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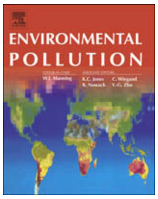

Occurrence and distribution of triclosan in the German Bight (North Sea)

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Investigation with coastal survey reveals distribution of triclosan in marine waters.

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ABSTRACT

The potential of triclosan (TCS) acting as an endocrine disruptor has led to growing concern about the presence of TCS in the environment. In this study, seawater samples were collected from the German Bight during sampling campaigns conducted with the German research ships *Gauss* and *Ludwig Prandtl*. TCS was determined both in the dissolved phase and in the suspended particulate matters with concentrations ranging 0.8–6870 pg L^{-1} and <1–95 pg L^{-1}, respectively. High concentrations of TCS were present in the estuaries of the Elbe and the Weser, indicating significant input of TCS by the river discharge. The correlation coefficient (R^2) between the dissolved concentration and salinity was 0.79 for the data obtained from the Gauss cruise, showing an obvious declining trend from the coast to the open sea.

1. Introduction

Triclosan (5-chloro-2-[2,4-dichloro-phenoxy]-phenol) (TCS) is widely used as antimicrobial agent in personal care products (Sabaliunas et al., 2003). It is also added to textiles to prevent odor, and to plastic products such as cutting boards. The production of TCS has been relatively stable in recent years, which is used ~350 t yr^{-1} in the EU (Environment Agency, UK). Studies have shown that TCS present in surface water may be toxic to certain algae species (e.g. *Scenedesmus subspicatus*) with a reported no-observer-effect concentration (72-h growth NOEC) of 500 ng L^{-1} (Orvos et al., 2002; Reiss et al., 2002). While Wilson et al. (2003) have shown that TCS may cause significant increase in *Synedra* and a significant reduction of the rare genus *Chlamydomonas* at 15 ng L^{-1} and 150 ng L^{-1}. Moreover, recent studies have shown that TCS blocks bacterial lipid biosynthesis by specifically inhibiting the enzyme enoyl–acyl carrier protein reductase, which led to concerns of the possible development of bacterial resistance to TCS (McMurry et al., 1998; Levy et al., 1999). Thus, it is necessary to investigate the occurrence and fate of TCS in the environment.

The Review studies (Heberer, 2002; Samsoe-Petersen et al., 2003) showed that TCS is degradable under aerobic conditions in wastewater treatment plants (WWTPs) and is extensively degraded and removed in activated-sludge systems. Results have shown about 70–90% of TCS is removed in the sewage and wastewater treatment plants (McAvoy et al., 2002; Singer et al., 2002; Bester, 2003; Ying and Kookana, 2007; Stasinakis et al., 2007), the remaining fraction is discharged with the effluent into the river and partly enters into the coastal water. Studies regarding photolysis of TCS in surface water have demonstrated that this may be a significant pathway in the upper layers of lakes, while TCS is quite stable for hydrolysis (Tixier et al., 2002; Aranami and Readman, 2007).

Studies in Europe, North America, Australia and China have reported the widespread occurrence of TCS with concentrations ranging 1000–10,000 ng L^{-1} for influent, 40–2000 ng L^{-1} for effluent (Singer et al., 2002; Bester, 2003; Weigel et al., 2004; Halden and Paull, 2005; Ying and Kookana, 2007) and <1–100 ng L^{-1} in the surface water (Lindström et al., 2002; Bester, 2005; Hua et al., 2005; Wu et al., 2007). TCS and its methyl metabolite have also been detected in fish in surface waters (Adolfsson-Erici et al., 2002; Böhmer et al., 2004; Balmer et al., 2004) and in algal in a north Texas wastewater treatment plant receiving stream (Coogan et al., 2007). Although studies have demonstrated the ubiquitous distributions of TCS, little information is available for coastal waters.

The objective of the present study was to understand distribution levels of TCS in surface seawater of the German Bight. Spatial variations of TCS in the coastal waters were discussed. This study provided reliable data to understand the fate of TCS in coastal waters.

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Table 1
Summary of results of TCS in dissolved ($[TCS]_{diss}$) and suspended particulate matters ($[TCS]_{SPM}$) measured in the German Bight during the NUTEX-5 cruise, particulate associated fractions (φ), and sample collecting data; water sample numbers of the sampling sites are marked on Fig. 2

Sample site	Longitude (E)	Latitude (N)	Depth (m)	Temperature (°C)	Salinity (‰)	pH	SPM (mg L^{-1})	f_{OC} (%)	$[TCS]_{diss}$ (pg L^{-1})	$[TCS]_{SPM}$ (pg L^{-1})	Φ (%)	$[TCS]_{SPM}$ (ng g^{-1})
1	6.96	53.4	11	18.6	25.2	7.84	18.4	18	110	27	19	1.5
2	6.5	53.62	11	16.7	32.5	8.02	8.7	24	89	3.5	4	0.4
3	7.08	53.82	20	15.2	33.7	8.32	2	50	12	<1	–	–
4	7.16	53.74	5	16.7	33.4	8.18	4.6	35	30	n.a.	–	–
5	7.42	53.87	23	15.6	33.5	8.29	2	70	18	<1	–	–
6	7.42	53.78	10	17	32.9	8.27	3	47	8	1.5	16	0.5
7	7.64	53.87	22	15	33.6	8.26	3.5	40	18	6	25	1.7
8	8.03	53.79	28	15.5	33.1	8.32	2	60	19	n.a.		–
9	8.23	53.47	2.5	18.9	31.8	8.05	58.2	15	100	95	48	1.6
10	8.18	53.51	13	18.5	32.8	8.07	18.7	17	140	29	17	1.6
11	8.17	53.59	17	17.1	31.8	8.34	9.9	23	120	9.3	7	0.9
12	8.29	53.71	2.5	19.7	22.3	7.79	25	18	620	29	5	1.2
13	8.02	53.92	12.5	14.9	34	8.26	6	23	33	2.3	7	0.4
14	8.22	53.98	22	14.2	34	8.26	2.6	46	10	n.a.	–	–
15	8.32	54.02	8	16	32.4	8.30	4.5	38	6870	58	0.8	13
16	8.70	54.12	13	18.2	27.9	8.40	13.2	26	1870	46	2.4	3.5
17	8.91	54.11	2	18.7	27.6	8.19	54.6	11	3200	88	3	1.6
18	8.73	54.12	9.5	18.6	28	8.42	14.2	24	1210	22	2	1.5
19	8.34	54.32	11	18	31.5	8.33	2	60	140	13	9	6.5
20	8.52	54.37	14.5	19.1	31.6	8.13	8.9	24	40	n.a.	–	–
21	8.22	54.47	10	19.1	31	8.28	4.7	30	1640	n.a.	–	–
22	8.27	54.70	15	18.4	31.9	8.16	21.3	17	1350	n.a.	–	–
23	8.40	55.07	7.5	20.5	31.2	8.10	3.7	32	1270	9.6	1	2.6
24	8.33	55.09	6	19.2	31.6	8.14	2.4	58	750	n.a.	–	–

2. Experimental section

2.1. Water sampling

Seawater samples were collected in the German Bight during BSH (Federal Maritime and Hydrographic Agency in Hamburg) routine monitoring cruise in 29/2–10/3/2004 with research ship '*Gauss*', and during a coastal survey 'NUTEX-5' with research ship '*Ludwig Prandtl*' in 27/6–15/7/2005.

Seawater samples were collected from a stainless steel pipe at 5 m-depth onboard *Gauss* (30–400 L) and at 1 m-depth onboard *Ludwig Prandtl* (10–50 L), using the ship's water circuit located in the keel. The average flow rate was about 0.7 L min^{-1}. Both suspended particular matter (SPM) and dissolved seawater phases were collected using a GFF (GFF 52 with 0.7 μm of nominal pore size) and a 95 mL PAD-2 resin column, respectively. Field blanks of water sampling were prepared onboard. Details of the seawater samples are shown in Table 1 and mean sampling position is shown in Figs. 1 and 2, respectively.

PAD-2 columns were pre-cleaned by consecutive Soxhlet extractions with acetone and dichloromethane, rinsed with copious amounts of water and were capped and sealed in polyethylene bags. GFFs were combusted at 450 °C for 12 h and wrapped in aluminum foil. After sampling, PAD-2 columns and GFFs were, respectively, stored at 4 °C and −20 °C until analysis. Samples were analyzed in clean-room facilities.

2.2. Extraction and clean-up

The PAD-2 columns and GFFs (Nutex-5) were spiked with 20 ng of AHTN D_3 after sampling (if the column was not spiked before sampling) and 20 ng of internal standard dibenzyl phthalate (Dr. Ehrenstorfer GmbH, Augsburg, Germany). They

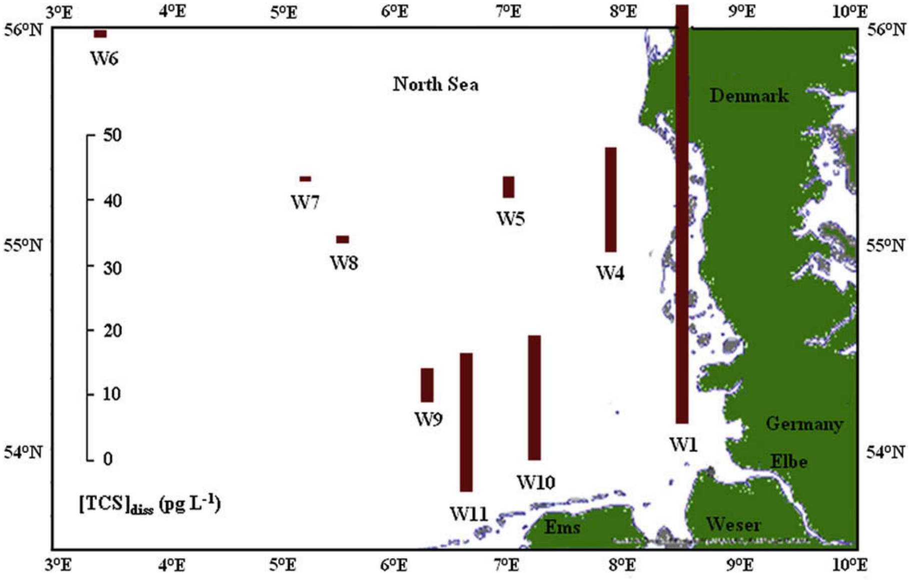

Fig. 1. Spatial distribution of the dissolved TCS in the German Bight determined from water sampling with *Gauss* cruise in 2004. The bars are placed on the average position for each water sample.