Supporting Information

Source Apportionment of Organotin Pollution in Different Types of Drinking Water from Megacity Communities Using Multiple Receptor Models: A Case Study in Shanghai, China

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S1. Reagents, standards and experimental materials of organotin compounds (OTCs).

Monomethyltin (MMT, 99.9%) was purchased from HAYASHI PURE CHEMICAL IND, LTD (Osaka, Japan). Dimethyltin (DMT, 99.9%), Monobutyltin (MBT, 95%), Dibutyltin (DBT, 97.2%), Tributyltin (TBT, 95%), Tetrabutyltin (TeBT, 96%), Triphenyltin (TPhT, 96%), and Diphenyltin (DPhT, 97%) were all obtained from Dr. Ehrenstorfer (Augsburg, Germany). Specific information on these OTCs is presented in Table S2. Methanol (CNW Technologies GmbH, Germany, chromatographic grade, 99.8%), Hexane (CNW Technologies GmbH, Germany, chromatographic grade, 95%), Sodium tetraethylborate (NaBEt4, CNW Technologies GmbH, Germany, 98%), acetic acid (Sinopharm Chemical Reagent Co., Ltd., China, analytical grade), sodium acetate (Sinopharm Chemical Reagent Co., Ltd., China, analytical grade), and sodium hydroxide (Sinopharm Chemical Reagent Co., Ltd., China, analytical grade) were used. Milli - Q (18.2 MΩ·cm) water was used for experiments (Beijing Purkinje General Instrument Co., Ltd., China).

Preparation of Sodium Tetraethylborate Solution (1% w/v): Add 2 g of KOH to 97 mL of deionized water. Place this solution in a refrigerator for 30 - 90 minutes until ice crystals form. Subsequently, add 1 g of $NaBEt_4$ to the KOH solution and shake it thoroughly to ensure homogeneity. Then, dispense equal - volume aliquots of the solution into clean Teflon vials (2 mL) and keep frozen.

Preparation of Acetic Acid/Sodium Acetate Buffer Solution (1 mol/L): Dissolve 82 g of sodium acetate in 1 L of deionized water. Then, slowly add acetic acid to the solution until the pH is stabilized at 4.5.

Preparation of Elution Solution: Add 10.79 g of solid ammonium chloride to 75 mL of deionized water. Make up the volume to 250 mL with methanol and mix well. At this stage, the concentration of ammonium chloride in the solution is 0.8 mol/L, and the solvent is a mixture of methanol/water = 7/3. Take the prepared solution and add glacial acetic acid to it, such that the glacial acetic acid / ammonium chloride solution is 10/90.

S2. Instrumental methods of organotin compounds (OTCs).

An Agilent GC-MS (7890-5975) with a DB-5MS column (30m × 0.25 mm × 0.25 μm) was used for analysis. The injector temperature was 280 °C, the column temperature was 30 °C, the injection volume was 1 μL under the non-split mode with a constant flow rate of 1 mL/min. The optimal oven temperature program was starting at 30 °C for 2 min, rising at 10 °C min^{-1} to 60 °C and then rising at 20 °C min^{-1} to 110 °C, finally to 280 °C at 40 °C min^{-1} for 3 min. The EI (70 V) ion source was used at 260 °C, with a solvent delay of 4 min. The mass spectrometry interface temperature was 300 °C, and the carrier gas was high-purity helium (99.999%).

S3. Recovery experiment of organotin compounds (OTCs).

Quality control and quality assurance were conducted using the spiked recoveries (n = 3) after adding standard substances at low (10 ng Sn/L), medium (70 ng Sn/L), and high concentrations (100 ng Sn/L) to blank water samples. The recovery results are presented in Table S4. For blank water samples, the spiked recoveries at low, medium, and high concentrations were all within the range of 70% - 130%, and the relative standard deviations (RSDs) were all less than 10%, meeting the experimental requirements.

Meanwhile, considering the potential matrix differences between actual water samples and blank water samples, which may lead to matrix interference during practical applications, 0.5 mL of an organotin mixed standard substance with a known concentration (100 μg Sn/L) was added to three groups of tap water samples from Tongji University to calculate the matrix recoveries. The measured concentrations of each substance with matrix spiking (n = 3) were 79 ± 2 μg DMT/L, 74 ± 4 μg MMT/L, 88 ± 2 μg MBT/L, 96 ± 3 μg DBT/L, 97 ± 7 μg TBT/L, 106 ± 4 μg DPhT/L, and 104 ± 2 μg TPhT/L, respectively. All these values fell within the reference concentration range, and the recovery rates were basically consistent with those of the blank water samples (Table S5). This indicates that this analytical and detection method can accurately quantify organotin substances in actual water samples.

Table S1. Details of tap water (TW), water vending machine (WVM), and water boiling machine (WBM HP & HT) sampling information.

Site No.	Sample	Site[a]		Site No.	Sample	Site[a]	Brand[b]
FM-TW-ek	TW	FM	Hospital	FM-WVM-EJ	WVM	FM	EJ
FM-TW-hl	TW	FM	School	FM-WVM-GH-1	WVM	FM	GH
FM-TW-hs	TW	FM	School	FM-WVM-GH-2	WVM	FM	GH
FM-TW-sj(m)	TW	FM	School	FM-WVM-HY	WVM	FM	HY
FM-TW-ss	TW	FM	School	FM-WVM-QJ	WVM	FM	QJ
FM-WBM HP-HL	WBM HP	FM	School	FM-WVM-QM-1	WVM	FM	QM
FM-WBM HP-HS	WBM HP	FM	School	FM-WVM-QM-2	WVM	FM	QM
FM-WBM HP-SJ(M)	WBM HP	FM	School	FM-WVM-YQ-1	WVM	FM	YQ
FM-WBM HP-SS	WBM HP	FM	School	FM-WVM-YQ-2	WVM	FM	YQ
FM-WBM HT-EK	WBM HT	FM	Hospital	N-WVM-EJ-138	WVM	N	EJ
N-TW-ch	TW	N	Hospital	N-WVM-EJ-3338	WVM	N	EJ
N-TW-xh	TW	N	Hospital	N-WVM-HY-169	WVM	N	HY
N-WBM HP-SC	WBM HP	N	School	N-WVM-HY-395	WVM	N	HY
N-WBM HT-CH	WBM HT	N	Hospital	N-WVM-QJ	WVM	N	QJ
N-WBM HT-FD(H)	WBM HT	N	School	N-WVM-QM	WVM	N	QM
N-WBM HT-GD	WBM HT	N	Hospital	N-WVM-YQ-357	WVM	N	YQ
N-WBM HT-XH	WBM HT	N	Hospital	N-WVM-YQ-90	WVM	N	YQ
PD-TW-df	TW	PD	Hospital	N-WVM-YSK	WVM	N	YSK
PD-TW-rj	TW	PD	Hospital	PD-WVM-EJ-3288	WVM	PD	EJ
PD-TW-sg	TW	PD	Hospital	PD-WVM-GH-1155	WVM	PD	GH
PD-TW-sy	TW	PD	School	PD-WVM-GH-1166	WVM	PD	GH
PD-WBM HP-RJ	WBM HP	PD	Hospital	PD-WVM-GH-2500	WVM	PD	GH
PD-WBM HP-SH	WBM HP	PD	School	PD-WVM-HY-573	WVM	PD	HY
PD-WBM HP-SK	WBM HP	PD	School	PD-WVM-QJ	WVM	PD	QJ
PD-WBM HT-DF	WBM HT	PD	Hospital	PD-WVM-QM	WVM	PD	QM
S-TW-ly	TW	S	Hospital	PD-WVM-YQ-2851	WVM	PD	YQ
S-TW-xk	TW	S	Hospital	PD-WVM-YQ-3905	WVM	PD	YQ
S-WBM HP-SJ(X)	WBM HP	S	School	S-WVM-EJ-111	WVM	S	EJ
S-WBM HT-BW	WBM HT	S	Hospital	S-WVM-EJ-937	WVM	S	EJ
S-WBM HT-CZ	WBM HT	S	Hospital	S-WVM-HY-368	WVM	S	HY
S-WBM HT-FD(F)	WBM HP	S	School	S-WVM-HY-487	WVM	S	HY
S-WBM HT-LY	WBM HT	S	Hospital	S-WVM-JB	WVM	S	JB
S-WBM HT-XK	WBM HT	S	Hospital	S-WVM-QJ-240	WVM	S	QJ
				S-WVM-QJ-550	WVM	S	QJ
				S-WVM-QM-1501	WVM	S	QM
				S-WVM-QM-333	WVM	S	QM
				S-WVM-YQ-1023	WVM	S	YQ

Site[a]: FM: Fengxian and Minhang Districts; N: North Water supply district; S: South Water supply district; PD: Pudong District. Brand[b]: EJ: Qing Lang; GH: Gang Hui; HY: Han Ying; JB; Jing Bo; QJ: Qi Jia; QM: Qin Mia; YQ: Yi Quan; YSK: Yi Sikai.

Table S2. Detailed information of OTCs.

Compounds	CAS number	Chemical Formula	Density (g/cm^3)	Solubility (mg/cm^3)	Melting Point (°C)	Boiling Point (°C)
MMT	993-16-8	$MeSnCl_3$	0.99	n.i.[a]	47	173
DMT	753-73-1	Me_2SnCl_2	1.40	20000[b]	106-108	188-190[d]
MBT	1118-46-3	$BuSnCl_3$	1.69	n.i.[a]	-63	93/1.3 kPa
DBT	818-08-6	Bu_2SnCl_2	1.36	4-50[b]; 92[c]	39-41	153/1.3 kPa
TBT	1461-22-9	Bu_3SnCl	1.21	50[b]; 5-17[c]	-16	172/3.3 kPa
DPhT	1135-99-5	Ph_2SnCl_2	n.i.[a]	n.i.[a]	41-43	333-337[d]
TPhT	639-58-7	Ph_3SnCl	n.i.[a]	n.i.[a]	103-108	240[d]

Notes: All data were obtained from PubChem (PubChem) and the CompTox Chemicals Dashboard (Chemicals Dashboard).

n.i.[a]: no information.

[b] Solubility in seawater.

[c] Solubility in distilled water.

[d] at standard atmospheric pressure.

Table S3. Properties and quantitative parameters of organotin standard materials.

Compounds	Mass (g/mol)	Retention Time (min)	Quantification ions m/z	Quantitative ions m/z
MMT	220.96	8.78	151, 179, 149	151
DMT	206.93	12.65	165, 135, 193	165
MBT	263.05	20.38	179, 177, 151	179
DBT	291.11	23.63	151, 149, 179	151
TBT	319.17	26.16	151, 207, 205	151
DPhT	331.07	31.59	303, 301, 197	303
TPhT	377.13	36.66	351, 349, 347	351

Table S4. Limit of detection (LOD), recoveries, relative standard deviations (RSD) and correlation coefficients of standard curves (R^2) of OTCs performed with spiked blank water.

Compounds	LOD (ng Sn/L)	10 ng Sn /L		70 ng Sn /L		100 ng Sn /L		R^2
		Recoveries	RSD	Recoveries	RSD	Recoveries	RSD	
DMT	2.23	71.6%	7.6%	75.6%	5.6%	79.4%	2.4%	0.993
MMT	2.40	70.2%	9.4%	73.8%	4.0%	77.5%	5.0%	0.996
MBT	3.23	106.0%	8.8%	80.8%	5.1%	83.8%	2.1%	0.996
DBT	2.56	79.2%	3.4%	92.2%	6.6%	96.7%	4.3%	0.996
TBT	3.75	73.1%	6.0%	95.9%	2.8%	103.2%	4.6%	0.994
DPhT	3.68	125.0%	1.2%	103.7%	3.8%	112.7%	5.8%	0.995
TPhT	1.51	118.8%	4.7%	112.7%	1.7%	101.8%	6.1%	0.998

Table S5. The standard recovery rate of tap water samples from Tongji University.

Compounds	Concentration (μg Sn /L)					Recoveries
	Group 1	Group 2	Group 3	Average	Standard deviation	
DMT	104	105	107	105	2	79%
MMT	78	74	70	74	4	74%
MBT	98	99	102	100	2	88%
DBT	93	95	99	96	3	96%
TBT	91	96	105	97	7	97%
DPhT	105	103	110	106	4	106%
TPhT	102	106	103	104	2	104%

Table S6. The toxicity data of OTCs.

Compounds	CAS number	Toxicity Data						TDI/(μg/kg·d)
		Organism	Route	Test Type	Dose	Test Type	Dose	
MMT	993-16-8	rat	oral	LD50	1370 mg/kg	LC50	600000 mg/m^3/1hr	/
DMT	753-73-1	rat	oral	LD50	73900 μg/kg	LC50	115 mg/m^3/4hr	/
MBT	1118-46-3	rat	oral	LD50	2140 mg/kg	LD50	2140 mg/kg	5.42
DBT	818-08-6	rat	oral	LD50	44900 μg/kg	/	/	1.75
TBT	1461-22-9	rat	oral	LD50	129 mg/kg	LD50	60 mg/kg	0.25
DPhT	1135-99-5	rat	oral	LDLo	410 mg/kg	/	/	3.50
TPhT	639-58-7	rat	oral	LD50	135 mg/kg	LD50	18 mg/kg	0.5

Notes: All data were obtained from PubChem (PubChem); TDI: tolerated daily intake

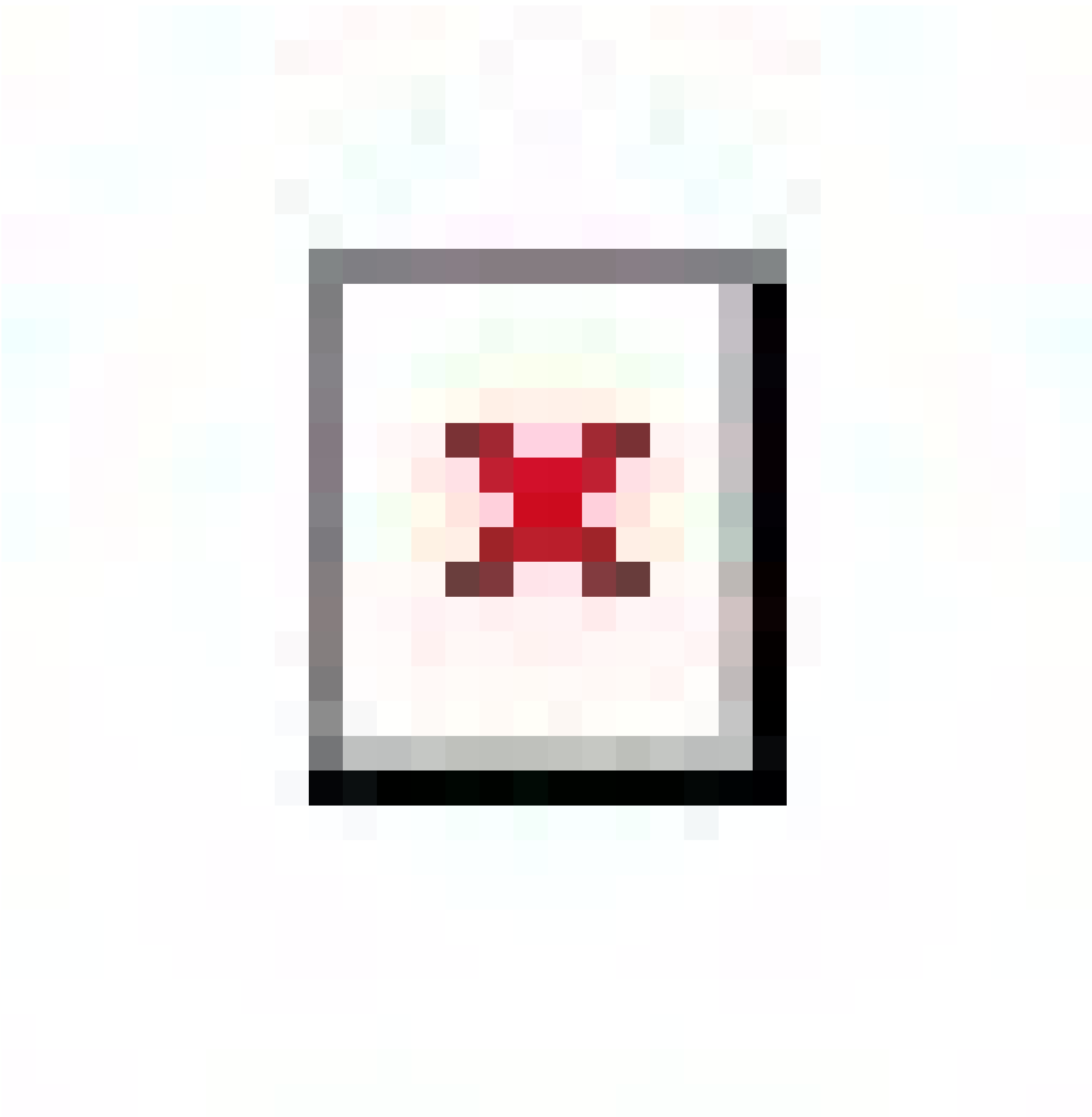

Fig. S1. Concentrations of OTCs in different drinking water samples.

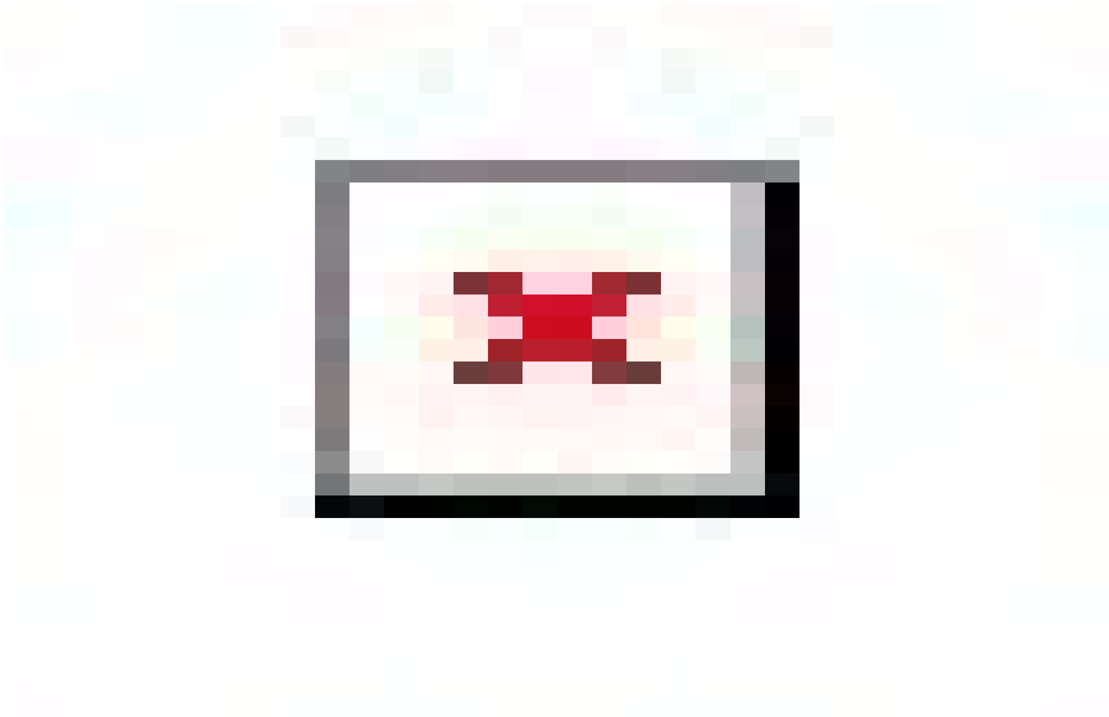

Fig. S2. Compositions of OTCs in different drinking water samples (excluding non-detected samples).

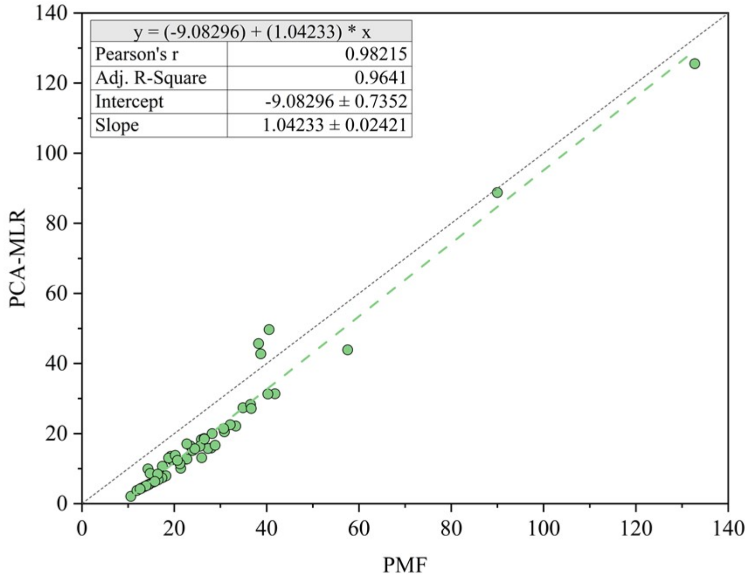

Fig. S3. Plots of fit for the predicted concentrations of PCA-MLR and PMF.

References

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