CONTAMINANTS: Budgets and Behaviour

Contaminant passage through the land-ocean interface as seen by ELOISE & IMPACTS

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Executive Summary

The individual character of coastal zones originates in the interplay between terrestrial, marine and atmospheric environments. These mould and change the sinks, sources and transport of contaminants. The great diversity of Europe's environment means great diversity in patterns of contaminant dispersal. With biogeochemical flows having local, regional and global roles, changes in the coastal environment can thus have significant consequences at all scales. Feedback with human society can be strong, the scale and effectiveness of anthropogenic influence arising from its immediate proximity.

Contaminants travelling through the coastal zone do so via three media:

1) rivers
2) atmosphere
3) groundwaters and sediments

Traditionally, river inputs to coastal waters were seen as the principal route of contaminant transport, and for a number of substances passage through estuary and river systems are indeed obligatory. Flows are one-way.

Within ELOISE, the topic of riverine contaminant supply to coastal waters focused largely on heavy metal inputs from historical mining regions - Rio Tinto and Rio Odiel discharging to the Spanish Mediterranean, 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. These issues are local in scale.

The long term legacy of these elevated metal discharges may be 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.

For specific other contaminants supply to the sea from the atmosphere dominates. This pathway is also common for general contaminants arriving at marine locations which are distant from land. Atmospheric transport is not uni-directional, and flows can occur:

1) from the land via the atmosphere to the sea
2) from the sea to the atmosphere, potentially subsequently to the land

As much as 85% of lindane reaching the Baltic, and 50% of metals reaching the Mediterranean, may arrive via the atmosphere. Chemical forms less prone to washout in precipitation may be transported far from their point of release, whilst those in or converted to more easily scavenged forms display specific patterns of input to the sea.

For atmospheric contaminants deposited to coastal waters, mercury and persistant-organic-pollutants (POPs) have received attention within ELOISE and IMPACTS. Both of these are semi-volatile with potentially reversible air/sea transfer, and both are bioaccumulative, adding complexity to biogeochemical cycling. These features create a regional/global issue.

Description of deposition, exchange, partitioning, sedimentation, uptake etc. has been improved within ELOISE and IMPACTS.

An opposing atmospheric transport, from the sea to the land, occurs with sulphur. The coincidence of determining factors can render coastal seas a sufficiently important atmospheric source that in certain areas these emissions can have dominate local terrestrial acidification. ELOISE observations have found higher coastal water concentrations of sulphurous compounds than previously recognised.

Relevant to global scale environmental change, river plumes transport the greenhouse gases methane, nitrous oxide (N2O), carbon dioxide etc. Estuaries/coastal zones also represent a coincidence of factors arising from their land use, drainage, population, industry, etc. which favoure the biological production of greenhouse gases.

The interplay of transport routes can be complex, e.g. with the riverine supply of contaminats to coastal waters which results in gas release to the atmosphere and subsequent transport back to land.

Contaminants moving from land to sea generally concerns substances of anthropogenic origin. Releases from the sea to the atmosphere conversely often involves naturally occurring contaminants, although their production may be subject to anthropogenic influence.

Movement in groundwaters of terrestrial origin with subsequent seabed release has received much less attention, but has illustrated the potentially long time scale of contaminant release – decades.

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1. Introduction

1.1 Various characteristics

Coastal zones by their nature are the product of the coincidence and interplay between their physical attributes. For contaminants the particular coincidence of biogeochemical transfers and processes is one of the defining features of the coast. Coastal zones are obligate components in the bi-directional pathways of contaminant transfer between terrestrial and marine environments, the sharp conjecture between land and sea leading to adaptation and to change in the predominant flows. Sink, source and transport functions react accordingly. This coincidence of biogeochemical processes and pathways imparts an individual character of significance to coastal regions. Furthermore, with these biogeochemical flows having local, regional and global roles, the coastal environment can have meaningful influence.

To shift perspective slightly, changes in the coastal environment can have significant consequences near and far, from the alteration of coastal sediment balances through to influence on ocean biogeochemistry and climate change resulting from changes in carbon and green house gas flows. Indeed, it may be said that a key feature of coastal environments compared to the open oceans is that they are more often and more greatly affected by anthropogenic factors. Inevitably geographical proximity is a significant factor, dictating the scale and effectiveness of anthropogenic influence. The gamut of social and economic factors frequently mentioned as relevant to the coast can here be brought forward, and advantage sought in that future socio-economic changes may be more straightforward to elucidate than changes in natural forces.

The diversity of Europe’s geography, including its coastal environments, is oft noted. Consequent upon the resultant diversity in patterns of contaminant dispersal, is a wide spread of topics of investigation within ELOISE and IMPACTS. Studies have extended from contaminant flows in the Barents Sea to those in the north African coastal waters, and from consideration of river catchment management in central Europe to the characteristics of airflow across the Atlantic. Anticipating the changes to coastal zone biogeochemical functioning and structure which will express the pressure exerted is the challenge for strategic ELOISE science based knowledge on inventories, fluxes, impacts, fates and diversity. In this digest of the outcomes of ELOISE research work, we seek to examine various facets of contaminant flows through the coast.

1.2. The contaminants addressed and the structure of this digest.

In this digest we are defining contaminants as both anthropogenic and naturally occurring substances with the potential to pollute, and we are considering the parameters which have the main impact upon fluxes of contaminants. We are not considering nitrogen and phosphorus when serving as nutrients. This distinction reflects that a separate digest on nutrient dynamics is being produced by the ELOISE secretariat, in which considerations of nitrogen as a nutrient will be better served. However, in certain forms nitrogen is not primarily a nutrient, and its potential polluting effects are unrelated to its role as a nutrient. The role of N_2O as a greenhouse gas is one such case, and this digest will review ELOISE and IMPACTS progress in understanding these biogeochemical flows. The second boundary is drawn where the focus of interest shifts from biogeochemistry to climate change itself, as at this point a separate digest being produced on climate change is the more appropriate forum.

Four broad groups contaminant have been considered by ELOISE and IMPACTS projects, being persistent organics and hydrocarbons, mercury, heavy metals in general, and biogenic gases. The issue to be considered in this digest is to what extent and in which way ELOISe and IMPACTS have added to our understanding of the factors affecting fluxes of these contaminant groups in the coast.

A large number of ELOISE and IMPACTS projects have also focused on nitrogen flows. Whilst evaluated in a separate ELOISE digest, the processes and factors behind nitrogen flows can have parallel relevance to understanding of contaminant fluxes as defined here. Where appropriate, this digest also picks out findings of nitrogen projects for mention. In the table below the distribution of ELOISE and IMPACTS projects between the four contaminant groups plus nitrogen, and between the various European coastal sea regions is given.

Table 1. ELOISE and IMPACTS projects addressing contaminants and nitrogen

	Mediterranean	North Sea	Kattegat-Skaggerak	Baltic Sea	Atlantic, etc	Black Sea	General
POPs/ hydrocrabons							
Air	ADIOS, AIRWIN			POPCYCLING			
River						EROS-21	
Substrate							
Biology	BEEP, COMMODE, MATBIOPOL	BEEP		BEEP	BIOCET, COMMODE, FAMIZ		ACE, MARA
mercury							
Air	MAMCS, MERCYMS, MOE		MOE	MOE	MOE		
River	EUROCAT, TOROS						
Substrate							
Biology	BEEP	BEEP		BEEP	BIOCET		
metals							
Air	ADIOS, AIRWIN						
River	METROMED, TOROS	COMET	COMET			EROS-21	
Substrate							
Biology	BEEP, KEYCOP	BEEP	KEYCOP	BEEP	BIOCET		
Carbon, GHG, other							
Air		BIOGEST, ESCAPE	ESCAPE	BASIC	BIOGEST		
River	DOMAINE, METROMED	ECOFLAT, EUROTROPH	DOMAINE, EUROTROPH	DOMAINE	DOMAINE		
Substrate		BIOGEST, F-ECTS			BIOGEST		
Biology	INTERPOL	BEAM	BEAM		DOMTOX		
nutrients							
Air		*ANICE*	*MEAD*	*SIGNAL*			
River	*CHABADA, DITTY, DOMAINE, EUROCAT, EUROTROPH, METROMED, NICE, STREAMES*	*EUROCAT, EUROTROPH, HIMOM, INCA, NICE, OAERRE*	*DOMAIN, NICE*	*DOMAIN, EUROCAT, MANTRA-East, SIGNAL*	*DOMAINE, OAERRE*	*DANUBS, EROS-21, EUROCAT*	
Substrate		*COSA, F-ECTS, PHASE*		*COSA, RANR, SUBGATE*	*F-ECTS, NICE*		
Biology	*BIOHAB, CYCLOPS, COMWEB, KEYCOP, NTAP, PHASE, ROBUST*	*DANLIM, ROBUST*	*KEYCOP*	*COMWEB, DANLIM, MOLTEN*	*COMWEB, EULIT, NTAP, ROBUST*		

There are thirty projects dealing directly with the contaminant groups selected, plus over twenty further projects dealing with nitrogen, and having potential relevance to understanding of systems or processes. This represents a sizeable fraction of the total of 56 ELOISE and 12 IMPACTS research projects

Contaminant issues often reduce to evaluating whether coastal zones are sources or sinks. Seeking to address this topic whilst collating the breadth of ELOISE/IMPACTS research in a succinct manner, this digest focuses solely on the transport routes of contaminants. The sections are thus:

1) fluvial supply to the coast
2) Atmospheric pathways: from land to sea
3) Atmospheric pathways: from sea to land

4) Ground and sediment waters

Very few ELOISE and IMPACTS projects have focused on societal factors, and of those that have, scant attention has been given to interrelations with contaminants as understood in this digest. Thus, there is no separate discussion of the contribution of IMPACTS and ELOISE to the social parameters of contaminant flows.

2: The pathways in and the pathways out of coasts

There are three principal transport pathways (rivers, atmosphere, ground/sediment water) and two directions (to and from the sea) to consider. Traditionally, the seaward riverine movement of contaminants has been seen as the major transport route. Indeed, for a number of components and for distinct marine locations not only is the coastal zone an obligate pathway for contaminant transfer, but estuaries in particular are obligate uni-directional routes for dissolved and particulate matter passing from the continent to the marine system. For others, and for locations in the coastal waters further from land, the atmosphere may actually be the principal route. Nevertheless, in both cases research into contaminant flow to the coast has commented on potential anthropogenic impact. The third transport route, ground and sediment water from below, has considered both anthropogenic and naturally occurring contaminants. Interest has been driven by a focus upon anthropogenic origins, whilst in release from sediments there has also been interest in natural contaminant sources.

The flow in the opposite direction, of contaminants from coastal waters to the atmosphere, and on occasionsto land, is highly significant in some cases. Very often concern is with naturally occurring substances, the release of which may or may not be subject to anthropogenic influence. In all cases, land-to-sea or sea-to-land, the significance of the topic lies in the distinct combinations of factors which occur, putting an emphasis on the very coastal nature of contaminant exchanges.

2.a) Fluvial Supply to the Coast

The focus of projects has been upon potential anthropogenic impacts, and upon the associated processes and circumstances. In general, focus was not primarily upon overall budgeting of the European coastal seas. With general riverine flux data widely collated by state agencies, and although somewhat dispersed and variable, data has always thus existed from which basic estimates of fluvial contaminant to the coastal zone could be made.

The ELOISE projects evaluating specifically riverine contaminant supplies are BIOGEST (estuarine biogas, northern Europe – commented in section *2.c) Atmospheric pathways: from sea to land*), EROS-21 (various, Black Sea), EUROCAT (various, various), and TOROS (metal contamination, S.Spain). In addition, of partial relevance are the fluvial projects dealing with nutrients DANUBS, Balck Sea), INCA (various), MANTRA-East (north-east) and STREAMES (Mediterranean).

Metal mining at present and as a legacy has provided one theme of research into riverine transport of metal contaminants to the coast. The TOROS project focuses on fluvial transport from the long term mining regions around Rio Tinto and Rio Odiel to the coast of southern Spain. Chronic release (2500 years), mine drainage, phosphogysum waste runoff and acute discharge (collapse of Los Frailes tailings dam, Aznalcollar) are features of this case study. Comparison of the total transport of some metals drawn with other European river systems is a measure of the severe situation. For example, mercury concentrations flowing into the Bay of Cadiz are put at ten times greater than those transported by the Seine to the Normandy coast, generally held to be a relatively contaminated river. Comparisons with mine impacted rivers in the UK also shows the Rio Tinto to be a much more heavily metal laden river (467). Acidic and metal rich waters exist down as far as salinities of 30 $^{o}/_{oo}$ in the estuary.

Physical characteristics of transport are explored, to examine the sink mechanisms operating. Removal of Hg, As, and U by sediment and also seasonally by algae fixation (466) during transport to the coast is contrasted with limited removal of other metals in the coarse of downstream transport. Precipitation of dissolved components and co-precipitation with iron is also observed, and the factors dictating suspended particulate-dissolved metal relationships explored (468).

The project also attempts to budget the estimates of metal inputs into the western Mediterranean Sea, by rivers, modelling dispersion along the coast, and through the Straits of Gibraltar (221, 462). These figures indicate that for components other than arsenic the Rio Tinto is far from an inconsiderable source of total inputs. That this Mediterranean river system is so much more heavily metal laden than other local rivers indicates the long term legacy which can result from mining in Europe. Simultaneoulsy, however, evaluation of the effects of acute release with the Los Frailes tailings dam collapse indicated no impact on coastal chemistry after six months (237). Of course, the high existing baseline should be recognised when interpreting such an assessment.

Table 2: Contaminant flows to Western Mediterranean, tonnes per year
From TOROS: (462)

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

Negative numbers indicate flow out of the Mediterranean

Another case with restricted long term consequences of elevated contaminant discharge is the Danube. The EROS-21 project demonstrated that despite high discharges in the past before recent economic changes, at present the fluvial flows of contaminants into the Black Sea from the Danube are not generally high (186). Only copper and lead show elevated inputs of metals in particulate form, with copper alone being elevated in dissolved form. The source of this is believed to be upstream mining. Release from surface runoff or contaminated sediments does not seem to be a particular issue. The Danube system, notable as a transnational river open to variable control strategies, does not appear to act as a conduit for elevated metal input to the Black Sea. Indeed, it appears able to ameliorate both any release of metals to the river system further upstream, and release from sediment Amongst organic contaminants, organochlorine levels were found to be ‘average’ for European rivers. HCH registers higher, this being evidence of continued lindane use within the catchment (188).

A further evaluation undertaken within the EROS-21 project was to review the rate of transmission of radioactive material along the river courses (189), radioactivity arising from the Chernobyl accident. A five-ten year delay was observed before the peak inflow of radioactivity occurred in the Danube and Dnieper.

Remnants of historical mining activities are found in a number of other catchments across Europe. A number of such cases were studied in the EUROCAT project, including the Humber, Rhine, Idrijca (Slovenia) and Vistula (Poland) catchments. Whilst primarily concerned with nutrient flows, some aspects of their contrasting characteristics were described. In the Humber continued contaminant release from sediment is the legacy of historical mining and industrialization, much in the same way as the Rio Tinto. In the Idrijca current mining and release from historical contamination both occur, whilst in the Rhine diffuse contaminant sources now exceed point sources, whether from direct discharge or from contaminated hotspots. In the Vistula historically contaminated sediments are prevented from reaching the coastal by extensive damming. However, damming cannot be necessarily considered a permanent prevention of contaminant release. Sediment contaminated with PCB still reaches the Rhine estuary (392). Scenario analysis with costing of options indicates the economic choices to be made if there is the political will for contaminant abatement.

The worlds second largest mercury mine operates in the Idrijca catchment. For such a specific case it has been possible to budget the total loss from mining operations - 37,000 tonnes, with a current annual river transport to the coast reduced to 1.5 tonnes into the Gulf of Trieste. Evaluation of the processes of mobilization has highlighted two principal routes, erosion of contaminated soil (also found by TOROS on the Rio Tinto – 237), and methylation. Hence, options for reducing the problem include soil erosion control, with reduction of organic loadings to rivers a possible means of restricting the methylation process (392).

Methylation is an issue researched by a number of ELOISE projects. This process will be considered in its own right in section *2.c) Internal flows and effects.* Of relevance to this section on riverine contaminant transport, brief attention is given here to the role of riverine organic material in promoting methylation. The MOE project found that mercury in particulates can be deposited on coastal shelves after riverine transport, and that mercury is often strongly associated with humic substances, subsequent methylation increasing the potential for bioaccumulation (209). Such binding of mercury influences different aspects biogeochemical cycling. As well as bioavailability increasing, evasion from seawater decreases compared to freshwater (209). In the Black Sea the deposit of organic material in proximity to river mouths is seen to be a recent feature (196) which increases the possibility for methylation. Bringing in understanding gained from nutrient projects at this point, for eaxample DOMAINE examined the transport of dissolved organic matter by rivers to the coast. It was found that the amount of dissolved organic matter exported is dependent on land use and vegetation in the catchment of origin. That the phenomenon in the Black Sea is a recent one is an indication that organic loading is related to economic factors, and the driver-pressure descriptors in the D-P-S-I-R classification could be readily applied if wished. In common with the Idrijca, the organic loading - methylation characteristic, and the knowledge gained from other projects as to underlying controls, open avenues to control, according to political desire and cost.

The scale of river inputs of contaminants vis-a-vis other transport routes has been estimated by ELOISE projects on some occasions, as in table 2. One contribution of ELOISE/IMPACTS in this respect has been a significant contribution to appreciation of non-riverine transport routes for pollutants. For substances long in the political arena, such as nitrogen (not a 'contaminant' in the context of this digest), the restricted contribution of river inputs has long been noted – the seas delivering maybe no greater than 60% of inputs to European northern regional seas. The ANICE project has recently confirmed such estimates (283). Evidence from TOROS, POPCYCLING and MERCYMS amongst other projects indicates still lesser riverine contributions to coastal water inputs for metals and persistent organics. From the estimates in table 2 from TOROS of metal supplies to the western Mediterranean, it can be seen that rivers supply only around 50% of inputs. More markedly, POPCYCLING estimated riverine lindane supply at 15% of total input to the Baltic Sea, and assumed γ-HCH to be representative for persistent organic compounds.

The hydrological processes behind contaminant transport are an aspect of understanding their supply by rivers. Processes operating will change with transport along the river, e.g. from stream driven turbulence and air-water exchange at higher eaces, and wind driven turbulence and exchange at lower estuarine reaches (213). One project addressing the representation of these was INCA. Although primarily concerned with nitrogen, the project did develop fluvial and hillslope hydrological modelling approaches. Elements such as modelling pre-existing soil moisture content and river bank storage (326) as factors in runoff generation, and estimating the hydrologically effective rainfall amounts (334) are factors which could improve management where runoff (as with possibly the Rio Tinto and Idrijca) and ground leaching were perceived as issues. The potential relevance of these investigations thus goes across projects and topics (392). Naturally, these tools would require tailoring to the contaminant in question given the differences in physical and chemical behaviour of components, but as this has already been undertaken on one occasion (extension from nitrogen to phosphorus – 347) the concept is not new. Specific site calibration is also always necessary, as INCA is not entirely generic.

Hydrological modelling was also undertaken by the RANR project. Although also concerned with nitrogen flows rather than contaminants as defined here, attention was given to understanding hydrology, and to separating the flow into baseflow, throughflow and runoff components at the catchment/hillslope stage (312). It was found that in sandy soils baseflow dominates the hydrograph, whilst in loamy soils throughflow and surface runoff were predominant. The speed of response, and the propensity to either surface erosion or increased solute loading of flow are factors of relevance to managing contaminated catchments. The lumped rainfall-runoff model used, NAM, is an empirical tool which requires calibrating to the particular catchment in order to obtain the hydrological characteristics of the chosen location, this being a standard procedure. If the ability to predict chemical flows was also required, the chemical characteristics of the contaminant of interest would need to be substituted.

The extent of penetration of river inputs into the receiving sea was evaluated by various projects. The dispersal of metal laden waters in the Bay of Cadiz from the Rio Tinto was examined by TOROS. In the Black Sea EROS-21 noted the widely observed phenomenon of deposit of particulate loads in the near estuary and shelf region, with transport of dissolved components into the open sea (090). Going further, the project also uncovered the penetration of the Bosphorus halocline by river flow, in so doing also demonstrating the potential limitations of modelling as sole data source. Prior general circulation modelling indicated extreme stratification, and limited deep water mixing. Modelling of ^{90}SR tracers arising from the Chernobyl accident had been able to mimic the penetration of the halocline in the Bosphorus only using time and space averaged data (074), individual events with short memory could not be simulated without higher resolution and higher quality function representation.

2.b) Atmospheric pathways: from land to sea.

The EOISE and IMPACTS projects have looked at a small number of contaminants following this pathway. For mercury, the majority of sources are located outside coastal zones, and moreover whilst contributing to both fluvial and atmospheric transport are generally unconnected with watersheds. Unlike most metals whose natural release is limited to erosion and leaching processes, the semi-volatility of mercury adds to the complexity of its biogeochemical cycling, lending it more in common with certain POPs than other metals. The lesser tendency of some mercury forms to precipitation scavenging from the atmosphere also gives candidacy for longer tropospheric lifetimes. Other forms, principally Hg^{II}, are soluble, have propensity to scavenging, and have a higher dry deposition velocity. There is thus an interplay between geological mercury sources, industrial exploitation, and the chemical and physical qualities of the contaminant which will influence coastal flows. Atmospheric transport and deposition of these forms of mercury to the coastal zone has been studied within the MAMCS project (287), indicating the significant role of meteorology in defining source regions for mercury supplied to the Mediterranean. Atmospheric transport is less site specific than the riverine pathway, with the scale of source-receptor relationships being regional-global. With coastal mercury flows having dependence on regional/global emission patterns, evaluation and management can reasonably be considered as rather independent of the traditional catchment concepts of water management as used in some European countries since for several decades. The focus on riverine catchment–coast interactions for fluvial contaminants might need supplementing with the concept of airshed-coast interactions. Such scales are often the domain of multilateral agreements, rather than local managers. When it is considered that inventorying of the contributions of various economic sources to total anthropogenic mercury emissions in Europe, as undertaken by the ELOISE projects MAMCS, MOE and MERCYMS, indicates that coal combustion (power plants and residential heat furnaces) generates more than half of the European emissions (203), the restricted role for local management is emphasised. Defining the interplay between atmospheric and riverine supply forms one aspect of the current project MERCYMS on Mediterranean mercury cycling, specifically the current construction of inputs databases (www.iia-cnr.unical.it/MERCYMS/project.htm).

Enlarging on the volatility of mercury, a characteristic feature of the environmental behaviour of certain persistent organic pollutants (POPs) are their tendency for undergoing reversible atmospheric deposition to various terrestrial and aquatic surfaces, as well as the propensity for reversible deposition between the aquatic and the sediment compartments. To model this, and to seek out source-recptor relationships, the POPCYCLING project built a multi-media model based on the fugacity of compounds (494).

The results lend support to the view that for marine waters, deposition across the air-sea interface is the main route of supply of persistent organic pollutants. The table below provides estimates of the split between riverine supply and atmospheric supply to the Baltic Sea (492). Evaluation within POPCYCLING has aided the description of the principle processes involved in sea-air exchange of POPs, as did the AIRWIN project 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 (069). Following AIRWIN, trophic status also has a role to play in determining chemical partitioning with implications for bioaccumulation. The pathways to bioaccumulation were the subject of the BIOCET and the FAMIZ projects, potential influence on reproduction of cetaceans the topic of the latter. The possible influence of POP's on eagles and seals was given some attention in POPCYCLING (063, 064). There is some suggestion from these projects of absorption of organic compounds in open waters, and conversely of volatilisation nearer to the coast. Whilst the exact parameterisation of such features will inevitably be case-specific, a valuable generic benefit is the understanding of process interactions which will apply to other substances of varying solubility.

Table 3.Relative importance (in percent) of riverine and atmospheric supply of lindane (γ- HCH) to various parts of the Baltic Sea (492).

	γ-HCH	γ-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

ANICE applied models to better determine the atmospheric nitrogen supply to coastal seas. The relevant feature for contaminants is the evaluation of close-coast meteorological processes and characteristics. These physico-chemical features have potential consequence for different contaminants than nitrogen.

2.c) Atmospheric pathways: from sea to land.

Coastal regions are an important source of atmospheric sulphur, and have influenced the policies developed between European countries to reduced acidification through emission control. For certain areas, such as northern Norway, marine natural sulphur can have overriding importance in acidification. Biogenic sulphur can also have influence on climate, e.g. through the creation of condensation nuclei for cloud formation. An exploration of the climate link was provided within the ESCAPE (149) and ROBUST (126) projects, these seeking out many of the relationships involved.

Emissions of sulphur to the atmosphere over sea areas is as dimethylsulphide (DMS), this subsequently oxising to sulphate (easily scavenged, with restricted dry deposition velocity). Phytoplankton community composition and biomass are primary determinants of DMS concentrations in seawater, and of its precursor dimethylsulphoniopropionate (DMSP). Whilst there is relatively little DMS production from actively growing phytoplankton (349), higher rates of DMS release result from zooplankton grazing pressure (215), which encourages breakdown of DMSP. Bacterioplankton are significant agents for this, with viral agents also able increase DMS production notably (73,148). *Phaeocystis* sp./zooplankton herbivory consumes particulate DMSP at rates equal to DMS production, and hence blooms can be sources of high DMS concentrations. The various interrelationships lead to correlations between locations of microzooplankton biomass, phytoplankton, and nearshore waters. Highest biomass is nearshore where zooplankton grazing of phytoplankton crops productivity (374).

The ROBUST project has also looked at the mechanisms behind DMS production. Anoxic sediments are proposed as a particular source of DMS, with eutrophic coastal or sedimentary environments favouring *Phaeocystis* sp., *Ulva* sp. and *Enteromorpha* sp. The establishment of high concentrations of DMS in coastal waters is a necessary step for emission to the atmosphere,and one which ELOISE projects have contributed to. The step to identifying the scale of atmospheric release requires consideration of sea-air exchange, and with this of existing air concentrations. This step for sulphur compounds was not taken by ELOISE or IMPACTS projects. Whilst not the purpose of this digest to conduct such research, it is interesting to compare the observed DMS concentrations observed by ELOISE projects in European coastal waters with values used for past estimates of the global sea-air flux of sulphur.

Table 4: Potential for coastal emissions of dimethylsulphide to the atmosphere

source	**sea**	**sea concentration** nM	**Flux to air** $\mu mol\ m^{-2}\ d^{-1}$
Erickson et al 1990	Northern Hemisphere, Jan - July	1.2 – 2.1	4.3-5.4
	North Pacific coast	1.8 – 5.0	6.4-13.8
ELOISE (148)	Mediterranean	1.3 – 7.3	4.0-22.6
	North Sea	0.8 – 65.0	2.5-201
	North Atlantic	2.7 – 9.8	8.4-30.3

The quoted values for sea concentrations and for sea to air fluxes from Erickson *et al* (1990) are here compared with the observed sea concentrations obtained by the ESCAPE project. The Erickson *et al* values are only an example of those available in literature, and are comparable to the overall global approximation provided by ROBUST of 3nM (126). Comparison is also made with approximate fluxes obtained by multiplying those concentrations with the sea to air transfer velocities estimated by Erickson *et al*. Clearly, the sea concentrations of DMS observed within ESCAPE are sufficient on their own to produce a far greater flux from European coastal waters than some global sea-air estimates have previously.

This remains a step from evaluating the flux to the land surface, there are reasons to believe that the coastal flux of sulphur to the land could be greater than previously estimated. Not only are ELOISE observations revealing greater concentrations of DMS, but they are doing so in the coastal waters, in closer proximity to the receiving land surface. Further, the atmospheric modelling traditionally used to estimate acid loading in Europe does not operate at fine enough spatial scale to reflect near-coast or estuarine sea concentrations. Finally, decreasing anthropogenic sulphur emissions in recent years will have increased the evasion from the sea surface of biogenic sulphur.

Several projects investigated greenhouse gases arising from the fluvial system. EROS-21 picked out the region subject to the Danube river plume as an important source of coastal dissolved CH_4 concentrations, competing for primacy with seeps from geology and with methanogenesis in sediments (192). More specifically, it was found that bacteria near the river mouth, shallow water bottom sediments and delta regions have greatest significance for atmospheric CH_4 releases. Other gases involved in climate impacts include N_2O which is dependent on the anoxic conditions encountered in the Black Sea.

In the Mediterranean, different projects have pointed to the significance of river plumes for CH_4 and N_2O production. The role of the deltaic sediments is seen in the findings of the PHASE project that biogeochemical cycles below the euphotic zone and pelagically driven. The METROMED project focused on the characteristic of estuaries and coasts that they often have high loads of organic and mineral particles, and of inorganic nitrogen, providing ideal conditions for CH_4 and N_2O production. That such coastal environments represent a key combination of factors behind the generation of these gases is indicated by observed concentrations in the Bay of Lions/Rhone estuary and the Gulf of Thermaikos/Axios estuary, which were 10x and 100x those of open ocean waters. It is to be anticipated that gases may be produced in sediments and thence transported to the surface waters.

The BIOGEST project focused precisely on biogas generation, having *phaeocystis* sytems at its centre. Taking a coupled estuary ecosystem-atmosphere approach, it seeks to determine and evaluate the atmospheric fluxes of biogases in European estuaries and their impact on the global budget. The biogases CO_2, CH_4, CO, non-methane hydrocarbons, N_2O, NH_3, DMS, COS, halogenated organic compounds and biogenic volatile metals were evaluated in several European estuaries, showing that these environments are important marine sources of a wide range of gases linked to biological systems. Estuaries/coastal zones represent a key coincidence of factors arising from their morphometry, land use, drainage, population, industry, river discharge and climate, which lead typically to factors favouring biogas production (088). These include high organic/nutrient loading to estuaries, sediment oxygen depletion, methylation, and high productivity. The example of CO_2 in the Scheldt shows that whilst river systems might themselves transport appreciable quantities of biogas produced by soil and river respiration, heterotrophic activity in the estuaries dominates, being responsible for 90% of dissolved carbon dioxide (260). This may be lower in more turbid less productive estuaries (e.g. Gironde, 086). Further, advective fluxes from the Scheldt to the estuary, and from the estuary to the North Sea were an order of magnitude smaller than estuarine water-air exchange. Thus, the estuary is annually a net source of atmospheric CO_2. Generalising beyond the Scheldt, estuaries are found to be supersaturated with carbon dioxide, and are estimated to be equivalent to 5-10% of European anthropogenic carbon dioxide release (037). This figure understates both the true anthropogenic influence, and the actual potential contribution of estuaries to total releases: not only is most respired labile carbon in polluted European estuaries itself anthropogenic in origin, but for the rest of the world where atmospheric anthropogenic emissions are lower and where significant fluvial organic carbon loading results from overpopulation, the estuarine contribution to the total will be higher.

Beyond investigation of estuarine and coastal carbon dioxide (139, 256, 260), studies were conducted of volatile metal(loid) species (019), volatilization of organotin compounds (112, 274), volatile selenium species and alkyl-iodides (026, 273), alkyl-metal(loid) species (170), mercury speciation in environmental matrices inc. estuarine muds (018, 219). Other biogeochemical trace contaminant atmospheric flows which may have regional and climatic roles and which can be influenced by the coastal environment are those of iodine and selenium. As well as local cycling, methyl iodine may operate as a replacement for methyl bromine, and estuaries may account for around 20% of European anthropogenic emissions (EROS-21). Organic substances can also be volatised from coastal environments, e.g. organotins from microbial/chemical methylation in anoxic estuaries. This is a contaminant with global scale cycling.

The land-to-sea transport of mercury has been discussed, and its volatile nature commented. Here the reverse sea-to-land transport is described. Simple observation data on the marine source can be found in the MOE project. Whilst essentially a model evaluation, paper (288) uses 3 coastal stations for the purpose, allowing areas supplying peak and background levels of observed atmospheric mercury to be estimated. Atlantic and Mediterranean waters are shown to be supersaturated with Hg and thus to emit mercury to the atmosphere (191). This agrees with other opinions that air from the Atlantic and Mediterranean have elevated Hg content, whilst air further from oceans (e.g. over Scandinavia) have low levels (206). The implication is the potential for sea to land transfer within a coastal strip. Mechanisms are not fully quantified, but a possibility is the over-sea formation of particulate $HgCl_2$. Not only would this form experience higher rates of dry deposition through its large particle size, but scavenging by precipitation would also not be inconsiderable (211). Coastal meteorology in particular could then be expected to be of consequence. A shift to more turbulent air flow as air masses make landfall would encourage dry deposition, and any increase in precipitation would enhance removal.

c) Ground and sediment waters

The attention to groundwater flows by ELOISE and IMPACTS has been restricted, but illuminating. Sampling by SUBGATE showed that contaminant deposits to terrestrial surfaces can emerge many decades later in coastal waters after transmission through groundwaters. CFC's used as tracers and observed in groundwaters below the Kiel Bight (264) suggest re-emergence of waters of terrestrial origin in the Baltic after over 50years. The great difficulty in generalising this information, however, is the high variability in such flows.

EROS-21 examined the transfer of compounds from sediments to the water column, and further to shelf sediments and to the deep water sediments (28). Whilst in the Black Sea processes are notably efficient due to sulphide scavenging and lateral transport of metal, and whilst there is limited mixing of deep waters in the special case of the Black Sea, the processes regulating dissolved iron migration, and the central role of anoxic conditions, are generic features with application beyond the Black Sea.

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