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STABLE POLLUTANTS IN THE EASTERN PART OF THE NORTH SEA.

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Abstract

The spatial distributions of most dissolved and particulate trace metals are, at least qualitatively, similar. Their concentrations decrease with distance from the coast and the mouth of the Scheldt estuary (particulates much stronger than dissolved species).The salinity gradient is similar (but opposite) to the trace metals and turbudity gradients. The five trace metals studied can be subdivided into two groups on the basis of their K_D's (ratio of particulate metal concentration to dissolved metal concentration). Hg and Pb are strongly associated with particulate matter (K_D from 0.5 to 1.5 offshore and from 7.9 to 40 in the coastal area), while Cd, Cu and Zn are predominantly in the dissolved phase (K_D from 0.1 to 0.2 offshore -up to 0.5 for Zn- and from 1 to 1.7 in the coastal area).

Offshore fluxes of dissolved Cd, Cu, Hg, Pb and Zn are calculated based on diffusive and mixing processes . They can explain the concentration increases in the coastal flow tube (315 $km^3 \cdot y^{-1}$), but are insignificant beyond that flow tube. The ratios of the Scheldt output to the offshore flux vary from 38 to 85%, depending on the kind of metal. The ratios of the Scheldt output to the metal flow parallel to the coast is smaller than 1%.

The steady-state concentration profiles of particulate metals versus salinity are fairly constant in the coastal-estuarine and marine watermasses, but decrease very abruptly from the first to the second watermass. Assuming a conservative behaviour of the particulate metals, offshore fluxes and the resulting concentration increases in the coastal flow tube, were deduced. These calculated concentration increases agree fairly well with the observed values. The contribution of the Scheldt estuary to the flows parallel to the coast ranges from 1.6 to 3.3%.

Biological productivity in coastal waters is often very high owing to the high levels of nutrients supplied via coastal inputs or through upwelling. In Belgian coastal waters for example the daily primary production ranges from 10 to 280 mg $N.m^{-2}day^{-1}$ corresponding to an annual primary production of about 25 g N $m^{-2}y^{-1}$ (Baeyens et al. 1983, Mommaerts et al. 1984, Baeyens et al. 1984). Shellfish and fish thus find there excellent spawning, nursing and feeding grounds. Coastal ecosystems are very sensitive to pollution which can perturb severely the biological processes. Heavy metals are amongst the major potential pollutants because of their toxicity .

Obtaining reliable, accurate trace metal data in sea water is still a cumbersome operation. In order to evaluate our analyzing procedures, we participated at several intercalibration exercises : (1) the intercalibration exercises for the analysis of cadmium and mercury in sea water requested by the Joint Monitoring Group of the Oslo and Paris Commissions and conducted by ICES. The results have been reported by Thibaud (1981) for cadmium and Olafsson (1981) for mercury ; (2) the intercalibration exercise of trace metals in sediments conducted by the "Centre Oceanologique de Bretagne, CNEXO". The results have been reported by Joanny et al (1980); (3) for the analysis of mercury in botanical substrates (BCR reference materials), we compared several mineralization-digestion methods. Results have been reported by Dehairs et al. (1982). Participation at these intercalibration exercises offered us the possibility to demonstrate that the analyzing procedures in use in our laboratories for Cd, Cu, Hg, Pb and Zn produce reliable data, at least on samples similar to those that served for the intercalibration exercises. Another encouraging fact is that our trace metal data obtained in the English Channel (Baeyens et al., 1987), the Southern Bight of the North Sea (see Table 1), the northern North Sea (Baeyens et al., 1987) and the Mediterranian Sea (our data are given in the section "Sampling of heavy metals",this paper; for example Mart et al., 1982 report data for the Western Mediterranian Sea) compare favourably with these obtained by other investigators in the same areas .

Analytical techniques and methods are described in detail in Baeyens et al. (1987).

Dissolved metals

In Table 1 a summary of previous determinations in the area

is given together with relevant data from the west coast of the Southern Bight. These comparisons should be carried out with care, however, for reasons mentioned in the introduction. The range of dissolved copper concentrations we observed, compares favourably with those observed by Duinker and Nolting (1982) and by Mart (1976), if we do not consider his stations 4 and 8 which show very high copper concentrations (8.5 and 15.05 $\mu g.L^{-1}$ respectively). The minimum values reported by Duinker and Kramer (1977) are a factor three to four higher than these observed during this study. For the west coast of the North Sea Basin, Taylor (1979) reports a mean dissolved copper concentration (2.1 $\mu g.L^{-1}$) which approaches our maximum values, while Balls (1985) observed a concentration range that is from two to four times lower than the concentration range in the Belgian coastal zone.

The dissolved lead values reported by Mart (1976) (with or without stations 4 and 8) and Duinker and Kramer (1977), for the present study area, are substantially higher than the values we observed. Duinker and Kramer (1977) attributed these high concentrations to particulate lead (lead oxide) passing the 0.45 µm pore size filters. This lead is released to solution following acidification. However, at the western side of the Southern Bight, Balls (1985) found dissolved lead concentrations which are comparable to our values. The dissolved lead concentrations show a tendency to decrease with increasing salinity. This is in contrast with the findings of Balls (1985), who observed lower dissolved lead concentrations in the Humber estuary (0.01-0.055 $\mu g.L^{-1}$) than in the coastal zone (0.015-0.135 $\mu g. L^{-1}$).

Dissolved cadmium values reported by Mart (1976) and Duinker and Nolting (1982) for the present study area are of the same magnitude, the minimum values reported by Duinker and Kramer (1977) are substantially higher. Taylor (1979) found a mean dissolved Cd concentration of 0.39 $\mu g.L^{-1}$ off the west coast of the Southern Bight, which is about three times higher than our maximum values, but Balls (1985) found still lower concentrations than the ones we observed off the Belgian and Dutch Coasts.

Dissolved zinc concentrations reported by Duinker and Kramer (1977) are substantially higher than our results but in a more recent study Duinker and Nolting (1982) observed a similar range. At the western side of the Southern Bight Basin, Taylor (1979) observed much higher values (mean value of 25.2 $\mu g.L^{-1}$).

Dissolved mercury data in the North Sea are scarce. Burton

and Leatherland (1971) reported values between 14 and 21 ng.L^{-1} in the English Channel on non filtered samples.Baker (1977) who measured reactive mercury in the Southern Bight reported values between 2.0 and 7.7 ng.L^{-1} with some individual high spots of 12 and 18 ng.L^{-1}. These values compare favourably with our offshore data (6 ng.L^{-1}) and coastal-estuarine data (19-22 ng.L^{-1}).

The dissolved metal concentrations observed in March 1983 in the mouth of the Scheldt estuary are of the same magnitude as those observed in the coastal watermass off the Scheldt mouth (coastal stations 14 and 15).

Particulate metals

The range of particulate copper concentrations we have measured compares favourably with those reported by Mart (1976) and Duinker and Nolting (1977), the range of zinc concentrations with those reported by Duinker and Nolting (1977). Mart (1976) observed significantly higher minimum values of particulate Cd and Pb, and a much narrower range of values for Pb.

Particulate to dissolved metal ratios (K_D)

Dissolved copper, cadmium and zinc proved to be the major portion of the total in offshore samples (K_D is 0.1 to 0.2 for Cu and Cd, 0.1 to 0.5 for Zn), but in the coastal-estuarine watermass dissolved and particulate fractions are of the same magnitude (K_D between 1 and 2).

Lead and mercury are much stronger associated with particulate matter and show a K_D of about 1 offshore, of ca. 40 (Pb) and 8.7 (Hg) in the coastal-estuarine watermass. Adsorption rate studies demonstrated the great affinity of mercury for suspended matter (e.g.Frenet-Robin and Ottmann, 1978 ; Baeyens et al., 1982).

The ratios (L.g^{-1}) of the concentrations in suspended solids (µg.g^{-1}) to the concentrations in solution (µg.L^{-1}) of Cu, Cd and Zn are respectively ca. 36, 31 and 67 offshore and ca. 22, 12 and 30 in the coastal-estuarine watermass. They are substantially higher for Pb and Hg, respectively ca. 183 and 260 offshore and ca. 382 and 122 in the coastal-estuarine watermass. Pb is thus the only metal showing a decrease of concentration in suspended solids relative to the dissolved

form with increasing salinity. The fact is that the iron and manganese hydrous-oxides with which a large fraction of Pb is probably associated do not reach the open sea zone.

Offshore transport of dissolved metals

Three processes may affect the concentrations in streamtube I (Figure 1) : (1) coastal inputs upstream the hydrodynamical front (2) diffusion through the front and (3) mixing with the coastal-estuarine watermass downstream the front.

Coastal inputs :they only seem to be important for Cu and Zn. However, no quantified data of these coastal inputs exist.

Diffusion :for the calculation of the concentration gradients at the hydrodynamical front, we made use of the salinity at the stations at each side of the front and the corresponding metal concentrations inferred from the metal-salinity best fit curve. The concentration gradients at the front then amount to 0.0014 (Cd), 0.022 (Cu), 0.065 (Zn), 0.0059 (Pb) and 0.00047 (Hg) in $tons.km^{-4}$. Using the diffusion coefficient deduced for salinity at the front (1018 $km^2.Y^{-1}$ or 32 $m^2.S^{-1}$), diffusion fluxes of 0.22 (Cd), 3.3 (Cu), 9.9 (Zn), 0.90 (Pb) and 0.072 (Hg) in $tons.Y^{-1}$, over the length of the front (about 10 km) are obtained. These diffusion fluxes represent between 40 and 65% of the dissolved metal supply by the river Scheldt. The resulting increases in streamtube I are 0.0025 (Cd), 0.039 (Cu), 0.12 (Zn), 0.011 (Pb) and 0.00085 (Hg), in $\mu g.L^{-1}$.

Mixing :the increases in streamtube I due to mixing with the adjacent coastal-estuarine watermass (4.4 $km^3.Y^{-1}$ and metal concentrations observed at S=31.46) yield 0.0047 (Cd), 0.057 (Cu), 0.14 (Zn), 0.014 (Pb) and 0.00067 (Hg), in $\mu g.L^{-1}$. These mixing fluxes represent between 52% and 79% of the dissolved metal supply by the river Scheldt.

When we compare now the concentrations observed at station 13 (S=31.72), increased by the contributions from diffusion and mixing calculated hereabove, with the concentrations at station 26 (S=31.5) we find a very good agreement (Table 2). In addition the input flux from the river Scheldt can almost account (between 73% and 90%) for the total

supply (diffusion + mixing) from the coastal-estuarine watermass to streamtube I.

Exchange of material between streamtube I (85 $km^3.Y^{-1}$) and streamtube II (230 $km^3.Y^{-1}$) is only possible through diffusion. Metal concentration gradients upstream station 13 are very small and diffusion negligible. The diffusion fluxes downstream station 13 amounts to 0.83 (Cd), 10.7 (Zn), 1.07 (Pb), 5.22 (Cu) and 0 (Hg), in $tons.Y^{-1}$. They are calculated analogous to the diffusion fluxes from the coastal-estuarine watermass to streamtube I, but now over a length of 30 km and using the concentration gradients between stations 13 and 33. The resulting increases in streamtube II are then 0.004 (Cd), 0.05 (Zn), 0.005 (Pb), 0.02 (Cu) and 0 (Hg), in $\mu g.L^{-1}$, and represent a concentration increase in streamtube II of less than 3%, except for Cd (8%). The input of the river Scheldt is between 1.4 and 1.7 times larger than the diffusion flux to streamtube II, except for Cd (only 0.7 times this diffusion flux).Since the total dissolved metal input of the Scheldt estuary is required to balance the metal supply to streamtube I an additional source (atmosphere, bottom,...) is thus necessary to support the flows to streamtube II. Diffusion to the outer streamtubes (III,IV,V) is insignificant as a result of negligible concentration gradients.

Transport of dissolved metals parallel to the coast

The total flow through streamtubes I to V (see Figure 1) equals 1260 $km^3.Y^{-1}$. This means about 25% of the flow entering the Southern Bight of the North Sea through the English Channel. The cumulative dissolved metal flow through streamtubes I to V equals 3000 (Zn), 250 (Pb), 900 (Cu), 59 (Cd) and 13 (Hg) in $tons.Y^{-1}$. The contribution of the Scheldt to this flow is very small and ranges from 0.6 to 0.9%.

To have an idea of the magnitude of these metal flows, we compared them with dissolved metal flows through the English Channel and the northern North Sea, normalized to a flow of 1260 $km^3.Y^{-1}$. Trace metal data are obtained from Jones in Topping et al. (1980), Duinker and Nolting (1982) and our measurements in the considered areas. It appears from Table 3 that the metal flows through the English Channel are always lower than the metal flows through the Belgian coastal area, which are of the same magnitude as those through the northern North Sea. Much larger inputs than the Scheldt estuary are, however, required to explain

the increase of dissolved metals in northern direction.

Offshore transport of particulate metals

The offshore flux of the particles can be estimated, assuming they behave in a similar way as the fluid in which they are suspended. Three processes affect the metal concentrations in streamtube I: coastal inputs (upstream area, south of the front), diffusion (at the hydrodynamical front), mixing (downsteam area, north of the front).

Coastal inputs : they only seem to be important for Cd, but there exists no quantified data.

Diffusion : the concentration gradients and the diffusion fluxes at the hydrodynamical front are calculated similarly to these of the dissolved metals. The diffusion fluxes amount to 0.45 (Cd), 31 (Zn), 20 (Pb) and 1.3 (Hg) in tons.Y^{-1}. These diffusion fluxes represent between 130 and 140% of the particulate metal supply by the river Scheldt except for Cd where the diffusion flux is balanced by the (high) river output. The resulting increases in streamtube I are : 0.0053 (Cd), 0.087 (Cu), 0.37 (Zn), 0.24 (Pb) and 0.015 (Hg), in $\mu g.l^{-1}$.

Mixing : the increases in streamtube I due to mixing with the coastal-estuarine watermass (4.4 $km^3.Y^{-1}$ and concentrations observed at S=31.46) yield : 0.0035 (Cd), 0.064 (Cu), 0.28 (Zn), 0.17 (Pb) and 0.0083 (Hg), in $\mu g.l^{-1}$. The mixing fluxes represent between 69 and 103% of the Scheldt supply.

When we compare the concentrations observed at station 13 (S=31.72), increased by the contributions from diffusion and mixing calculated hereabove, with the concentrations at station 26 (S=31.5) ,we find for most metals a good, however not that excellent agreement as for dissolved metals (see Table 4). The metal supply (diffusion and mixing) from the coastal-estuarine watermass to streamtube I is, although too low, not supported by the Scheldt input (between 41 and 58%).

Exchange of material between streamtube I (85 $km^3.Y^{-1}$) and streamtube II (230 $km^3.Y^{-1}$) is only possible through diffusion. Upstream of station 13 metal concentration gradients are very small and diffusion is negligible. The diffusion fluxes downstream of station 13 amounts to 0.20 (Cd), 14 (Zn), 17 (Pb), 3.3 (Cu) and 1.9 (Hg), in tons.Y^{-1}. They are calculated analogous to the diffusion fluxes of dissolved metals from streamtube I to streamtube II. The resulting increases in

streamtube II are then, 0.0009 (Cd), 0.06 (Zn), 0.07 (Pb), 0.01 (Cu) and 0.008 (Hg), in $\mu g.l^{-1}$, and represent a concentration increase in streamtube II of 4 to 5% for Cd, Cu and Zn, of 13% for Pb and of 26% for Hg. These increases fall in the range of observed increases. The coastal values of Pb and Hg are increased more drastically over the marine watermass levels than Cu, Zn and Cd and result in much larger diffusion fluxes and concentration increases. When we compare the particulate metals input of the river Scheldt to the diffusion flux to streamtube II, we find the Scheldt input larger than the diffusion flux for Cd, Zn and Cu (between 1.6 and 2.2 times), but smaller for Pb and Hg (between 0.5 and 0.9 times). Diffusion to the outer streamtubes (III,IV,...) is insignificant, due to the negligible concentration gradients. In any case, if particulates move like the bulk of the fluid, in the way thus we have calculated mixing and diffusion processes, an additional source (atmosphere, bottom,...) is required to balance these flows. The deficit expressed in terms of Scheldt input flux varies from one time this flux for Cd, over two times for Zn, Cu and Pb to three times for Hg.

Transport of particulate metals parallel to the coast

The cumulative particulate metal flow through streamtubes I to V equals 1400 (Zn), 520 (Pb), 340 (Cu), 21 (Cd) and 27 (Hg), in $tons.Y^{-1}$, considering particulate metals as conservative substances. The contribution of the Scheldt to these flows is somewhat higher than in the case of dissolved metals and ranges from 1.6 to 3.3%.

The particulate metal flows through the English Channel and the northern North Sea have been calculated in a similar way as it has been done for dissolved metals. Both particulate metal flows are of the same magnitude, but are about three times lower than the particulate metal flows through the Belgian coastal zone (Table 5). Either additional sources or a non-conservative behaviour of the particulate metals (e.g. erosion-sedimentation cycles) must explain this result.

Conclusion

The impact of the metal discharge by the river Scheldt is limited to a fairly small watermass (streamtubes I and II in Figure 1 , corresponding together to 315 $km^3.Y^{-1}$), and does not reach more offshore zones. Transport (diffusion and mixing) from the coastal-estuarine

watermass (bound by the hydrodynamical front and streamtube I,see Figure 1) to streamtube I and then further to streamtube II can explain the observed increases in metal concentration in the latter streamtubes, but these offshore transport fluxes are not completely balanced by the river input. For the dissolved species is the deficit of the same magnitude as the river input, but for the particulates is it a few times this input.

Additional sources such as atmosphere, bottom, dumping, can be possible explanations and should be considered in future. On the other hand, if in the Belgian coastal area, flows of particulate metals are only one fourth of those we have calculated (their advective and diffusive velocity is thus four times smaller than the fluid in which they are suspended), then (1) the flows of particulate metals parallel to the coast are of the same order as those observed in the English Channel and the northern North Sea and (2) the river input will show now a small excess compared to the offshore flux, excess which satisfies the small deficit of the dissolved metals offshore flux.

Testing the assumption concerning the transport velocity of particulate metals requires information about the size distribution and density of the particles, their settling rates and the size distribution of the particulate metals, while testing the assumption concerning the transfer of metals from the particulate to the dissolved phase requires information about adsorption-desorption processes in the watercolumn and mobilization processes in the sediments.

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Table 1 : Trace metal data reported for the eastern and western side of the Southern Bight of the North Sea.

	Zn ($\mu g.L^{-1}$)	Cu ($\mu g.L^{-1}$)	Cd ($\mu g.L^{-1}$)	Pb ($\mu g.L^{-1}$)	Hg ($ng.L^{-1}$)
Dissolved phase					
Belgian and Dutch coasts					
Mart 1976 - all stations	-	0.34 - 15.05	0.022 - 0.390	0.079 - 7.50	-
- all stations minus stations 4 and 8	-	0.34 - 2.20	0.022 - 0.110	0.079 - 2.60	-
Duinker and Kramer, 1977	3.00 - 20	1.00 - 2.50	0.100 - 0.300	1.700 - 3.30	-
Duinker and Nolting, 1982	0.30 - 12	0.20 - 2.40	0.020 - 0.120	-	-
This study - october 1982	0.25 - 8.8	0.28 - 2.50	0.014 - 0.110	0.045 - 0.260	6 - 19
- march 1983	0.90 - 7.2	0.38 - 1.80	0.030 - 0.170	0.090 - 0.660	6 - 24(a)
Mouth of the Scheldt					
This study - march 1983 : 25.8‰ S	4.4	1.6	0.13	0.35	-
18.7‰ S	7.1	1.4	0.17	0.26	-
East coast of Britain					
Taylor, 1979(b)	(25.2)	(2.1)	(0.39)	(-)	-
Balls, 1985	-	0.12 - 0.58	0.010 - 0.060	0.015 - 0.135	-
Particulate phase					
Belgian and Dutch coasts					
Mart, 1976	-	0.11 - 1.4	0.023 - 0.086	0.47 - 3.86	-
Duinker and Nolting, 1977	0.3 - 10(c)	0.11 - 3.0	-	-	-
This study - october 1982	0.07 - 9.0	0.08 - 2.6	0.002 - 0.13	0.03 - 8.3	4 - 160
- march 1983	0.16 - 8.3	0.06 - 1.8	0.010 - 0.060	0.07 - 4.2	-
Mouth of the Scheldt					
This study - march 1983 : 18.7‰ S	9.3	2.6	0.13	3.6	-

(a) May 1981 cruise results. (b)Taylor, 1979, reports total mean values. The mean dissolved concentrations presented here are calculated after the percentages of dissolved metals reported in his paper : 85% for Zn and Cd, 74% for Cu. (c) The particulate Zn values presented here are only between 60% to 90% of the total particulate Zn amount (Duinker and Nolting, 1977).

Table 2 Comparaison of observed and calculated dissolved metal concentrations in streamtube I.

	Observed concentration at S=31.5 (station 26) $\mu g.L^{-1}$	Calculated concentration after diffusion and mixing (S=31.5) $\mu g.L^{-1}$	Ratio of Scheldt flux/ Diffusion + Mixing flux %
Cd	0.0885	0.0892	90
Cu	1.085	1.096	86
Zn	2.70	2.76	82
Pb	0.270	0.275	73
Hg	0.0130	0.0135	85

Table 3 Dissolved metal flows parallel to the coast (in $tons.Y^{-1}$), for a water flow of 1260 $km^3.Y^{-1}$.

	Scheldt	Belgian Coast	English Channel			northern North Sea		
			(1)	(2)		(1)	(2)	
Zn	18	3000	760	980	930	3400	1300	2500
Pb	1.5	250			180			300
Cu	7.0	900	280	430	530	330	1300	690
Cd	0.55	59	13	52	38	88	38	39
Hg	0.11	13			16			

(1) Jones, in Topping et al. (1980).

(2) Duinker and Nolting (1982).

Table 4 Comparaison of observed and calculated particulate metal concentrations in streamtube I.

	Observed concentrations at S=31.5 (Station 26) $\mu g.l^{-1}$	Calculated concentrations after diffusion and mixing at S=31.5 $\mu g.l^{-1}$	Ratio of Scheldt Flux /Diffusion+ Mixing Flux %
Cd	0.035	0.035	58
Cu	0.78	0.65	41
Zn	3.0	2.7	44
Pb	2.1	1.7	45
Hg	0.15	0.13	45

Table 5 Particulate metal flows parallel to the coast (tons.Y^{-1}) for a water flow of 1260 $km^3.Y^{-1}$.

	Scheldt	Belgian coast	English Channel	nothern North Sea
Zn	24	1400		
Pb	15	520	190	140
Cu	5.3	340	120	78
Cd	0.44	21	6.0	5.4
Hg	0.88	27		

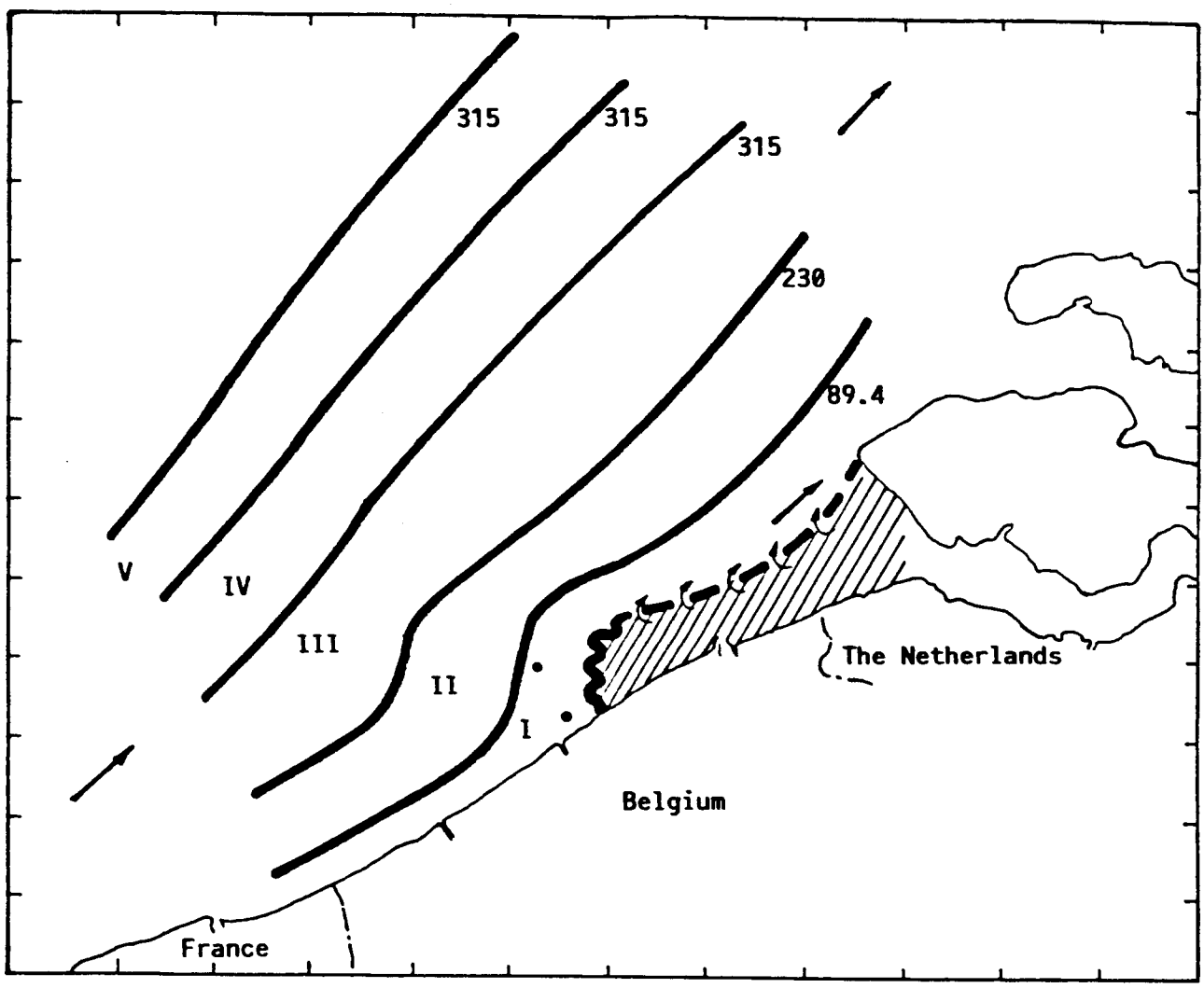

Figure 1: Residual flows through the Belgian coastal area ($km^3.y^{-1}$). The shaded area indicates the coastal-estuarine watermass and the undulating line the residual hydrodynamical front.