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Eutrophication and Contaminants in Aquatic Ecosystems

Eutrophication and persistent pollutants are two of the main environmental problems in European marine and freshwater ecosystems. As they tend to co-occur, interactive processes between eutrophication and contaminants are suggested, that may lead to environmental effects that cannot be predicted from each process alone. In order to predict the consequences of remedial measures (changing the input of organic matter, nutrients and contaminants) it is important to understand mechanisms that alter the bioavailability and fate of contaminants. The environmental risks will depend on the speciation of contaminants and their association to media and matter and by that means affect exposure. Furthermore, the risks will depend on the mobility of the substances and their pathways in food chains. In 1995, the Swedish Environmental Protection Agency initiated a 5-year research program *Interactions between EUtrophication and CONtaminants* (EUCON). A background document was prepared listing a number of relevant questions and hypotheses. On the basis of this document a program was launched, addressing the problems related to the interaction between eutrophication and contaminants (persistent organic compounds and trace metals) in the marine environment, with focus on the Baltic Sea, and in lakes. This paper summarizes the state-of-the-art, hypotheses and highlights from the research program with emphasis on the implications and applications of the results.

INTRODUCTION

The aquatic environment acts as a recipient of urban wastewater which contains large quantities of nutrients (phosphorous and nitrogen) and oxygen demanding substances, i.e. organic matter. In recent years, there has been political pressure to implement treatment systems for sewage to reduce the input of nutrients and organic matter, in order to avoid extensive primary production and anaerobic bottom environments, eutrophication, in sensitive coastal areas and lakes.

In addition to eutrophication, the aquatic ecosystems are exposed to persistent pollutants like PCBs, DDT, PAHs, dioxins, and toxic trace metals. The primary input of these contaminants in the western world has decreased considerably during the last decade due to strict national and international regulations. Despite efforts to reduce inputs, secondary sources like contaminated sediments, dump sites, contaminated land and the technosphere supply the aquatic environment with contaminants. Additionally, volatile contaminants are atmospherically transported long distances and deposited in the aquatic environment, far away from the source areas.

Traditionally, the environmental effects of eutrophication and contaminants have been studied separately without a holistic approach. Consequently, most environmental decisions are based on our knowledge of separate effects, assuming that interactive processes which may influence the overall effects are non-existent. There are studies, however, that show that the trophic status may influence the bioavailability and the mobility (cycling) of contaminants (1, 2). Additionally, the level of contamination may also influence the primary production and indirectly the status of eutrophication (3, 4).

Knowledge about potential interactive mechanisms is desirable both with respect to remedial action and as a basis for the development of environmental criteria and classification of contaminants with respect to risks. It is important to understand the basic processes taking place in ecosystems that will influence the fate of contaminants with respect to transport, biogeochemical cycling, persistence, bioavailability, bioaccumulation, biomagnification and direct effects on the organisms; e.g. toxicity (5).

In 1995, the Swedish Environmental Protection Agency initiated a research program (EUCON) (6) to increase knowledge about interactions between eutrophication and contaminants in the aquatic environment. A total of 10 research projects received financial support, covering the following scientific topics:

– The impact of eutrophication on the distribution and sedimentation of lipids and organic contaminants in the marine environment.
– Development of analytical methods for analysis of lipids in suspended particulate matter and bottom sediments in the marine environment.
– The effect of eutrophication on the transport of organic contaminants and trace metals in lakes.
– Eutrophication and contaminants - effects on plankton mediated processes in lakes.
– Modelling of processes related to the interaction between trophic status and contaminants.
– Trend analysis of contaminants in fish in the Baltic Sea.
– Implications of changed nutrient loading for the bioaccumulation of hydrophobic organic contaminants in Baltic Sea blue mussels.
– Accumulation of contaminants in Baltic Sea sediments.
– The effects of changes in the species composition and nutrient limitations on the accumulation of organic contaminants in marine phytoplankton.
– Benthic processes related to contaminants and marine eutrophication.

The topics included both the marine and freshwater environments, pelagic as well as benthic systems, field and laboratory studies, methods, ecological processes and effect studies. A number of publications from this research program already exist and several papers will be published elsewhere in the near future.

This paper summarizes important results obtained within the EUCON-projects in relation to overall objectives and hypotheses. The implications with respect to the use of the results are also emphasized.

OBJECTIVES AND RESEARCH QUESTIONS

The main objective of the EUCON research program was to assess and predict interactions between contaminants and eutrophication in aquatic ecosystems and to elucidate their effects. The main objectives were to provide the scientific basis for future remedial measures in ecosystems with both eutrophication and contaminant problems, and to provide a scientific basis for interpretation of long-term monitoring data on contaminants, taking into account the importance of eutrophication.

A spin-off effect of this work is that the results may be useful in the process of establishing water and sediment quality criteria for different aquatic systems, taking into account interactive processes.

The EUCON program established in its initial phase some questions that needed to be addressed:

i) Are the concentrations of bioaccumulative substances in marine biota likely to decrease as a result of dilution in a larger biomass caused by eutrophication?

ii) Has eutrophication caused decreasing concentrations of contaminants in the water mass and pelagic biota in the Baltic Sea?

iii) Has eutrophication caused an increased sedimentation of contaminants?

iv) Has eutrophication caused an increased trapping of contaminants in sediments in lakes and in the Baltic Sea during the last decades?

v) Are the concentrations of contaminants in the water mass and pelagic biota likely to increase if the nutrient input to the sea is reduced, without a simultaneous reduced input of contaminants?

Important processes and mechanisms related to the interaction between contaminants and eutrophication are illustrated in Figure 1.

The extensive questions above could not be fully answered during a 5-year research program. Some results obtained were not conclusive and some results were obviously site-specific and could not lead to generalizations. Some fundamental questions related to biomass dilution, growth dilution, effects of dissolved organic carbon, sedimentary processes, sediment-water interactions, atmospheric exchange and lipid and food-web structure have been addressed.

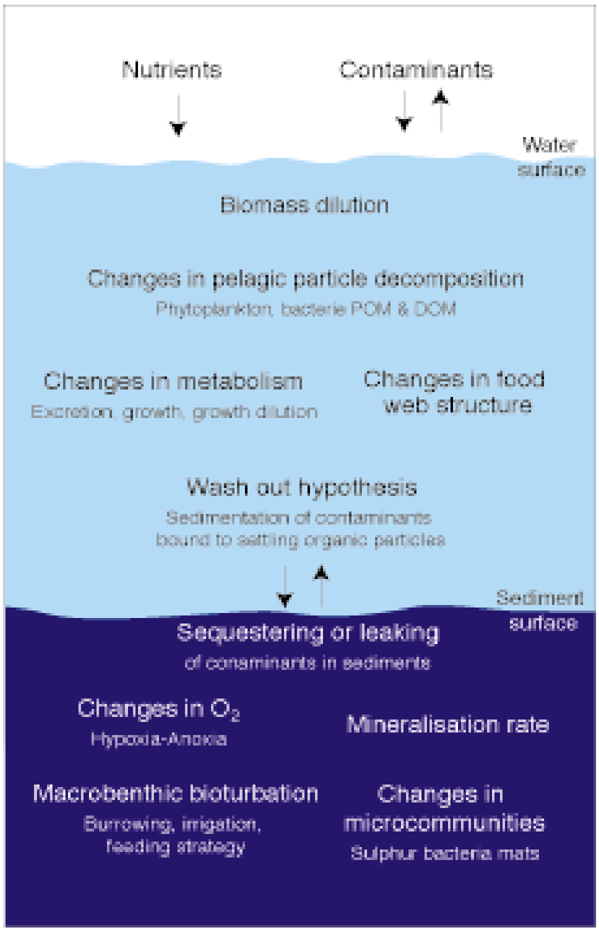

Figure 1. Processes and mechanisms related to the interaction between contaminants and eutrophication in the aquatic environment (from Gunnarsson et al. 5).

INTERACTION BETWEEN CONTAMINANTS AND EUTROPHICATION: STATE-OF-THE-ART

Effects of Eutrophication

Eutrophication in freshwater and coastal marine areas includes a series of processes that alter the ecosystems significantly. An increased nutrient input causes a stepwise change in productivity, with phytoplankton biomass increase being one of the most obvious effects. Eutrophication initiates a web of processes, including changes in the physical environment like reduction in light climate and a consequent stepwise change in the transformation of light and heat transport in the water.

Phytoplankton biomass may cause increased sedimentation of organic matter following a bloom that may result in enrichment in the sediment. As a result, anoxia may develop in stratified and stagnant areas. The increased productivity may not only increase the biomass, it will also change species composition and diversity of the phytoplankton community. The reduced light conditions caused by the phytoplankton, might negatively affect the vertical distribution and productivity of other primary producers such as attached algae or submersed macrophytes.

The negative effects of a plankton bloom in the Baltic are presented in Figure 2.

Altered ecological conditions of primary producers in a lake or in marine coastal areas may cascade to other trophic levels. The first link of consumers, the zooplankton, may change mainly in population structure as a result of the changing species composition of their predators, the second link consumers. In lakes, cyprinid species like roach (*Rutilus rutilus*), and juvenile bream (*Abramis brahma*) may be dominant, and as these species are effective as zooplanktivores, the zooplankton community is subjected to high predation pressure. The community is likely to turn

Figure 2. Discolouring of water in the Baltic Sea due to dense plankton bloom. Photo: P. Jonsson.

to smaller species and the larger forms of, e.g. Cladocera, which are effective grazers, are suppressed (7).

The third link consumers, piscivorous fish, change from visual hunters such as pike (*Esox lucius*) and perch (*Perca fluviatilis*) to a dominance of predators, walley (*Stizostedion lucioperca*), using other predation strategies, due to the reduced light conditions. The benthic invertebrate fauna is initially favored by eutrophication, but at later stages shows loss in diversity and a dominance of species that can tolerate low oxygen conditions. The eutrophication scenario described is general, but may include other species in coastal areas or in particular geographical regions. The general ecological effects of eutrophication can be summarized as:

i) Altered food-web structure, often with fewer links in the food chains.
ii) Changed population structure, where the base of juvenile individuals is broadened and the life span of the species is reduced.
iii) Loss of species diversity, but increased abundance of species that tolerate or benefit from a changing environment.
iv) Eutrophication-induced hypoxia or anoxia that may cause dramatic changes particularly in the benthic system.

The Fate of Contaminants in Eutrophic Systems

The eutrophication scenario has implications for the fate of hydrophobic organic compounds (HOC). Increasing nutrient loads on aquatic ecosystems, result in effects on pollutant transport, uptake in biota and possibly effects on the ecosystem.

The first study that linked eutrophication to the fate of HOCs was carried out in the archipelago of Stockholm, Sweden (8). That study concluded that an increasing biomass in an aquatic system, due to increasing nutrient amounts, dilutes HOCs and the result is lower concentrations of pollutants in individual organisms. The hypothesis was valid under certain circumstances but, as for all hypotheses the conclusion has been questioned, and several other explanations exist for why HOC concentrations are lower in zooplankton and fish in eutrophic, aquatic ecosystems compared to nutrient-poor systems.

One key to an understanding of the interactions between HOCs and eutrophication is the primary producers, phytoplankton, that form the base for many food webs in aquatic environments. Under favorable conditions—high nutrient availability and a suitable water temperature—the growth rate of phytoplankton is high, resulting in a high biomass. The high rate of cell division may possibly exceed the rate of uptake of hydrophobic pollutants (9,10). The uptake kinetics of HOC in phytoplankton has been proposed to follow a two step process: a fast adsorption from the water to the cell walls, followed by a slower transfer to the interior of the cell. The second step substantially increases the concentration in the plankton cell (6). Under these conditions, the HOC concentration does not reach equilibrium in phytoplankton, and levels become lower in the fast growing biomass of eutrophic compared to oligotrophic systems.

Transport of pollutants to the consumers results in a lower exposure of HOC via the food, thereby explaining the difference in biota concentrations between the systems. Phytoplankton species composition differs considerably between nutrient-rich and nutrient-poor environments. Cyanobacteria dominating the eutrophic systems contain lower amounts of lipids and have another type of cell wall than eucariotic phytoplankton, such as diatoms and green algae. HOC accumulation is related to the amount of lipids and a higher fat content gives a higher amount of HOC in biota. Thus, the species composition may be crucial for further transport in the food web (11, 12). A dominance of diatoms in an aquatic system may result in a higher food-web transport of HOC, resulting in higher concentrations in consumers (13).

The initial adsorption in the uptake of HOC in phytoplankton can be affected by the chemical composition of the cell wall. The muramine acid of cyanobacteria causes a lower uptake of HOC than the phospholipid cell wall of eucaryotic phytoplankton, and an analog difference in food-web transport as mentioned above. The lipid quality is another possible factor that influences HOC uptake in phytoplankton, e.g. diatoms are known to contain high amounts of triglycerides, a lipid group that has high affinity for HOC (14).

High biomass of phytoplankton in nutrient-rich environments also has other effects on the fate of HOCs. The increased sedimentation of organic matter scavenges HOC from the water mass, and if the fraction of nonmetabolized organic matter increases, the load of organics and HOC to the sediment and to sediment living organisms may increase (15, 16). Several possible scenarios of HOC input to the benthic fauna are then possible. First of all, the increased load of HOCs may result in increased concentration of HOCs in benthic biota. A study from running waters has indicated that HOC concentration increased in trout by increasing the trophic level in the water (the amount of nutrients) (17). Similar results have been found in studies using US mussel-watch data (18). However, as has been pointed out by the authors, both these studies might have been influenced by local discharges of HOCs in parallel with the nutrients. Consequently, these observations cannot conclusively be interpreted as a result of increased scavenging resulting in increased load of HOC in benthic biota and their predators. Another possibility is that enrichment of organic matter in the sediment counteracts the bioavailability of HOCs, as the compounds are more firmly attached to the organic particles, compared to minerogenic matter (e.g. 19). Experimental studies by Gunnarsson et al. (2), however, indicated that the addition of labile organic matter increased the bioaccumulation of organic contaminants (PAHs) in benthic organisms.

Should eutrophication lead to anoxia in the bottom water, transport of HOC out of the sediment may occur, having an overall impact on HOC cycling in the system (12). It has been shown that there are several processes affecting this transport such as (*i*) organic matter content of the sediment; (*ii*) bioturbation; and (*iii*) development of reduced gases (19–21). These processes will all directly or indirectly affect the transport of HOC across the sediment/water interface, and thus remobilize the HOC during hypoxia. However, quantification of such processes in the field is still to be carried out. The impact of anoxia on trace metal exchange across the sediment-water interface and on bioaccumulation of these metals has been illustrated experimentally (1).

Sedimentation and transport of HOC across the sediment/water interface seem to be important for the cycling of pollutants in aquatic environments (16). Several studies (13, 16, 22) indicate that recycling processes of HOCs within lakes have a major impact on concentrations in biota and for the overall budget. The mechanistic process involved includes mineralization of organic matter just above or at the sediment surface and a subsequent release of HOCs, when the organic carbon content or lipids of the biogenic matter reaches low levels, probably as a result of microbial attack. This has been shown by studying sedimenting matter, where HOC concentration increases during particle settling through the water column, as a result of organic carbon or lipid loss (mineralization) during settling (13, 22). Sedimentation rates of HOC in aquatic systems are considerably higher than wet deposition of the compounds to lakes, further indicating the importance of internal processes (10).

Phytoplankton biomass may also have an effect on input of HOC to the aquatic system. The transport of HOC, apart from catchment area input, across the surface microlayer is driven by atmospheric deposition (dry and wet deposition, gas-phase exchange), and counteracted by water to air volatilization (23). Recently, Dachs et al. (24) proposed that a large biomass of phytoplankton may "purge" gaseous HOCs from the atmosphere to a lake. As HOCs in the dissolved state are accumulated by phytoplankton and later sedimented in association with the crash of the plankton bloom, the HOC levels in the water are reduced. The reduction will lead to a change in the air to water equilibrium, and a consequent flow of HOC from the atmosphere to the lake. As a result, gaseous transport of HOC is higher to eutrophic than to oligotrophic systems. It has further been suggested that the higher amount of organic particles, e.g. phytoplankton, in nutrient-rich lakes reduces volatilization, as a higher proportion of HOC is particle-bound and only HOCs in the dissolved state are subjected to volatilization (24).

In a trophic gradient of lakes from oligotrophic to moderately eutrophic, HOC concentrations in zooplankton and predatory fish decrease (25, 26). In addition to the explanations given above, physiological and/or ecological variables may govern this relationship. One factor is suggested to be growth rate of, e.g. fish in the systems. Higher individual growth rates and food conversions, possibly in connection with reduced pollutant assimilation, may result in lower concentrations of HOCs (26, 27). However, that a major proportion of fish species has an increased growth rate in a eutrophic system compared to that in nutrient-poor systems is not always the case. Some species certainly have, but others may suffer from increased intra- and interspecific competition, reducing individual growth rate. An altered population structure, with an increasing base of younger individuals will affect pollutant concentrations, resulting in overall lower concentrations. However, this may also be argued.

Some results, but not all, indicate that fish age is a significant variable for pollutant concentrations as older individuals have higher concentrations of pollutants. This, in turn, may be an effect of longer exposure time but could also be due to the fact that older fish have higher amounts of lipids (lipid depots, e.g. salmonids, *Stizostedion* sp., anguilid eels, herring). Consequently, an altered population structure of fish under eutrophication (younger individuals) would tentatively influence pollutant concentrations. If biomagnification explains the concentrations found in fish, an altered population structure of the prey (food consists of younger age classes of prey) will result in lower pollutant concentration in the predator. A strong year class of, e.g. alewife will remain for a long time in Lake Ontario, North America, and the HOC concentration increases as the exposure time becomes longer and, consequently, lake trout concentration of HOCs is significantly affected (28).

Several authors have suggested that food-chain length has a significant impact on pollutant levels, especially for the last consumer link (29). A longer food chain results in higher concentrations, as shown in Canadian Shield lakes, where piscivorous fish have higher concentrations of HOCs, when the opossum shrimp (*Mysis relicta*) was present at an additional trophic level, compared to lakes where it was absent (30). Food chains in eutrophic lakes are thought to be shorter and simpler than food-chains in oligotrophic environments (7). In conclusion, food-web interactions may well be a significant factor for HOC concentration in fish, but the relationships are by no means fully understood today.

CONTAMINANTS AND EUTROPHICATION: PROCESSES AND EFFECTS

The EUCON research program aimed at increasing our understanding of the most important processes taking place in the pelagic and benthic compartments with respect to interaction between eutrophication and contaminants. Emphasis has been on governing factors concerning mobilization of contaminants and transfer from one compartment to another. A focal point has been to what extent a change in trophic status influences the scavenging of contaminants in the water column and transfer of contaminants to the sediments. Additionally, a major task has been to assess the role played by the amount and the quality of organic matter in the bottom sediments, with respect to mobilization or fixation of contaminants in the sediments. These processes are of importance when predicting the environmental changes which occur when contaminants and organic matter interact.

The EUCON research program also aimed at filling gaps in knowledge related to biological effects as a result of interactions of eutrophication and contaminants. One of the main issues is to predict the changes in levels of contaminants in pelagic organisms, including fish, assuming that the input of contaminants to the aquatic environment is constant, but the level of eutrophication changes. This is an issue of concern for environmental policy makers and managers. Furthermore, an important question to answer is how the quantity and quality of organic matter, both in the pelagic and the benthic compartments, influence the bioavailability and bioaccumulation of contaminants.

PELAGIC ECOSYSTEMS

When contaminants such as HOCs and metals, are transported to the Baltic, whether it is by runoff from the watersheds via tributary rivers, primary input of effluents from point sources, discharges from ships or input from atmospheric fallout, the pelagic compartment of the Baltic environment is exposed. The environmental fate of the contaminants depends on a number of processes in the pelagic zone. These factors include the partitioning between water and particles as well as the lipophilic compartments within the water that could be dissolved, accumulated in organisms or bound to organic or inorganic particles. Another important factor is the time period under which these forces can act and establish equilibrium partitioning. The time available depends on the length of the various biological cycles (e.g. growth and degradation) taking part in the partitioning processes (e.g. adsorption, absorption, and desorbtion) as well as the rate of sedimentation.

A number of studies have demonstrated the importance of partitioning and scavenging HOCs from the pelagic to the demersal zone and the sediments (9–12, 15, 31–33). Some of the studies within the EUCON program have focused on the particle-bound vertical transport of HOCs and metals from the photic zone to the sediments (22, 34–36).

Field studies and experimental studies, have provided information on the role of pelagic organisms, particles and lipids for food-web transfer via processes like bioconcentration, bioaccumulation and biomagnification for metals and HOCs (27, 29, 37–39).

Interestingly, results have indicated that instead of biomagnification, the water/lipids partitioning determines the concentration of lipophilic compounds in the predator of the aquatic food web (33, 40–43). One of the studies that supports the importance of biomagnification in the aquatic food web was carried out by Kidd et al. (29) in Lake Laberge. They demonstrated biomagnification of toxaphene in the aquatic food web (plankton—insect larvae—muscle tissue of fish—fat tissues of the top carnivores, i.e. muscle of lake trout (*Salvelinus namaycush*) and liver of burbot (*Lota lota*) (fresh weight basis). The fat content of burbot liver exceeds 40%. Data are also available on another top carnivore, the lean pike (*Esox lucius*) from the same lake and collected the same year (44), but not presented in the paper by Kidd et al. 1998 (29). The concentrations found in pike from Lake Laberge were low and contradict the suggestion of biomagnification as an important factor in explaining concentrations in a predator. However, on a lipid weight basis the concentrations were very similar irrespective of species or position in the food web. This finding stresses the role of the lipids in explaining the concentrations found in fish. Other results (43) support the hypotheses of lipid control of chlorinated compounds in fish at various levels in the food chain. Data on Baltic salmon, feeding on herring and sprat, show almost identical concentrations of PCB on a lipid weight basis as the long-term monitoring data on herring obtained from the Swedish Environmental Monitoring Programme (45, 46).

These contradictory results, biomagnification or lipid control, are good examples of the uncertainty that we often have to deal with when we try to interpret ecological processes.

The importance of the pelagic part of the Baltic can easily be recognized when we realize that the most important international fishing occurs in the open sea, directed either to pelagic herring or salmon or demersal cod. Effluents containing nutrients as well as contaminants have combined effects on the commercial Baltic fishery. The increased nutrient load causing anoxic conditions in the spawning regions of the Baltic together with over-fishing have caused decreasing cod stocks and periodic bans on cod fishing. Simultaneously, the lipid-rich cod liver is banned on the Swedish market because of elevated concentrations of lipophilic pollutants like PCB and dioxins (47). The components of the pelagic food chain of the Baltic are lipid rich, including the zooplankton feeding herring and the piscivorous salmon. Because of high concentrations of lipophilic contaminants dioxins and PCBs, salmon and herring are among fish species that according to the Swedish food administration should be avoided in the diet of pregnant women, and not regularly eaten by women of reproductive age.

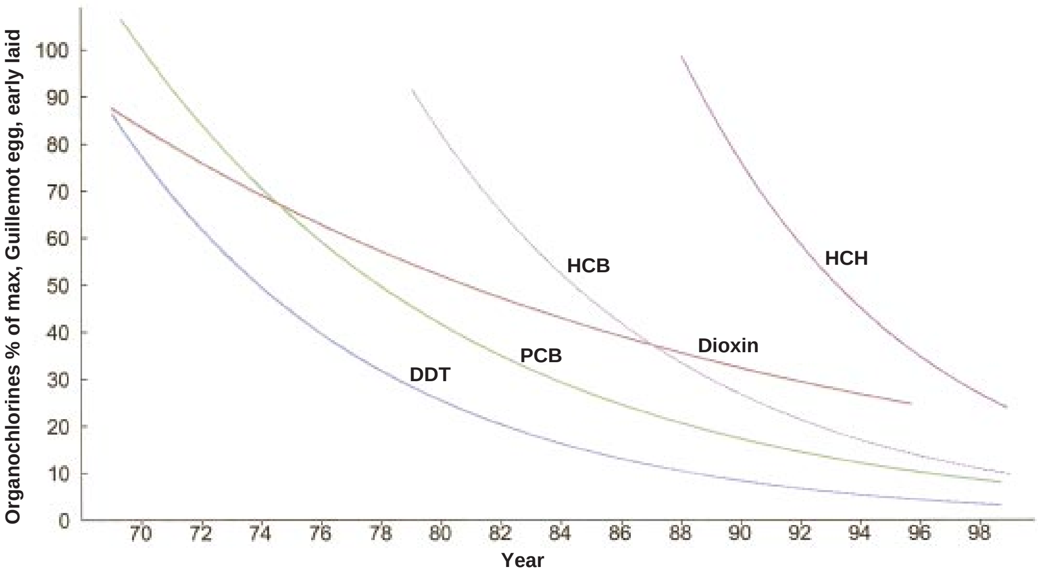

Figure 3. Changes in concentrations of various contaminants over time in Baltic guillemot egg. The lines show the log-linear regression lines for annual geometric means of measured concentrations of DDT (blue), dioxins (red), PCB, (green), HCB (lilac) and HCHs (black). All lines have been normalized to 100 for the highest concentration of the various contaminants to indicate the timing and relative change over time.

Temporal Changes.

The long-term monitoring of concentrations of HOCs in Baltic biota has revealed a decrease in concentrations during the last 30 years. This is illustrated in Figure 3. It has been suggested that at least part of this decrease can be explained by biodilution during a period of Baltic history when the nutrient load increased (40). Furthermore, some available results indicate that a higher trophic status of the water implies lower concentrations of bioaccumulative compounds (8, 13, 25). However, recent studies both in Baltic biota and Swedish freshwater biota, also representing waters where no eutrophication has occurred, reveal a remarkable consistence with respect to the declining rate of both DDT and PCB concentrations over time (46). This suggests either that the gradual increase in eutrophication of the Baltic during the period 1968–1998 has had no major effect on the concentrations or that other processes explaining the decrease, such as international and national measures to halt pollution, have been even more influential. When concentrations of nutrients appear to have increased by a factor of 2.5–4 during the last 25 years (48) the simultaneous decrease in contaminant concentrations in Swedish biota including Baltic biota have decreased by a factor of 10 or even more (46).

Based on long-term trend monitoring it has been suggested that the major reason for the similarities in the decreasing rate is degradation in the atmosphere (49, 50). Irrespective of environments studied (terrestrial, freshwater or marine, temperate or subarctic, eutrophic or oligotrophic), matrices used (fish, birds or mammals) and even chemical compounds (HCH and DDT) studied, concentrations decrease in a very similar way (49–51). Thus, the major explanation for the decrease in concentrations during the last 30 years is not scavenging in the water, biological degradation in the environment, global chromatography moving the compounds from the warmer areas of the world to colder. A possible explanation could be a comparatively fast degradation in a media common to all environments; the atmosphere. In the atmosphere, degradation by OH-radicals and UV radiation can, possibly, degrade HOCs.

It seems important to distinguish between concentrations found in biota at a certain time and the relative rate at which concentrations decrease over time. The concentration of contaminants found in the pelagic zone, in a certain environment at a certain time is determined by current discharges, biodilution, dilution by growth, scavenging in the water column, etc. Any new release of persistent, bioaccumulative persistent compounds to the Baltic might have a more serious effect on the ecosystem if the amount of nutrients decreases in future. However, the change in concentrations over time also depends on how the amount of discharge changes over time as well as the processes determining the long-term environmental sink of the pollutants (not only the aquatic environment) and the rate at which the compounds are degraded.

BENTHIC ECOSYSTEMS

Eutrophication-related Benthic Changes in the Baltic Sea

Since the mid-20th century, great changes have occurred in the benthic system of the Baltic proper as a consequence of spreading oxygen deficiency. Laminated sediments, indicative of no benthic faunal activity, date back to at least the 1930s in some areas, but spread rapidly in the central Baltic during the 1960s and 1970s (52). From the 1960s up to the present, 50–100 000 km^2 of bottoms below the pycnocline at about 80 m have, for most of the time, been devoid of bottom fauna (53, 54). Above the pycnocline, however, an increased food supply due to eutrophication may have caused a general increase in biomass of the benthic fauna (55). In the archipelagoes (e.g. St. Anna and Stockholm archipelagoes (56, 57), bottoms as shallow as 20 m have laminated sediments and little or no fauna. In some areas,

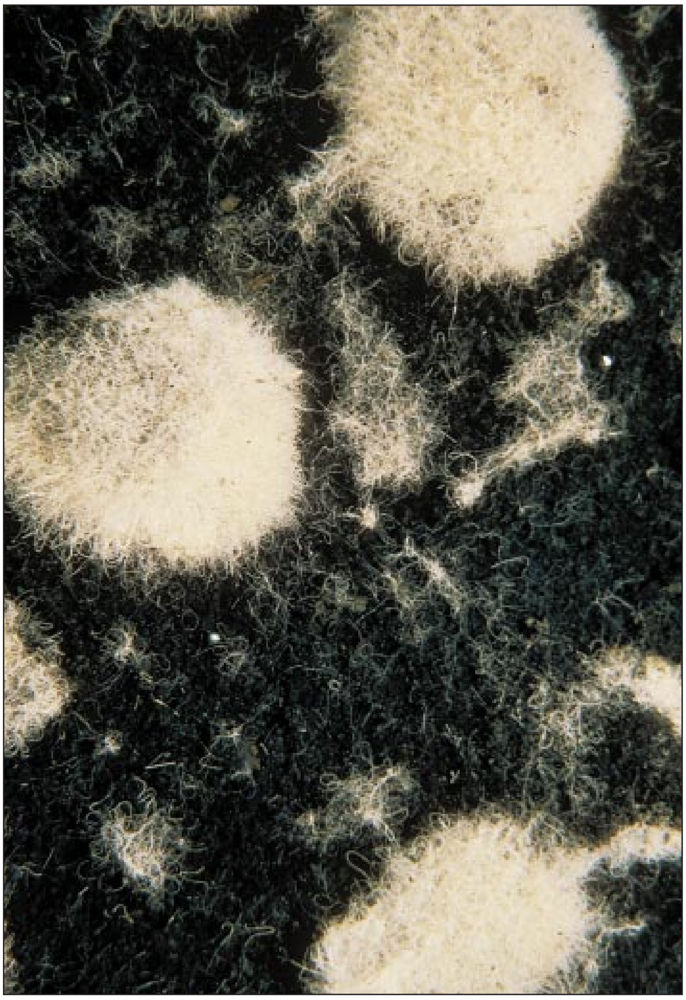

Figure 4. Mats of *Beggiatoa* from an oxygen deficient sea bottom. Photo: R. Rosenberg.

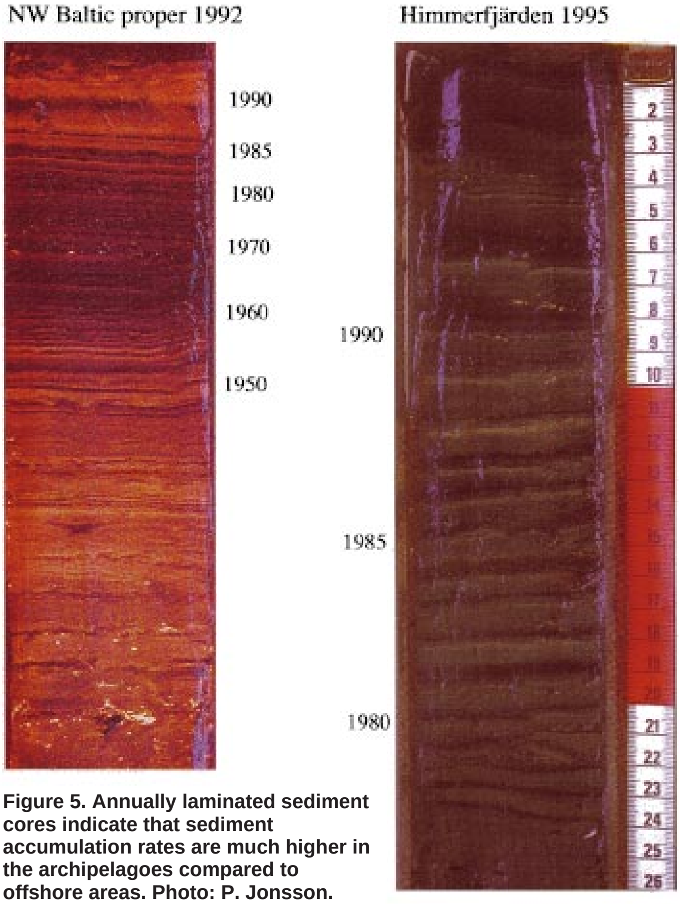

Figure 5. Annually laminated sediment cores indicate that sediment accumulation rates are much higher in the archipelagoes compared to offshore areas. Photo: P. Jonsson.

sulfur bacteria, *Beggiatoa* spp., occur at the sediment surface indicating the presence of H_2S at the sediment-water interface (Fig. 4). It has been estimated that about one-third of the bottoms of the Baltic proper were laminated and devoid of life in the late 1980s.

Sediment Carbon Content as a Tracer of Eutrophication?

It is most likely that the increased eutrophication of the Baltic has resulted in an increased sedimentation of organic material (58). One hypothesis that constituted the basis for extensive studies of archipelago sediments was that the carbon content in surficial sediments reflects the eutrophication situation in the area. However, in the archipelago areas, only very small differences in carbon content in surficial sediments have been detected for the different bays (59) and these differences were not correlated with eutrophication status. This is most likely due to morphometric differences in the bays causing different levels in the accumulation sediments, e.g. for recently primary produced organic matter, and resuspended/eroded old glacial and post-glacial clays.

As the organic content in surficial accumulation sediments could not be used as a tracer of eutrophication, other techniques have been tested to distinguish the eutrophication gradient. One method that appeared useful was the application of the isotopic composition of $^{15}N/^{14}N$ and $^{13}C/^{12}C$ (59) to elucidate the eutrophic gradient in the Stockholm archipelago. Elevated PCB levels correlated well with the changed isotopic composition within 50 km from Stockholm. Although it was shown that the Stockholm watershed is still a major source of PCB and nutrients to the Baltic Sea, the results do not support any enrichment or dilution of $\sum$PCB-7/C ratios caused by eutrophication.

Resuspension/Erosion a Dominant Process in the Baltic Sea

In the Baltic Sea, the allochtonous particle input via rivers (60) is of minor importance compared to the total sediment accumulation in the deep depositional areas (52), suggesting that erosion/resuspension processes are of major importance with respect to sediment accumulation. Furthermore, compared to the total sediment accumulation in deep areas, organic matter derived from recent primary production constitutes on average only some 5% of the sediment accumulation in the Baltic proper (58).

Recent investigations of laminated sediments in offshore areas (61) and archipelagoes (62) have shown that the gross deposition rate to a high degree is governed by the frequency of high wind speeds. This implies that the particles that finally settle in areas with continuous deposition of fine material are characterized by a highly variable age of carbon, ranging from days to thousands of years.

Particle-associated PCBs may be retained on some transportation bottoms until strong energy input from waves, currents or submarine slides resuspend the sediments years or decades after the first deposition (63). This delay mechanism is important to consider, not only in retrospective sediment studies, but also when to expect substantial improvements in the ecosystem after remedial measures. During this time lag, the organic matter containing PCBs has been exposed to oxic as well as anoxic conditions, leading to a uniform PCB congener pattern when it finally settles in the deep depositional areas (63).

Recent studies (56, 62) have shown that the sediment accumulation rate is on average 6–8 times higher in the Stockholm archipelago compared to offshore areas (Fig. 5). These results are supported by cesium-134 measurements (59). Jonsson et al. (56) demonstrated that the carbon accumulation rate is governed by the gross sediment accumulation rate. This is most evident in archipelago areas with high gross accumulation rates, whereas in offshore areas, with 6–8 times lower gross sediment accumulation rates, autochtonous carbon constitutes a relatively larger part of the total sediment accumulation. Therefore, temporal changes in sedimentation of autochtonous carbon are more easily detected in offshore compared to archipelago areas.

Comparison between different data sets showed that surficial sediment concentrations of PCBs are remarkably similar in archipelagoes and offshore areas in the NW Baltic proper. Jonsson et al. (56) suggested this to be due to the fast water exchange (weeks-months) between different parts of the Baltic and between offshore and archipelago areas.

The uniform PCB concentrations in different areas in combination with a much higher gross sediment accumulation rate in the archipelagoes implies a manifold higher sediment burial of PCBs in the archipelago compared to offshore areas. Unexpectedly, it also indicates an import of contaminants from the open sea to the archipelago. The obtained linear relationship between gross sediment accumulation rate and sediment burial of PCBs (56) is further supported by a mass-balance calculation for the Baltic Sea indicating an average residence time for PCBs of less than one year (63).

The Importance of Carbon Quality

Hydrophobic contaminants bind to organic particles, and the amount of particles in the water will increase in eutrophic areas. Thus, organic contaminants will be scavenged and accumulate on the bottom. The benthic fauna ingest organic particles and accumulate parts of the associated contaminants. Gunnarsson et al. (64) have demonstrated that not only the quantity of organic matter, but also the quality plays a significant role for the bioaccumulation of lipophilic contaminants. Consequently, in eutrophic areas, such as the Baltic Sea and the Kattegat, with an enhanced phytoplankton biomass and high abundance of "fresh food", benthic organisms such as blue mussels (*Mytilus edulis*) and brittle stars (*Amphiura filiformis*) appear to increase their burden of contaminants during the productive season in comparison with oligotrophic areas.

In contrast to the equilibrium partition theory (65) uptake of organic contaminants was not found to be inversely related to the organic carbon content of the food particles, but rather proportional to the quality. Most deposit feeding animals are selective feeders, and their search for nutritious food is coherent with the fact that associated contaminants will also be ingested.

With respect to sediment burial of HOCs in Baltic sediments it is indicated that processes related to sediment dynamics are of major importance for the burial rate of HOCs in the Baltic Sea and that modelling of sediment burial cannot be done by simply calculating the distribution from the equilibrium partitioning theory (65). In the Baltic, where the age of the recently accumulated carbon in the deep areas varies from days to thousands of years, the carbon quality is most likely of major importance regarding the behavior of HOCs in the ecosystem.

Bioturbation Scenario

Due to the large areas of erosion and transportation bottoms (2/3 in the Baltic proper according to Jonsson et al. (52), a PCB carrying particle, finally buried in the laminated depositional areas, normally has passed a number of resuspension events before being trapped in the anoxic sediments perhaps years to decades after the first deposition. During this transport it has been subject to oxic as well as anoxic degradation, which leads to very slow degradation of the PCBs once the particle has been deposited in the laminated sediments. The registered small changes in congener pattern with increasing sediment depth are assumed to be an indication of this (63).

Concentration of contaminants can be up to 10 times greater in burrow linings in the sediment compared to that in adjacent sediment, enhanced in faeces, and seems also to adsorb to *Beggiatoa* spp. when present on the sediment surface (64). Bioturbation has been shown to significantly increase the release of organic contaminants to the water column, especially when the sediment is enriched in contaminants. Evidently, in the top sediment, where the benthic fauna is active, the organic contaminants are redistributed, and a significant proportion may be cycled back to the pelagic system. Part of this can occur through benthic predation by diving ducks and demersal fishes. A significant part of the contaminants is metabolized by the benthic invertebrates. In this way, some of the toxic substances either change chemical structures or become detoxified.

Most benthic species in the Baltic rework the sediment down to a few centimeters. Eckhéll et al. (61) found recolonizing fauna to bioturbate laminated sediment down to about 3 cm, which most probably will increase the flux of contaminants out of the top sediment.

One of the dominant species occupying most sublittoral and oxic bottoms in the Baltic, the amphipod *Monoporeia affinis*, burrows down to a depth of about 5 cm in the sediment (66). The polychaete *Marenzelleria viridis* has since the mid-1980s invaded the Baltic and has now spread from the south up to the Bothnian Sea. This species occurs in numbers up to several hundred per m^2 in many of the invaded areas (67). *M. viridis* is a deep-burrowing species, down to 40 cm, even in sulfide-rich sediments with concentrations up to 3 mmol L^{-1} of sulfide (68). This polychaete may have a significant impact on the flux of contaminants as it penetrates deeper down than any other benthic species in the Baltic. The main depth distribution of *M. viridis* in the Baltic seems so far to be restricted to above the pycnocline, but it cannot be excluded that it may colonize deeper bottoms when the next bottom water renewal occurs. If so, presently buried organic contaminants can also be remobilized at greater depths.

It has been demonstrated that the highest concentrations of PCBs in the Baltic Sea are normally found in the upper 10 cm of the sediment column (63). However, in the NW Baltic proper clear concentration maxima are found a few centimeters below the sediment surface (56, 69–71).

Normally, the sediment accumulation rate in Baltic Sea surficial sediments is in the range 1–4.3 mm yr^{-1} (61). The annual sediment burial of ∑PCBs in the anoxic sediments of the Baltic proper has been estimated to 735 kg yr^{-1}. Assuming, firstly, that the upper 5 cm of the sediment column may be bioturbated in case of improved oxygen conditions in the bottom water, secondly an average sediment accumulation rate in this layer of 2 mm yr^{-1} an amount of 23.4 tonnes of PCB could, theoretically, be remobilized.

However, taking into account recent findings that a turnover from bioturbated to laminated sediments enhances sediment concentrations of PCBs in the order of 50% (P. Jonsson, Swedish Environmental Protection Agency, Stockholm, Sweden, pers. comm.), a possible total release of PCBs may be approximated to 1/3 of 23.4 tonnes, i.e. ca 8 tonnes. This is a significant amount of PCB that most likely will enhance concentrations in benthic and pelagic biota.

Since the 1970s all investigated HOCs except polybrominated compounds have shown decreasing concentrations over time in biota samples from the Baltic (46,72–74). The concentrations of dioxins, PCBs and DDT compounds as well as HCHs and HCB, have all decreased at a rate of more than 5% annually since the 1970s. For HCHs, the decrease was very fast during the 1990s and the annual rate of decrease often exceeds 20%. For the polybrominated compounds concentrations increased during the 1970s, but seem to decrease again during the recent time period (75).

Since the middle of the 1980s sediment cores have been used in studies of the temporal trend of contaminants in the Baltic environment (76). A number of the studies published indicate stable or even increasing concentrations of PCB over time in the sediment cores during the last decades (63, 71, 77–79). On the contrary, other studies based on sediment cores predominantly from the northwestern part of the Baltic proper show decreasing concentrations over time since the 1970s (63, 69, 70).

Bignert et al. (80) compared the temporal trends for DDT compounds and PCBs in biota samples and lamina of sediment cores. As a representative of biota matrices Guillemot eggs (*Uria aalge*) were used. This matrix has been used in temporal trend monitoring, and analyzed annually since the late-1960s (46, 80). Two cores from the northwestern part of the Baltic proper were used, one covering the time period 1940–1998 and the other 1968–1998. The original data from these cores are presented in Jonsson et al. (56).

The data from the two cores are unique since they provide a possibility to study the between-year variations on the basis of analysis of the annual lamina. The results show the same fairly large between-year variation that has been found in studies of biota matrices (80). This variation implies that interpretations of temporal trends based on scattered lamina in a sediment core might be very uncertain (81).

In the sediment core covering the period 1940-1998 a concentration peak (dry weight basis) was found at the end of the 1960s and in the early 1970s, corresponding to the time of the maximum use of the PCB and DDT compounds. In both cores, a decrease was found after this period although the rates of the decrease were substantially lower than in the biota samples. Interestingly, concentrations of both DDT and PCB were remarkably high already during the 1940–1960s and the PCB concentrations of the 1940s corresponded to concentrations found at the end of the 1970s on a dry weight basis, which appears very unlikely. An interpretation based on carbon normalized data for PCB concentrations indicates a concentration peak prior to the peak of the industrial production of DDT and PCB. Jonsson (77) pointed out that the circulating carbon found in the Baltic, to a large extent, comprises post glacial carbon and only a limited part has recent biological origin. This shows that carbon normalization does not improve our possibilities to interpret the temporal trend.

Fairly high proportions of nonmetabolized DDT were found in deeper lamina representing the 1940s, whereas the relative amount of nonmetabolized DDT increased over time after the 1960s. This is contrary to what can be seen in biota samples where the relative amount of nonmetabolized DDT decreases over time since the 1960s (46). In fact, the composition of metabolised DDT and nonmetabolized DDT was very similar in the most recent sediment lamina and the fish samples of corresponding years. The findings indicate an ongoing degradation of the fairly persistent DDT in the sediments since the 1960s. The high concentrations of PCB in the deeper sediment lamina representing the 1940s could partly be due to an uncertain dating of the deeper layers. However, the presence of nonmetabolized DDT in these layers indicates that the pesticide has been transported vertically in the core by diffusion processes, down to layers of the sediment core where the bacteria composition is unable to degrade the pesticide. Diffusion processes may also explain the vertical distribution of PCB in the core.

The discrepancy between the time trend data of sediments and biota may be explained by diffusion processes in the sediments, driven by partitioning equilibrium of nonpolar compounds as well as pore-water advection in the sediments. These explanations are so far tentative and we shall keep in mind that a number of earlier studies have failed to show any decrease in the concentrations of the contaminants discussed here. Apparently, we are lacking a simple verified model from which we retrospectively can interpret the temporal trend of HOCs in sediments. Important pieces of information are missing to explain the retention mechanisms of HOCs in the sediment, although we have a lot of information that has improved our capability to critically examine our possibilities. We have also gained information on the relative importance of the climate for resuspension of old sediments that might partly explain why concentrations decrease more slowly in the sediments than in biota (61).

IMPLEMENTATION AND APPLICATION OF THE RESULTS

Applied environmental research programs are initiated to enlighten specific environmental problems, to provide knowledge on relevant issues to environmental authorities and managers in industry. Aquatic systems are very complex and to implement new knowledge on the relevant issues it is necessary to stratify the research and to simplify the questions. A simplification is burdened by risk of false information and/or misinterpretations. However, the alternative is to base environmental decision-making on political views rather than on scientific information. Consequently, the researchers in applied environmental research have to clarify and present the results in a way that can be used for management decisions. At any time the best advice given should be based on the best available knowledge and by using the precautionary principle.

The EUCON program focussed on the interaction between contaminants and eutrophication. It is clear from the results obtained within the EUCON research program that interactive processes between contaminants and eutrophication exist in the aquatic environment. Such a process is the association between contaminants and organic matter. This implies that it is necessary to evaluate the fate of contaminants if the input of nutrients is changed and this results in a concomitant change in primary production. It is necessary to apply a holistic view and to consider the two environmental problems in parallel; the contaminants and eutrophication.

As for most studies of biological processes knowledge of the history is often very useful. Knowledge about the effects of environmental pollution as well as an understanding of the dimension of this pollution problem often needs a historical or retrospective view. Long time-trend series of contaminant concentrations in fish coupled to data of input of contaminants and nutrients may give indications of possible interactions between accumulation of contaminants in fish and the eutrophic status of the waterbody. Based on this knowledge, predictions may indicate the effects on contaminant concentrations in biota when the input of nutrients and organic matter is reduced due to remedial actions. The results from time-trend studies on biota clearly show that concentrations of most investigated HOCs in Baltic biota (DDT, PCB, HCHs, HCB, and dioxins) have decreased during the last 30 years (46, 74). Concentrations have decreased often to less than 10% of the levels during the 1970s. During the same time period the levels of nutrients in the Baltic Sea have increased by a factor of 2.5–4. Consequently, the trend may also to some extent be explained by increased biological production and a larger biomass (biodilution). However, the decreases are well-correlated to the international measures that have been taken to protect the environment (46).

We also know that the implication of switching from a eutrophic status to an oligotrophic status reduces scavenging efficiency and decreases the rate of sedimentation of organic matter and associated contaminants, and we would expect also a lowered biodilution. This implies that decisions regarding the input of nutrients, organic matter and contaminants to the aquatic environment should be considered carefully. The ideal situation appears to be an intermediate trophic situation were the input of nutrients is sufficient to maintain a healthy plankton population and a high biodiversity in the pelagic and benthic ecosystem as well as a sustainable fish stock. A certain degree of nutrient enrichment and high biological production will transfer contaminants from the water to the sediments and/or dilute the contaminants. It is generally accepted that the bioavailability of sediment-bound contaminants is less than the bioavailability of contaminants present in the water phase. However, it is a delicate balance. If the degree of eutrophication or nutrient enrichment exceeds a certain threshold, negative effects like uncontrolled plankton blooms and oxygen reductions in the deep water, anoxic conditions near the sediment surface may occur. The approach with respect to the input of contaminants is simpler. The ultimate goal is to reduce the input of harmful substances to zero. However, based on the available information and considering that all measures in the short-term perspective are expensive, we as scientists must stress that if measures are taken to reduce the output of nutrients to the Baltic it is imperative to reduce simultaneously the discharges of persistent, bioacumulative contaminants to the environment. If available resources are only used for reduction of nutrients there is an obvious risk that contaminant concentrations will increase in the biota of the Baltic, a risk for both mankind and wildlife.

The parallel time-trend studies on biota samples and sediment cores suggest that retrospective studies based on sediment cores cannot, at present, be interpreted accurately. Most probably, the information stored in the sediment cores represents an important value for understanding the mechanisms of retention, although at present we are unable to obtain this information. This calls for further research to improve our understanding of the important sediment retention and burial process.

At present the bottom sediments represent a large pool of contaminants. Even a small contribution of contaminants from this pool to the ecosystem may play a significant role. Large bottom areas of the Baltic Sea have laminated sediments, devoid of life due to lack of oxygen. If the oxygen conditions at the sediment-water interface are improved in the future the sediments will be colonized by benthic organisms. Due to the burrowing activity of the benthic animals, an increase in physical mixing of the contaminated sediment will take place, also involving subsurface sediments with higher levels of contaminants. This will inevitably increase the likelihood of elevated levels of contaminants in benthic organisms. Another mechanism, which is independent of man-made activity, is water renewal in the Baltic. In enclosed seas like the Baltic the deep-water exchange is irregular, and for long periods the bottom water in the deep basins is stagnant and the bottom sediments anoxic. A sudden deep-water exchange will create a new situation at the sediment-water interface both with respect to physiochemical conditions as well as biological activity. A severely contaminated sediment will inevitably become a source of pollution. This knowledge calls for caution and observation in the future, and demands monitoring and in-depth studies to evaluate the potential risks if changes from anoxic to oxic conditions take place. The exchange of trace metals at the sediment-water interface during variable redox conditions has been studied experimentally in other programs addressing the interactions between eutrophication and contaminants (1).

The EUCON program has focused on processes influencing transport, fate and effects of contaminants under different regimes of eutrophication in freshwater and marine environments. The substantial increase in the understanding of the ecological problems related to eutrophication and contaminants will be beneficial in future advice to environmental managers, despite gaps in our knowledge. These gaps imply two things. *i*) The identification of gaps helps when we stratify future research activities and thereby focus on the most cost-effective studies. *ii*) The identified gaps will imply a more appropriate use of the precautionary principle.

GAPS IN KNOWLEDGE

A lot of questions, which were raised in the formulation of the EUCON research program, have been addressed successfully. Our knowledge has increased substantially on topics such as bio- and growth dilution, sedimentation and sediment sequestering, the importance of lipids, food quality, organism characteristics and food-web structure. Of course, as is common in scientific research, the results are not always straightforward and instead new questions have been raised on some of these topics. There are also areas which have not been addressed and which need further research before a more complete understanding of the interactions between eutrophication and pollutants is achieved.

As a consequence of altered eutrophic status the possible importance of shifts in planktonic composition has been documented, as mentioned above. Limited efforts have, however, been directed towards changes in the dissolved and colloidal fraction in water. This fraction, which is available for pollutant interaction, frequently comprises far more than 90% of the total content of organic matter in natural waters. There is still limited knowledge on whether and how eutrophication processes affect this pool and how this possibly can explain the effects found. One can speculate whether the dissolved and colloidal pool characteristics in eutrophic waters favor pollutant binding in comparison to this pool in oligotrophic waters.

A deeper knowledge is fundamental not only for interpreting pelagic pollutant availability, but also biotic and abiotic degradation. For metals the binding to organic matter is more complex, although the dissolved and colloidal fractions in water are fundamental. Since most of the research has deliberately focused on organic pollutants in the EUCON program, new knowledge gained on the interaction between metals and eutrophication has been limited and further investigations are needed.

A growing area of interest in environmental chemistry is the importance of various anthropogenic particulate matrixes and nonaqueous phase liquids, e.g. soot, surfactants, hydrocarbon fuel residues, etc. For example, recent studies (82), show that soot increases the particulate–water partition coefficients for POPs by several orders of magnitude. Temporal and spatial input, sources and transport routes of these anthropogenic matrices and eutrophic substances are most likely correlated. A future challenge would be to evaluate the combined significance of these anthropogenic emissions.

One of the most, and in many environments the single most, important input route of organic and inorganic pollutants to the aquatic systems is through the air-sea interface. Important rate-limiting processes governing this exchange are physical, i.e. precipitation, water surface wind speed, etc. However, the characteristics of the organic-rich surface layer and the organic particulate, colloidal and dissolved matrixes below the surface are probably not insignificant. Further knowledge of if and how these compartments are affected by increasing eutrophication and its possible significance for the interactions between eutrophication and pollutants is also of future interest.

Within the EUCON project it has been indicated that eutrophication-induced scavenging of PCBs and DDTs may be of minor importance in the Baltic Sea. Climate-induced resuspension/erosion and subsequent scavenging of particle-bound HOCs from the water mass seem to blur the relative importance of distribution due to fugacity, and more research is urged to enlighten this possibly very important indication. Furthermore, different time trends in concentrations and burial of HOCs are indicated from different parts of the Baltic Sea and need more attention in future research.

Also, in the EUCON program it was suggested that HOC concentrations may be enhanced in laminated sediments compared to bioturbated mainly due to lack of benthic fauna. Although enhanced concentrations are indicated in the order of 50%, this type of investigation deserves more attention in the future to allow predictions of whether a large-scale recolonization of benthic fauna in the Baltic proper may enhance concentrations in benthic and pelagic biota and thus cause undesired effects of the measures taken.

Another very important interface, where full understanding of the processes and the effects of increasing eutrophication are lacking, is the sediment-water interfase. Even if this research area has been dealt with in the EUCON program it needs further attention, particularly concerning quantification of fluxes. This holds true also for the indirect effects of increasing eutrophication on the biotic and abiotic processes in the sediments. Here, the indirect effects resulting in sediment anoxia and its possible potential to alter sediment-water exchange, degradation and burial of pollutants, are especially of concern. The role of the benthic animal activity for the flux of both contaminants and nutrients in and out of the sediment may be key factors for ecological effects in the ecosystem. These fluxes need to be quantified under different environmental conditions.

The restructuring of food webs and changes in organism abundance and diversity as well as physiological alterations on organism levels as a response to eutrophication are also intriguing fields of research that need further attention. The EUCON program has contributed to increasing our knowledge in these fields but it is clear that more research is needed. As mentioned in the section on the state-of-the-art above, a broad change in the ecosystem structure is a natural cause of eutrophication; although the variation between different systems with different regulating factors is far from fully understood. Therefore, it is not possible to evaluate how today's knowledge on a given eutrophic ecosystem generally can be extrapolated to others. This area is of high relevance not only for ecotoxicological risk assessment but also in human risk assessment. Under a constant pollutant load the POP concentration in top predators used as human food can vary considerably depending on, e.g. lipid status and age (size) of the organisms as well as length and structure of the foodwebs. These are all factors that can be significantly altered in a system under changing eutrophic conditions.

Finally, other large-scale anthropogenic activities have modified the initial conditions for many small and large waterbodies. Examples are the large-scale ditching of forests to drain-off precipitation, the regulation of rivers as a prerequisite for hydroelectrical power generation, the liming of lakes, and the development of industries using large volumes of organic-rich process water. All these man-made activities have run parallel to the increasing eutrophication, and a future challenge is to evaluate the combined effects on the transport and fate of pollutants.

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