Mass balance analysis of triclosan, diethyltoluamide, crotamiton and carbamazepine in sewage treatment plants

N. Nakada, M. Yasojima, Y. Okayasu, K. Komori and Y. Suzuki

ABSTRACT

The behavior of antibacterial triclosan, insect-repellent diethyltoluamide (DEET), anticonvulsant carbamazepine, and antipruritic crotamiton was investigated at two sewage treatment plants (STPs) to clarify their complete mass balance. Twenty-four-hour flow-proportional composite samples were collected from the influent and effluent of primary and final sedimentation tanks, a biofiltration tank and disinfection tanks. Sludge samples (i.e., activated and excess sludge) and samples of the return flow from the sludge treatment process were collected in the same manner. The analytes in both the dissolved and particulate phases were individually determined by a gas chromatograph equipped with mass spectrometer. Triclosan was dominantly detected in the particulate phase especially in the early stage of treatment (up to 83%) and was efficiently removed (over 90%) in STPs, mainly by sorption to sewage sludge. Limited removal was observed for DEET (55 $\pm$ 24%), while no significant removal was demonstrated for crotamiton or carbamazepine. The solid-water distribution coefficients (K_d, $n = 4$) for triclosan (log K_d: 3.7–5.1), DEET (1.3–1.9) and crotamiton (1.1–1.6) in the sludge samples are also determined in this study. These findings indicate the limitations of current sewage treatment techniques for the removal of these water-soluble drugs (i.e. DEET, carbamazepine, and crotamiton).

Key words | mass balance, personal care products, pharmaceutical, sludge, solid-water distribution coefficients (K_d)

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INTRODUCTION

The occurrence and fate of pharmaceutically active compounds (PhACs) in the aquatic environment have been demonstrated over the past decade in terms of potential adverse effects on humans and aquatic organisms (Ternes 1998; Kolpin *et al.* 2002). Although PhACs, especially those found in pharmaceuticals and personal care products (PPCPs), are necessary for maintaining our health and for stable stockbreeding, some PPCPs are directly and indirectly discharged into the aquatic environment (Ternes 1998; Kolpin *et al.* 2002; Nakada *et al.* 2008), even under appropriate usage conditions. In fact, numerous studies have reported the occurrence of PPCPs in conventional sewage treatment facilities (Ternes 1998; Nakada *et al.* 2006*a*; Kim *et al.* 2007), rivers and groundwater (Kolpin *et al.* 2002; Nakada *et al.* 2008), seawater (Weigel *et al.* 2002) and waterworks (Heberer 2002). These studies have also documented the persistence of certain PPCPs (e.g., diethyltoluamide (DEET), carbamazepine, crotamiton, iopromide, and clofibric acid) in the environment and current water treatment processes.

Although some articles have reported the presence and removal efficiency of PhACs during sewage treatment, there have been few reports detailing their fate in conventional sewage treatment plants including their

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distribution in sludge, except for a few surveys on triclosan (Heidler & Halden 2007) and antibiotics (Golet *et al.* 2003; Göbel *et al.* 2005; Lindberg *et al.* 2006). In addition, only a few studies have considered the contribution of the input of PPCPs via a dewatered process and recycling streams in sewage treatment plants (STPs) (Nakada *et al.* 2005). Detailed information on the fate of PPCPs during the treatment process is crucial as regards to selecting the appropriate treatment. Additionally, such information should inspire the pharmaceutical industry to shift to other ingredients (i.e., voluntary control) in the same drug categories, because there are considerably more alternative drug ingredients than treatment methods available to wastewater engineers.

The present study surveyed the fates of persistent amide-type drugs (i.e., DEET, crotamiton and carbamazepine) and triclosan (Figure 1a) in conventional STPs. Triclosan (2,4,4'-trichloro-2'-hydroxyphenyl ether) is used throughout the world as a bactericide in various personal care and consumer products. Diethyltoluamide (*N,N*-diethyl-m-toluamide), commonly known as DEET, is also widely used as an insect repellent. DEET has frequently been observed in non-target nationwide screening in the USA (Kolpin *et al.* 2002). Carbamazepine (5*H*-dibenzepine-5-carboxamide) is prescribed for psychiatric purposes such as for treating epilepsy. Crotamiton (*N*-ethyl-*o*-crotonotoluidide) is incorporated in certain skin lotions to treat scabies and itching.

The aim of the present study is to clarify detail the fates of triclosan, DEET, crotamiton and carbamazepine in municipal STPs in Japan and to demonstrate the limitation as regards the removal for these compounds in detail. This study estimates complete mass balances from the detected concentrations and measured aqueous/ sludge flows.

MATERIALS AND METHODS

Chemicals

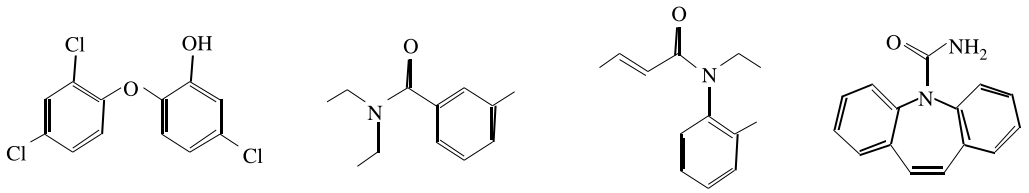
Triclosan and surrogate standard carbamazepine-d_{10} were purchased from Wako Pure Chemical Industries Ltd. (Osaka, Japan). DEET and carbamazepine were purchased from Across Organics (Geel, Belgium). Crotamiton was purchased from Sigma-Aldrich (Tokyo, Japan). All solvents were obtained from Wako Pure Chemical Industries Ltd. (Osaka, Japan) or Kanto Chemical Inc. (Tokyo, Japan) and were of special grade for PCB analysis. Chemical structures and physicochemical properties of target compounds in this study are shown in Figure 1a.

Description of sewage treatment plants and sampling

Surveys were conducted at two sewage treatment plants (STP-1 and STP-2) in Kanagawa Prefecture, Japan in July and September of 2004, respectively (Figure 1b). We have already described the mass balance of five anti-inflammatory drugs in these two STPs (Nakada *et al.* 2005) employing the surveys used in this study. Additionally, the mass balance of estrogen and its conjugates, nonylphenol and its acidic metabolites, and estrogenic activity in STP-1 in winter and summer have been determined (Nakada *et al.* 2006*b*). STP-1 and STP-2 serve populations of approximately 30,500 and 70,600, respectively. Both STPs treat mainly domestic sewage, and activated sludge treatment with low nitrification activity is carried out. The aeration tank consists of a long-channel plug-flow reactor, followed by a final sedimentation tank. In STP-1, the secondary effluent is disinfected with sodium hypochlorite before discharge, but in STP-2, the secondary effluent flows through a biological filtration system followed by an ultraviolet (UV) treatment tank for disinfection (Figure 1b). The filtration system contains ceramic particles (6 mm in diameter) with a 27 m^2 surface area and a 2 m bed height. The UV treatment is provided by a 31.3 mJ/cm^2 middle pressure mercury lamp with a maximum emission spectrum of 260 nm. The sewage flow rate and sludge volume are measured by means of flow meters equipped at appropriate locations in each STP. During the sampling period, the plant effluent flow and the hydraulic retention time (HRT) and solid retention time (SRT) were 10,140 and 19,320 m^3/day, 9.1 and 12 h, and 5.8 and 9.3 d on average for STP-1 and STP-2, respectively. In both STPs, part of the secondary effluent (before disinfection) was drained as reuse water for various purposes in each plant (e.g., backwashing and flushing) (Figure 1b). Three-quarters of the water flows into the plants as plant influent.

In STP-1, twelve samples were collected every 2 h for 24 h (from 9 a.m. to 7 a.m.) from the influent

(a) Target compounds



Triclosan
Log k_{ow}: 4.76

DEET
Log k_{ow}: 2.18

Crotamiton
Log k_{ow}: 2.73

Carbamazepine
Log k_{ow}: 2.45

(b) Sampling points

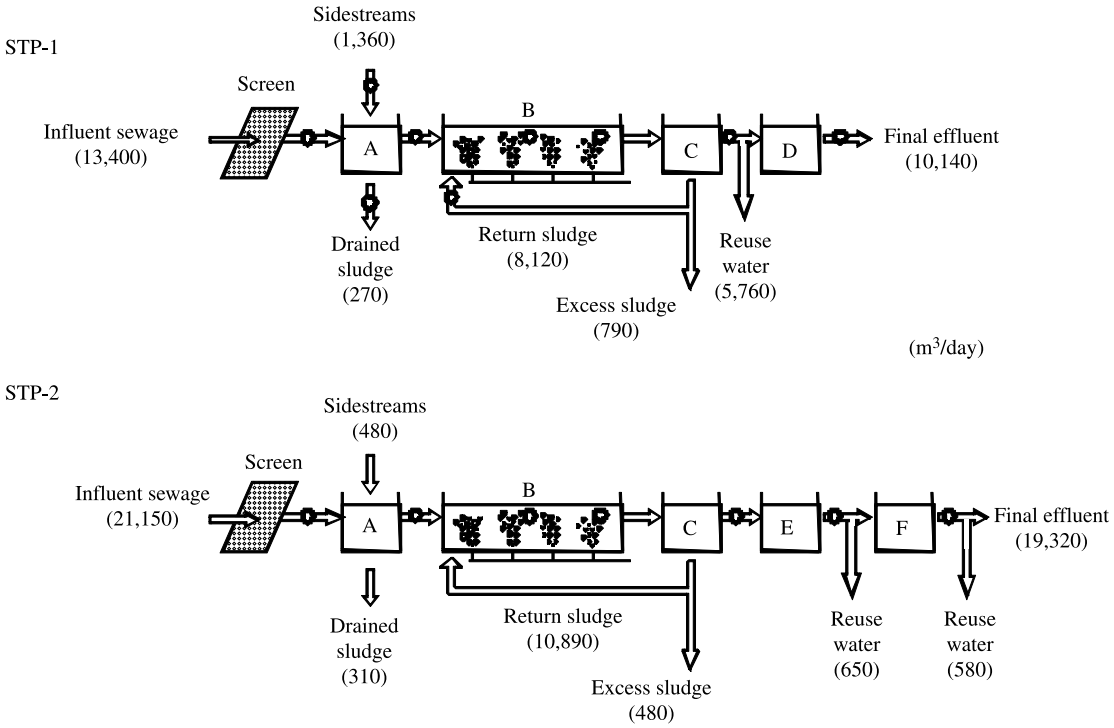


Figure 1 | Chemical structure and log Kow (PHYSPROP) of triclosan, DEET, crotamiton and carbamazepine (a) and sampling locations (open circles) in municipal sewage treatment plant (STP) (b). A: Primary sedimentation tank; B: Aeration tank; C: Final sedimentation tank; D: Chlorination tank; E: Biofiltration tank; F: Ultraviolet radiation tank. The primary settling tanks receive sidestreams from the sludge dewatering process. Numbers in parentheses indicate water/sludge flow data (m^3/day) at each location at the sampling time.

(after screening), primary and secondary effluent, final effluent, activated sludge in the aeration tank, return sludge from the final sedimentation tank, sidestreams (i.e., return flow) from the sludge dewatering process, and drained sludge from the primary sedimentation tank (Figure 1b). From each sample set, twenty-four-hour flow-proportional composite samples were prepared. In STP-2, influent, primary and secondary effluent, final effluent, activated sludge in the aeration tank and effluent from the biological filtration system were collected as the same manner. Activated sludge samples were collected from two locations in each STP, namely from the mid-distance point and from the end of the tank (Figure 1b). Although the surveys in both plants were conducted on weekdays to minimize a daily fluctuation of water, there is a possibility that errors in calculation of mass balance analysis might occur due to the surveys without taking into account the HRT.

All the samples were stored in a refrigerator or in a chest containing ice water during sampling and were then immediately transported to the laboratory. Five hundred

mL of effluent from the final sedimentation tank, biological filtration system and disinfection process, or 100 mL of other samples were filtered through glass fiber filters (GF/B, pore size: 1.0 μm, Whatman) within 24 h of collection. The filters were wrapped with clean aluminum foil and stored at −30°C until the analytes were extracted.

Analytical methods

The filtered sewage (dissolved phase) samples were concentrated by using an octadecyl silica cartridge (Sep-Pak tC18 long, Waters) within 2 days according to a previous report (Nakada *et al.* 2006*a*) with minor modifications. Prior to extraction, carbamazepine-d_{10} solution was added to the filtered samples as a surrogate for DEET, crotamiton and carbamazepine. Then, the sample was loaded onto the cartridge at 15 mL/min. The compounds retained on the cartridge were eluted with 20 mL of methanol. The eluent was evaporated just to dryness in a gentle stream of nitrogen. The residue was redissolved in hexane and then purified by a silica gel column (Sep-Pak Si, Waters; 900 mg silica weight) that had previously been washed with 20 mL of hexane. The compounds in the column were eluted successively with 5 mL of hexane/dichloromethane (75:25; v/v), 5 mL of dichloromethane, and 10 mL of dichloromethane/acetone (70:30; v/v) in turn. Triclosan was eluted in the dichloromethane fraction, while the amide PPCPs were eluted in the dichloromethane/ acetone fraction. These fractions were analyzed by gas chromatography employing a mass spectrometer after appropriate preparation (i.e., derivatization) as fully described in a previous report (Nakada *et al.* 2006*a*).

Suspended solids and sewage sludge on the filter (particulate phase) samples were freeze-dried followed by ultrasonic extraction using methanol (15 min × 2) and acetone (15 min). Prior to extraction, carbamazepine-d_{10} solution was added to the filter. The filter extracts were mingled and filtered again using a new GF/B filter. The filtrate was evaporated just to dryness in a gentle stream of nitrogen. The residue was redissolved, purified and analyzed as described above.

A procedural blank was run with the analysis sample for each survey. The limit of quantification (LOQ) for PPCP analytes was defined as three times the procedural blank value. The precision, recovery ($n = 3$) and LOQ of the method used for triclosan, DEET, crotamiton and carbamazepine in raw sewage and treated effluent are summarized in Table 1. The recovery of surrogate carbamazepine-d_{10} ranged from 65–205% with an average of 125% ($n = 33$).

To confirm the operational conditions of the surveyed sewage works, the water temperature, suspended solids, dissolved organic compounds, nitrogen species, biochemical oxygen demand (BOD) and chemical oxygen demand (COD) were measured for each composite sample, according to the standard method (Testing methods for industrial wastewater 1998) (Table 2).

RESULTS AND DISCUSSION

Analytes concentrations in dissolved and suspended phases

All analytes (i.e., triclosan, DEET, crotamiton and carbamazepine) were detected in each sample from the two

Table 1 | Recovery, reproducibility and limit of quantification (LOQ) of target compounds

	Influent					Effluent		
Sample	Dissolved phase		Particulate phase			Dissolved phase		
Compound	Recovery (%)[a]	RSD (%)[a]	Recovery (%)[a]	RSD (%)[a]	LOQ (ng/L)	Recovery (%)[a]	RSD (%)[a]	LOQ (ng/L)
Triclosan	79	6	89	1	15	86	3	3
Diethyltoluamide	107	8	69	2	20	115	11	4
Crotamiton	111	9	62	1	9	85	14	2
Carbamazepine	98	1	96	3	21	113	3	4

RSD: relative standard deviation, a: $n = 3$.

Table 2 | Concentrations of target compounds in dissolved phase (D) and particulate phase (P) of wastewater samples (ng/L) an characterization of wastewater samples analyzed in the present study*

		Triclosan		Diethyltoluamide		Crotamiton		Carbamazepine		
Plant ID	**Sample†**	**D**	**P**	**D**	**P**	**D**	**P**	**D**	**P**	
STP-1	Influent	470	2,210	1,490	63	893	< LOQ	97	< LOQ	
	Primary effluent	773	1,070	925	105	680	< LOQ	76	< LOQ	
	Activated sludge (M)	211	2,250	757	26	600	10	75	< LOQ	
	Activated sludge (E)	232	1,670	901	188	771	12	95	< LOQ	
	Secondary effluent	211	8	847	< LOQ	922	< LOQ	127	< LOQ	
	Final effluent	243	19	935	23	1,020	< LOQ	132	< LOQ	
	Sidestreams	232	921	1,150	20	924	< LOQ	119	< LOQ	
	Excess sludge	396	19,100	663	213	530	58	97	< LOQ	
	Return sludge	215	3,870	800	48	669	61	98	< LOQ	
STP-2	Influent	2,060	9,830	1,030	33	668	< LOQ	113	< LOQ	
	Primary effluent	2,170	4,920	1,390	27	944	< LOQ	99	< LOQ	
	Activated sludge (M)	351	60,400	1,190	78	882	56	94	< LOQ	
	Activated sludge (E)	243	46,700	1,140	77	1,100	63	139	< LOQ	
	Secondary effluent	396	193	413	6	1,420	< LOQ	93	< LOQ	
	Biofiltration effluent	401	79	344	5	1,100	< LOQ	92	< LOQ	
	Final effluent	233	36	291	5	899	< LOQ	110	< LOQ	
Plant ID	Sample†	TW‡ (°C)	SS (mg/L)	VSS (mg/L)	DOC (mg/L)	NH_4-N (mg/L)	NO_2-N (mg/L)	NO_3-N (mg/L)	BOD (mg/L)	COD_{Mn} (mg/L)
STP-1	Influent	26.7	160	138	34.1	18.7	0.00	n.d.	169	112
	Primary effluent	–	68	59	31.8	15.7	0.18	n.d.	114	62
	Activated sludge (M)	–	1,160	965	8.33	10.8	0.38	0.42	–	–
	Activated sludge (E)	–	1,260	1030	7.96	4.43	0.05	3.23	–	–
	Secondary effluent	–	3.0	2.0	7.07	4.72	0.33	5.88	9.0	11
	Final effluent	–	3.0	2.0	7.38	7.53	0.83	2.69	0.9	10
	Sidestreams	–	200	172	31.6	11.6	0.89	0.86	–	–
	Excess sludge	–	3,980	3480	91.8	23.9	0.03	n.d.	–	–
	Return sludge	–	3,280	2700	8.58	6.37	0.02	0.05	–	–
STP-2	Influent	26.5	176	–	–	–	–	–	249	105
	Primary effluent	–	84.4	–	–	–	–	–	186	67
	Activated sludge	–	1,510	–	–	–	–	–	–	–
	Secondary effluent	–	1.3	–	–	–	–	–	6.1	14.5
	Final effluent	–	0.3	–	–	–	–	–	1.8	9

* < LOQ: below the limit of quantification (Table 1); n.d.: not detected; –: not measured.

†Activated sludge samples were collected from two points in each STP (see text).

‡T_W: sample temperature.

surveyed STPs. All compounds except for triclosan were dominantly found in the dissolved phase and were detected at the same levels in both STPs (Table 2).

Triclosan predominated in the particulate phase, especially in sewage samples before final sedimentation and in sludge samples. The total triclosan concentration (i.e., dissolved and particulate phases) in the plant influent was 2.7 μg/L for STP-1 and 11.9 μg/L for STP-2, while the concentrations in the effluent were 0.26 and 0.27 μg/L, respectively. These values are comparable to those reported from the USA with concentrations of 3.8–16.6 and 0.2–2.7 μg/L (McAvoy *et al.* 2002), and from Switzerland with concentrations of 0.58–1.30 and 0.07–0.65 μg/L (Lindström *et al.* 2002), respectively. Limited information is available about the aquatic toxicity of triclosan; however, the effective concentration on algae has been estimated at several μg/L (Orvos *et al.* 2002), which is only one order of magnitude higher than the triclosan concentration in the effluent observed in this study. Additionally, Tatarazako *et al.* (2004) reported the acute toxicity of triclosan to aquatic organisms such as bacteria, crustacea and fish with an inhibiting concentration ranging from 0.07 to 0.29 mg/L as a reduction of 25% in survival or reproduction. These values that induced acute toxicity were significantly higher than the triclosan concentration in the effluent.

Among the analytes, DEET showed the highest concentration, over 1 μg/L, in the dissolved influent phase measured in this study. The high concentration is comparable to the reported in a study undertaken at five STPs in Tokyo (Nakada *et al.* 2006*a*), in which DEET was detected at a higher concentration in summer (1,045 ng/L on average ($n = 4$)) than in other seasons (267 ng/L for spring ($n = 4$), 695 ng/L for autumn ($n = 4$) and 35 ng/L for winter ($n = 4$)). DEET is an insect repellent and is therefore more commonly used in warmer seasons when harmful insects are active, resulting in the high DEET concentration in the influent in Japan. The DEET concentration in sewage was two orders of magnitude higher than that observed in Korea (Kim *et al.* 2007), although the sampling seasons were unclear.

Crotamiton was detected in the dissolved phase of all samples with concentrations ranging from 530 to 1,420 ng/L, while it was detected at a very low level in the particulate phase (< LOQ to 63 ng/L). The concentration in the dissolved phase is comparable to that of a previous survey undertaken in Tokyo, Japan (Nakada *et al.* 2006*a*). Although information on the crotamiton concentration in sewage and the aquatic environment has not been reported for other countries despite its worldwide usage, its stable nature and high concentration even after treatment (up to 1 μg/L) results in frequent detection in the aquatic environment in Japan (Nakada *et al.* 2008).

Carbamazepine was detected only in the dissolved phase of the influent, with concentrations of 97 ng/L for STP-1 and 113 ng/L for STP-2, and almost the same concentration was found in the final effluent (110–132 ng/L). The observed carbamazepine concentration in the influent is comparable to or several times lower than the values reported from Canada (0.7 μg/L: median) by Metcalfe *et al.* (2003), Finland (285 ng/L) by Vieno *et al.* (2007) and Germany (1.78 μg/L on average), which may be due to the usage amount in each country (Nakada *et al.* 2006*a*). The concentration in the effluent in this study falls into the range reported from Canada, Korea (Kim *et al.* 2007), Finland, and other European countries (Paxéus 2004) with the exception of Germany (Ternes 1998). Additionally, the observed carbamazepine concentration in the effluent was sufficiently lower than the acute (>13,800μg/L) and chronic toxicity (>25 μg/L) levels of carbamazepine with respect to aquatic organisms (Ferrari *et al.* 2003).

To facilitate estimations of the fates of PPCPs during the treatment process and in the aquatic environment, solid-water distribution coefficients (K_d) were calculated for the selected PPCPs in primary, activated, and return sludge (Table 3). The log K_d values for DEET and crotamiton in the sludge samples ranges from 1.3 to 1.9, implying that removal by sorption is not efficient for these compounds during the treatment process and in the aquatic environment. On the other hand, the value for triclosan is relatively high, ranging from 3.7 to 5.1, indicating that triclosan would be removed from the dissolved phase into sludge. A similar value (4.3) was determined for deactivated sludge (Reiss *et al.* 2002). The K_d values for triclosan in activated sludge from STP-2 was one order of magnitude higher than that from STP-1 (Table 3). This could be caused by a difference between the SRTs of STP-2 (SRT: 9.3 d) and STP-1 (5.8 d), although the data are limited. Further research is needed (e.g. batch experiments).

Table 3 | Log K_d values for the selected PPCPs in primary, activated and return sludges

	STP-1			STP-2
Compounds	**Primary sludge**	**Activated sludge**	**Return sludge**	**Activated sludge**
Triclosan	4.1	3.9 ± 0.1	3.7	5.1 ± 0.03
DEET	1.9	1.8 ± 0.5	1.3	1.6 ± 0.01
Crotamiton	1.4	1.1 ± 0.05	1.4	1.6 ± 0.03
Carbamazepine	n.a.	n.a.	n.a.	n.a.

n.a.: not available because the concentration in the suspended phase was below the limit of quantification.

PPCP removal ratio

The removal ratio of the analytes during treatment was calculated by comparing the influent and effluent concentrations. The respective removal efficiencies in STP-1 and STP-2 were as follows: 90% and 98% for triclosan, 38% and 72% for DEET, −14% and −35% for crotamiton, and −36% and 3% for carbamazepine. The almost complete removal of triclosan, limited removal of DEET, and the insignificant or negative removal of crotamiton and carbamazepine correspond well with the reported values. The excellent removal of triclosan may be due to sorption to solids as discussed below, and is comparable to that reported from the USA (Federle *et al.* 2002; McAvoy *et al.* 2002). The removal of DEET was higher in STP-2 than in STP-1. Metcalfe *et al.* (2003) reported that an HRT of over 12 h was a key factor in PPCP removal (e.g., ibuprofen and naproxen) and that the SRT was unrelated to removal. The same authors also reported that the removal of carbamazepine was not dependent on HRT. The HRT in STP-1 (9.1 h) and STP-2 (12 h) could account for the difference in DEET removal. On the other hand, both STPs showed excellent removal rates for suspended solids of 98 - 100%, with BOD and COD rates of 99% and 91 - 92%, respectively (Table 2).

Mass balance analyses

The mass balance at the different sampling points in STP-1 (Figure 2a) and STP-2 (Figure 2b) was calculated based on the detected concentration of each analyte and the daily water/sludge flow monitored at several points (Figure 1b). In STP-2, the mass balance in return sludge was not calculated because the analytes in the return sludge were not measured. For triclosan in STP-1, a total of 36 g/d (6 g/d in the dissolved phase and 30 g/d in the suspended phase) flowed into the primary sedimentation tank, where part of the mass balance was reduced, and 26 g/d of triclosan was mixed with the return sludge containing 30 g/d of triclosan to be treated in an aeration tank. After aeration and final sedimentation only 3.5 g/d remained in the secondary effluent. Chlorine disinfection did not achieve significant removal, and 2.6 g/d of triclosan was discharged into the aquatic environment. The slight removal of triclosan in the aeration tank was observed (~23%), which could be explained in terms of biotransformation under aerobic conditions (McAvoy *et al.* 2002). The total mass balance of the 37.6 g/d inflow and 10.3 g/d outflow indicates some degree of triclosan biodegradation. A much greater triclosan load in STP-2 than in STP-1, as well as a longer SRT (9.3 d) in STP-2, could have caused the higher accumulation ratio of triclosan in the activated sludge. The agricultural use of excess sludge that could contain a high concentration of triclosan should be considered in terms of its toxicity as regards soil biota, although groundwater pollution by triclosan in the sludge may not occur due to its hydrophobicity (log Kow: 4.76 (PHYSPROP)). The main removal of DEET occurred between the aeration tank and the final sedimentation tank (18% in STP-1 and 70% in STP-2). For crotamiton and carbamazepine, no significant removal was observed in the STPs. The results clearly demonstrate that these water-soluble compounds (i.e., log Kow range 2.18–2.73: (PHYSPROP) were not susceptible to removal in STPs employing conventional sewage treatment processes.

The treatment plants surveyed in this study deal mainly with domestic sewage discharged by populations of 30,500 and 70,600 living in individual areas served by STP-1 and STP-2, respectively. There is less than a twofold difference

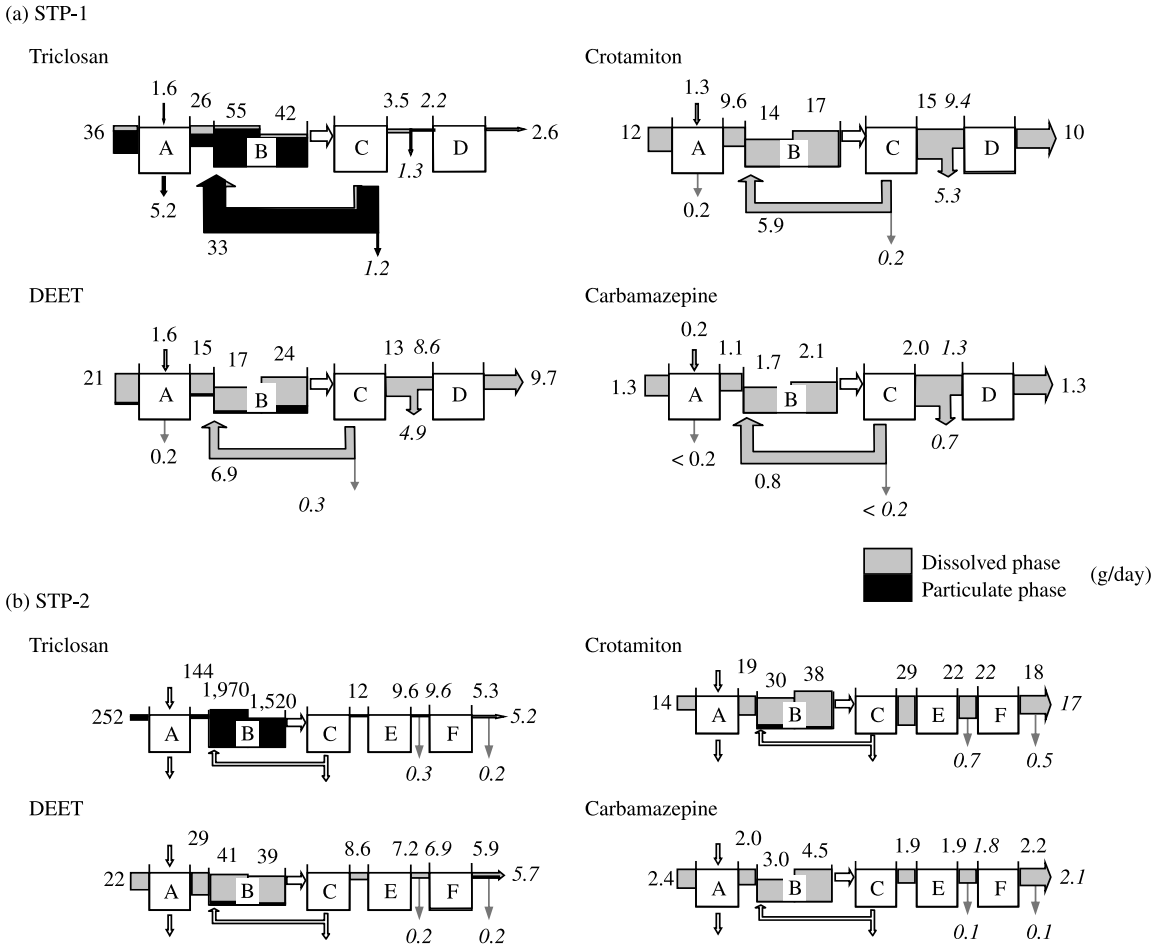


Figure 2 | Mass balance of triclosan, DEET, crotamiton, and carbamazepine during physicochemical treatment (A and C), biological treatment (B and E), and the chlorination process (D) in STP-1 (a) and STP-2 (b). A to D are shown in Figure 1. Values in italics were estimated in proportion to water/sludge flow.

between the mass inflows of DEET, crotamiton and carbamazepine with respect to the two STPs, while that of triclosan is seven times higher in STP-2 than in STP-1. Triclosan is an antibacterial and antifungal agent that has a wide variety of uses and a wide range. STP-2 treats approximately 500 m^3/day from two hospitals in the serviced area, while STP-1 deals with approximately 90 m^3/day from one hospital. This difference may explain the high load of PPCPs in STP-2 (i.e., triclosan in this study and four anti-inflammatory drugs (Nakada *et al.* 2005)).

CONCLUSIONS

Detailed surveys were conducted at two STPs to clarify the fates of triclosan, DEET, crotamiton and carbamazepine, which are reportedly persistent in water treatment facilities and the aquatic environment. The results illustrate their distribution in dissolved and suspended phases and their fates in the STPs. Triclosan exhibited its highest concentration in raw sewage and excellent removal (~90%) among analytes, while the accumulation of triclosan was observed in the sewage sludge. DEET, crotamiton and carbamazepine passed though the STPs without significant removal or biodegradation, and could also persist in the environment. This is the first report of the fates of DEET and crotamiton in STPs, although these fates might include uncertainties caused by small number of survey and samplings without consideration of HRT. An investigation of their behavior in sediment in an aquatic environment is needed. The findings of the present study suggest that

persistence and biodegradability during sewage treatment and in the environment together with the possible toxicity of ingredients should be considered with respect to the design, sale, prescription and use of these drugs.

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