Cadmium and mercury in Seine Estuary flounders and mussels: the results of two decades of monitoring

K. Nakhlé, D. Cossa, D. Claisse, B. Beliaeff, and S. Simon


The flounder (Platichthys flesus) is a flatfish that inhabits marine coastal environments, especially estuaries. It is an alternative quantitative biological indicator to the common marine mussel (Mytilus spp.), which is currently used as a sentinel species to monitor chemical contamination in numerous monitoring programmes. Findings from two decades of monitoring cadmium (Cd) and mercury (Hg) using both sentinel species in the Seine Estuary (France) are reported. For comparison, time-series of water concentrations for the same two metals at the mouth of the River Seine are given. Cd concentrations in the liver of the fish and in the soft tissue of mussels show similar temporal trends, consistent with the major temporal variations of Cd concentrations recorded in river water and with changes in industrial discharge of Cd (phosphogypsum waste) within the Seine Estuary. On the other hand, Hg concentrations in the muscles of flounders show temporal variations with no link to that observed in mussels or fluvial Hg contributions, which are in fact nearly covariant. It is concluded that optimization of the use of flounders as sentinel organisms for monitoring temporal trends of metal contamination in estuarine environments requires in-depth knowledge of its ecology within the area studied. An adapted sampling strategy based on this knowledge should provide results that are easier to interpret.

Keywords: cadmium, estuary, flounder, mercury, monitoring, mussel watch, Platichthys flesus.

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K. Nakhlé, D. Cossa, D. Claisse, and B. Beliaeff: IFREMER, Centre de Nantes, BP 21105, F-44311 Nantes Cedex 03, France. S. Simon: Cellule de Suivi du littoral Haut-Normand, 16, quai C. de la Vigne, F.76000 Le Havre, France. Correspondence to D. Cossa: tel: +33 240 374176; fax: +33 240 374075; e-mail: d.cosse@ifremer.fr

Introduction

Despite the complexity of the processes involved in “bio-accumulation” of a substance or element in a living organism, the use of quantitative bio-indicators appears to be a reliable method for evaluating the bio-availability of contaminants and for monitoring contamination in the environment (Phillips and Rainbow, 1993). The organisms used for such bio-monitoring must meet certain conditions, among which are their capacity to bioconcentrate the substance being studied and their abundance and mobility, to allow good coverage and geographic representation (Cossa, 1989). For these reasons, the French Mussel Watch, the “Réseau National d’Observation” (RNO), uses the soft tissue of mussels and oysters for monitoring chemical contaminants along the French coasts (Claisse, 1989; www.ifremer.fr/envlit). However, certain international programmes also recommend the use of fish such as flatfish. This is the case of the flounder (Platichthys flesus), a fish common to coastal regions and estuaries in the monitoring zones of the eastern North Atlantic covered by the Oslo–Paris Convention (OSPAR; http://www.ospar.org) (Masson, 1987). The life history of P. flesus is characterized by three stages. The pelagic larval stage is found offshore and the juveniles develop in brackish and freshwater environments until they become sexually mature (Wheeler, 1969). Once reproduction has been achieved, larvae disperse around the coast or return into the estuary. Despite these migrations, flounders are considered to be good indicator, or sentinel, species for monitoring contamination by metallic and organic contaminants (Jensen and Cheng, 1987; Cossa et al., 1992; Leah et al., 1992; Voight, 1999, 2002). Those working on the organotropism of metals in flounders have shown that mercury (Hg) tends to accumulate in muscles, and cadmium (Cd) preferentially in the liver (Julshamn and Grahl-Nielsen, 1996). Other studies have looked at allometric relationships between the size of fish and the concentration of contaminants. In the case of Hg in the muscle of flounders, positive correlations with size or age of the fish have been documented by Jensen (1982), Luten et al. (1987), Marthinussen and Staveland (1990), and Collings et al. (1996). However, no clear trend has been ascertained for Cd in liver.

Here, we describe the findings from 18 y of data acquisition relating to the concentration of Hg in the muscle and Cd in the liver of flounders from Seine Bay (adjacent to the English Channel) collected between 1986 and 2003. Flounders in Seine Bay and the Seine Estuary are abundant, and their ecology has been well documented (Cellule de suivi du littoral Haut-Normand, 1997; Miramand et al., 1998; Minier et al., 2000). Two specific objectives have been pursued: (i) variations in the

Present address of K. Nakhlé: Centre National des Sciences Marines, PO Box 534, Batroun, Lebanon.

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concentrations of Cd and Hg in the flesh of flounders of different size and (ii) possible temporal patterns in concentrations, and their attempted interpretation. Achieving the first objective should maximize the possibility of attaining the second. Indeed, finding a way to minimize the effect of size on metal concentrations in fish tissue will favour the possibility of indicating potential temporal trends in metal concentration. The results are compared with the temporal trends in Cd and Hg concentrations in mussels and water measured in the Seine Estuary. The survey is part of the RNO, which provides the data required to monitor the quality of coastal water to the Working Group on Concentrations, Trends and Effects of Substances in the Marine Environment (SIME) of the OSPAR Convention (Anon., 1994; www.ospar.org).

Survey site
Seine Bay (Figure 1) covers a total surface area of ~4000 km². Open to the English Channel, its depth does not exceed 30 m and tidal currents are strong. The bay receives freshwater from the River Seine, at an average rate of flow of 490 m³ s⁻¹. The Seine basin, ~75 000 km², is home to 15 million inhabitants and drains areas that are very much subjected to agricultural, industrial, and urban activities. Contamination of the estuary and the bay by mercury has been described by Coquery et al. (1997) and Cossa et al. (2003), and Cd contamination by Chiffoleau et al. (2001). In short, Cd contamination in the Seine Bay is largely attributable to local inputs of phosphogypsum, a byproduct of the phosphoric acid industry. Three phosphoric acid plants were present in the estuary. Two located near Rouen discharged their wastes into Seine Bay during the period 1974–1987. The other, located at Le Havre, discharged its waste through a pipe (Figure 1), and was closed in 1992. Diffuse sources of Cd from the drainage basin have also been revealed by analysing the Seine waters upstream from Rouen (Idlafkih et al., 1995; Thévenot et al., 1998). For Hg, the diffuse inputs from the drainage basin dominate. According to Cossa et al. (2003a), although the annual Hg riverine inputs varied between 500 and 1500 kg, the local discharge within the estuary did not exceed 150 kg.

Material and methods
Sampling and pre-treatment
Flounders
Flounders were caught by bottom trawlers operating in the Seine Estuary between Le Havre and Honfleur. Small fish were caught close to the Channel, opposite the river mouth on clay substrata, and larger fish in water to the south in areas of sandy substrata (Figure 1). Both sites were trawled during the 18-y monitoring period (1986–2003).

Sampling followed the guidelines of the OSPAR monitoring programme (Anon., 1994). Three trawls were carried out annually within a period of 2 d in November or December, at the same locations (Figure 1). A sample of 25 fish was taken from the catch to obtain a homogenous spread of individuals within the widest size range possible and to provide size-dependent stratified sampling. To avoid contaminating samples, each fish was placed alone in a polyethylene bag inside an insulated box, and transported at +4°C. At the laboratory, each fish was measured and weighed, then samples were frozen and kept at −20°C until dissection. Under these conditions, the time elapsed from the date of sampling and analysis was not a critical factor (Lafleur, 1973). To minimize the risk of contamination, dissection was carried out under a laminar airflow hood fitted with an HEPA filter (0.2 μm). Fish were defrosted just before dissection, and the dissection was carried out with titanium, stainless steel, or polyethylene utensils, rinsed with Milli-Q® water. During the operation, the fish was laid on a polyethylene plate that was rinsed between each sample with Milli-Q® water. Subsamples were stocked in glass vials that had been washed in diluted hydrochloric acid beforehand (1:10; v:v) and rinsed with Milli-Q® water. The dorsal fillet and the liver were separated and placed in an identified vial. All samples taken were freeze-dried, kept at +4°C, and held in the dark until analysis.

Mussels
Mussels (Mytilus edulis) were collected in the Seine Estuary four times each year for the past 20 y within the RNO programme at

Figure 1. Seine Estuary and Seine Bay. The black arrows indicate the location of trawls for collecting flounders. The asterisk marks the location of the outlet of the submarine phosphogypsum discharge pipe.
three stations (Villerville, Le Havre, La Hève, Figure 1) (Claisse, 1989). At each site, the soft tissues of 50 mussels from 30 to 60 mm shell length were pooled and analysed. Sampling was carried out according to the guidelines of the OSPAR monitoring programme, described in detail by Claisse (1989).

Water
Surface water was sampled twice each month at the entrance of the Seine Estuary using clean procedures (Chiffoleau et al., 2001). The water samples were collected in acid-cleaned polyethylene bottles, then filtered in the laboratory (0.45 μm, LCR® or Nucleopore® membranes) under a laminar flow hood. The filtrate was stored acidified (0.4% v/v, HCl Suprapur®) in acid-cleaned Teflon (FEP) bottles until analysis.

Analyses
Biota
Hg determination in biota was carried out on an aliquot section of the dried muscle by atomic absorption spectrophotometry, using an automatic mercury analyser (AMA—254, Altec) after dry digestion according to the technique described by Cossa et al. (2002). The accuracy and the reproducibility of the method were established using certified fish muscle reference material (DORM–1; National Research Council of Canada). The certified values (0.80 ± 0.07 mg kg⁻¹) were reproduced (0.85 ± 0.01 mg kg⁻¹) within the confidence limits. Repeatability varied from 1% to 7% depending on the concentration of the sample. The detection limit was 0.007 mg kg⁻¹ [dry weight (d.w.)]. Cd determination was based on measurement by graphite furnace atomic absorption spectrophotometry with Zeeman correction (SpectrAA600, Varian) after wet mineralization of tissues by concentrated HNO₃, following the method described by Cossa and Bourget (1980). The analytical conditions were those published by Chiffoleau et al. (2003). The accuracy and the reproducibility were determined on certified fish liver reference tissue (DOLT–1; National Research Council of Canada). The coefficient of variation (CV = s.d. × 100/mean) on 19 replications was 5%, and the reproduced value average obtained was 4.2 ± 0.2 mg kg⁻¹ for a certified value of 4.18 ± 0.28 mg kg⁻¹. The detection limit was 0.02 mg kg⁻¹ (dry weight).

Water
Metal determinations in water and particulate material were performed according to the analytical procedures described by Chiffoleau et al. (2001) and Cossa et al. (2003b) for Cd and Hg, respectively. Detection limits, defined as 3 × s.d. of the blanks, were 1 ng l⁻¹ for Cd and 0.1 ng l⁻¹ for Hg, with repeatabilities of both better than 12%. The accuracies of the dissolved metal determinations were successively verified using SLRS–2, –3, and –4, and ORMS–3 certified reference materials (National Research Council of Canada) for Cd and Hg, respectively. For the particulate phase, certified sediment samples (MESS–2; National Research Council of Canada) were used. For Cd (using the current SLRS–4), the reproduced value averaged 14 ± 2 ng l⁻¹ for a certified value of 12 ± 2 ng l⁻¹. For Hg using ORMS–3, the average value for replicate analyses was 12.8 ± 1.5 ng l⁻¹ for a certified value of 12.6 ± 1.1 ng l⁻¹. Using MESS–2 for the particulate phase, the repeatabilities were 8% for Cd and 7% for Hg. The calculated average of the determinations obtained for Cd on MESS–2 was 0.28 ± 0.02 mg kg⁻¹ for a certified value of 0.25 ± 0.04 mg kg⁻¹, and for Hg 0.089 ± 0.002 mg kg⁻¹ for a certified value of 0.092 ± 0.009 mg kg⁻¹. The detection limits in the particulate matter were 0.01 and 0.007 mg kg⁻¹ for Cd and Hg, respectively.

Results
Flounder
Over an 18-y period between 1986 and 2003, 354 flounders were sampled, processed, and analysed in the course of 14 annual samplings. There was no fishing in 1997, 1998, 2000, or 2001.

The lengths of the fish studied measured between 170 and 430 mm. The CV of average sizes of fish sampled was ~5%. The lowest annual average was 237 mm (1993), the highest 297 mm (1987), and the general average was 272 mm (Table 1). The annual series were differentiated by their maximum size. As an example, the largest flounder in 1993 measured 310 mm, in 1994 it measured 430 mm.

The relationship commonly used to describe the allometric variation in length (L) against weight (W) is $W = aL^b$. The regression coefficients ($b$) of the allometric relationship for males (2.85, Figure 2) were similar to the $b$ value of the

<table>
<thead>
<tr>
<th>Year</th>
<th>Length (mm)</th>
<th>Cd in liver (mg kg⁻¹, d.w.)</th>
<th>Hg in muscle (mg kg⁻¹, d.w.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1986</td>
<td>284 ± 64 (185–410)</td>
<td>0.50 ± 0.46 (0.07–1.83)</td>
<td>0.64 ± 0.59 (0.15–3.15)</td>
</tr>
<tr>
<td>1987</td>
<td>297 ± 71 (185–405)</td>
<td>1.15 ± 1.82 (0.15–8.11)</td>
<td>0.60 ± 0.49 (0.16–2.41)</td>
</tr>
<tr>
<td>1988</td>
<td>283 ± 67 (185–415)</td>
<td>1.08 ± 0.74 (0.22–3.02)</td>
<td>0.93 ± 0.73 (0.10–2.54)</td>
</tr>
<tr>
<td>1989</td>
<td>282 ± 68 (180–395)</td>
<td>0.93 ± 0.86 (0.13–3.73)</td>
<td>0.61 ± 0.52 (0.17–2.41)</td>
</tr>
<tr>
<td>1990</td>
<td>254 ± 57 (175–355)</td>
<td>1.42 ± 1.98 (0.30–9.56)</td>
<td>0.54 ± 0.53 (0.18–2.67)</td>
</tr>
<tr>
<td>1991</td>
<td>275 ± 54 (190–370)</td>
<td>0.82 ± 0.69 (0.11–3.62)</td>
<td>0.41 ± 0.63 (0.10–3.27)</td>
</tr>
<tr>
<td>1992</td>
<td>272 ± 62 (180–395)</td>
<td>0.58 ± 0.32 (0.13–1.46)</td>
<td>0.26 ± 0.22 (0.06–0.93)</td>
</tr>
<tr>
<td>1993</td>
<td>237 ± 35 (190–310)</td>
<td>0.33 ± 0.29 (0.01–1.16)</td>
<td>0.55 ± 0.25 (0.07–0.99)</td>
</tr>
<tr>
<td>1994</td>
<td>273 ± 67 (170–430)</td>
<td>0.63 ± 0.85 (0.04–2.44)</td>
<td>0.35 ± 0.36 (0.01–1.20)</td>
</tr>
<tr>
<td>1995</td>
<td>259 ± 51 (195–375)</td>
<td>0.29 ± 0.33 (0.09–1.75)</td>
<td>0.33 ± 0.17 (0.10–0.95)</td>
</tr>
<tr>
<td>1996</td>
<td>249 ± 35 (195–305)</td>
<td>0.34 ± 0.26 (0.09–1.18)</td>
<td>0.42 ± 0.17 (0.13–1.00)</td>
</tr>
<tr>
<td>1999</td>
<td>275 ± 58 (185–380)</td>
<td>0.52 ± 0.76 (0.06–3.03)</td>
<td>0.56 ± 0.41 (0.24–2.16)</td>
</tr>
<tr>
<td>2002</td>
<td>268 ± 52 (186–359)</td>
<td>0.26 ± 0.19 (0.08–0.81)</td>
<td>0.47 ± 0.23 (0.16–1.34)</td>
</tr>
<tr>
<td>2003</td>
<td>276 ± 60 (182–405)</td>
<td>0.32 ± 0.20 (0.11–0.90)</td>
<td>0.55 ± 0.34 (0.22–1.61)</td>
</tr>
<tr>
<td>1986–2003</td>
<td>272 ± 59 (170–430)</td>
<td>0.66 ± 0.95 (0.01–9.56)</td>
<td>0.52 ± 0.46 (0.01–3.27)</td>
</tr>
</tbody>
</table>

Minimum and maximum values are given in parenthesis.
relationship obtained for females (2.96, Figure 2), both close to 3, the value determined by Deniel (1981).

**Mercury**

The concentrations of mercury in the dorsal muscle of the flounder ranged from 0.01 to 3.27 mg kg\(^{-1}\) in dry tissue weight (d.w.). This concentration range for flounder muscles from the same site is larger than the ranges previously published for the species (Table 2). The maximum permitted mercury concentration for sale of this type of fish was set by European Directives 466/2001 and 93/351/EEC at 0.5 mg kg\(^{-1}\) (wet weight), ~2.5 mg kg\(^{-1}\) (d.w.). Only 4 of the 362 samples studied surpassed this level, and these abnormal amounts were in large flounders (>347 mm) (Figure 3).

The frequency distribution of mercury concentrations was asymmetric, with a “Skewness” asymmetry coefficient of 2.94. Log\(_{10}\) transformation of the data produced a normal distribution for the Hg data. Consequently, calculations were carried out on concentration logarithms. Figure 3 illustrates the statistically significant correlation that existed between mercury concentrations and the length of flounders (\(r^2 = 0.28, n = 346\)). The concentrations in mercury remained stable up to a size of 300 mm, and above this, there was a considerable increase in concentrations in individual fish, but no difference in this relationship with the sex of the fish (Figure 3). To circumvent Hg concentration vs. length relationships when studying temporal patterns, we used only data from fish <300 mm in the geometric averages (\(m_g\)) calculation. Figure 4 indicates the temporal variations of annual \(m_g\) of mercury concentrations in the muscle of flounders (length <300 mm) between 1986 and 2003, with 95% confidence intervals. The temporal evolution shows relative stability of the concentrations, with minimum concentrations in 1991 and 1992.

**Cadmium**

The minimum concentration observed in the liver of the flounders was 0.01 mg kg\(^{-1}\) (d.w.) and the maximum was 9.56 mg kg\(^{-1}\) (d.w.). There is no quality standard for Cd in the liver of fish in the European Directive covering cadmium and mercury (466/2001). If we take the value established for shellfish (1 mg kg\(^{-1}\) wet weight), taking into account the percentage of water within the samples of 75% (the average calculated from all liver samples after freeze-drying), this “guide value” expressed in d.w. would be 4 mg kg\(^{-1}\). Four samples out of a total of 327 exceeded this value (Figure 5), and again they corresponded to large specimens. However, big fish did not systematically have high Cd concentrations (Figure 5; \(r^2 = 0.09, n = 327\)).

Comparisons with similar measurements performed in flounder livers from other European regions monitored in the ICES programme (Jensen and Bro-Rasmussen, 1992) indicate that the flounders from the Seine Estuary were more contaminated. This

![Figure 2. Relationships between total length and weight of the sample flounders.](https://academic.oup.com/icesjms/article-abstract/64/5/929/640482/510256/4481871a.png)

**Table 2.** Range of Cd and Hg concentrations in flounders from various regions of the North Atlantic.

<table>
<thead>
<tr>
<th>Location</th>
<th>Hg in muscle (mg kg(^{-1}))</th>
<th>Cd in liver (mg kg(^{-1}))</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>North Atlantic</td>
<td>0.01 – 2.60</td>
<td>–</td>
<td>ICES (1989)</td>
</tr>
<tr>
<td>North Atlantic</td>
<td>0.10 – 2.00</td>
<td>–</td>
<td>Franklin (1987)</td>
</tr>
<tr>
<td>Baltic Sea</td>
<td>–</td>
<td>0.04 – 3.39</td>
<td>Voight (1999)</td>
</tr>
<tr>
<td>English Channel and Bay of Biscay</td>
<td>0.12 – 2.18</td>
<td>–</td>
<td>Cossa et al. (1992)</td>
</tr>
<tr>
<td>Seine Estuary and Seine Bay</td>
<td>0.01 – 3.27</td>
<td>0.01 – 9.56</td>
<td>This study</td>
</tr>
</tbody>
</table>

All data have been converted to mg kg\(^{-1}\) dry weight (d.w./wet weight = 0.2).
statement is consistent with knowledge of Cd contamination in this estuary (Chiffoleau et al., 2001). Similar to our Hg analysis, we proceeded with two series of data processing to try to detect temporal trends in the Cd concentration of fish livers. First, we selected fish <300 mm, given that large fish were not homogeneously represented in annual sample sets. Second, as Cd data were not normally distributed (“Skewness” = 5.11), a log\(_{10}\) transformation was applied to the Cd results. Figure 6 illustrates the temporal variations of annual averages of concentrations of Cd in the liver of flounders (length <300 mm) between 1986 and 2003, accompanied by their 95% confidence intervals. There was a consistent increase in concentrations between 1986 and 1990, a reduction between 1990 and 1995, then stability from 1995 on (Figure 6).

Mussels and water
The historical trends in contamination of the Seine Estuary and Seine Bay by Cd and Hg are well documented, both by the results of monitoring the River Seine inputs, which is performed upstream of the tidal influence at the Poses Dam (upstream Rouen), and the RNO Mussel Watch Programme (www.ifremer.fr/envlit).

Figures 7 and 8 illustrate the temporal variations in Cd and Hg concentrations in freshwater from the River Seine determined...
from monthly sampling at its mouth. For Cd, the most striking
feature is the large decrease by a factor of five within 17 y
(Figure 7). Particulate Cd ranged from around 10 mg kg\(^{-1}\)
in 1986 to 2 mg kg\(^{-1}\) in 2003, and dissolved Cd decreased from 0.1
to 0.02 \(\mu g\) l\(^{-1}\) during the same period. The Hg concentrations
on particles and dissolved mercury show parallel variation, but
with a distinct pattern compared with Cd. There was an increase
in concentrations from the outset of the observations between
1990 and 1993, up to 10 mg kg\(^{-1}\) and 10 ng l\(^{-1}\) for particulate
and dissolved Hg, respectively, followed by a rapid decrease.

Monitoring of Cd and Hg in soft mussel tissue (Figures 9 and 10) from three stations of the Seine Estuary (Villerville, Le
Havre, and La Hève; Figure 1) revealed high-frequency variations
superimposed on smoother long-term variations. The high-
frequency signal had been noticed previously for Hg (Laurier
et al., 2006) and other metals (Claisse, 1989), and was partially
attributed to seasonal physiological changes in the mussels. The
long-term variations peaked between 1990 and 1993 for Cd, and
between 1992 and 1994 for Hg. The amplitude of the reduction
factor in Cd concentrations was ten between the maximum

Figure 5. Cadmium concentrations in liver (Cd) plotted against fish length (TL). Concentrations are expressed per unit of d.w. The correlation coefficient is statistically not significant \((r^2 = 0.09, n = 346)\).

Figure 6. Temporal trend (1986–2004) of cadmium (\(\log_{10}Cd\)) concentration (annual average ± s.d.) in flounder liver from fish of length < 300 mm.
values and the current minimum, but closer to five for Hg concentrations.

**Discussion**

Comparing the pattern of variability in Hg concentrations in River Seine water (Figure 8) with those of mussels sampled from the estuary (Figure 10) seems to indicate a causal link. However, the chronological series of Cd in mussels does not simply reflect the concentration changes in river inputs. The reduction in inputs from the River Seine is continuous (Figure 7), whereas the distribution of Cd in mussels increased between 1988 and 1990 (Figure 9). As indicated in the “Survey site” section, in addition to the Cd borne by River Seine waters, there were various direct Cd discharges into the Seine Estuary from 1974 to 1992 (Chiffoleau et al., 2001). According to those authors, the Cd contamination of the Seine Estuary was largely a consequence of the discharge of phosphogypsum wastes from three phosphoric acid plants, which ceased operating in 1992. At the beginning of our sampling period, the phosphogypsum waste discharge within the estuary, 200 000 t y$^{-1}$ according to Chiffoleau et al. (2001), was made through a pipe on the bed of the estuary, opening southeast of Le Havre (Figure 1). Additionally, according to the same authors, the phosphogypsum wastes were Cd-enriched during the period.

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**Figure 7.** Temporal trend (1986–2004) of cadmium (Cd) concentrations in the River Seine near its mouth. Dissolved and particulate Cd refers to fractions smaller or larger than 0.45 μm, respectively.

**Figure 8.** Temporal trend (1986–2004) in mercury (Hg) concentrations in the River Seine near its mouth. Dissolved and particulate Hg refers to fractions smaller or larger than 0.45 μm, respectively.
1987–1992. Therefore, with a coincidental maximal Cd concentration, the mussels obviously trace the historical Cd discharge within the Seine Estuary.

Within this context, we ask how should the variations in Cd and Hg in flounders be interpreted? In the case of Cd, an increase followed by a decline can be observed in the concentrations in the livers of flounders in the Seine Estuary and Seine Bay (Figure 6), similar to the temporal changes in Cd concentrations in mussel tissue (Figure 9). However, the significant increase in concentrations in the flounder livers had been noted since 1988, 1 y before that in the mussels, and the decline also appears to have started earlier (Figure 9). Does this time-lag reflect ecological differences between the two species (habitat, diet, growth, etc.) or, more simply, the gap between the sampling periods? Indeed, the flounders were sampled in November and December, and the mussels were collected four times a year, from February to November. Another difference between the temporal trend of Cd concentrations in flounders and those in mussels is the roughly stable values in flounders after 1993, whereas the Cd in mussels continued to decrease until 1998. Although Cd discharges into

**Figure 9.** Temporal trend (1986–2004) in cadmium (Cd) concentrations in soft mussel (*Mytilus edulis*) tissue from the Seine Estuary. Concentrations are expressed per unit of d.w. Annual average concentrations (± s.d.; full-size graph) are expressed on a logarithmic scale.

**Figure 10.** Temporal trend (1986–2004) in mercury (Hg) concentrations in soft mussel (*Mytilus edulis*) tissue from the Seine Estuary. Concentrations are expressed per unit of d.w. Annual average concentrations (± s.d.; full-size graph) are expressed on a logarithmic scale.
the sediments stopped in 1992, Cd concentrations in the River Seine continued to decline (Figure 7). This suggests that flounders, unlike mussels, are more reactive to Cd changes in the sediments than in the water column.

In the case of Hg in the muscles of flounders (Figure 4), we did not observe any temporal variation linked to that of mussels or fluvial Hg contribution (Figures 8 and 10). During the period of high concentrations of Hg in mussels and in River Seine water, large variations were observed in flounders, with the lowest average Hg concentrations in 1992 and the highest in 1993 (Figure 4). If we assume that the sediment is the main source of metal contamination for flounders, the migrations by this species within the estuary and Seine Bay can be seen as a possible explanation for the high variability of mercury concentrations in the fish. Indeed, the spatial heterogeneity of Hg contamination in the Seine Estuary is high: in Seine Bay, concentrations averages 0.25 μg g⁻¹, whereas they attained 10 μg g⁻¹ upstream near Rouen (Cossa et al., 2003). However, in the absence of more in-depth knowledge of the migrations and the feeding habits of flounders, this interpretation must remain speculative.

In summary, monitoring of Cd using flounder livers since 1986 has revealed a temporal pattern that can be related to the temporal changes in the Cd-rich waste inputs to estuarine sediments, while monitoring of mussel tissue also recorded these internal estuarine input variations, but superimposed with a continuously decreasing trend in Cd concentrations in the River Seine. For Hg, although the temporal trends for mussel tissue and river concentration are similar, the Hg trend recorded using flounder muscle does not follow the same pattern. The discrepancy between the responses of the two bio-indicators may be due to their different niches. Therefore, the use of flounders as sentinel organisms for monitoring Cd and Hg contamination temporal trends in estuarine environments requires in-depth knowledge of their ecology within the study site. An adapted sampling strategy based on this knowledge should provide results that are easier to interpret.

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References
Laurier, F., Cossa, D., Beucher, C., and Brévière, E. 2006. The impact of
groundwater discharges on mercury partitioning, speciation and
bioavailability to mussels in a coastal zone. Marine Chemistry,

(Platichthys flesus L.) from estuaries and coastal waters of
the northeast Irish Sea. Environmental Pollution, 75: 317–322.

Luten, J. B., Bouquet, W., Riekel-Booy, G., Rauchbaar, A. B., and
Scholte, M. W. M. 1987. Mercury in flounder, Platichthys flesus,
cod, Gadus morhua, and perch, Perca fluviatilis, in relation to
their length and environment. Bulletin of Environmental
Contamination and Toxicology, 38: 318–323.

Marthinsen, I., and Staveland, G. 1990. Levels of mercury in flounder
(Platichthys flesus L.) and cod (Gadus morhua L.) caught during the
year 1988 in the Hvaler Archipelago, southern Norway. ICES,
Copenhagen. 8 pp.

Masson, G. 1987. Biologie and écologie d’un poisson plat amphihalin,
le flet (Platichthys flesus flesus, Linneé, 1758) dans l’environnement
ligérien: distribution, démographie, place au sein des réseaux tro-
phiques. Doctoral thesis presented to the University of Western
Brittany. 344 pp.

Minier, C., Levy, F., Bocqueneé, G., Godefroy, D., Burgeot, T.,
and Leboulenger, F. 2000. Flounder health status in Seine Bay. A
multibiomarker study. Marine Environmental Research, 50: 69–73.

Miramand, P., Guyot, T., Rybarczyk, H., and Bessineton, C. 1998. Cd,
Cu, Pb, Zn dans les tissus et in the réseau trophique benthique du
bar et du flet de l’estuaire de la Seine. Programme Scientifique
Seine Aval, Thème Edifices Biologiques, Rapport 1997/Fín–4:
171–181.

Aquatic Contaminants. Environmental Managment Series. Alden

Métaux: sources multiples et accumulation. In La Seine en son
Bassin: Fonctionnement Écologique d’un Système Fluvial
Anthropisé. 391–437. Ed. by M. Meybeck, G. de Marsily, and E.

Voight, H-R. 1999. Concentration of heavy metals in fish from coastal
waters around the Baltic Sea. ICES Journal of Marine Science,
56(Suppl.): 140–141.

Forschung, 15: 234–239.


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