FROM LAND TO SEA VIA THE SKY: WHAT WE KNOW OF AIRBORNE CONTAMINATION OF EUROPEAN SEMI-ENCLOSED SEAS

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Abstract. The atmosphere is often overlooked as a component of the coastal zone, despite the very convergence of terrestrial, marine and atmospheric biogeochemical pathways lending a defining character to the coast. The magnitude of pollutant supply through the atmosphere is also often overlooked. Investigations, however, reveal that the atmosphere contributes substantial proportions of total coastal inputs. Source regions for these pollutants are typically found to be large, potentially larger than the watersheds which are traditionally taken as the appropriate geographic scale for management. This puts emphasis on international collaboration. Yet whilst we understand the general magnitude of the issue, our routine assessment of marine atmospheric pollution is often insufficient to allow us to know the true effectiveness of air pollution abatement measures for coastal and regional seas. At best we can estimate no more than the expected general tendencies in pollutant input for many European coastal and regional seas, including the Black Sea.

Key Words: marine pollution, atmospheric pollution

INTRODUCTION

Coastal zones are, by their nature, the product of the coincidence and interplay between the terrestrial, marine and atmospheric environments. Their significance in Europe reflects a geography characterised by enclosed and semi-enclosed seas. These surround and dissect the continent from the high arctic to the margins of the Sahara, and from the Atlantic seabord to the inland seas of the Asian plains. The continent is also densely populated by economically and culturally diverse societies. Together, the very heterogeneity of the coastal zone in physical, chemical, biological and human dimensions produces an extensive matrix of natural ecosystems and habitats and a multiplicity of human uses, making the region also one of complexity.

Anthropogenic driving pressures upon coastal resources, for example urbanisation, industrial and agricultural development, and mass tourism, are as much in evidence in Europe as in any part of the world. Often it is their simple proximity to coastal waters which determines the magnitude of impact of these local pressures. Pollutant discharges are amongst these, with source, transport and sink functions reacting to the sharp coastal conjecture between land and sea. Indeed, the very coincidence and diversity of biogeochemical processes and pathways impart their own individual character to coastal regions.

Focus is often given to riverine influences upon seas, and to the significance of drainage basins for coastal environments. However, in addition to aquatic biogeochemical transfers occurring on (and even below) the earths surface, large contaminant flows travel above it through the atmosphere, some of these moving in both directions between sea and land. The issue of scale is pertinent. Biogeochemical flows through the atmosphere commonly have global and regional scale perspectives in addition to the local issues associated with proximity, and frequently have little connection to the local drainage basins. As flows can be bi-directional, the coastal zone can both be subject to, and can have, significant influence at all scales. One example would be the riverine supply of dissolved greenhouse gases with subsequent re-emission in coastal and marine waters.

This article will discuss the status and tendencies in Europe of contaminant loading from the atmosphere. Information will be drawn from research and governmental sources concerning atmospheric pollutant flows to and from European regional seas. Significant in the research community has been the ELOISE – European Land Ocean Interaction Studies - initiative of the European Union comprising a thematic grouping of 60 projects over the decade from the mid-1990s. Anticipating the changes to coastal zone biogeochemical functioning and structure which will arise from the future pressures exerted by society has been a challenge for ELOISE, and the real diversity in coastal contaminant dispersal generated a wide spread of topics. Alongside these are the intergovernmental initiatives for monitoring and cooperative management of European regional seas, some of which give attention to atmospheric pollution. An emphasis on Europe's northern seas is evident, and examples herein are largely drawn from the North and Baltic Sea regions.

Whilst information for the Black Sea is essentially missing, the spatial scale of atmospheric flows and the picture for other parts of Europe suggest limited recent abatement of airborne pollutant loadings in the Black Sea. Overall, the broad picture painted is that despite the gamut of policies which address contaminant flows to Europe's coasts, it is doubtful that airborne pollution of regional seas has or is being tackled as effectively as has been the case for drainage basin discharges.

MAGNITUDES OF AIR INPUTS

Traditionally, directional river inputs to coastal waters have been seen as the principal pathway for contaminant flows. Within ELOISE, the theme of riverine contaminant supply to the coast considered largely heavy metal inputs from historical mining regions - Rio Tinto and Rio Odiel discharging to the Spanish Mediterranean (TOROS project), the Danube exiting to the Black Sea, the Humber and Rhine flowing to the North Sea, the Vistula in the Baltic basin and the Idrijca entering the Adriatic (EUROCAT project). The evidence is that these issues are generally local in scale, and their long term legacy restricted. Contaminants are removed from the river en route by biological or physical processes, this removal continuing after the river reaches the sea. There are also options for active management to limit initial transfer to river water via attention to surface runoff characteristics within catchments. TOROS sought to establish the comparative magnitude of fluvial contaminant flows in these mining areas, and in so doing provided insight to the significance of the atmospheric route. Table 1 quantifies the various pollutant flows to the western Mediterranean downstream of the Rio Tinto mining region in Spain. This reveals that despite this being a heavily mined area, rivers supply only around 50% of metal inputs to coastal waters (Elbaz-Poulichet *et al*., 2001). In non-mining/non-industrial areas the riverine contribution can be expected to be smaller.

Table 1 Contaminant flows to Western Mediterranean, tonnes p.a.

	Cu	Zn	Cd	As
Atlantic waters	1140	1010	45	24000
Rio Tinto etc./Gulf of Cadiz	2000	6300	100	6300
Other rivers	230	130	3	200
Atmosphere	600-1200	3200-5100	35-60	
Ouflow to Atlantic	-1300	-6900	-	-33000

Negative numbers indicate flow out of the Mediterranean (Elbaz-Poulichet *et al*., 2001)

The characteristics of individual contaminants naturally influences their propensity for a given transport route. For some, riverine inputs have been found to be of still lesser significance. The POPCYCLING project estimated riverine lindane supply at only 15% of total input to the Baltic Sea, with as much as 85% reaching the Baltic via the atmosphere (Breivik and Wania, 2002) as given in Table 2. It was assumed that ϒ-HCH is representative of the behaviour of other persistent organic compounds.

For substances which have been of political interest for some time, the evidence from research projects, such as ANICE (Frank de Leeuw *et al*., 1003), for substantial atmospheric supplies confirms the message long given by inter-governmental activities. For example, the atmosphere has been estimated as delivering nearly 30-40% of anthropogenic inputs of nitrogen to the European northern regional seas (Table 3). ANICE substantiated these magnitudes applying

Table 2 Relative importance (in percent) of riverine and atmospheric supply of lindane (ϒ - HCH) to various parts of the Baltic Sea (Breivik & Wania, 2002).

	ϒ - HCH	
	Atmospheric deposition	**Riverine inflow**
Bothnian Sea	93	7
Gulf of Finland	68	32
Gulf of Riga	52	48
Baltic Proper	88	12
Kattegat	89	11
Skagerrak	91	9
Whole Baltic Sea	85	15

models to better determine the atmospheric nitrogen supply to North Sea coastal seas using high resolution representation of close-coast meteorological processes. This atmospheric proportion is actually believed to have steadily increased since the mid-1980s; although complicated by variable interannual river flows it is believed that riverine and directly discharged nitrogen inputs to the North Sea have fallen by around 30%, whilst atmospheric inputs have remained static (OSPAR, 2000).

THE IMPORTANCE OF CONTAMINANT PROPERTIES

Chemical transformations can render transport and deposition patterns complex. Forms less prone to washout in precipitation may be transported far from their point of release. Conversely, those which are in, or which are converted to, more easily scavenged forms display specific deposition patterns, often reflecting the particular nature of coastal meteorology. Hence, the distance any given pollutant will travel is dependent on its characteristics and resulting atmospheric lifetime. Nitrogen is an example, its various forms having different rates of dry deposition and varying solubilities. After emission, gaseous oxidised nitrogen transforms over time from less to more soluble forms, but simultaneously increases its particulate fraction which dry deposits more slowly. Reduced nitrogen is always very soluble, but similarly transforms from gaseous to a slower depositing particulate form with time.

Mercury is another such contaminant, whose forms with a lesser tendency to precipitation scavenging are candidates for longer transport distances and lifetimes in the atmosphere. Other forms, such as Hg^{II}, are soluble, do have propensity to scavenging by precipitation and will also more readily deposit to the earths surface whilst in a dry state due to their higher dry deposition velocity. The role of meteorology in defining source regions for mercury deposition is then crucial. Deposition to the Mediterranean has been studied within the MAMCS project (Pirrone *et al.*, 2003), which indicated that the scale over which source-receptor relationship exists is regional-to-global. Emission inventorying within the ELOISE projects MAMCS, MOE and MERCYMS, showed that coal combustion (power plants and residential heat furnaces) generates more than half of the European emissions (Pacyna *et al.*, 2001), these lying outside coastal zones. Management is then less a local river catchment scale matter, but rather is the domain of airshed-coast interactions, and of multilateral agreements.

Complexity is added, however, by semi-volatility. Unlike other metals mercury release is not limited to erosion and leaching processes. A possibility of volatilisation and re-emission is a feature shared with many persistent organic pollutants (POPs) for which there is also two-way movement between 'compartments' of the environment, *e.g.* water and sediment. The AIRWIN project has aided description of air-sea exchange *i.e.* diffusive vapour exchange, precipitation scavenging of vapours and particles, direct particle dry deposition, aerosol-vapour partitioning, and partitioning and sedimentation in the water column (Wania *et al.*, 1998). Complexity is further intensified for components which bioaccumulate, as for mercury and POPs creating a truly regional/global dimension. The pathways of bioaccumulation were the subject of the BIOCET and FAMIZ projects, potential influence on cetacean reproduction the topic of the latter. The possible influence of POP's on eagles and seals was given some attention in POPCYCLING (Koistinen, 1997a & b).

Table 3 Estimates from OSPAR and HELCOM of the proportion of nitrogen inputs to the North Sea/North East Atlantic and to the Baltic from atmospheric transport

Sea area	contaminant	Atmospheric proportion of total
NW Atlantic [(a)]	reduced nitrogen	32%
	oxidised nitrogen	37%
[(c)]	cadmium	29%
	mercury	20%
	lead	32%
Baltic [(b)]	total nitrogen	31%
	lead	48%

a) OSPAR (2000) b) HELCOM (1996) c) EEA (2001)

WHERE DOES IT COME FROM?

Since the late 1970s there has been coordinated intergovernmental effort to estimate the atmospheric flows of pollutants between European countries. This has lead to formal reporting of national emission totals for various pollutants, has established coordinated monitoring programmes, and has developed regional scale air pollution modelling to link the two and to describe source-receptor relationships. Initially used in acid rain abatement, the model capability now exists for a variety of pollutants. With sufficient validity indicated from comparisons with monitoring, the dispersion and deposition of pollutants across Europe can be described, allowing 'blame matrices' to be calculated of the contribution of one pollutant source upon receptors.

The source contributions to depositions in receptor areas are regularly published for some pollutants (*e.g.* EMEP, 2004). Table 4 is an extract of such estimates, showing the quantities of oxidised nitrogen believed deposited in the North and Baltic Seas in 1996. These are derived by computer modelling utilising reported emission totals. The table is not surprising in indicating large contributions from source countries such as the United Kingdom and Germany. It is perhaps more interesting to note that, despite their distance, Austria, Romania and Italy together contribute a similar quantity of oxidised nitrogen to the North Sea as does Norway with its long coastline. The message is of transcontinental transport and influence, and of limited local influence.

Table 4 gives results for country allocations of pollutants deposited. Commonly, such models actually have resolutions of no better than several tens of kilometres, with the model dividing the earth's surface into a grid of that resolution. Country totals are then the summation of many grid squares. Thus, it is only a matter of alternate data handling to give allocations down to the spatial resolution of the model regardless of boundaries, and to estimate the 'source region' for pollution received at any given place. Calculated from dispersion models, the source region for nitrogen arriving in the Baltic via the atmosphere is shown in figure 1, together with the source region for nitrogen arriving via rivers.

Table 4 Matrix of attribution of oxidised nitrogen deposited to two sea areas. Units: 100 tonnes nitrogen in 1996 (EMEP, 2001)

Source\Receptor sea	North Sea	Baltic Sea
Albania	0	0
Austria	11	6
Belgium	90	19
Bulgaria	1	1
Denmark	89	86
Finland	9	57
France	259	41
Germany	407	183
Greece	0	0
Hungary	9	8
Iceland	1	0
Ireland	27	3
Italy	14	13
Luxembourg	4	1
Netherlands	202	42
Norway	33	24
Poland	141	157
Portugal	5	0
Romania	6	4
Spain	24	3
Sweden	39	88
Switzerland	6	2
Turkey	0	0
United Kingdom	963	98

The 'watershed' is an understood concept dependent on geomorphology and useful in managing riverine pollutant flows; the 'airshed' or 'atmospheric source region' are less straightforward concepts. Here they have been defined according to model results. The Baltic have first been ranked by sources of atmospheric nitrogen which contribute to depositions in the Baltic have been ranked by the magnitude of their contribution. The minumum number of source regions (at model resolution – in this case 150km) then needed to supply 50% of the atmospheric nitrogen reaching the sea are assumed to represent as the 'actual 50% source region'. Importantly, this definition departs from that of the watershed in that it is dependent on the distribution of nitrogen emitted, and not simply on geophysical factors. To remove this effect and achieve a pure geophysical definition, the same calculations have been made assuming equal emissions of nitrogen at all points. This source region is then described as the 'airshed'. These source regions in figure 1 can be seen to be ignorant of political or drainage basin boundaries. They can also be seen to be large.

There are a number of points to be considered when defining/targetting source regions and considering the potential for management. Firstly, from the discussion concerning table 4 it is evident that maps such as figure 1 are component specific – in this case, total nitrogen. Focusing on either oxidised or on reduced nitrogen alone would have given a different picture. Secondly, the definitions chosen require consideration of geography, economics, social factors, etc. The actual distribution of emissions, the distribution of various economic sectors constituting to emissions, and the future potential source region are all amongst the range of possible issues. Thirdly, the management scale will almost always be large. By definition, issues of transboundary pollution are at least bilateral, and the first widespread international abatement measures were multilateral under the auspices of the United Nations 1979 Geneva Convention on Long Range Transboundary Air Pollution (LRTAP). Today in Europe it may be argued that the European Union is driving air quality management both within and outside the Union. The source regions are transboundary, and implementation of abatement strategies without neighbours will seemingly fail.

ARE WE CHANGING POLLUTANT EMISSIONS AND DEPOSITIONS TO THE SEAS?

Strategies exist for achieving both discharge and environmental quality standards. Dealing firstly with emissions, the European Marine Strategy is just one example in which it is stated the objective is "....to ...reduce ...emissions, and losses ...to the marine environment ..." (European Commission, 2002). Review of legislative and policy objectives is not the intention here, the important point being that the approach to management holds a focus upon emission control to abate input to the sea. Such an approach is long established across the spatial scales of governance in Europe.

In figure 2 are the figures for emissions of oxidised and of reduced nitrogen from signatories to the OSPAR Convention (North Sea countries) during the 1990s. Nitrogen is of interest as a nutrient and its potential contribution to eutrophication. Reported emissions show a fall of around 30% for oxidised nitrogen (combustion sources), but relatively unchanging reduced nitrogen emissions (agricultural sources). Overall, the decline in total nitrogen release is estimated at over 15%.

Either model estimates or monitoring results can be used to estimate marine deposition changes. The difficulty with monitoring is that it is often infrequent and isolated. Onshore coastal monitoring must then be interpreted to evaluate open sea depositions. Modelling, on the other hand, offers the possibility of numerical estimation beyond terrestrial observation. There have been considerable international efforts over the past two decades to model transboundary at-

Fig. 1 Watershed, airshed, and actual source regions for nitrogen flowing to the Baltic Sea

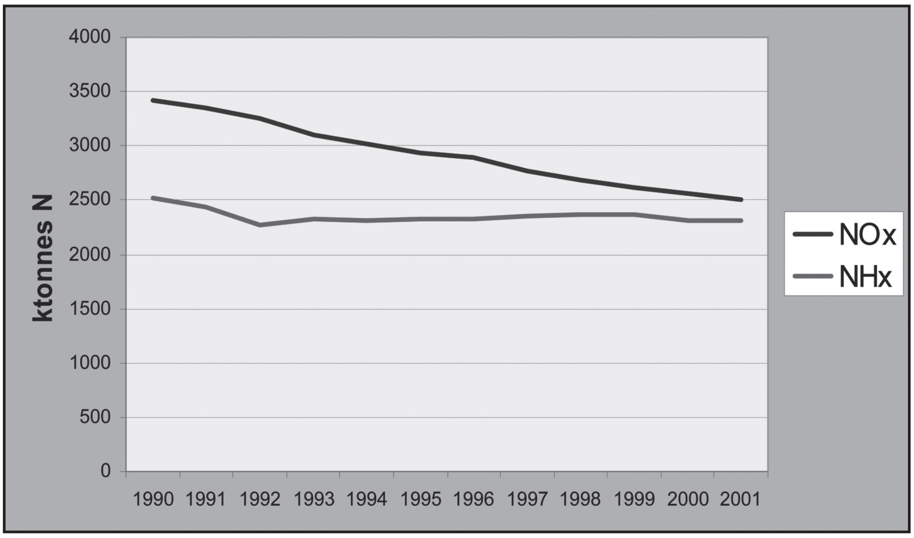

Fig. 2 Emissions of nitrogen from OSPAR countries 1990-2001. Compiled from reported emissions: http://www.emep.int

mospheric flows of nitrogen as a consequence of its roles in acidification, eutrophication and ozone production. Model estimates for nitrogen may then be considered reasonably reliable, the downside being that deposition estimates are driven by the input emission data, and hence cannot be an independent verification of reported emission change. In figure 3 the model estimates of nitrogen deposition for the various parts of the North Sea are shown.

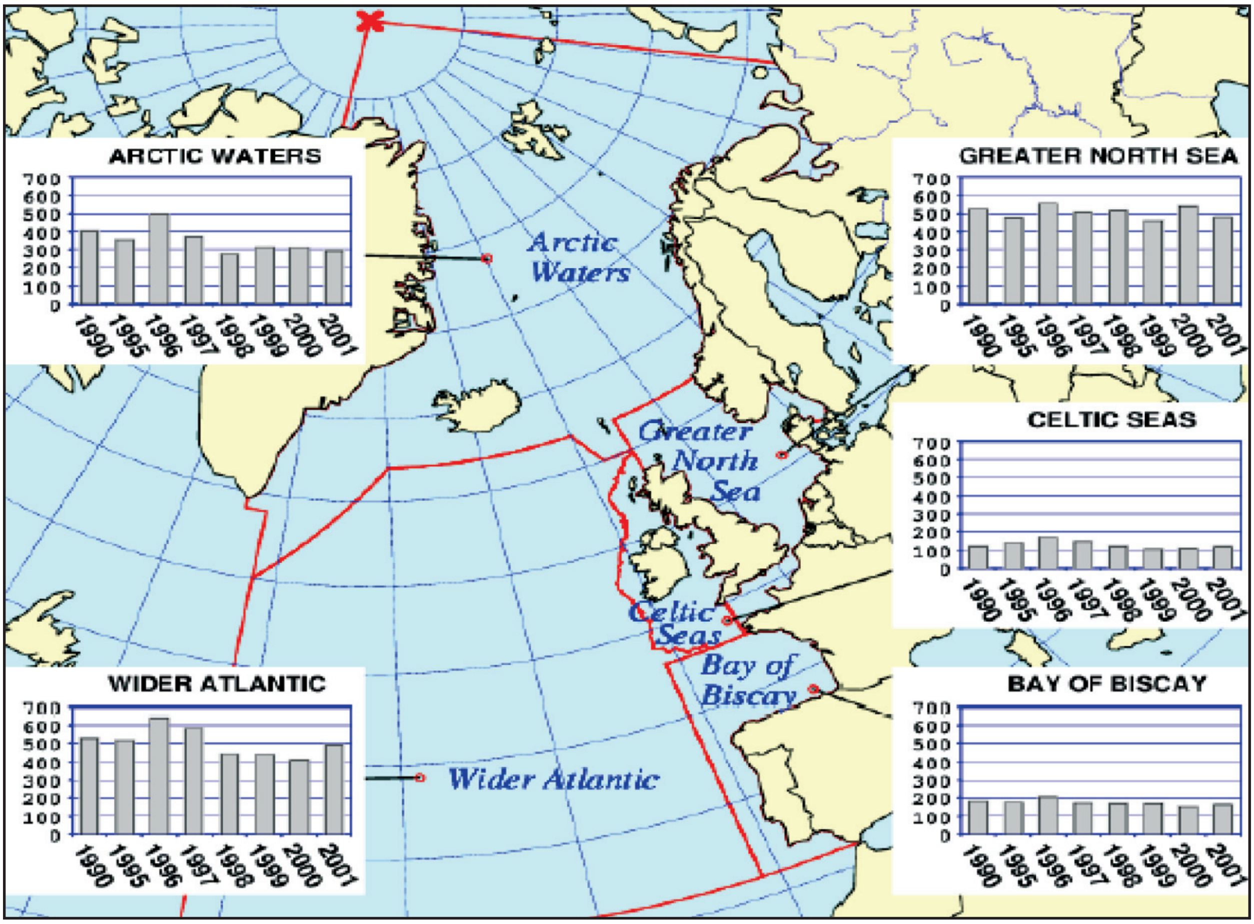

Fig. 3 Model estimates of nitrogen deposition to the North Sea, 1990-2001 (EMEP, 2004b)

Remembering the inextricable link with reported emission figures, it is noteworthy that the reported 15% fall in total nitrogen emissions is not obviously translated into a similar decline in nitrogen deposition to the north-east Atlantic and North Sea. Although uncertainties and approximations in parametreisations together with interannual meteorological variability may play a role in this, the restricted transformation after emission from slow to faster depositing forms of nitrogen can also be important. The benefits of emission reductions in large source countries near the North Sea, such as the UK or Germany, may then only be experienced at greater distances. If this is so, then the central tenet of the European Marine Strategy that emission control is the key to controlling pollutant loading clearly needs some care in application if objectives are to be met.

Deposition of toxic substances to the marine environment is also of concern, reflected in the coordinated monitoring of a variety of metal compounds by OSPAR countries. This offers the opportunity to explore the use of observed information in evaluating the state of, and rate of change in, the environment. Figure 4 shows reported changes in emissions of lead, zinc and arsenic during the 1990s, these decreasing by over 80%, around 50% and circa 25%, respectively. Figures 5, 6 and 7 display selected reported changes in depositions in precipitation during the same period. For each, observations from two countries are shown, selected from stations for which data quality has not been questioned (CAMP, 2005). For each of these the Sen trend line is displayed.

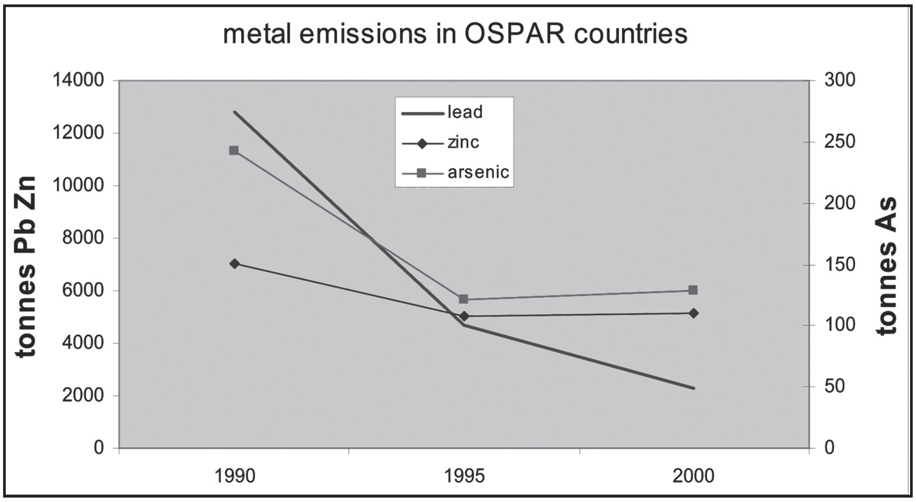

Fig. 4 Changing emissions of metals from OSPAR countries during the 1990s.

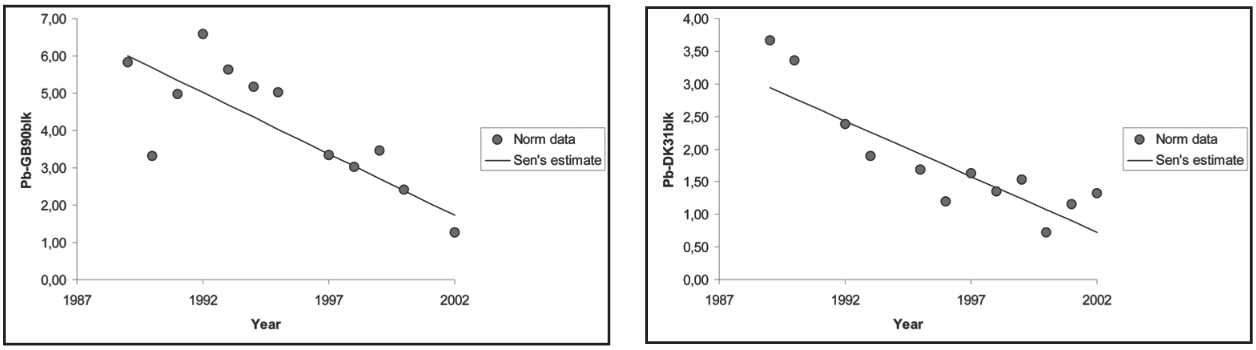

Fig. 5(a) & (b) Lead observations in UK (a) and Denmark (b). CAMP (2005)

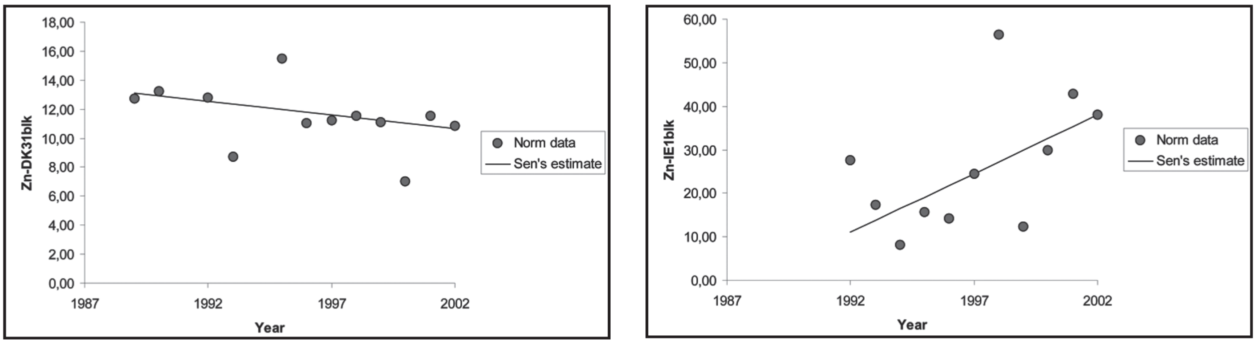

Fig. 6(a) & (b) Zinc observations in Denmark (a) and Ireland (b). CAMP (2005)

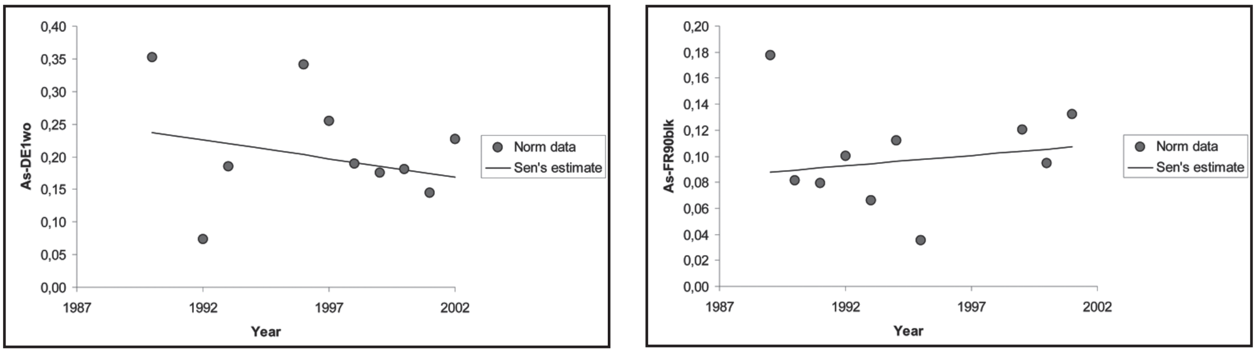

Fig. 7 (a) & (b) Arsenic observations in Germany and France. CAMP (2005)

Observed changes in deposition of lead in precipitation at the Danish and UK stations (Fig. 5) are in accordance with reported emission changes in the region, and indeed beyond. Lead release to the atmosphere has declined across Europe with the adoption of lead free fuel, and monitoring reveals a steady decline in inputs to the North Sea on both east and west coastlines. This correlation is not repeated for zinc. Atmospheric releases are reported to have declined by around 25%, but observed quantities in precipitation from Irish and Danish coasts are starkly inconsistent. The fall in precipitation zinc content in Denmark appears to be less than emission decreases, whilst on the Irish Atlantic coast there would appear to have been a rise in deposition. Possible explanations for the mismatch with emission figures include genuine environmental factors, such as transport from distant influential emission sources and/or the obvious very variable meteorology, or difficulties either in estimating emissions or in the sampling and analysis of zinc. Similar difficulty exists for arsenic. Emissions are believed to have declined by around a half within the OSPAR region, and yet observed precipitation concentrations do not indicate a corresponding reduction in atmospheric marine inputs. French and German observations are both variable, and the tendency in opposite directions. As before, transport from beyond the region, or difficulties either in estimating emissions or in the sampling and analysis of arsenic can lie behind this. It also reveals the difficulty in relying on simple statistical interpretations of 'trend'. In both cases the trend was found statistically significant at the 99% level, yet the spatial reality of transboundary transport renders it most unlikely that the trend can actually be opposite.

The difficulty in determining environmental status on the basis of current programmes has been encountered recently by the European Environment Agency. Data compilations of the direct or riverine discharge of metals and organic pollutants to the North Sea to produce FactSheets show declines in input of zinc and lead during the 1990s (arsenic not evaluated) (EEA, 2001). Similar FactSheet compilations for atmospheric inputs, however, were found to be simply not reliable due to the limited quantity and the variability in available data (EEA, 2004). Further difficulty arises in that the information there is for European marine areas tends to be focused upon northern and western seas. Figure 8 displays available inter-governmental data taken from databases held at NILU for cadmium deposited on the coasts from the atmosphere. Whilst North and Baltic Sea coverage is sufficient for conclusions to be drawn, there is simply no similar data available for Europe's other marine areas. Overcoming such a shortfall by use of research programme data is problematic. Such data is typically not immediately available, and furthermore cannot readily achieve the multiannual and geographical intercomparability which government sponsored programmes can seek.

SUMMARY

Supply of pollutants to marine areas from the skies in Europe is complex, reflecting the diversity in pollutants and their characteristics, the variation in sources, and the heterogeneity of Europe's physical environment. That inputs are not inconsequential has been well understood for some time, the atmospheric pathway frequently contributing a majority of pollutant supply to coastal waters. The large geographic scale of source regions for atmospheric pollution inputs is also known. This puts a solid emphasis on international collaborative action if management is to be effective, sometimes in contrast to riverine inputs which can be more local in character.

But the precision of our estimates is sometimes no better than 'order of magnitude'. For some regions, particularly the North and Baltic Seas, monitoring and modelling programmes go some way to permitting evaluation of the success of abatement measures. However, these programmes remain generally minimalist, one consequence of which is difficulty in distinguishing genuine changes in depositions from the multitude of factors which can contribute to natural spatial and temporal variability.

Over much of the rest of Europe's marine areas there is an absence of routine assessment. On the basis of the large geographical scales involved, and the 'order of magnitude' estimates we have, our best evaluation for regions such as the Black Sea is to surmise that patterns are anticipated to be similar to those elsewhere.

That is, falling inputs of nutrients, metals and organic pollutants, although the indications are that decline has not been as definite as it has been for riverine inputs.

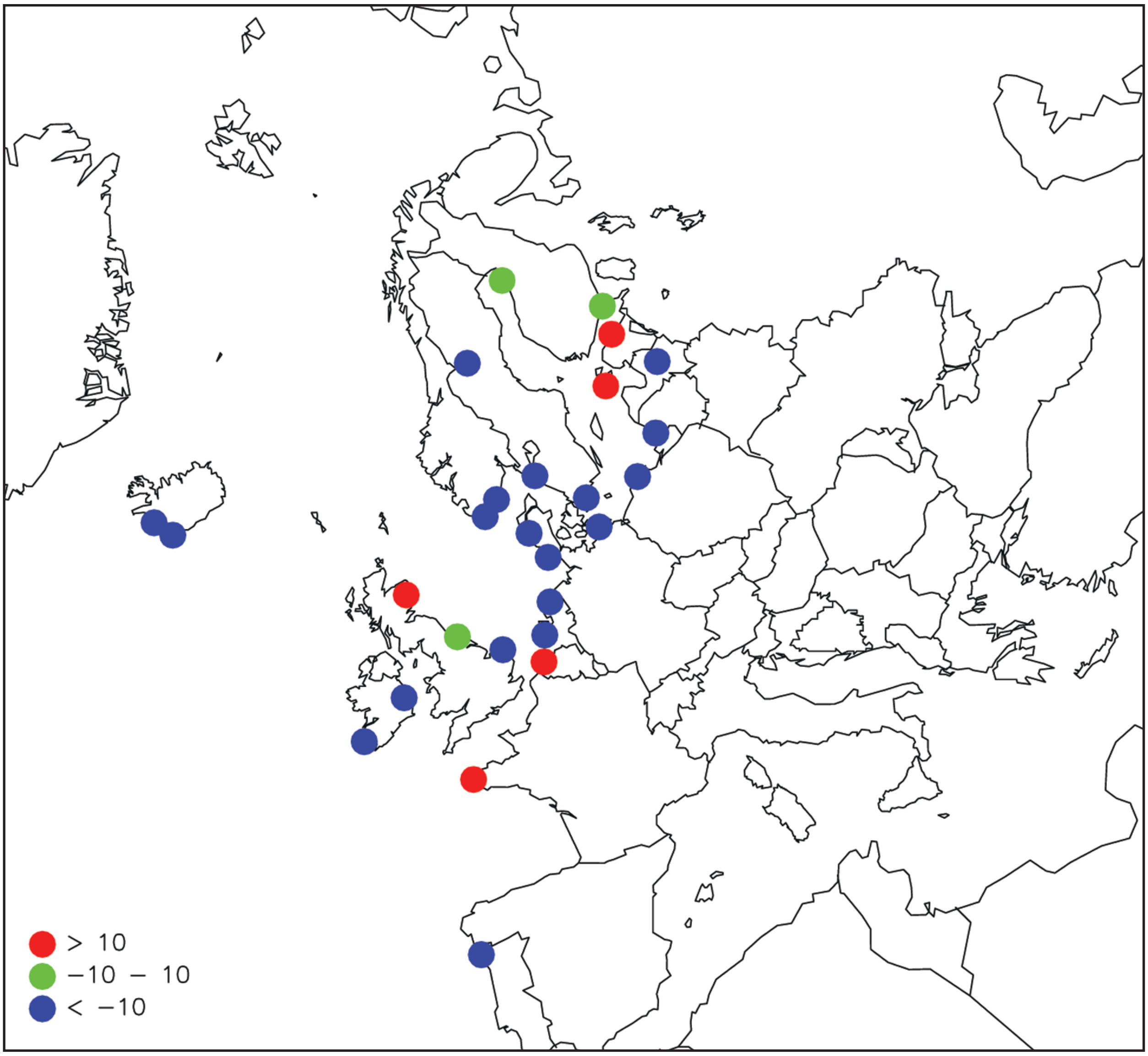

Fig. 8 Percentage changes in cadmium input to European marine areas from the atmosphere, 1995-2001. Compiled from cooperative intergovernmental programmes. Shows the focus on northern Europe, and lack of coverage for the south and east

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