

Organic halogens in unpolluted waters and large bodies of water receiving bleach plant effluents

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ABSTRACT *We reviewed and updated recently performed studies of organic halogens in unpolluted waters and two large bodies of water receiving bleach plant effluents—Lake Vättern in Sweden and the Baltic Sea. All water samples contained measurable amounts of adsorbable organic halogens (AOX); the highest concentrations (up to 200 µg Cl/L) were observed in humic lakes not exposed to any industrial discharges. Analysis of chlorophenols revealed that there is a long-distance transport (> 100 km) of chloroguaiacols from bleach plants to remote parts of receiving waters. However, there was no evidence of chlorinated organics from bleach plants accumulating over several years in the water phase. One chlorophenol, 2,4,6-trichlorophenol, and its methylated analogue, 2,4,6-trichloroanisole, were also detected in surface waters considered to be unpolluted. Mass balance calculations showed that different processes in terrestrial environments make large contributions of AOX; enzyme-mediated chlorination of humic substances is a plausible explanation to the widespread occurrence of organic halogens.*

KEYWORDS

Analysis
Bleach plants
Chlorine compounds
Effluents
Surface water
Testing
Toxicity

The history of toxic organic substances in the environment is closely related to the industrial production of organic halogens. Large quantities of chlorinated compounds are released into the environment through the use of halogenated organic solvents. Substances such as DDT (1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane), PCBs (polychlorinated biphenyls), and chlorinated dibenzo-*p*-dioxins are globally spread, and traces of bleach plant effluents have been found far from the point of discharge.

The natural production of organic halogens has been documented in several studies (1). However, environmental scientists have given such reports little attention. The mere presence of organic halogens in the environment has often been considered evidence of industrial pollution.

Group parameters, such as adsorbable organic halogens (AOX), have been suggested as indicators of general pollution (2).

Our purpose is to put the large-scale dispersion of chlorinated organics from bleach plants into perspective by comparing concentrations and chemical characteristics of organic halogens in bleach plant recipient waters and unpolluted surface waters. Special attention is given to concentrations of AOX and chlorophenols and to new evidence of considerable natural production of organic halogens in terrestrial environments (3, 4). We will also discuss the persistence of chlorinated organics from bleach plants.

Evidence of a large natural production of organic halogens was first obtained from studies of brominated algal metabolites in marine environ-

ments and halomethanes in the atmosphere (5, 6). Recently performed studies have emphasized the role of humic substances; enzyme-mediated chlorination of such substances may be a major natural halogenation mechanism (7-9). Long-range transport of bleach plant effluents has been studied by several researchers (10-12). Much of the empirical data cited here has been reported elsewhere (13).

Study areas

Two bleach plant recipient waters were studied in detail: Lake Vättern in Sweden and the Baltic Sea. Unpolluted surface waters were sampled in remote areas of southern Sweden, primarily in the counties of Jönköping and Halland.

Lake Vättern has a surface area of

1860 km² and a water volume of 74 km³. The average outflow from the lake is 1.25 km³/year (40 m³/s), which corresponds to an average residence time of approximately 60 years. In 1955, chlorbleaching was introduced at a pulp mill located close to the northern end of the lake. There are no other large point sources of organic halogens in the drainage area.

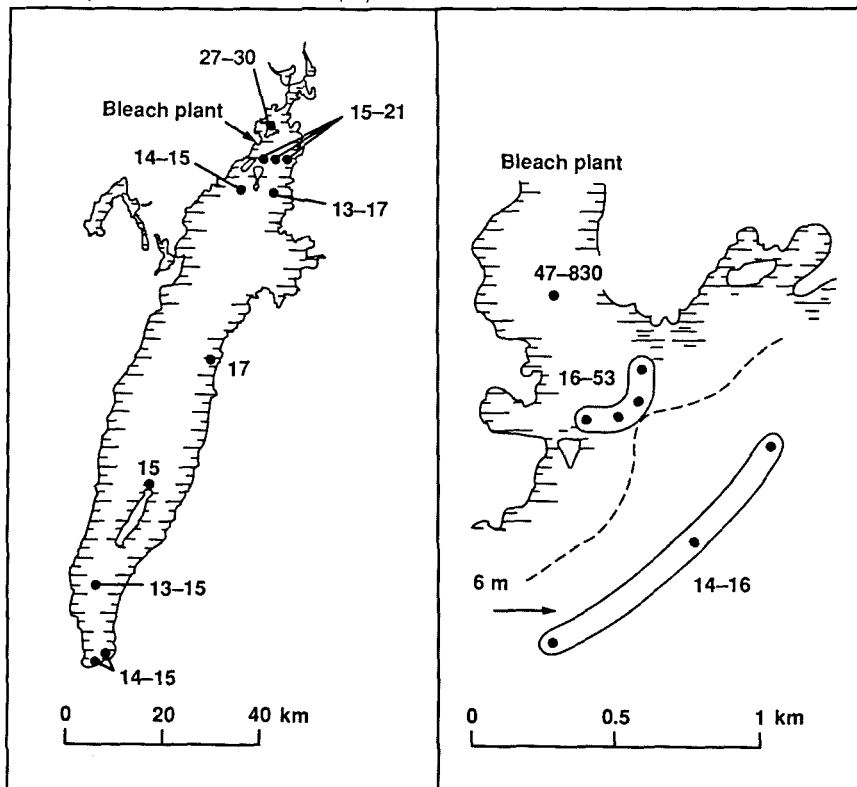
The Baltic Sea has a surface area of 375,000 km² and a water volume of 20,700 km³. The average outflow from the Baltic Sea through the Straits has been estimated to be 950 km³/year, which corresponds to an average residence time of approximately 20 years. The water residence time in two of the major sub-basins, the Bothnian Bay and the Bothnian Sea, is 3–5 years (14). Ten of Sweden's fifteen bleached kraft mills are located along the Baltic Sea coast, and another two are on inland waters connected to the Baltic. Most of the Finnish bleached kraft mills are located on inland waters connected to the Bay of Finland, which, in this study, was considered to be part of the Baltic Proper. The Finnish discharges of chlorinated organics to the Bothnian Sea are much smaller than the Swedish discharges, whereas the Bothnian Bay is more heavily loaded by Finnish bleach plants (15). Discharges of chlorinated organics from other Baltic Sea nations are smaller.

Analytical procedures

Analysis of AOX in water involved adsorption on activated carbon, followed by the removal of inorganic halides, combustion of the carbon, and then micro-coulometric titration of the formed halides (16). For samples of brackish water, inorganic halides were removed from the activated carbon by an extended washing procedure including two nitrate washes and ultrasonic treatment (17). Sediment samples were analyzed with respect to AOX in a similar way. Inorganic halides were removed by a nitrate wash, the sediment was burnt in the AOX analyzer, and the halides formed were determined by micro-coulometric titration.

Extractable organic halogens (EOX) were analyzed in concentrates obtained by hexane extraction of large water samples (40 L) followed by stepwise evaporation of the hexane

1. Observed AOX concentrations, in $\mu\text{g Cl/L}$, in water sampled in different parts of Lake Vättern (left) and in the vicinity of the Aspa mill (right). The concentration range shows the temporal variability of the AOX concentration (13).



extract to 2 mL, addition of 150 μL of hexadecane, and evaporation to a final volume of 150 μL . The amount of not-very-volatile, extractable organic halogens was determined by the combustion, at 1000°C, of 70 μL of the hexadecane extract, applied to silica in a crucible, followed by micro-coulometric titration of the formed halides. Very volatile compounds, such as chloroform, are lost during the final evaporation and were therefore analyzed separately according to a method involving GC/ECD (gas chromatography/electron capture) analysis of pentane extracts (18).

Chlorophenols in water were analyzed by extractive acetylation and GC/ECD or GC/MS in the selected ion monitoring mode (SIM) of the acetates formed; this method was originally developed by Voss (19). In our study, the detection level was lowered by including an XAD-8 adsorption step (17). Chlorophenols in sediments were analyzed by a method described by Remberger *et al.* (20).

The 2,4,6-trichloroanisole in water was determined using a modified stripping technique (21) and GC/MS(SIM) detection.

Occurrence of organic halogens in water

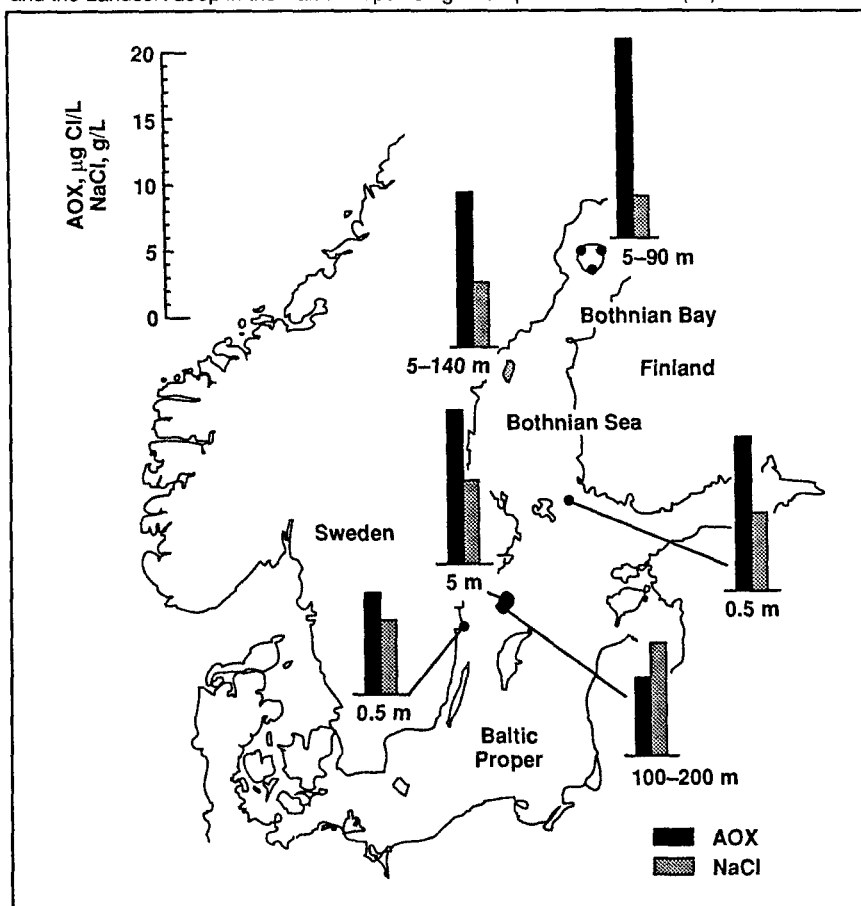
Adsorbable organic halogens (AOX)

As Fig. 1 illustrates, measurable amounts of AOX were present in all water samples from Lake Vättern. High concentrations of AOX were observed in the immediate vicinity of the bleach plant. However, at a distance of less than 2 km from the point of discharge, the amount of AOX had decreased approximately 15 $\mu\text{g Cl/L}$. This concentration was then maintained throughout the lake.

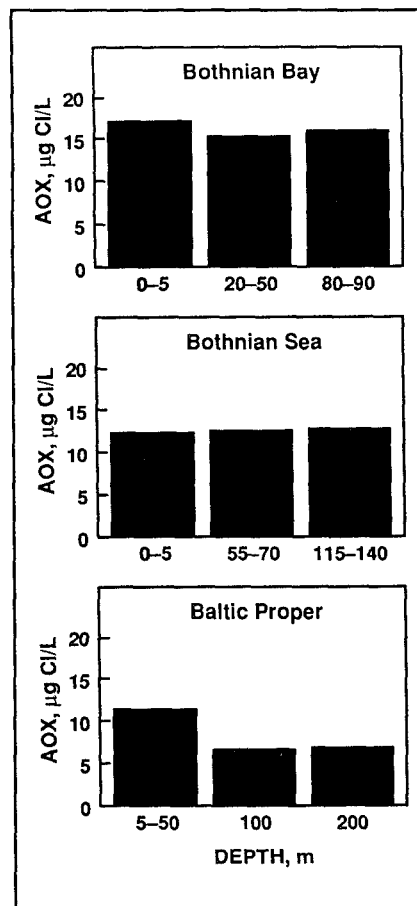
Measurable concentrations of AOX were also present in all analyzed water samples from the Baltic Sea. The highest AOX concentrations were observed in the Bothnian Bay, and the lowest were observed in the Baltic Proper, as illustrated in Fig. 2. The salinity showed an opposite pattern, with an upward trend from north to south. An inverse relationship between AOX and salinity was also observed at sampling sites where salinity increased with increasing depth (Fig. 3).

Surveys of AOX in unpolluted waters showed that higher concentra-

2. Observed AOX concentrations and NaCl concentrations in water samples from different parts of the Baltic Sea. Mean values of 5-10 samples in the Bothnian Bay, the Bothnian Sea, and the Landsort deep in the Baltic Proper. Single samples at other sites (13).



3. AOX concentrations at different depths in the three major basins of the Baltic Sea (17)



tions can be found in waters not exposed to any industrial discharges than in Lake Vättern and the Baltic Sea. The results presented in Fig. 4 show that the average AOX concentration ($32 \mu\text{g Cl/L}$) in the industrially unpolluted tributaries of Lake Vättern was higher than in the lake ($15 \mu\text{g Cl/L}$). Even higher concentrations (up to $200 \mu\text{g Cl/L}$) were observed in a survey of unpolluted lakes in the counties of Jönköping and Halland in southern Sweden (Fig. 5). In fact, some of these lakes had AOX concentrations that exceeded the highest concentrations reported from the industrially polluted Rhine River (22).

Measurements of AOX in precipitation have shown that the average concentration in Sweden is approximately $15 \mu\text{g Cl/L}$ (23). In throughfall, however, the AOX concentration can be much higher, and concentrations as high as several hundred $\mu\text{g Cl/L}$ have been found (24).

Chlorophenols

Four phenolic compounds, 2,4,6-trichlorophenol (2,4,6-TrCP), 3,4,5-trichloroguaiacol (3,4,5-TrCG), 4,5,6-trichloroguaiacol (4,5,6-TrCG), and tetrachloroguaiacol (TeCG), were selected as potential indicators of bleach plant discharges. Analysis of water from Lake Vättern showed that these compounds were present in all samples and that their concentrations decreased with the distance from the mill (Fig. 6). In the southern part of the lake, more than 100 km from the mill, the individual chlorophenols had concentrations around or below 1 ng/L .

In the Baltic Sea, samples were taken at sites that could be expected to be little influenced by bleach plant effluents. Still, one of the studied compounds, 3,4,5-TrCG, was detected in all samples. The other chlorophenols were only detected in some of the samples, and the concentrations were normally below 1 ng/L (Fig. 7).

In industrially unpolluted surface

waters, we detected only one of the four analyzed chlorophenols, 2,4,6-TrCP. This compound was sometimes present in concentrations up to 10 ng/L . A related compound, 2,4,6-trichloroanisole, which can be analyzed at concentrations as low as 0.1 ng/L , was detected in practically all of the analyzed water samples. The 2,4,6-trichloroanisole is probably formed by microbial methylation of 2,4,6-TrCP (25).

The results for precipitation samples were similar to those for unpolluted surface water samples. Traces of 2,4,6-TrCP and the corresponding anisole were found in some samples, but there was no evidence of other chlorophenols.

Volatile, extractable, and high-molecular-weight organic halogens

The major fraction of the AOX in bleach plant effluents consists of compounds of rather high molecular weight (26). As shown by Wigilius *et al.* (27) and Grøn (4), this is also true for AOX in unpolluted surface waters.

Only a very small fraction of the AOX in unpolluted surface waters can be extracted by hexane. In an on-going sampling program, EOX concentrations from 10 ng Cl/L to 45 ng Cl/L have been measured using the procedure mentioned for not-very-volatile EOX. GC/ECD analyses of pentane extracts indicate that the concentrations of very volatile organic halogens are also far below 1 $\mu\text{g Cl/L}$.

The concentration of extractable organic halogens (EOX) in bleach plant effluents has been reported to vary from 0.5% to 1.5% of the AOX concentration (28). However, the major fraction of the EOX consists of volatile compounds that easily evaporate during treatment in an aerated lagoon or in receiving waters. Other extractable compounds can be rapidly removed from the water phase by sedimentation. Measurements presently being performed at a bleach plant on the Motala River in Sweden indicate that, after passage through a lake (average residence time of about five weeks), the concentration of not-very-volatile EOX is only about 0.2% of the AOX concentration. This EOX-to-AOX ratio does not differ significantly from the ratios observed in unpolluted surface waters.

Mass balance calculations and estimated persistence

The historical development of AOX discharges from Swedish bleach plants to Lake Vättern and different sub-basins of the Baltic Sea has been estimated from production statistics and specific AOX discharges for different combinations of wood and bleaching processes. By combining this information with simple water exchange models for Lake Vättern and the Baltic Sea, we calculated the potential influence of bleach plant effluents on the average AOX concentration in receiving waters. Lake Vättern was assumed to consist of one undivided reservoir with complete mixing, whereas the Baltic Sea was considered to be divided into the Bothnian Bay, the Bothnian Sea, and the Baltic Proper.

The curves in Fig. 8A show the impact of the bleach plant, if AOX is removed from receiving waters only by water export—meaning no degradation, evaporation, or sedimentation of organic halogens. Under this as-

sumption, the AOX concentration in Lake Vättern would presently exceed 100 $\mu\text{g Cl/L}$. However, the AOX concentration actually observed was only about 15 $\mu\text{g Cl/L}$ (Fig. 1).

Further calculations using different assumptions regarding the removal of bleach plant AOX from the water phase of Lake Vättern showed that the average half-time was at most about two years. The curves in Fig. 8B show that this half-time would yield an average concentration close to the AOX concentrations actually observed. If the contributions from sources other than bleach plant effluents are also taken into account, the estimated half-time of bleach plant AOX will be lower.

So far, we have shown only that a considerable fraction of the AOX in bleach plant effluents is removed fairly rapidly from the water phase of Lake Vättern. This actuality does not exclude the possibility that there is a long-term accumulation of AOX in sediments. To test this hypothesis, AOX was analyzed in sediments from the accumulation bottoms of Lake Vättern. The results showed that the ratio of AOX to loss-on-ignition (during analysis) was on the order of 0.001 in sediments from the major accumulation bottoms in the lake (Table I). Combining this information with existing data about the total amount of organic matter in bottom sediments, we concluded that less AOX is stored in the sediments than in the water phase. Therefore, the removal of bleach plant AOX is not solely a matter of physical transport to the sediments, but major fractions of the AOX are also degraded either in the sediments or in the water phase.

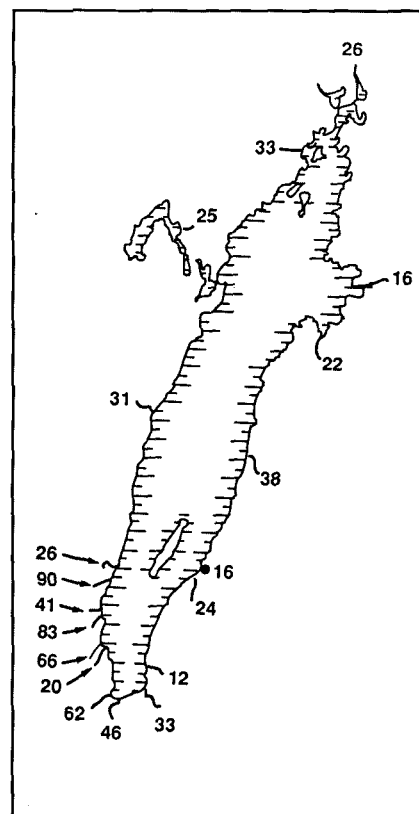
Because of its comparatively greater water exchange, the Baltic Sea is less exposed to bleach plant effluents than Lake Vättern. If AOX in receiving waters is removed by water export only, the bleach plant contribution that is theoretically possible will be at approximately the same level as the AOX concentrations actually observed (Fig. 2 and Fig. 8A). However, it is unrealistic to assume that there is no degradation, sedimentation, or evaporation. In Lake Vättern, the half-time of bleach plant AOX is at most two years; in the Baltic Sea, it is probably even shorter, because of the higher salinity in receiving waters. Higher salinity facilitates floccu-

lation and sedimentation of high-molecular-weight organics (29). In our study, there was an inverse relationship between AOX and salinity.

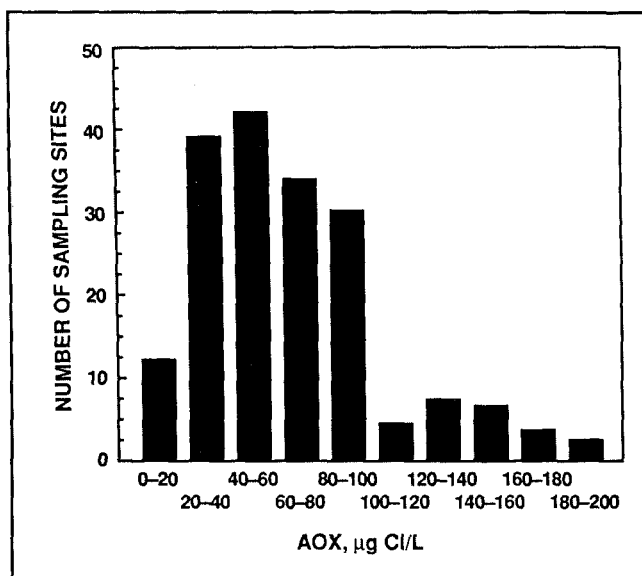
The calculated results shown in Fig. 8B indicate that the bleach plant contribution of AOX to the Baltic Sea probably does not exceed 3 $\mu\text{g Cl/L}$. In other words, the observed concentrations (7–15 $\mu\text{g Cl/L}$) in receiving waters are higher than the theoretically possible contribution from Swedish bleach plants. If discharges from nations other than Sweden are taken into account, the calculated contribution from bleach plant effluents will be slightly higher. However, this consideration cannot jeopardize the conclusion that, even in recipient waters heavily loaded with bleach plant effluents, sources other than bleach plants can contribute a major fraction of the AOX.

Even though the average half-time of AOX from bleach plants does not indicate long-term accumulation, certain fractions of AOX can be more persistent in water or sediments. Sediment studies have shown that traces of bleach plant effluents can be

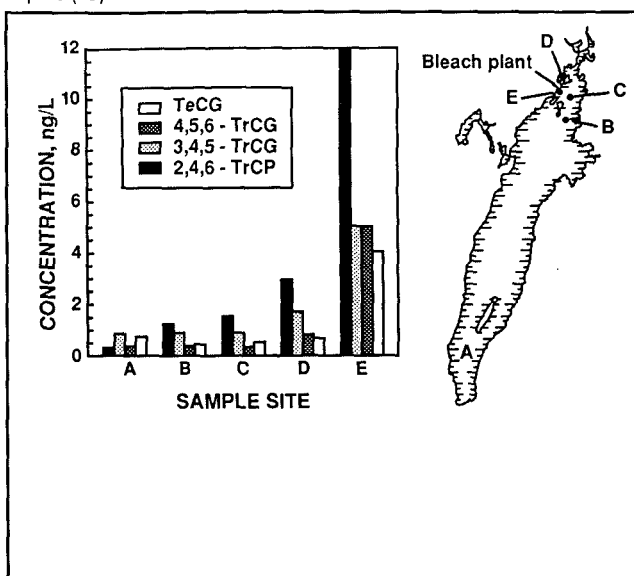
4. Average AOX $\mu\text{g Cl/L}$ concentrations in the major tributaries of Lake Vättern (13)



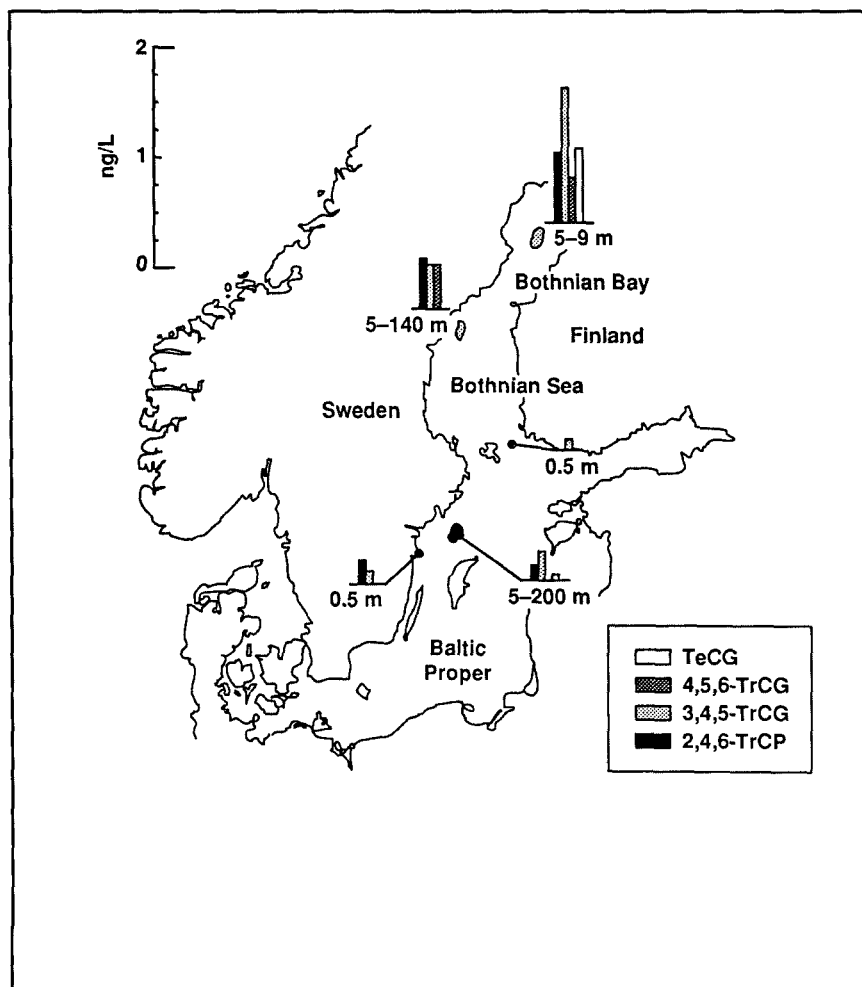
5. Observed AOX concentrations in lakes in the counties of Jönköping and Halland in southern Sweden. Sampling in March–April 1990.



6. Observed concentrations of 2,4,6-trichlorophenol, 3,4,5-trichloroguaiacol, 4,5,6-trichloroguaiacol, and tetrachloroguaiacol in Lake Vättern. Each value is the mean of two samples taken at different depths (13)



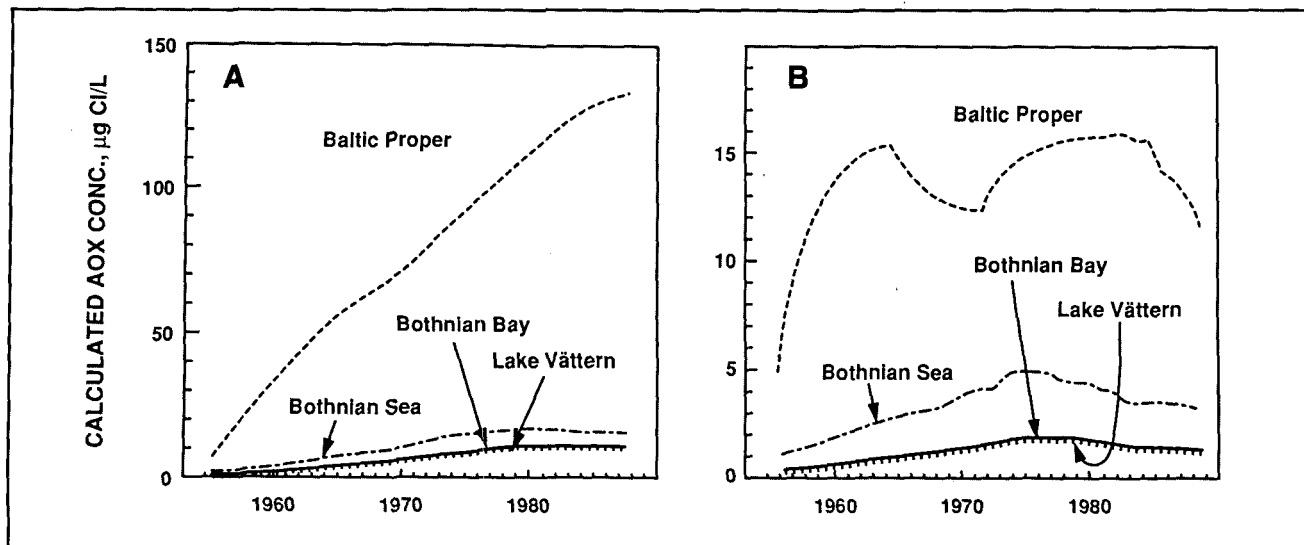
7. Observed concentrations (ng/L) of 2,4,6-trichlorophenol, 3,4,5-trichloroguaiacol, 4,5,6-trichloroguaiacol, and tetrachloroguaiacol in the Baltic Sea. Each value is the mean of two samples taken at different depths (13).



detected far from the points of discharge and that considerable amounts of EOX are being accumulated in the vicinity of bleach plants (10, 30). There may also be a considerable accumulation of EOX in remote parts of the Baltic Sea, but evidence of this is so far incomplete.

To estimate the half-time of chlorophenols in water, we also performed mass balance calculations for compounds of this type. Assuming that the chlorophenol-to-AOX ratio in the bleach kraft discharges has been constant, the expected chlorophenol concentrations in receiving waters can be calculated by multiplying the concentrations in Fig. 8 with the chlorophenol-to-AOX ratio in the effluent. Measurements at a bleach plant with an oxygen stage followed by a (C+D)(EO)DED sequence and chlorine doses of 35 kg/ton Cl_2 and 21 kg/ton ClO_2 indicated that this ratio can vary between 0.0003 and 0.003 for the analyzed chlorophenols (28). In Lake Vättern, a ratio in this range would give concentrations from 40 ng/L to 400 ng/L, if the analyzed chlorophenols were completely stable in water. The concentrations observed in Lake Vättern are much lower (Fig. 6). For example, not more than a few percent of the cumulated discharges of 3,4,5-TrCG can now be found in the lake water, and this corresponds to a half-time of a few months. Similar

8. Calculated impact of Swedish bleach plant discharges on the concentration of AOX in Lake Vättern and the Baltic Sea. Half-time of the AOX in water: (A) infinitely long and (B) 2 years. (13)



half-times have previously been reported for chloroguaiacols in large bodies of freshwater in Canada (12).

The persistence of chlorophenols in sediments is presently being studied. Preliminary results indicate that a considerable fraction of the cumulated 3,4,5-TrCG discharges can still be found in the bottom sediments. However, differences in the procedures for analysis of wastewater, surface water, and sediment samples make the results difficult to interpret: chlorophenols in water were analyzed by a direct solvent extraction procedure, whereas chlorophenols in sediments were analyzed after a treatment with NaOH that can release bound phenols.

Further evidence of a large natural production of organic halogens

The high concentrations of AOX in remote lakes in Sweden show that sources other than discharge point sources can make substantial contributions of organic halogens. Such contributions may be attributable to long-range atmospheric transport of industrial pollutants. However, the fact that the AOX concentration in surface water can be up to 10 times higher than the average AOX concentration in precipitation indicates that this is not the only pathway for AOX.

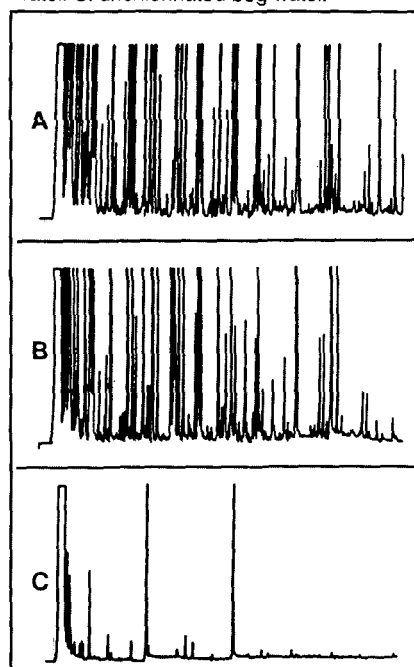
More solid evidence of a large natural production of organic halogens was recently obtained from a mass balance study of AOX in a raised

bog in Sweden (3). This study showed that the total pool of AOX in the bog corresponded to several hundred years of atmospheric wet deposition, and there were no local sources in the study area. Additional evidence of naturally produced organic halogens was obtained from the analysis of fulvic acids from groundwaters. AOX in measurable amounts was present in organic matter with an average age of 1300-5300 years.

In search of a mechanism that may explain the widespread occurrence of organic halogens in unpolluted environments, we tested the hypothesis of enzyme-mediated halogenation of humic substances. First, we used a commercially available chloroperoxidase. In the presence of hydrogen peroxide and chloride, this enzyme can incorporate up to 5% of chlorine into fulvic acid. Thereafter, we performed halogenation experiments using soil extracts obtained according to a modified procedure for extracting peroxidases from soil. These experiments showed that, in the presence of hydrogen peroxide and chloride, certain soil extracts were able to transform monochlorodimedone to dichlorodimedone. This ability was impaired when the extracts were heated (7).

Further studies revealed that soil extracts and the commercially available chloroperoxidase responded in practically the same manner to the addition of organic compounds known to inhibit the activity of haloperoxi-

9. ECD chromatograms of stripping extracts (31). A: enzyme-mediated chlorination of bog water. B: hypochlorite chlorination of bog water. C: unchlorinated bog water.



dases (9). This finding indicates that enzyme-mediated halogenation of humic substances occurs in soil.

Experimental studies of natural and technical chlorination

In a recently performed study, the reaction products obtained by enzyme-mediated chlorination were compared to such products from

I. AOX to loss-on-ignition at different depths and distances from the Aspa mill at Lake Vättern

Site number	AOX to loss-on-ignition, $\mu\text{g Cl/g}$	Distance from mill, km	Depth, m
1	17400	1.3	4.5
2	750	1.5	21
3	9200	1.7	7.0
4	3400	2.0	12
5	1900	3.0	40
6	1500	4.5	85
7	1300	5.5	95
8	1100	8.5	73
9	1200	11	55
10	860	30	82
11	870	100	118

nonenzymatic chlorinations (31). More precisely, a bog water sample was chlorinated by the addition of: (a) chloroperoxidase, hydrogen peroxide, and chloride, (b) hydrogen peroxide and chloride but no chloroperoxidase, or (c) hypochlorite. The ECD chromatograms of stripping extracts corresponding to the different chlorinations (Fig. 9) show that chloroperoxidase and hypochlorite yielded practically identical spectra of volatile, chlorinated reaction products. The addition of hydrogen peroxide and chloride alone resulted in very few chlorination by-products.

Analysis of chlorophenols in the chlorinated bog-water showed that 2,4,6-TrCP was a major reaction product of both enzyme-mediated chlorination and hypochlorite addition. This finding strengthens the conclusion that 2,4,6-TrCP can be formed by natural chlorination of humic substances. Furthermore, it provides additional evidence of similarities between natural and technical chlorination processes.

Conclusions

The presence of chloroguaiacols in water from remote parts of large lake and sea recipients showed that there is a measurable long-distance transport (> 100 km), in water, of bleach plant effluents. Estimates of average half-times of AOX and chlorophenols in water gave no evidence of several-year-long accumulation of organic

halogens in the water phase.

Even in heavily loaded bleach plant recipient waters, such as the Baltic Sea, diffuse sources (including natural halogenation processes) can contribute the major fraction of AOX. Enzyme-mediated halogenation of humic substances in soil is a plausible explanation of high AOX concentrations in unpolluted surface waters and observed background levels of 2,4,6-TrCP. □

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