie, University of Liège

<sup>2</sup> Laboratorium voor Analytische Scheikunde, Vrij Universiteit te Brussel

<sup>3</sup> Laboratoire d'Océanographie chimique, Université libre de Bruxelles

## Abstract.

Continental shelves and coastal areas are sites of several processes which can drastically influence the surface seawater  $p\text{CO}_2$  distribution and result in relatively important air-sea  $\text{CO}_2$  fluxes. We present here some data obtained in the English Channel, in the Gulf of Biscay and in the Celtic Sea which illustrate the effect of those coastal processes. It is in particular pointed out that upwelling and riverine input, which generate relatively important primary production, can also be sources of  $\text{CO}_2$  to the atmosphere.

## 1.- Introduction.

Air-sea  $\text{CO}_2$  exchange is an important process in the global carbon cycle because it determines the amount of anthropogenic carbon which enters the ocean. The buffering capacity of the ocean with respect to the atmospheric  $\text{CO}_2$  excess is in fact a subject of controversy (see Tans *et al.*, 1990; Watson *et al.*, 1991). While oceanic water is most often close to the atmospheric equilibrium, in terms of  $\text{CO}_2$  level, and then site of quite low air-sea exchange, it is now often suggested that coastal areas could be sites of relatively important fluxes (Walsh, 1988; Wollast, 1989; Walsh, 1991; Frankignoulle and Gattuso, 1993; Frankignoulle *et al.*, submitted). Indeed, coastal ocean has particular properties (riverine inputs, coastal upwellings, high productivity, ecosystems diversity, ...) which can induce a drastic change in surface water  $\text{CO}_2$  level and then generate very high local  $\text{CO}_2$  fluxes. In spite of this observation, the coastal ocean is never considered when modelling the oceanic carbon cycle, mainly due to the deficiency of field data.

The  $\text{CO}_2$  flux at the air-sea interface is given by

$$(1) \quad F = K\alpha(p\text{CO}_2(\text{Water}) - P\text{CO}_2(\text{Air})) = K\alpha \Delta p\text{CO}_2$$

where  $K$  is the gas exchange coefficient (piston velocity),  $\alpha$  is the  $\text{CO}_2$  solubility coefficient in seawater,  $p\text{CO}_2(\text{Water})$  and  $P\text{CO}_2(\text{Air})$  are the partial  $\text{CO}_2$  pressure in both phases. While the direction of the flux is then given by the sign of the gradient through the interface, its magnitude is function of both the gradient and the exchange coefficient. The latter depends on physical environment, that can be very complex [wind speed, turbulence, bubbles, surface film, ... see Liss (1983)] and the role of which is not yet fully understood. Wind speed is nevertheless recognised as the preponderant parameter controlling the gas exchange coefficient value (Liss and Merlivat, 1986; Wannikhof, 1992). Because the  $\text{CO}_2$  level in the low atmosphere is quite homogeneous throughout the world, the partial  $\text{CO}_2$  pressure in surface seawater is a key parameter which determines the direction of the air-sea  $\text{CO}_2$  flux.

In this paper, we briefly review the effect that biological activity could induce on seawater  $p\text{CO}_2$  distribution, we discuss some  $p\text{CO}_2$  data which have been obtained in the Gulf of Biscay together with chlorophyll and primary production ones and we finally present air-sea  $\text{CO}_2$  exchange measurements made in the Celtic Sea.

## 2.- Material and methods.

$p\text{CO}_2$  is calculated from experimental determination of both pH and total alkalinity (Talk). The later is obtained from classical Gran electrotitration on GF/C filtered samples. Talk calculation is made with corrections for fluoride and sulphate according to Hansson and Jagner (1973). The accuracy has been checked to be 0.05% ( $1\ \mu\text{eq/kg}$ ).

pH is measured using an electrochemical cell calibrated to take into account the effect of the ionic strength. The pH sensor, built to minimize and keep constant the junction potential, is calibrated by titrating  $\text{NaCl}-\text{Na}_3\text{PO}_4$  solutions and is finally tested against NBS buffers (Frankignoulle and Distèche, 1984; Frankignoulle *et al.*, in preparation). The error on the absolute pH value is estimated to be about 0.006 pH unit.

$p\text{CO}_2$  and the complete speciation is calculated according to the Mole/kg NBS pH scale using the  $\text{CO}_2$  acidity constants from Mehrbach *et al.* (1973), the  $\text{CO}_2$  solubility coefficient from Weiss (1974), the borate acidity constant from Hansson (1973) and the water dissociation constant from Lyman (1957). The total borate molality is calculated using the Culkin (1973) ratio to salinity. The error on  $p\text{CO}_2$  has been established to be less than 8 ppmV.

Chlorophyll *a* concentrations are determined using the spectrophotometric method (Lorenzen and Jeffrey, 1978).



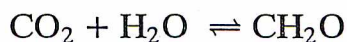
Primary production is determined as explained by Elskens *et al.* (this volume).

$\delta^{13}\text{C}$  on the total suspended matter (collected as explained by Dauby *et al.*, this volume), expressed relative to the PDB standard, is measured by Isotope Ratio Mass Spectrometry after combustion of the sample with the method described by Sofer (1980).

Air-sea  $\text{CO}_2$  exchanges are estimated using the bell method, which consist to deploy an air incubator on the sea surface and to detect the initial variation of  $\text{CO}_2$  in the air phase (Frankignoulle, 1988).

## 2.- Seawater $p\text{CO}_2$ distribution versus biological activity.

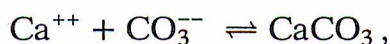
Biological activity can be roughly divided into two main processes, both of which having an important effect on the  $\text{CO}_2$  level in surrounding water: the organic carbon metabolism (photosynthesis/respiration) and the inorganic carbon metabolism (calcification/dissolution). The first process can be summarized by



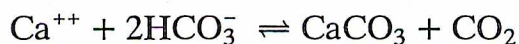
and the production of organic matter results in an uptake of dissolved  $\text{CO}_2$  level and a decrease of the partial  $\text{CO}_2$  pressure which can be simply calculated using the so-called REVELLE factor

$$\beta = \frac{\delta \ln T\text{CO}_2}{\delta \ln p\text{CO}_2} \simeq 10,$$

(see Sundquist, 1981). The calcification process can be written



or



and the uptake of carbonate ion (first equation) or the release of dissolved  $\text{CO}_2$  (second equation) results in an increase of the partial  $\text{CO}_2$  pressure in surrounding water. Contrary to organic metabolism, calcification is a process which favours the escape of  $\text{CO}_2$  from seawater to the atmosphere (Ware *et al.*, 1992; Frankignoulle and Gattuso, 1993). Ware *et al.* (1992) have estimated, using speciation calculation, that the ratio *released*  $\text{CO}_2$  / *precipitated*  $\text{CO}_3^{--}$  is about 0.6 and have suggested that coral reefs, which are sites of intensive calcification, could act globally as source of carbon dioxide to the atmosphere. Gattuso *et al.* (submitted) have recently performed air-sea  $\text{CO}_2$  measurements on a Polynesian coral atoll and did observe a diel variation of the sign of the flux, with fluxes from air to sea during the day and fluxes from sea to air during the night, and



with a daily budget as a net flux from sea to air. Using buffer factors expressions developed by Frankignoulle (submitted), Frankignoulle *et al.* (submitted<sup>1</sup>) have proposed an analytical expression which allows computation of the *released* CO<sub>2</sub> / *precipitated* CO<sub>3</sub><sup>2-</sup> ratio and have shown that it increases with the dissolved CO<sub>2</sub> level in seawater and that the increasing atmospheric CO<sub>2</sub> level has a positive feedback with marine calcification.

When calcification occurs, it is obviously coupled to organic metabolism and the final impact on partial CO<sub>2</sub> pressure depends on the proportion of both processes. Frankignoulle (submitted) has proposed a REVELLE factor for simultaneous organic and inorganic metabolisms

$$\beta = \frac{\delta \ln p\text{CO}_2}{\delta \ln \text{TCO}_2} = -7.02 + 0.186\% C_{\text{org}}$$

where % C<sub>org</sub> is the percentage of carbon uptake for organic metabolism.

As observed before, when calcification does not occur, primary production results in a decrease of seawater *p*CO<sub>2</sub>. Watson *et al.* (1991) have recently plotted *p*CO<sub>2</sub> versus chlorophyll data obtained in the North Atlantic during Spring 1989. Depending on the investigated area, they have observed three kinds of linear relationship between both these parameters depending on present and past phytoplanktonic activity: the peak stage (bloom) linear regression is characterized by the fact that *p*CO<sub>2</sub> nearly intercepts the atmospheric equilibrium value as chlorophyll reaches 0 (*p*CO<sub>2</sub> = 358 - 16.8 [Chl *a*], *r* = -0.79), while the late stage of phytoplanktonic activity linear regression yields a lower intercept for 0 chlorophyll level (*p*CO<sub>2</sub> = 299 - 6.6 [Chl *a*], *r* = -0.70) because CO<sub>2</sub>, which can not be provided from deeper water through the seasonal thermocline, has to invade the surface layer from the atmosphere to recover the equilibrium value, and this is a relatively slow process.

Frankignoulle *et al.* (submitted<sup>2</sup>) have also determined the surface *p*CO<sub>2</sub> pattern in the English Channel in June 1992 and have obtained a distribution versus chlorophyll *a* which agrees well with the relationship proposed by Watson *et al.* (1991) for phytoplanktonic peak stage. On another hand, data they have obtained in the Southern Bight of the North Sea clearly show that riverine input can produce locally very high production but associated to a high *p*CO<sub>2</sub> level: in the area under the influence of the Scheldt Estuary, while the chlorophyll *a* concentration is as high as 7.99 µg l<sup>-1</sup>, *p*CO<sub>2</sub> is 407 ppmV, due to the high alkalinity value of Scheldt water. This coastal site thus acts together as an important sink of carbon, via primary production, and as a source of CO<sub>2</sub> to the atmosphere. Riverine input can be studied by the determination of the δ<sup>13</sup>C in the total suspended matter and the figure 1 gives an example of results obtained in the English Channel in June 1991 (see Dauby *et al.*, submitted). In this area, the natural carbon sources are:

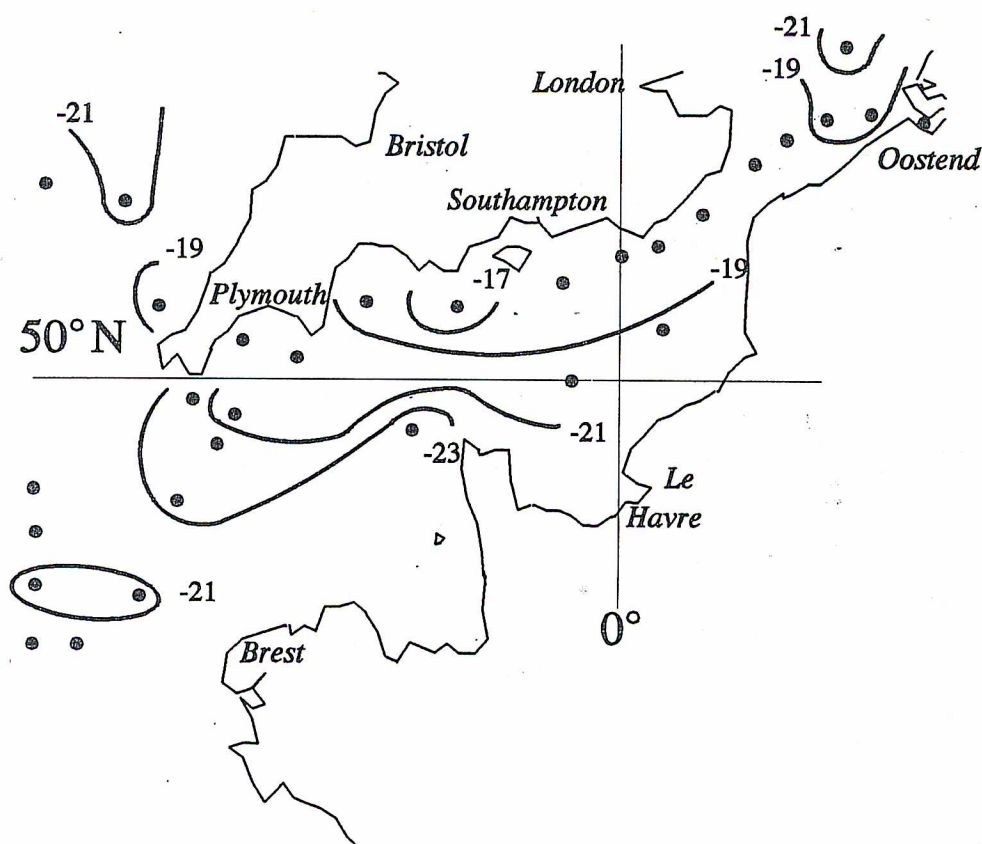


Fig. 1.— Distribution of  $\delta^{13}\text{C}$ , in ‰, observed in June 1991 in the English Channel.

- 1) phytoplankton, with temperature dependant  $\delta^{13}\text{C}$  values [about  $-23\text{‰}$  in our conditions (Fontugne and Duplessy, 1981)],
- 2) marine plants, with higher  $\delta^{13}\text{C}$ , ranging from  $-10$  to  $-20\text{‰}$  [seaweeds and seagrasses (Dauby, 1989)],
- 3) terrestrial plant material ( $\delta^{13}\text{C} \simeq -27\text{‰}$ ).

As can be seen on figure 1, maximum  $\delta^{13}\text{C}$  values (about  $-17\text{‰}$ ) are observed near Solent River Mouth. This could be explained by an input of carbon originating either from marine macrophytes or from human industrial activities, which are particularly developed in the Southampton area. This “pollution effect” could moreover explain the gap in dissolved oxygen values observed in this area (Frankignoulle *et al.*, submitted<sup>2</sup>). On both sides of that maximum,  $\delta^{13}\text{C}$  in suspended matter progressively decreases to reach values (especially Southwesterly) expected for a phytoplankton-based oceanic system. Data measured in the Southern Bight of the North Sea reflect a mixture of planktonic production, anthropogenic and terrigenous inputs.

To illustrate the influence of biological activity and coastal processes on the distribution of surface seawater  $p\text{CO}_2$ , we present here data which have been obtained during transects along the Gulf of Biscay in 1989 and 1990. Table 1 gives, for 6 stations from the Ushant front to Vigo (Spain), the primary



Table 1

Distribution of surface  $p\text{CO}_2$  (in ppmV) along a transect through the Gulf of Biscay, together with surface chlorophyll  $a$  contents (in  $\mu\text{g l}^{-1}$ ) and integrated over depth and time primary production (in  $\mu\text{g C m}^{-2} \text{d}^{-1}$ )

Date	Site	Location	Chl. $a$	Pr. Prod.	$p\text{CO}_2$
13/9/89	Ushant front	N 490332 W 051230	1.41	884	268
7/7/90	Ushant front	N 485911 W 060138	1.18	900	291
3/7/90	Gulf of Biscay	N 510489 W 011996	0.33	153	338
3/7/90	Gulf of Biscay	N 503026 W 003862	0.13	141	337
15/9/89	Gulf of Biscay	N 471950 W 083130	0.11	47	310
10/7/90	Vigo	N 424095 W 092214	4.91	1431	444

production, the chlorophyll  $a$  contents and the partial  $\text{CO}_2$  pressure of surface water. Primary production is integrated over time and depth, taking into account the depth of the mixed layer (density), the nutrients concentration (2 incubation depths), the solar radiation profile (quantameter profile, production calibrated *versus* radiation) and a correction factor (production in dark condition) to calculate production over a 24 hours period. Results given in table 1 can be discussed as follows:

- the Ushant front is characterized by a relatively high chlorophyll  $a$  concentration and production rate. The associated  $p\text{CO}_2$  values are quite far from atmospheric equilibrium (presently about 355 ppmV) as a consequence of dissolved  $\text{CO}_2$  use for organic metabolism. This frontal area then acts as sink for atmospheric  $\text{CO}_2$ .
- the Gulf of Biscay itself has low chlorophyll  $a$  content and primary production. The observed  $p\text{CO}_2$  are higher than those obtained in the frontal area and nearer the atmospheric equilibrium value.
- the near-shore station (Vigo) is characterized by high chlorophyll  $a$  content and important primary production. In spite of this important biological activity, surface  $p\text{CO}_2$  is also very high and far from atmospheric equilibrium. This is obviously due to the important coastal upwelling which takes place along the Spanish and Portuguese Atlantic coast. Such high primary production site is then also an important source of  $\text{CO}_2$  to the atmosphere. As discussed by Frankignoulle *et al.* (submitted<sup>2</sup>), some other coastal areas are also characterized by high production rate associated to high surface  $p\text{CO}_2$  values: the Southern Bight of the North Sea which is influenced by water coming from the Scheldt displayed, in June 1992, chlorophyll concentration as high as  $7.55 \mu\text{g l}^{-1}$ , due to eutrophication, and  $p\text{CO}_2$  value of 407 ppmV, due to the alkalinity of Scheldt water.

### 3.- Air-sea $\text{CO}_2$ exchange measurements.

Air-sea  $\text{CO}_2$  fluxes have been estimated, during diel cycles, using the bell method in June 1992 in the Celtic Sea, above both the continental shelf and slope. Obtained data are reported in figure 2. At the slope station, important fluxes, especially the first day, are observed from air to sea in good agreement with the sign of the  $\text{CO}_2$  gradient. At the shelf station, fluxes are about five times lower than the first day at the slope station. During those measurements, the pH has been monitored in surface water and, in both cases the partial  $\text{CO}_2$  pressure is quite constant ( $\pm 5$  to 10%) during the whole experiment. The observed  $\text{CO}_2$  gradients can thus explain neither the variation of measured fluxes observed on the slope station from the first day to the second, nor the difference between both stations. According to the  $\text{CO}_2$  gradients, fluxes at the slope station

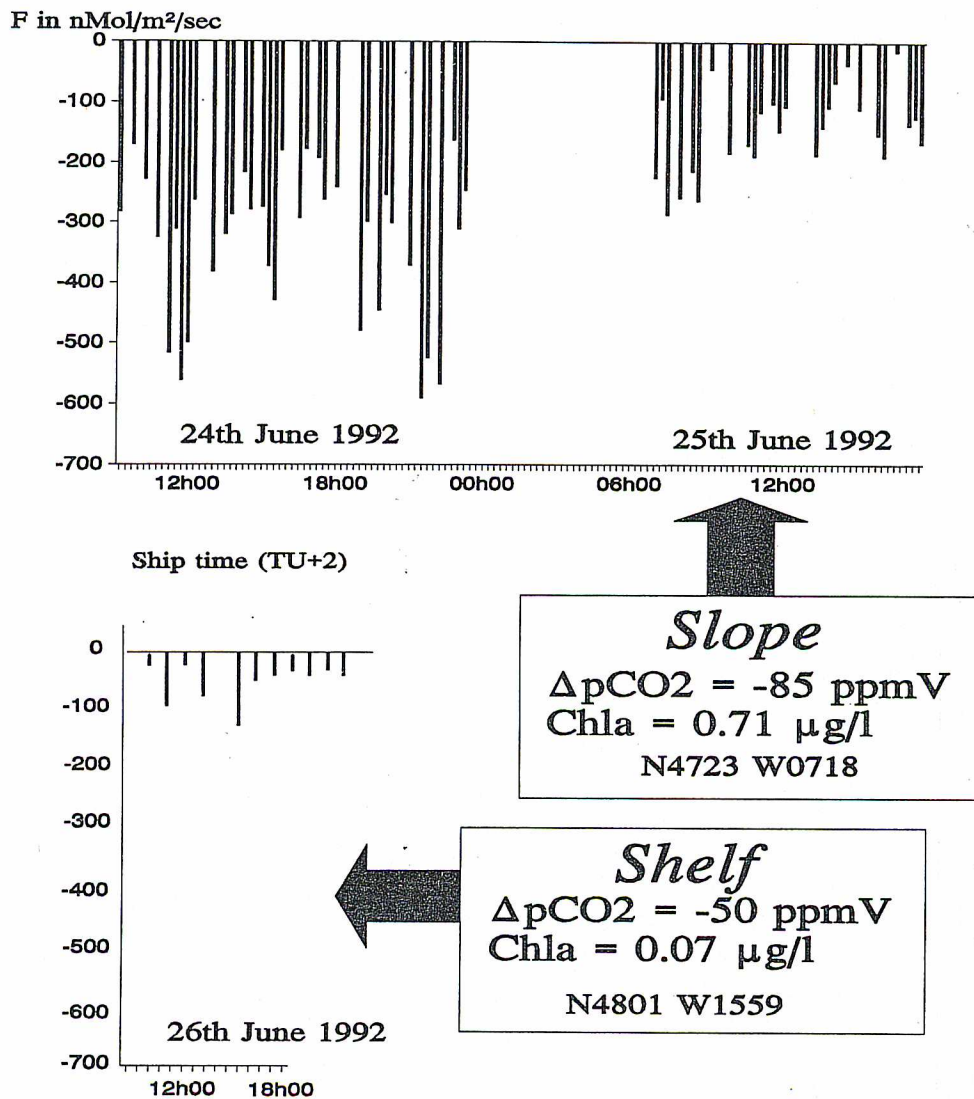


Fig. 2.- Measured air to sea  $\text{CO}_2$  fluxes in the Celtic Sea in June 1992. Negative flux values do correspond to air to sea flux [cf. equation (1)]



should be about twice those at the shelf station. This is roughly observed by considering only the second day at the slope station. As shown on figure 3, high fluxes detected at the slope station are in fact related to the wind speed

### *Air-sea CO<sub>2</sub> exchange (Celtic Sea, Continental slope)*

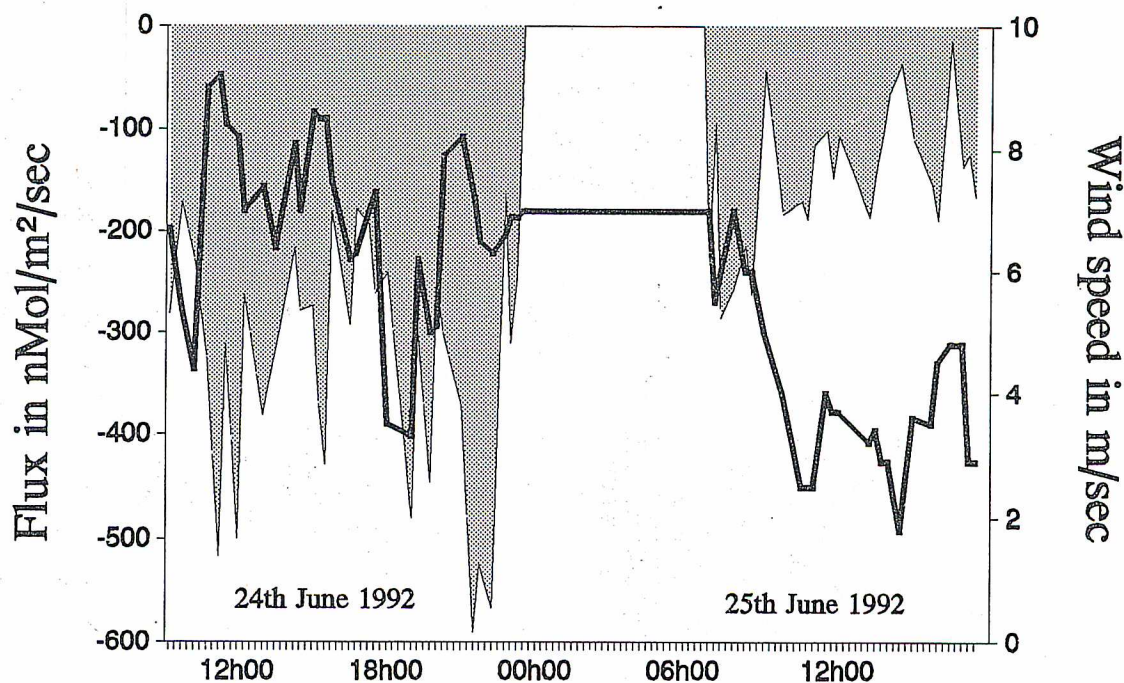


Fig. 3.— Measured CO<sub>2</sub> fluxes (area) at the slope station as a function of wind speed line

which is nearly always higher than  $5 \text{ m s}^{-1}$ , with peaks up to  $9 \text{ m s}^{-1}$ , at the slope station from the beginning of the experiment till about 9 a.m. the second day. Wind speed at the shelf station is lower than  $3 \text{ m s}^{-1}$ . Figure 3 clearly shows that air-sea flux maxima are correlated to wind speed ones. The effect of wind on air-sea exchange have been investigated by wind-tunnel experiments (see *e.g.* Liss and Merlivat, 1986; Wanninkhof, 1992), according to which the gas exchange coefficient [*cf.* equation (1)] is quite constant for wind speed lower than  $5 \text{ m s}^{-1}$  but increases exponentially for higher values. The variation of wind speed during the experiment can thus explain the one observed for air-sea CO<sub>2</sub> flux and results presented here illustrate well the relative importance of both the gradient and the wind speed on the exchanges. It is worth noticing that the used method to detect air-sea fluxes does suppress the direct wind effect on the exchange, due to the presence of the bell. Frankignoulle (1988) has anyway pointed out that the bell takes into account the global turbulence induced by wind in the upper layer and results obtained in the North Sea have shown that flux obtained using the bell is in good agreement with results from wind-tunnel within about 30%.



#### 4.- Conclusions.

Coastal areas can be sites of processes which have a drastic effect on the surface seawater  $p\text{CO}_2$  distribution and hence on air-sea  $\text{CO}_2$  exchange. Data obtained in the Gulf of Biscay show that the Atlantic coastal upwelling (Spain) induces both important biological activity and high  $p\text{CO}_2$  value in the surface layer. In other locations of the Gulf, primary production is associated to low  $p\text{CO}_2$  level in good agreement with the expected effect of production alone. Depending on the river type, riverine input can also generate high production and  $p\text{CO}_2$  oversaturation. The use of  $\delta^{13}\text{C}$  as a tracer for terrestrial input should be improved by the knowledge of  $\delta^{13}\text{C}$  associated to anthropogenic compounds. Air-sea  $\text{CO}_2$  flux measurements carried out in the Celtic Sea have pointed out that wind speed has, as expected, a preponderant effect on the magnitude of the flux.

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