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Occurrence of the Pharmaceutical Drug Clofibric Acid and the Herbicide Mecoprop in Various Swiss Lakes and in the North Sea

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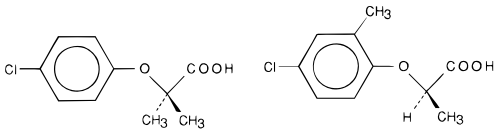
Clofibric acid (CA) and the herbicide mecoprop (MCPP) were detected in Swiss lakes from populated areas and in the North Sea in the low nanograms per liter range. Gas chromatography/mass spectrometry (GC/MS) and MS/MS were used to distinguish the two isomeric compounds (analyzed as the methyl esters). The concentrations of CA in the North Sea (1−2 ng/L) reach levels of those of "classical" environmental pollutants such as α- and γ-hexachlorocyclohexane. The data suggest a high mobility of CA in the aquatic environment. The changed relative concentrations of CA and MCPP in the North Sea as compared to those in the lakes and in comparison to production figures indicate a much more persistent behavior for CA than for MCPP. The data also indicate that some pharmaceutical compounds need evaluation on ecotoxicological grounds in very much the same way as do agricultural chemicals.

Introduction

The aquatic environment acts as a sink for many persistent compounds. The lakes and the sea thus receive inputs of anthropogenic compounds from agricultural, industrial, and other human activities via rivers and the atmosphere. The compounds, once dispersed in the environment, undergo various chemical, physical, and biological processes with continuing degradation and alteration. Pesticides, in particular herbicides, have a considerable potential for contaminating waters because they are applied directly to soil and may then leach into groundwater, rivers, and lakes and are eventually transported to the sea. Not surprisingly, various pesticides have been detected in the aquatic environment.

When we analyzed various natural waters for the presence of herbicides, we observed a consistently present contaminant from the group of the phenoxyalkanoic acids. The compound was subsequently identified as clofibric acid (CA; 2-[4-chlorophenoxy]-2-methylpropionic acid), a pharmaceutical drug and a structural isomer of the phenoxyalkanoic acid herbicide 2-[4-chloro-2-methylphenoxy]propionic acid (mecoprop, MCPP) (see Chart 1 for structures). CA is a high-volume chemical with an estimated annual production in the low kiloton range. It is mainly used in the form of the ethyl ester (clofibrate) in human medical care as a blood lipid regulator (*1*). It has relatively high therapeutic doses of 1−2 g/day per person and is used by patients for extended periods of time, sometimes life-long. The effective metabolite is the phenoxyalkanoic acid. CA appears to be metabolically and environmentally more stable than the related agricultural phenoxyacetic and 2-phenoxypropionic acids (MCPP; 2,4-dichlorophenoxyacetic acid, 2,4-D; 2-[2,4-dichlorophenoxy]-propionic acid, DCPP) (*2*).

CHART 1. Structure of Clofibric Acid (CA) and the Isomeric Mecoprop (MCPP; Chiral, Absolute Configuration of the *R* Enantiomer Indicated)



clofibric acid (CA) mecoprop (MCPP)

Over 20 years ago, CA was identified in effluents from domestic wastewater treatment plants (*3*, *4*). In 1991, the compound was detected in surface water, groundwater, and drinking water from areas formerly used as sewage farms in and around the city of Berlin and since then in rivers outside Berlin (*5*−*7*). These findings suggested this contamination to be not only a local problem from improper waste disposal but also likely to be a general environmental problem. We now report the presence of CA in various lakes in Switzerland and in the North Sea, thus the wide-spread occurrence of this pharmaceutical compound in the environment. The study shows that not only agricultural but also certain pharmaceutical compounds have to be considered as environmental contaminants, thus adding a new facet to environmental chemistry.

Experimental Section

Waters Sampled. Waters from four lakes situated in the central and eastern regions of Switzerland and from the North Sea were analyzed. The lakes studied were Zürichsee (Lake Zurich), Walensee, Greifensee, and Sempachersee (for specific details on these lakes, see refs 8 and 9). The North Sea (surface area, approximately 5.8×10^5 km^2; average depth, 94 m; volume, approximately 4.8×10^4 km^3) is situated on the continental shelf of northwest Europe (see map in Figure 1). It opens into the Atlantic Ocean to the north and via the English channel to the southwest. It is bounded by the coastlines of England, Scotland, Norway, Sweden, Denmark, Germany, the Netherlands, Belgium, and France. Approximately 164 million of people live within its catchment area (841 500 km^2). Especially, the catchment areas of the rivers Elbe, Weser, Rhine, Meuse, Scheldt, Seine, Thames, and Humber are densily populated, highly industrialized, and regions of intense farming. All these rivers discharge (164−194 km^3/year) into the southern part of the North Sea and are thus among the largest sources of contaminants in the North Sea. General circulation in the southern North Sea is mainly influenced by the inflow of Atlantic water through the Channel and prevailing westerly winds, causing mean transport in an eastward direction along the continental coast and northward along the Danish west coast. Due to these facts, the southeastern part of the North Sea, the German Bight, is one of the most severely contaminated areas of the North Sea. The water residence time in the German

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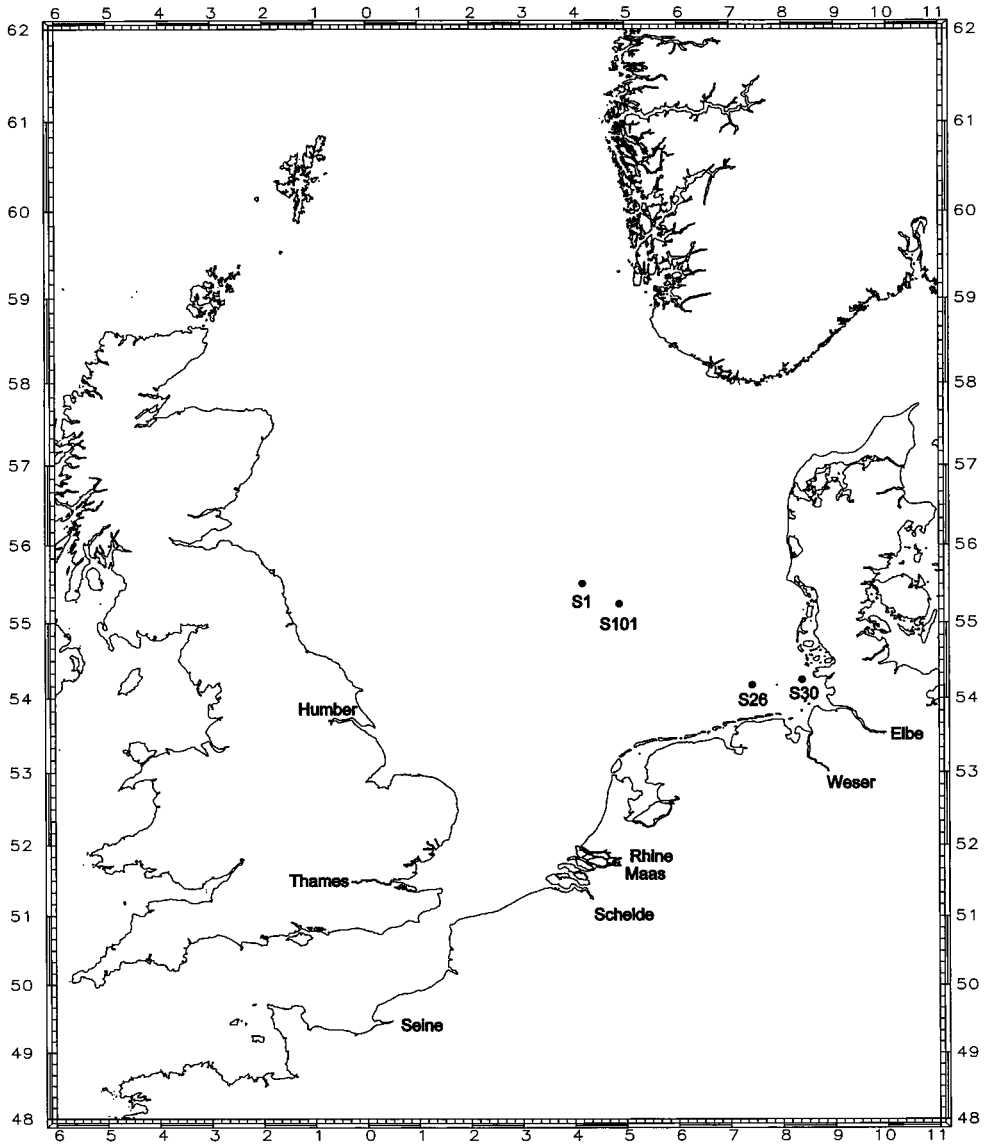


FIGURE 1. Map of the North Sea with sampling locations.

TABLE 1. Clofibric Acid (CA) and Mecoprop (MCPP) in Swiss Lakes and in the North Sea

sample, location	salinity	depth (m)	date	compound, concn (ng/L) CA	MCPP
North Sea, S1	34.403	5	Sep 4, 1996	1.2	≈0.6
North Sea, S101	34.725	6	Dec 1, 1996	2.4	≈0.6
North Sea, S26	33.770	5	Dec 3, 1996	1.3	1.2
North Sea, S26	31.059	5	Apr 20, 1997	1.5	2.0
North Sea, S30	29.739	3	Sep 1, 1996	7.8	2.7
North Sea, S30	28.434	5	Apr 19, 1997	≈0.5	≈0.9
North Sea, Helgoland, ref 15		?	1991	7[a]	11
Zürichsee[b]		1	June−Dec 1996	1−4.5	8−10
Sempachersee[b]		1	July−Dec 1996	1−5.5	11−15
Greifensee[b]		1	July−Dec 1996	2−9	25−45
Walensee		1	July 1996	<1	<1

[a] Reported as another isomer as MCPP. [b] Range of values reported.

Bight is estimated at 1−3 months. The flushing time of the North Sea is estimated at 1−2 years (*10*).

For this study, four sites in the North Sea (see Figure 1) were sampled on collection cruises conducted with the RV *Gauss* of the Federal Maritime and Hydrographic Agency, Hamburg, in September and December 1996 and in April 1997. Stations S26 and S30 are located in the coastal zone and are influenced by riverine inputs; they have salinities ranging from 28.4 to 33.7 (see Table 1), which means freshwater contents of 18−2.6%. Stations S1 and S101 lie in the outer German Bight (near the central North Sea) and have salinities of 34.4−34.7, typical values for southern and

central North Sea water. The four Swiss lakes were sampled on various occasions from July to December 1996.

Sampling and Analytical Procedures. Surface water from the lakes and the North Sea was collected with standard equipment and filled on-site into methanol-rinsed 1-L glass bottles. The samples were fortified with 50 μL of a 0.4 ng/μL $^{13}C_6$-(*R*/*S*)-DCPP (Cambridge Isotope Laboratories, Cambridge, MA; courtesy C. Zipper, Swiss Federal Institute of Environmental Science and Technology, EAWAG, Dübendorf) in methanol (spike level, 20 ng/L), shaken vigorously, and then kept at 4 °C until extracted and analyzed, usually within a few days. The water samples were not filtered; however, course particles were removed by sedimentation. The samples were extracted using solid-phase extraction with a reusable Bio-Rad SM-2 (Bio-Rad Laboratories, Hercules, CA) column as previously described for acetamide pesticides (*11*) except that the sample and the aqueous phases during partitioning were adjusted to pH $\approx$ 2 with dilute H_2SO_4. The extracts were methylated with diazomethane and then passed through a small silica column (0.7 g of silica gel 60, Merck, Darmstadt, FRG; deactivated with 5% water; 5 mm i.d. Pasteur pipet) topped with 10 mm of sodium sulfate. The analytes (as methyl esters, ME) were eluted with 10 mL of *n*-hexane–methylene chloride (1:1), the eluate was concentrated to 20–100 μL, and an 1–2-μL aliquot was used for analysis by high-resolution gas chromatography/mass spectrometry (HRGC-MS).

HRGC-MS analysis was performed on a VG Tribrid mass spectrometer (Micromass, Manchester, England) with electron-impact ionization (EI, 70 eV) under full-scan (*m/z* 35–435, 1.16 s/scan; mass resolution $M/\Delta M = 500$), selected ion monitoring (SIM), or MS/MS under conditions previously described (*12, 13*). In the MS/MS mode, the formation of specific daughter ions generated from selected parent ions was monitored (selected reaction monitoring, SRM), in particular using the ion transitions $228^+ \rightarrow 128^+$ (CA-ME), $228^+ \rightarrow 142^+$ (MCPP-ME), and $254^+ \rightarrow 168^+$ ($^{13}C_6$-DCPP-ME). An enantioselective 20-m OV1701-TBDM (TBDM, heptakis-[2,3-dimethyl-6-*tert*-butyldimethylsilyl]-β-cyclodextrin) fused silica (0.25 mm i.d.) HRGC column with 35% of the chiral selector (amount relative to OV1701) was used for analysis. The column was temperature programmed as follows: 50 °C, 2-min isothermal, 20 °C/min to 120 °C, then at 2.5 °C/min to 240 °C, followed by an isothermal hold at this temperature. Enantiomeric ratios (ER) for MCPP were defined as $ER = p_R/p_S$ whereby p_R and p_S are the peak areas of the earlier eluted (*R*)- and the later eluted (*S*)-MCPP-ME, respectively.

The amounts of an analyte were determined from peak area ratios relative to the internal standard ($^{13}C_6$-DCPP, summed concentrations of the enantiomers) and in reference to suitable standard solutions (see ref 13 for the source of the reference compounds). The solid-phase extraction procedure allowed the detection of these compounds at concentrations of 0.2–1 ng/L with acceptable recoveries (>50%) when surface waters from lakes and rivers were analyzed . Initially, low recoveries for $^{13}C_6$-DCPP were experienced with water from the North Sea for unknown reasons; sampling and analysis were then repeated. For this study, the total concentration of MCPP was calculated as the summed concentration of both enantiomers.

Results and Discussion

GC/MS Analysis and Distinction of CA and MCPP Using MS/MS. The GC/MS analysis of the reference materials yielded an earlier eluted single peak for the achiral CA and two later eluted peaks for the two enantiomers of MCPP, when analyzed as the methyl esters on the enantioselective OV1701-TBDM column. Single peaks for both compounds were obtained when HRGC columns with an achiral stationary phase (DB-1) were used. The EI mass spectra of the two isomeric compounds, CA-ME and MCPP-ME (*R* enantiomer), are shown in Figure 2. The mass spectra of both compounds show molecular ions ($M^{\bullet+}$) at *m/z* 228 and a major fragment ion at *m/z* 169 [M – $COOCH_3$]$^+$. However, the two compounds differ in the further fragmentation in such that the "chlorophenol ions" $(ArOH)^{\bullet+}$ are at *m/z* 128 for CA-ME and at *m/z* 142 for MCPP-ME, respectively. These ions are formed via a four-ring mechanism, as pointed out in Chart 2 and discussed in a previous report (*12*). The ions selected to monitor CA-ME and MCPP-ME in SIM analyses were the $M^{\bullet+}$ ions at *m/z* 228, the major fragment ions at *m/z* 169, and their Cl isotopic ions.

In Figure 3, EI SIM chromatograms (*m/z* 228) show the elution of these compounds (as methyl esters) in extracts of water samples from Sempachersee (August 1996) and from the North Sea (sampling station S30, September 1996). In Figure 3, upper panel, the SIM chromatogram for the North

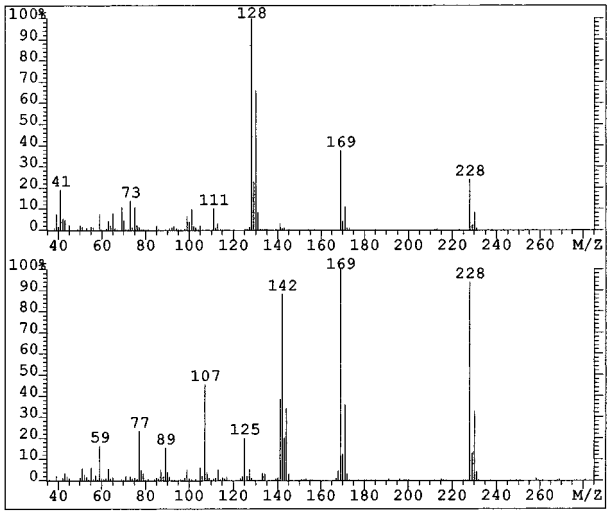
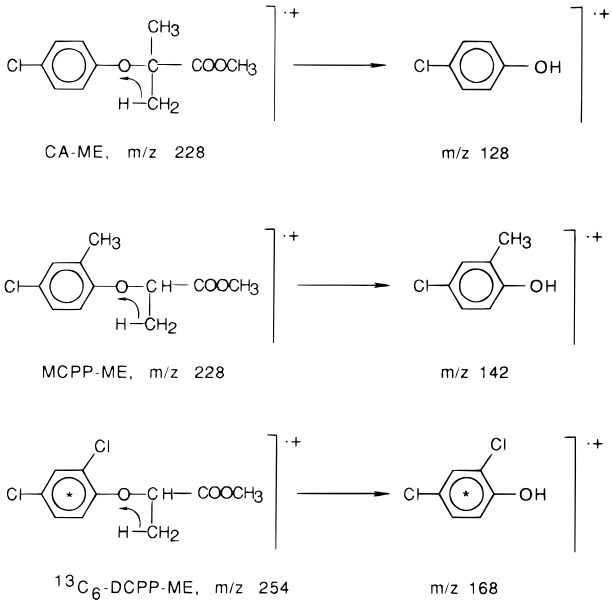


FIGURE 2. EI mass spectra of CA-ME (upper panel) and MCPP-ME (*R* enantiomer; lower panel). Note presence of isobaric $M^{\bullet+}$ and $(M - 59)^+$ ions and the diagnostically important "chlorophenol" ions at *m/z* 128 and 142 for CA-ME and MCPP-ME, respectively.

CHART 2. Formation of "Chlorophenol" Ions ($ArOH^{\bullet+}$) and Ion Transitions (SRM) Monitored for the Detection of CA (Top), MCPP (Middle), and $^{13}C_6$-DCPP (Internal Standard, Bottom; Asterisk Denotes the ^{13}C Labeling), Analyzed as Methyl Esters



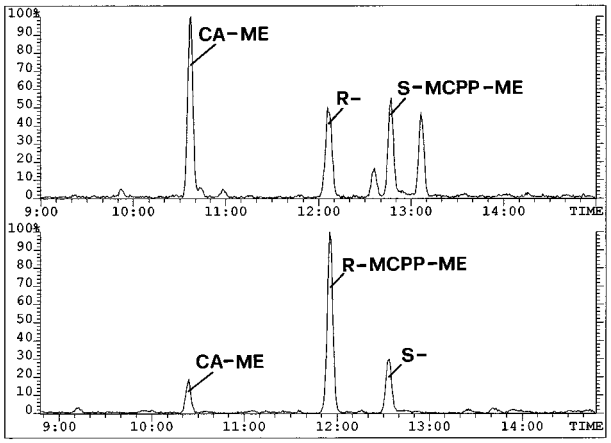


FIGURE 3. EI SIM chromatograms (*m/z* 228) showing elution of CA, (*R*)- and (*S*)-MCPP in a sample from the North Sea (sampling station S30, September 1996; upper panel) and in a sample from Sempachersee (lower panel) analyzed as the methyl esters using the enantioselective OV1701-TBDM HRGC column. Note presence of additional components in the North Sea sample.

Sea sample indicates the presence of 7.8 and 2.7 ng/L of CA and MCPP, respectively, with a similar abundance of the two enantiomers of MCPP (ER ≈ 0.9). This chromatogram indicates the presence of peaks from additional, not further identified, components. In Figure 3, lower panel, the SIM chromatogram for the Sempachersee sample indicates the presence of 5 and 14 ng/L CA and MCPP, respectively; this time with a higher abundance of (*R*)-MCPP (ER = 2.5). The presence of CA and MCPP in these samples was confirmed when additional ions were monitored. However, the amounts present were too low to allow recording of full-scan mass spectra for these compounds. Although just the *R* enantiomer, designated as mecoprop-P, is registered in Switzerland, the presence of both enantiomers in the Swiss lakes is not unexpected since (*R*)-MCPP is partly enantiomerized in soil to (*S*)-MCPP in a biologically mediated reaction (*13*).

The samples were further analyzed using MS/MS. MS/MS previously indicated several ion transitions of high signal abundance for both compounds (see ref 12). Of particular importance here were the transitions from $M^{\bullet+}$ to $(ArOH)^{\bullet+}$ via the loss of the side chain, $228^+ \rightarrow 128^+$ for CA-ME (loss of $C_5H_8O_2$) and $228^+ \rightarrow 142^+$ for MCPP-ME (loss of $C_4H_6O_2$), respectively (see Chart 2). The isomeric compounds were thus distinguished by monitoring different daughter ions generated from parent ions with the same mass. The internal standard ($^{13}C_6$-DCPP) was monitored as the methyl ester via the analogous transition $254^+ \rightarrow 168^+$. In Figure 4, SRM chromatograms show the elution of these compounds for the North Sea sample (September 1996). In Figure 4, upper panel, the SRM chromatogram $228^+ \rightarrow 128^+$ shows the elution of CA-ME but no signals for MCPP-ME and the other, previously under SIM conditions, detected compounds. In contrast, the SRM chromatogram $228^+ \rightarrow 142^+$ (middle panel) shows the elution of MCPP-ME (both enantiomers; ER ≈ 0.8) but no signals for CA-ME. The two SRM chromatograms document the high selectivity for the detection of these isomeric compounds. Finally, the SRM chromatogram $254^+ \rightarrow 168^+$ (lower panel) shows the elution of $^{13}C_6$-DCPP-ME, confirming its (approximate) racemic composition.

The presence of CA and MCPP in these samples was thus confirmed by the following findings: (1) occurrence of the acidic compounds in the appropriate analytical fractions prior to and after methylation; (2) co-elution with the authentic compounds on HRGC columns with chiral and achiral stationary phases (data not shown); (3) simultaneous signals for characteristic ions with the expected response ratios using

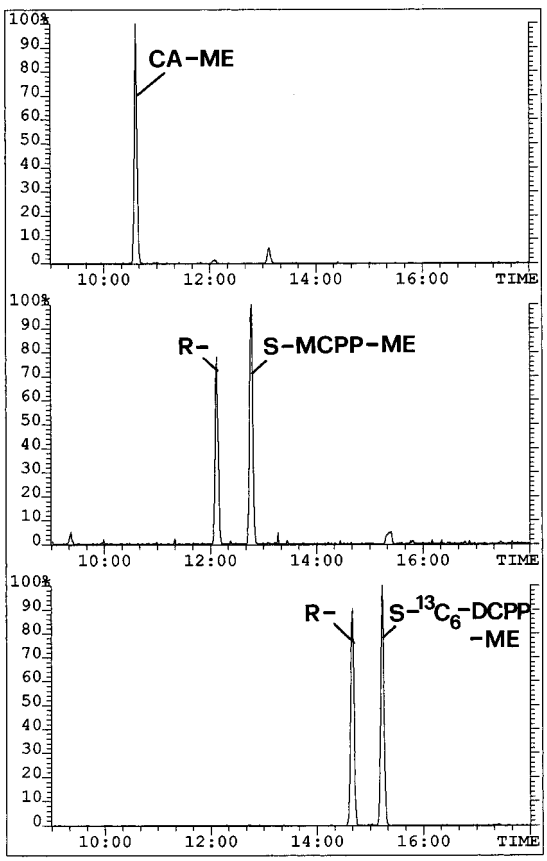


FIGURE 4. SRM chromatograms confirming the presence of CA ($228^+ \rightarrow 128^+$; upper panel), and (*R*)- and (*S*)-MCPP ($228^+ \rightarrow 142^+$; middle panel) in the North Sea sample (sampling station S30, September 1996). Also shown, SRM chromatogram $254^+ \rightarrow 168^+$ for $^{13}C_6$-DCPP (internal standard, lower panel). All compounds were analyzed as the methyl esters using the enantioselective OV1701-TBDM HRGC column.

different HRGC columns; and (4) MS/MS confirmation using characteristic ion transitions for both of the compounds.

CA and MCPP in the Various Swiss Lakes. The concentrations of CA and MCPP in the various lakes are listed in Table 1. The concentrations of CA in the Zürichsee, the Sempachersee, and the Greifensee were in the range of 1−9 ng/L and for MCPP were up to 45 ng/L. For the Zürichsee, the largest of these lakes (estimated water volume, ≈ 3.8×10^9 m^3), concentrations of 1−5 ng/L amount to a total of 3.8−19 kg CA in the lake. With a water export of ≈2.7×10^9 m^3/year about 2.7−13.5 kg of CA are removed by export. Assuming a steady-state situation, a similar amount of CA must enter the lake per year, corresponding to 1350−13 500 daily doses/year of CA from a total population of 315 000 in the catchment area (note that wastewater treatment plants of the city of Zurich do not discharge into the lake). CA and MCPP were not detected in the Walensee, a lake situated in a more mountainous, less populated region.

CA and MCPP in the North Sea. The concentrations of CA and MCPP from the locations in the North Sea are also reported in Table 1. CA and MCPP were detected at concentrations in the range of 0.5−7.8 ng/L and 0.6−2.7 ng/L, respectively. The highest concentrations of both compounds were found in a sample taken in the plume of the river Elbe (sampling station S30, September 1996). Therefore, it is almost certain that this river is a source of these compounds, although the very different concentrations observed in September 1996 and April 1997 indicate very unsteady inputs. Previously, CA was detected in the Elbe (27−157 ng/L, see ref 7) and other rivers. Based on the limited

data available so far, a detailed interpretation of this distribution is hardly possible. Nevertheless, the data already available now suggest a different environmental behavior of the two compounds as outlined below.

The data suggest a quite uniform distribution of CA over the whole German Bight (concentrations 0.5−2.4 ng/L; excepting the value of 7.8 ng/L at station S30 in September 1996); even near the central North Sea with a salinity of >34, concentrations of 1−2 ng/L are observed. Such an ubiquitous occurrence can be explained either by atmospheric inputs (which is not very likely for this compound) or by widespread inputs, i.e., by inputs from all coastal areas and a quite persistent behavior. Stations S1 and S101 may be influenced by inputs from the U.K. coast, as has been observed, for example, with α-HCH and by inputs from the river Rhine (*14*). Ludwig *et al.* (*15, 16*) reported the presence of a not further specified, earlier eluted "MCPP" isomer at a concentration of 7 ng/L in a water sample taken east of Helgoland in the North Sea in 1991 (HRMS SIM data), supposedly from a location not too far from our sampling station S30. It is almost certain that this earlier eluted isomer is in fact CA. This would imply that the concentrations of CA 6 years ago, when the former study was made, was of the same magnitude as today. Assuming an average concentration of 1−2 ng/L in the North Sea and an estimated water volume of 4.8×10^{13} m^3, the total amount of CA in the water is calculated as 48−96 ton. Assuming a steady state over the years and a water residence time of 1−2 years, the annual inputs are estimated to be in the range of 50−100 ton. CA-containing drugs were prescribed in Germany in amounts of about 30 ton per year (1992 and 1993); simple extrapolation to the population of the whole North Sea catchment area would yield some 70 ton. It is highly remarkable that the concentrations of CA in the North Sea are of the same magnitude as in the Swiss lakes, which are much closer to human activities. This and the apparent absence of a concentration gradient from the coast to the open sea indicate a very persistent behavior of CA in the environment. It should be noted that these concentrations are similar to those of classical environmental pollutants such as γ-HCH (lindane) and α-HCH (*14*).

The data for MCPP suggest a relatively clear concentration gradient from the coast (1−2.7 ng/L at stations S26 and S30, with a salinity of 28.4−33.7) to the open sea (≈0.6 ng/L at stations S1 and S101 with a salinity of 34.725). This can be explained by inputs from the rivers Elbe and Weser and the dilution with uncontaminated seawater and/or a decrease by degradation. As expected, concentrations in most Swiss lakes are higher than those in the coastal region of the North Sea (see Table 1). Concentrations of MCPP in the open sea are lower than those of CA, although MCPP is used in far greater quantities [e.g., in 1995, mecoprop-P was used in Germany in amounts of 200−500 ton; in the whole area of the Oslo and Paris Convention, in 1993/94, mecoprop was used in amounts of ≈2000 ton, and mecoprop-P in amounts of ≈1400 ton (*17*); 50−70% of the total quantity was applied in the catchment area of the North Sea]. These observations and considerations point to a much higher environmental persistence of CA.

Concluding Remarks. The data confirm that pharmaceutical compounds can be present in environmental samples at concentrations similar to those of pesticides. CA appears to be highly mobile and persistent in the aquatic environment. CA has been previously detected in source-specific locations and in effluents from a wastewater treatment plant (*3, 4*). The wide spread occurrence of CA, however, is relatively new. Since CA is not manufactured in Switzerland, its presence in this country must reflect human consumption. The presence of CA in the lakes points to inputs from the therapeutic use of this compound via wastewater treatment plants (≈80% of Swiss households connected to such plants). The likely presence of CA in the North Sea since at least 1991 suggests a long-time contamination. This and previous studies (*5−7, 18*) revealed that pharmaceuticals may lead to detectable concentrations in the environment. It can be speculated that other high-volume pharmaceutical compounds with appropriate physicochemical properties may also lead to detectable concentrations and possible environmental contamination. This also means that such compounds should be evaluated on the grounds of their ecotoxicological properties, as has been done for a long time with agricultural chemicals.

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