Evaluation of dissolved and particulate arsenic flux in the Dover Strait (*Fluxmanche* programme)

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ABSTRACT

In order to estimate arsenic flux from the English Channel toward the North Sea, 168 samples were obtained from September 1990 to November 1991 along a transect between Boulogne and Folkestone comprising six stations and two depths. Organic and inorganic arsenic were measured in dissolved phase and total arsenic in suspended particles. The mean concentration of dissolved total arsenic in water was 1.38 ± 0.12 $\mu g.l^{-1}$. The variability of measured concentrations was low, particularly for the four intermediate stations. Results were slightly more sensitive for English (1.38 ± 0.16 $\mu g.l^{-1}$) and French (1.35 ± 0.11 $\mu g.l^{-1}$) coastal stations due to exchanges with shallower sediments. Dissolved organic arsenic was nearly negligible from December to March (< 1 %) and maximal from July to September, essentially in the form of dimethylarsinic acid. It constituted about 10 % of dissolved total arsenic and was related to phytoplankton activity. The lowest concentrations of dissolved total arsenic (1.26 ± 0.03 $\mu g.l^{-1}$) were also measured in July. Arsenic in suspended particles (0.16 ± 0.19 $\mu g.l^{-1}$), which accounted for only a small portion (*i.e.* 8.5 %) of the total arsenic in the water column, was more elevated (20 %) near the English coasts (0.30 ± 0.25 $\mu g.l^{-1}$). Mean concentrations for particles were 22.2 $\mu g.g^{-1}$, and the distribution coefficient with water was $\log(K_d) = 4.14 \pm 0.24$. The flux from the English Channel toward the North Sea from 1 October 1990, to 1 October 1991, was 4,442 tons of dissolved arsenic and 412 tons of particulate arsenic.

Oceanologica Acta, 1993. **16**, 5-6, 585-591.

RÉSUMÉ

Évaluation du flux d'arsenic dissous et particulaire dans le détroit du Pas-de-Calais (programme *FluxManche*)

Afin d'estimer le flux d'arsenic qui transite de la Manche vers la Mer du Nord, 168 échantillons ont été prélevés de septembre 1990 à novembre 1991 sur un transect comportant six stations et deux profondeurs, entre Boulogne et Folkestone. On a mesuré l'arsenic organique et inorganique dans la phase dissoute ainsi que l'arsenic total dans les particules en suspension. Dans l'eau, la concentration moyenne en arsenic total dissous est de 1.38 ± 0.12 $\mu g.l^{-1}$. La variabilité des concentrations mesurées est faible, en particulier pour les quatre stations intermédiaires ; elle est un peu plus sensible dans les stations côtières : côte anglaise (1.38 ± 0.16 $\mu g.l^{-1}$), côte française (1.35 ± 0.11 $\mu g.l^{-1}$). Ces variations côtières peuvent être attribuées aux échanges avec les sédiments les moins profonds. L'arsenic organique dissous est presque négligeable de décembre à mars (< 1 %) ; sa présence atteint un maximum de juillet à septembre, essentiellement sous forme d'acide diméthylarsinique. Il avoisine

10 % de l'arsenic total dissous et doit être relié à l'activité phytoplanctonique. C'est aussi au cours du mois de juillet que l'on mesure les teneurs les plus faibles en arsenic total dissous (1.26 ± 0.03 µg.l⁻¹). L'arsenic associé aux particules en suspension (0.16 ± 0.19 µg.l⁻¹) ne représente qu'une faible part de l'arsenic total contenu dans la colonne d'eau (8,5 %). Cette contribution est plus importante à proximité des côtes anglaises (0.30 ± 0.25 µg.l⁻¹), soit environ 20 % du total. Les concentrations exprimées relativement aux particules sont en moyenne de 22.2 µg g⁻¹ et le coefficient de partage avec l'eau est : $log(K_d) = 4.14 ± 0.24$. Le flux annuel de la Manche vers la Mer du Nord, du 1er octobre 1990 au 1er octobre 1991, est de 4 442 tonnes d'arsenic dissous et de 412 tonnes d'arsenic particulaire.

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INTRODUCTION

The estimation of contamination levels in the North Sea is one of the current major concerns of the European scientific community. Due to a residual current, essentially from the English Channel towards the North Sea, elementary fluxes in the Dover Strait play an essential role in this situation. The main objective of the *Fluxmanche* programme, in which scientists with complementary activities work together, is to evaluate residual fluxes entering the North Sea.

The present work, concerning only fluxes of dissolved and particulate arsenic, benefited from parallel studies on the modelling of hydrodynamic (Salomon *et al.*, 1993) and sedimentary (Lafite *et al.*, 1993) fluxes. These studies, based on discrete data, provide daily quantitative estimations of fluxes over a period of one year.

Arsenic is a potentially toxic substance, naturally present in the oceanic environment but likely to be increased by high

inputs from human sources. Its biogeochemical cycle is particularly complex since it involves more than fifteen organic or inorganic arsenical molecules. It accumulates in marine organisms, essentially in the form of arsenosugars in algae and arsenobetaine in fish (Edmonds and Francesconi, 1987). Although localized studies in estuary waters have been published (Howard *et al.*, 1984; Howard and Comber, 1989), the present work is the first to assess the usual arsenic levels and corresponding fluxes of the English Channel.

Our work provides a statistical basis for determining dissolved and particulate arsenic levels in the Dover Strait as well as short- and long-term geographical and temporal variabilities. The analytic method used to measure dissolved arsenic provides data on the chemical forms of arsenic in this environment.

SAMPLING

From September 1990 to November 1991, samples were collected every two months within the scope of the *Fluxmanche* programme. Six stations were sampled regularly, both at the surface and in depth, along a transect from Folkestone to Boulogne (Fig. 1). In addition, in October 1990 and July 1991, multiple samples were obtained in coastal stations in order to determine the influence of tidal cycles. In total, 168 samples were collected and analyzed. Sampling was conducted using a pumping system entirely in teflon (PTFE) and a polyethylene tubing. Water samples were filtered through a Whatman GFC fiberglass filter, placed in 250 ml polyethylene flasks acidified by 0.5 ml of H_2SO_4 20M, stored at 4°C in the dark and analyzed within the following month. All flasks and material required for sampling, filtration and analysis were cleaned beforehand by a 10 % nitric acid solution and then washed in ultrapure deionized water.
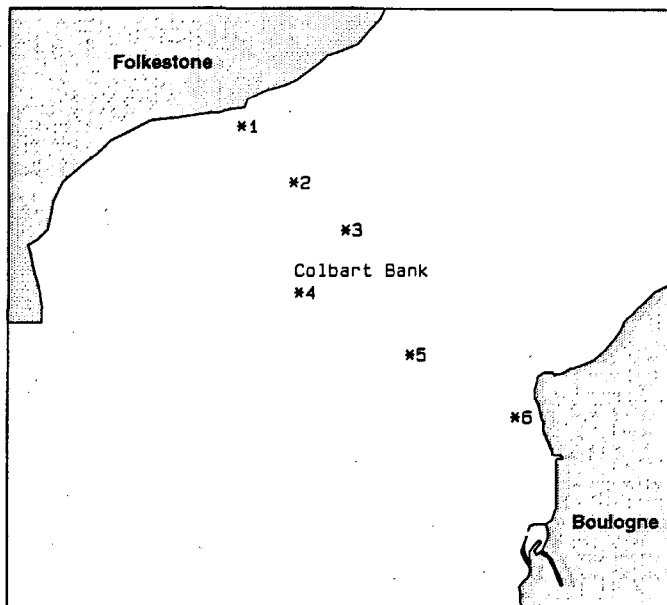

Figure 1

Geographical location of the sampling stations.

Situation géographique des stations de prélèvement.

ANALYSIS

Dissolved arsenic

Dissolved arsenic was analyzed in seawater samples by atomic absorption spectrometry (AAS) according to a

Table 1

Mean data calculated for each survey and each station. Values in italics correspond to the standard deviation of corresponding means (N = number of samples, As(i) = inorganic arsenic, MMA = monomethylarsonic acid, DMA = dimethylarsinic acid, As(t) = dissolved total arsenic, As(p) = particulate arsenic, log(Kd) = log[1 000 *As(p)/As(t)]).

Données moyennes calculées pour chaque campagne et chaque station. Les valeurs en italiques correspondent à la Standard Deviation des moyennes correspondantes (N = nombre d'échantillons, As(i) = As organique, MMA = acide monométhyl arsonique, DMA = acide diméthyl arsinique, As(t) = As total dissous, As(p) = As particulaire, log(K$_d$) = log[1 000 *As(p)/As(t)].

	N	As(i) µg.l⁻¹	MMA µg.l⁻¹	DMA µg.l⁻¹	As(t) µg.l⁻¹	As(p) µg.l⁻¹	As(p) µg.g⁻¹	log(K$_d$)
23 September 1990	29	1.36 *0.11*	0.01 *0.01*	0.07 *0.04*	1.45 *0.12*	0.11 *0.06*	*	*
5 October 1990	6	1.32 *0.03*	0.01 *0.00*	0.07 *0.03*	1.40 *0.02*	0.09 *0.06*	*	*
8 November 1990	12	1.35 *0.04*	0.01 *0.00*	0.04 *0.01*	1.39 *0.05*	0.09 *0.06*	19.8 *7.9*	4.12 *0.20*
4 December 1990	12	1.47 *0.13*	0.00 *0.00*	0.01 *0.01*	1.48 *0.12*	0.13 *0.10*	14.8 *8.9*	3.92 *0.30*
12 January 1991	12	1.44 *0.06*	0.01 *0.01*	0.01 *0.01*	1.46 *0.05*	0.18 *0.13*	19.4 *7.2*	4.10 *0.16*
22 March 1991	12	1.42 *0.06*	0.00 *0.00*	0.01 *0.01*	1.43 *0.06*	0.15 *0.07*	23.8 *3.4*	4.19 *0.17*
16 May 1991	12	1.29 *0.08*	0.00 *0.00*	0.04 *0.04*	1.33 *0.04*	0.31 *0.33*	43.8 *31.2*	4.39 *0.36*
11 July 1991	42	1.11 *0.03*	0.00 *0.00*	0.15 *0.03*	1.26 *0.03*	0.14 *0.12*	19.3 *8.2*	4.14 *0.20*
25 September 1991	12	1.37 *0.13*	0.02 *0.01*	0.12 *0.05*	1.51 *0.10*	0.22 *0.38*	17.4 *8.0*	4.03 *0.15*
21 November 1991	12	1.41 *0.12*	0.00 *0.01*	0.03 *0.01*	1.44 *0.12*	0.30 *0.31*	29.1 *10.3*	4.28 *0.15*
Station 1	39	1.31 *0.18*	0.01 *0.01*	0.07 *0.05*	1.38 *0.16*	0.30 *0.24*	19.9 *10.1*	4.11 *0.25*
Station 2	19	1.38 *0.14*	0.00 *0.00*	0.05 *0.06*	1.43 *0.10*	0.20 *0.15*	24.6 *12.2*	4.20 *0.18*
Station 3	19	1.33 *0.11*	0.00 *0.01*	0.06 *0.06*	1.40 *0.07*	0.14 *0.08*	29.8 *19.2*	4.27 *0.22*
Station 4	33	1.25 *0.15*	0.00 *0.01*	0.10 *0.07*	1.35 *0.10*	0.08 *0.05*	28.4 *18.7*	4.26 *0.25*
Station 5	19	1.37 *0.11*	0.01 *0.01*	0.06 *0.05*	1.44 *0.08*	0.06 *0.05*	18.0 *7.7*	4.06 *0.17*
Station 6	39	1.24 *0.14*	0.01 *0.01*	0.11 *0.06*	1.35 *0.11*	0.13 *0.21*	15.6 *7.1*	4.03 *0.21*
TOTAL	**168**	**1.30** *0.15*	**0.01** *0.01*	**0.08** *0.06*	**1.38** *0.12*	**0.16** *0.19*	**22.2** *14.0*	**4.14** *0.24*

previously described method (Michel *et al.*, 1992). This technique involves the generation of volatile hydrides, low temperature trapping on a liquid nitrogen-cooled chromatography column and elution at a programmed temperature allowing arsines to be separated according to their degree of methylation before AAS detection and quantification.

This technique permitted separate measurement of inorganic arsenic (As^{III} + As^{V}), monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) which, under the action of sodium borohydride, produce respectively arsine, monomethylarsine and dimethylarsine. Although other species of arsenic are probably present at times in estuary seawater (Howard and Comber, 1989), it is generally recognized that these four chemical species constitute the near totality of dissolved arsenic in the oceanic environment, including our study area.

The quality of results obtained with this method was regularly checked during the *Fluxmanche* programme by analysis of the CASS-1 and NASS-2 certified standards of the National Research Council of Canada (NRCC). The typical values for these samples were 1.04 ± 0.07 and 1.65 ± 0.19 µg.l⁻¹ for the respective certified values of 0.98 ± 0.08 and 1.51 ± 0.10 µg.l⁻¹.

Particulate arsenic

The suspended particles were trapped on fiberglass filters (Whatman GFC, 47 mm). Up to 4 l of seawater were used

in order to obtain a minimum of 5 mg of SPM. These samples were mineralized by a 5 ml mixture of nitric, sulphuric and perchloric acids (8:1:1 v/v - quality Merk Suprapur) at a final temperature of 205°C on a dry bath, in borosilicate glass vessel. An aliquot of the mineralized solution was analyzed after redissolution into 10 ml of deionized water, using the same technique as for seawater. Blank values were obtained using the same procedure on clean filters. They were low and taken into account for the calculations. With respect to the mineralization used, the measurement obtained was that of particulate total arsenic.

RESULTS AND DISCUSSION

Results for the 168 samples are presented in statistical form in Table 1. Their classification by survey date or sampling station indicates seasonal and geographical variations. Global mean concentrations were 1.30 ± 0.15 µg.l⁻¹ for inorganic arsenic, 0.005 ± 0.008 µg.l⁻¹ for MMA and 0.079 ± 0.063 µg.l⁻¹ for DMA. These concentrations are quite comparable with those measured in Atlantic waters by Waslenchuk (1978), Burton *et al.* (1980) and Cossa *et al.* (1992). Concentrations measured in the Seine River in 1990 and 1991 are also quite comparable, though with a greater range of variations related to differences in river flow (Michel *et al.*, 1992)

The mean arsenic concentration in suspended particles was 22.2 ± 14.0 µg.g⁻¹. With respect to the proportion of

particulate matter, the mean contribution was 0.16 ± 0.19 $\mu g.g^{-1}$ of sea water. This great variability was due to the quality of the suspended matter and probably to the presence of plankton in greater or lesser proportion. Weather and hydrologic conditions may also have affected these parameters.

Geographical variations

Individual concentrations of dissolved total arsenic are indicated in Figure 2 for each station and sampling level. No significant differences were noted between samples taken at the surface and at depth. Nor did there appear to be any significant variations with respect to the geographical location of each station. The results of the statistical analysis (Tab. 1) confirmed this homogeneity since mean concentrations of dissolved total arsenic for each station were all between 1.25 ± 0.10 and 1.44 ± 0.08 $\mu g.l^{-1}$. The same homogeneity (no notable geographical variations) was apparent in inorganic arsenic, MMA and DMA concentrations considered separately.

Particulate arsenic was more markedly differentiated between the stations. All measurements are indicated in Figure 3 for each station and sampling level. It may be noted that the coastal stations had maximum concentrations markedly above mean values. It is also noteworthy that these elevated values were measured in the spring and summer for English coastal stations 1 and 2, and in winter for station 6 near the French coast. The higher means for particulate arsenic in coastal waters were due to the presence of greater quantities of suspended matter. In fact, the opposite is true for arsenic levels considered in terms of $\mu g.g^{-1}$ of suspended particulate matter (SPM), with higher concentrations for stations 2, 3 and 4. This clearly indicates the preponderance of arsenic-poor mineral particles in coastal areas and, conversely, of plankton particles in the middle of the Dover Strait.

Variations during a tidal cycle

In the context of the September 1990 and July 1991 surveys, samples were taken during an entire tidal cycle at stations 1 and 6. In general, no significant variations were noted in concentrations of dissolved total arsenic within this short time period. For surface and in-depth measurements, mean values were 1.28 ± 0.02 $\mu g.l^{-1}$ for station 1 in July 1991, and 1.47 ± 0.04 and 1.25 ± 0.03 $\mu g.l^{-1}$ for station 6 in September 1990 and July 1991 respectively. However, there was a notable abnormality for station 1 in September 1990, with a mean concentration of 1.45 ± 0.21 $\mu g.l^{-1}$. This far greater variability is apparent in Figure 4, which shows a very marked decrease in concentrations immediately after low-tide slack.

This variability noted during a tidal cycle on the English coast was reflected in the concentrations measured for this station throughout the study period. The sampling procedure used provided no satisfactory explanation for this phenomenon, which may have been due to a resuspension of sediments in an area where water depth is not greater than 15 m during low tide. This could have resulted in a process of adsorption or release not apparent at the other stations.
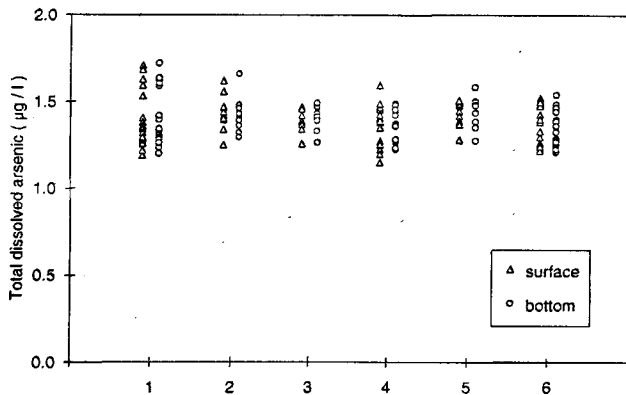

Figure 2

Dissolved arsenic concentrations according to station and sampling level.

Concentrations en arsenic dissous en fonction de la station et du niveau de prélèvement.
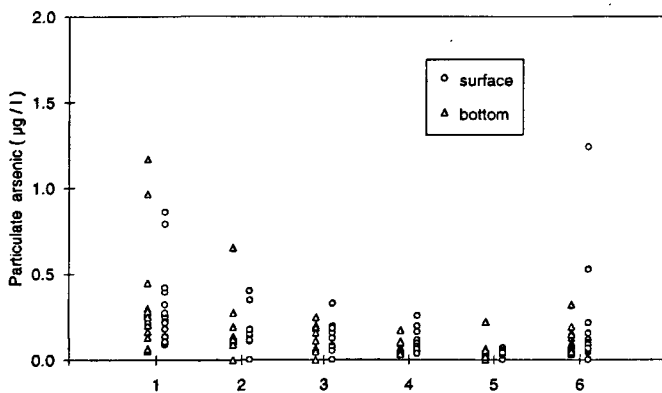

Figure 3

Particulate arsenic concentrations according to station and sampling level.

Concentrations en arsenic particulaire en fonction de la station et du niveau de prélèvement.
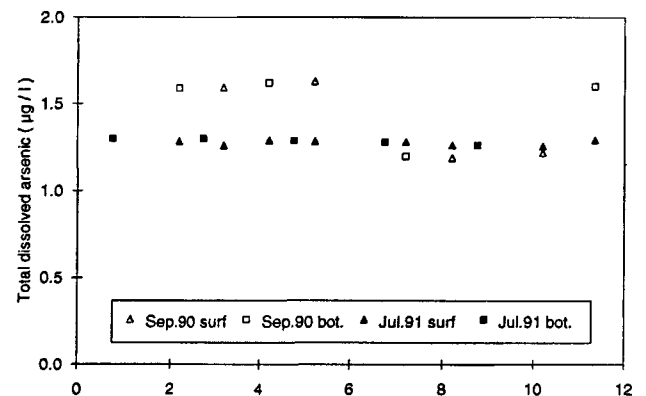

Figure 4

Variations in dissolved arsenic concentrations at station 1 during two tidal cycles.

Variations de la concentration en arsenic dissous à la station 1 au cours de deux cycles de marée.
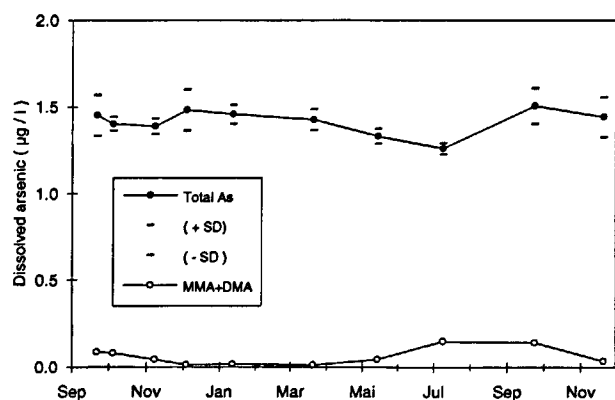
Figure 5

Seasonal variations in mean concentrations of organic arsenic and dissolved total arsenic in the Dover Strait from September 1990 to November 1991.

Variations saisonnières de la concentration moyenne en arsenic organique et en arsenic total dissous de septembre 1990 à novembre 1991, dans le détroit du Pas de Calais;

Variations during an annual cycle

The mean concentration in dissolved inorganic arsenic was maximal in winter (1.47 ± 0.13 µg.l^{-1}), decreased very markedly in May and became minimal in July (1.12 ± 0.03 µg.l^{-1}). For organic arsenic, the situation was just the opposite. Mean DMA was minimal in December (0.01 ± 0.01 µg.l^{-1}), increased in May and became maximal in July (0.15 ± 0.03 µg.l^{-1}). As it is well known that phytoplankton produce MMA and DMA (Andreae, 1978; Howard *et al.*, 1982), the variations related to the spring and summer activity of these organisms are entirely normal.

As a result of these opposed variations in inorganic and organic arsenic, there was a certain stability in total dissolved arsenic during the annual cycle. Mean dissolved arsenic concentration was minimal in summer (1.26 ± 0.03 µg.l^{-1}) and maximal in winter (1.51 ± 0.10 µg.l^{-1}; Fig. 5). The summer deficit was certainly related in part to the accumulation of arsenic by plankton. It is also possible that a still unidentified portion of organic arsenic cannot be analyzed by classical methods (Howard and Comber, 1989), although this hypothesis remains to be confirmed.

Distribution coefficient between dissolved and particulate phases

It was difficult to compare our values for total arsenic concentration in particulate matter with those previously reported for estuary zones. The mean concentration (22.2 ± 14.0 µg.g^{-1}) was higher than expected. The highest concentrations were reached in May when phytoplankton activity was most intense, with exceptional levels then of 90 µg.g^{-1} for stations 3 and 4 where dilution by nonbiogenic sediments was least intense. Moreover, these maximal concentrations were obtained for surface waters. The means per station for the entire study period also reflect this dilution effect related to the presence of terrigenous particles at stations nearest to the coast. For the

station on the French coast, the mean concentration was only 0.13 ± 0.21 µg.g^{-1}.

The variability in the distribution coefficient (K_d) between dissolved and particulate phases was thus rather elevated. Mean log(K_d) was 4.14 ± 0.24, with the highest values occurring at stations 2, 3 and 4 (Tab. 1). For arsenic, the notion of a distribution coefficient should be considered with due caution since plankton metabolization occurs and not simple adsorption equilibrium.

Arsenic distribution between the particulate and dissolved phases accounts for the modest contribution of particles to global arsenic flux in our data for the Dover Strait.

FLUX CALCULATIONS

To account for the geographical variability of hydrologic and sedimentary parameters in flux calculations, as well as the arsenic concentrations in dissolved and particulate phases, the Dover Strait was divided into ten parallel sections (Tab. 2). Dissolved and particulate arsenic concentrations for each section were interpolated as indicated in Table 2 where mean concentration in the water column = (surface concentration + near-seafloor concentration)/2.

Data acquisition was not as frequent for arsenic measurements as for hydrologic or sedimentary parameters. Salomon *et al.* (1993) have indicated the importance of daily variations in residual hydrologic flow in the Dover Strait in relation to tidal amplitude and weather conditions. Lafite *et al.* (1993) have shown that to a lesser degree particle fluxes were also variable at short term for the same reasons. Since arsenic fluxes as a function of these two

Table 2

Definition of the geographical sections used for flux calculations (after Salomon et al., 1993).

Définition des sections géographiques utilisées pour le calcul des flux (adapté de Salomon *et al.*, 1993).

#Cell	Latitude deg.dec.	Longitude deg.dec.	Station from which As data used
1	51.0598	1.1894	1
2	51.0431	1.2411	(1+2)/2
3	51.0264	1.2929	2
4	51.0097	1.3188	(2+3)/2
5	50.9763	1.3188	3
6	50.9429	1.3706	(3+4)/2
7	50.9262	1.3965	(4+5)/2
8	50.8928	1.4483	5
9	50.8594	1.5000	(5+6)/2
10	50.8260	1.5518	6

parameters should be evaluated at least daily, it proved essential to estimate the most probable arsenic concentrations between the surveys conducted every two months. We chose a linear interpolation of the missing data for concentrations in dissolved and particulate phases, basing this procedure on the variability results during the annual cycle discussed above.

Evaluation of daily fluxes was performed according to the following equations

$$\text{Dissolved flux} = \sum_{i=1}^{i=10} W_i \cdot CD_i$$

$$\text{Particulate flux} = \sum_{i=1}^{i=10} P_i \cdot CP_i$$

where W_i = daily residual water flux in the section (i) (Salomon *et al.*, 1993)

CD_i = mean concentration of dissolved total arsenic in the section (i)

P_i = daily residual particle flux in the section (i) (Lafite *et al.*, 1993)

CP_i = mean concentration of particulate arsenic in the section (i)

The results obtained gave the changes in dissolved and particulate arsenic fluxes for the entire study period (Fig. 6 and 7). There was a very marked variation in these daily fluxes, mainly resulting from differences in water and particulate flows related to shifts in dominant winds. However, it is noteworthy that the highest concentration of particulate arsenic in May 1991 was due to the high arsenic concentration in particulate matter at that time.

Annual dissolved and particulate arsenic fluxes were also calculated by simply totaling the corresponding daily fluxes for the period from 1 October 1990 to 1 October 1991. The flux of dissolved total arsenic from the English Channel towards the North Sea during this period was 4,442 tons, whereas the corresponding flux of particulate arsenic was only 412 tons (*i.e.*, 8.5 % of total flux) The accuracy of these calculations for the annual period considered can be estimated as within 20 %. However, as these estimations depend essentially on weather conditions and water flows which can vary considerably (Salomon *et al.*, 1993), their reproducibility for other annual periods would not be as exact.

These flux measurements can also be compared with those calculated more simply on the basis of annual water or particulate fluxes multiplied by the corresponding annual mean concentrations. For the same annual period (1 October 1990 to 1 October 1991), dissolved arsenic flux would thus be 4,371 metric tons (W = 3,163 km³ and CD = 1.38 µg.l⁻¹). For particulate arsenic, annual flux would be 424 tons (P = 1.91 E + 10 tons and CP = 22.2 µg.g⁻¹). It may be noted that the fluxes calculated in this manner do not differ significantly from those determined by the preceding method. This similarity is attributable to the fact that both calculations were based on the same water and particulate fluxes and that the mean concentrations in dissolved and particulate arsenic were determined on a very significant basis and show only slight variability.

CONCLUSIONS

This study provides the first statistical measurements of dissolved and particulate arsenic concentrations (respectively 1.38 µg.l⁻¹ and 22.2 µg.g⁻¹) in English Channel waters. Variability during the tidal cycle was sometimes considerable, especially on the English coast. Seasonal variability in dissolved total arsenic concentrations was clearly apparent, although never greater than a mean 16 %. This variability is essentially attributable to phytoplankton production parallel in spring and summer to an increase in MMA and DMA concentrations and a mobilization of arsenic by biological matter. Geographical variability was of little importance.

The integration of the present work into the global programme of flux studies in the Dover Strait enabled us to benefit from data acquired simultaneously for water and particulate fluxes and to develop systematic deductions about dissolved and particulate arsenic fluxes. Besides providing a daily representation of
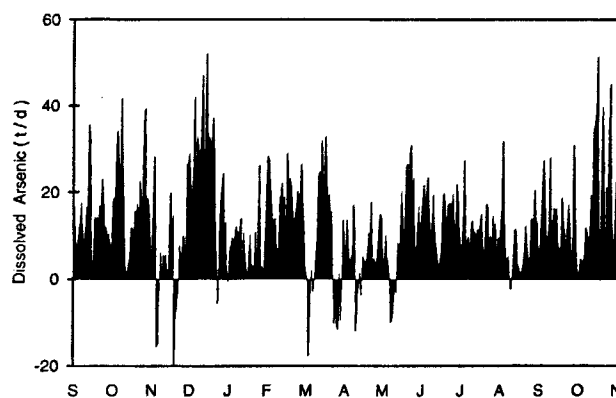

Figure 6

Daily variations in dissolved total arsenic flux in the Dover Strait from September 1990 to November 1991.

Variation journalière du flux d'arsenic total dissous de septembre 1990 à novembre 1991, dans le détroit du Pas de Calais.
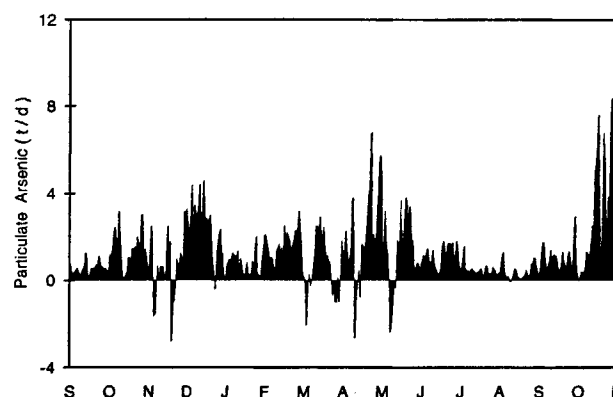

Figure 7

Daily variations in particulate arsenic flux in the Dover Strait from September 1990 to November 1991.

Variation journalière du flux d'arsenic particulaire de septembre 1990 à novembre 1991, dans le détroit du Pas de Calais.

fluxes, we were able to calculate the annual fluxes during the study period. It is thus estimated that 4,442 and 412 tons of dissolved and particulate arsenic respectively transited through the English Channel toward the North Sea between 1 October 1990 and 1 October 1991.

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