

Supporting Information

Human exposure to phthalic acid esters via ingestion of municipal drinking water from automatic water purifiers: levels, sources, and risks

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S1. Experimental method of purification materials

Approximately 2 g filter material including high-precision micro-processing filter (HPMF) and granular activated carbon (GAC) spiked with surrogate standards (i.e., Butyl benzyl phthalate (BBP)-D4, DEHP-D4 250 ng, and then spiked with extracted by Soxhlet extraction with 150 mL hexane (HEX) lasting for 12 hours. After that, the extraction was rotary evaporated to 1mL, and then loading in the Florial cartridge. Briefly, the cartridges were preconditioned with 25 mL hex: acetone (v: v = 1:1). Then, samples were evaporated to 1 mL under a gentle nitrogen flow. The residue was dissolved with n-hexane (Hex) for gas chromatography-mass spectrometry (GC-MS) analysis.

Table S1 Compound dependent operation parameters of phthalate esters (PAEs).

Compounds	Retention Time (min)	Quantification ions	Quantitative ions	LOD (ng/L)	LOQ (ng/L)
DBP	7.87	149, 150	149	1.13	1.14
DIBP	7.24	149, 150	149	1.13	1.14
DEP	5.65	149, 177	149	0.14	0.16
DMP	4.82	163, 77	163	0.18	0.19
DEHP	14.21	167, 167	167	0.47	0.52
DMEP	8.12	59, 58	59	0.70	1.30
DNP	21.05	149, 293	149	0.02	0.05
DHP	14.22	41, 149	41	7.38	8.13
DHXP	11.52	149, 150	149	0.86	0.87
DINP	18.97	149, 207	149	1.86	2.04
DPP	9.43	149, 153	149	0.68	0.70
BMPP	8.73	84, 149	85	1.83	1.99
DnOP	17.46	149, 279	85	0.93	0.95
DCHP	14.02	149, 167	149	0.44	0.46
DPhP	14.40	225, 77	225	0.72	0.74
DBEP	13.23	149, 57	149	5.76	5.88
DIPP	8.71	149, 71	71	2.52	2.65
DAP	6.53	149, 189	149	4.83	4.87
BBP	11.68	149, 91	149	2.11	2.15
DIPrP	6.01	149, 150	149	1.18	1.19
DPrP	6.68	149, 150	149	1.02	1.03
DEEP	9.04	72, 149	72	2.71	2.79
DEHP-D4	14.18	153, 149	153	-	-
BBP-D4	11.65	153, 149	153	-	-

LOD: limit of detection; LOQ: limit of quantification.

-not

detected.

Table S2 Details of tap water (TW), water vending machine (WVM), and water boiling machine (WBM) sampling information.

Site No.	Site	Time	Sample	Site No.	Site	Time	Sample
PD-WVM-GH-1	PD	09.21	WVM	N-WBM-1	N	06.09	WBM
PD-WVM-GH-2	PD	09.21	WVM	N-WBM-2	N	06.09	WBM
PD-WVM-GH-3	PD	09.21	WVM	N-WBM-3	N	06.09	WBM
FM-WVM-GH-4	FM	10.28	WVM	S-WBM-1	S	09.08	WBM
S-WVM-JB-1	S	09.08	WVM	S-WBM-2	S	09.08	WBM
S-WVM-JB-2	S	09.08	WVM	S-WBM-3	S	09.08	WBM
N-WVM-YSK-1	N	10.20	WVM	S-WBM-4	S	09.08	WBM
N-WVM-YSK-2	N	10.20	WVM	S-WBM-5	S	09.08	WBM
N-WVM-YSK-3	N	06.05	WVM	S-WBM-6	S	09.08	WBM
N-WVM-QM-1	N	06.05	WVM	PD-WBM-5	PD	09.21	WBM
PD-WVM-QM-2	PD	09.21	WVM	N-WBM-5	N	09.17	WBM
S-WVM-QM-3	S	09.08	WVM	PD-WBM-1	PD	09.21	WBM
S-WVM-QM-4	S	09.08	WVM	PD-WBM-2	PD	09.21	WBM
FM-WVM-QM-1	FM	10.28	WVM	PD-WBM-3	PD	09.21	WBM
FM-WVM-QM-2	FM	10.28	WVM	PD-WBM-4	PD	09.21	WBM
N-WVM-EJ-1	N	06.05	WVM	N-WBM-6	N	10.20	WBM
N-WVM-EJ-2	N	06.05	WVM	FM-WBM-1	FM	10.28	WBM
PD-WVM-EJ-3	PD	09.21	WVM	FM-WBM-2	FM	10.28	WBM
S-WVM-EJ-4	S	09.08	WVM	FM-WBM-3	FM	10.28	WBM
S-WVM-EJ-5	S	09.08	WVM	FM-WBM-4	FM	10.28	WBM
FM-WVM-EJ-6	FM	10.28	WVM	FM-WBM-5	FM	11.03	WBM
FM-WVM-EJ-7	FM	10.28	WVM	N-TW-1	N	06.05	TW
N-WVM-CG-1	N	06.05	WVM	N-TW-2	N	06.09	TW
FM-WVM-CG-5	FM	10.28	WVM	N-TW-3	N	06.09	TW
PD-WVM-CG-2	PD	09.21	WVM	N-TW-4	N	06.09	TW
PD-WVM-CG-3	PD	09.21	WVM	N-TW-5	N	06.09	TW
S-WVM-CG-4	S	09.08	WVM	S-TW-1	S	09.08	TW
FM-WVM-CG-6	FM	10.28	WVM	S-TW-2	S	09.08	TW
N-WVM-AM-1	N	06.05	WVM	PD-TW-1	PD	09.21	TW
PD-WVM-AM-2	PD	09.21	WVM	PD-TW-2	PD	09.21	TW
S-WVM-AM-3	S	09.08	WVM	PD-TW-3	PD	09.21	TW
S-WVM-AM-4	S	09.08	WVM	PD-TW-4	PD	09.21	TW
FM-WVM-AM-5	FM	10.28	WVM	FM-TW-1	FM	10.28	TW
N-WVM-HY-1	N	06.05	WVM	FM-TW-2	FM	10.28	TW
N-WVM-HY-2	N	06.05	WVM	FM-TW-3	FM	10.28	TW
PD-WVM-HY-3	PD	09.21	WVM	FM-TW-4	FM	10.28	TW
S-WVM-HY-4	S	09.08	WVM				
S-WVM-HY-5	S	09.08	WVM				
FM-WVM-HY-6	FM	10.28	WVM				

Table S3 Detection frequency of 22 phthalates in TW, WVM, and WBM samples

DF (%)	TW (n=15)	WBM (n=21)	WVM (n=39)
DBP	87	95	100
DIBP	80	95	100
DEP	80	80	77
DMP	33	80	74
DEHP	40	75	74
DMEP	47	50	74
DHP	40	55	77
DNP	20	35	49
DHXP	0	10	13
DINP	7	0	8
DPP	0	0	8
BMPP	10	50	5
DnOP	0	10	3
DCHP	0	0	0
DPhP	20	0	8
DBEP	0	15	3
DIPP	7	0	0
DAP	0	0	0
BBP	0	0	0
DIPrP	0	0	0
DPrP	0	0	0
DEEP	87	0	0

Table S4 Detailed information of 22 phthalates

Compounds	CAS number	Molecular Formula ^a	Mass ^b (g/mol)	log K _{ow} ^b	Vapor Pressure ^c	Water Solution ^c
DBP	84-74-2	C ₆ H ₂₂ O ₄	278	4.50	2.01E-05	4.68E-05
DIBP	84-69-5	C ₆ H ₂₂ O ₄	278	4.11	4.76E-05	2.23E-05
DEP	84-66-2	C ₁₂ H ₁₄ O ₄	222	2.47	2.10E-03	4.86E-03
DMP	131-11-3	C ₁₀ H ₁₀ O ₄	194	1.60	3.08E-03	2.10E-02
DEHP	117-81-7	C ₂₄ H ₈ O ₄ ^a	390	7.60	1.42E-07	1.08E-07
DMEP	117-82-8	C ₁₄ H ₁₈ O ₆	282	1.11	2.28E-04	3.00E-02
DNP	84-76-4	C ₂₆ H ₄₂ O ₄	418	9.52	5.14E-07	1.73E-05
DHP	3648-21-3	C ₂₂ H ₃₄ O ₄	362	7.56	2.07E-06	5.10E-06
DHXP	84-75-3	C ₂₀ H ₃₀ O ₄	334	6.82	1.40E-05	1.49E-07
DINP	28553-12-0	C ₂₆ H ₄₂ O ₄	418	9.37	5.40E-07	4.78E-07
DPP	131-18-0	C ₁₈ H ₂₆ O ₄	306	5.62	1.50E-05	2.60E-06
BMPP	84-63-9	C ₂₀ H ₃₀ O ₄	334	5.76	3.46E-04	3.50E-05
DnOP	117-84-0	C ₂₄ H ₃₈ O ₄	390	8.10	1.00E-07	5.64E-08
DCHP	84-61-7	C ₂₀ H ₂₆ O ₄	330	6.20	8.69E-07	1.21E-05
DPhP	84-62-8	C ₂₀ H ₁₄ O ₄	318	4.18	1.61E-07	2.57E-07
DBEP	117-83-9	C ₂₀ H ₃₀ O ₆	366	4.06	2.17E-03	8.19E-04
DIPP	605-50-5	C ₁₈ H ₂₆ O ₄	306	5.17	1.27E-04	2.20E-05
DAP	131-17-9	C ₁₄ H ₁₄ O ₄	246	3.23	1.16E-03	7.39E-04
BBP	85-68-7	C ₁₉ H ₂₀ O ₄	312	4.73	8.60E-06	8.61E-06
DIPrP	605-45-8	C ₁₄ H ₁₈ O ₄	250	3.27	2.63E-03	6.78E-04
DPrP	131-16-8	C ₁₄ H ₁₈ O ₄	250	3.63	1.32E-04	4.32E-04
DEEP	605-54-9	C ₁₆ H ₂₂ O ₆	310	2.30	2.54E-05	2.33E-03

^a Molecular formula of PAEs is from (Gani et al., 2016).

^b Mass (g/mol), logK_{ow}, were obtained from PubChem (PubChem).

^c Vapor pressure (mmHg) and water solution (mol/L) were obtained from the CompTox Chemicals Dashboard (Chemicals Dashboard).

Table S5 Water treatment processes of 24 h water vending machine

Brand ^a	Water treatment processes ^b
GH	PAF-CB-HPMF-ROF-RIF-UVS
JB	PAF-GAC-BAC-RIF-ROF-HPMF-EMM-OS
YSK	PPFC-GAC-GACF-PPFC-ROF-ACF-OS
QM	PPFC (5 μm)-GAC-CB-ROF-OS-UVS-CB-CSTR
EJ	PPFC (5 μm)-GAC-GACF-PPFC (1 μm)-ROF-UVS
CG	PAF-ACF-HPMF-ROF-HPMF
AM	PPFC (5 μm)-ACF-UF (1 μm)-ROF-NCF-AOS-UVS
HY	PAF-ACF-HPMF-ROF-ACF-OS

^aBrands: GH: Gang Hui; JB; Jing Bo; YSK: Yi Sikai; QM: Yi Lian; EJ: Qing Lang; CG: Yi Quan; AM: Qi Jia; HY: Han Ying.

Pretreatment filter: PAF: pretreatment automatic filter; PPFC: polypropylene filter core (5 μm and 1 μm); HPMF: high-precision micro-processing filter.

Activated carbon adsorption (AC): ACF: activated carbon filter; CB: carbon block; GAC: granular activated carbon; BAC: bar activated carbon; CACF: compressed activated carbon filter.

Filtration: ROF: reverse osmosis filter; NCF: nano crystal filter.

Sterilization: UVS: UV sterilization; AOS: activated oxygen sterilization; OS: ozone sterilization.

Other processes: EMM: energy mine magnetize; CSTR: coconut shell taste regulator, RIF: rear impurity filter.

Table S6 Exposure parameters of men and women for the intake of drinking water

IR (L/kg bw/day)	Mean
<1 months	0.052
1-3 months	0.048
3-6 months	0.052
6-12 months	0.041
1-2 years	0.023
2-3 years	0.023
3-6 years	0.018
6-11 years	0.014
11-16 years	0.010
16-18 years	0.009
18-21 years	0.009
<21 years	0.013
21-65 years	0.014
All years	0.014

Ingestion rates (IR, L/kg bw/day) were obtained from USEPA Exposure factors handbook (USEPA, 2011).

Table S7 Comparison of PAE concentrations in source water of the Yangtze River and Taihu lake from previous studies.

River basin	Predominant congener	Range (ng/L)	Median(ng/L)	Reference
Yangtze river	DBP	94.2-5.76×10 ⁶	473	Zhang et al., 2022
Taizhou section	DEP	5.6-194	16.1	
	DMEP	11.7-212	76.2	
Yangtze river	Σ ₁₆ PAEs	2.63×10 ³ -2.29×10 ⁴	-	Zhang et al., 2020
	DIBP	-	-	
	DnBP	-	-	
	DEHP	-	-	
Yangtze estuary				Deng et al., 2021
Wetland sediment	Σ ₈ PAEs	3.9-253	-	
Hangzhou Bay	Σ ₁₀ PAEs	2.68×10 ⁴ -4.24×10 ⁶	-	Wang et al., 2021
	DiBP, DnBP, DEHP	-	-	
Yangtze river	DBP, DEHP	61-2.86×10 ⁴		Zhang et al., 2012
Taihu lake				
Surface sediment	Σ ₁₀ PAEs	1.2×10 ³ -1.87×10 ⁴	-	Zhang et al., 2021
Core sediment	Σ ₁₀ PAEs	0.23×10 ³ -5.22×10 ⁴	-	
	DBP, DIBP			
Taihu lake				Luo et al., 2021
Surface water	DBP, DEHP, DIBP	ND-1.61×10 ⁴	-	
Taihu lake	DBP, DEHP	7.4×10 ² -1.3×10 ⁴	-	Gao et al., 2019
Taihu lake	Σ ₄ PAEs			Shi et al., 2012
	DBP	2.8×10 ³ -1.6×10 ⁶	-	
	DEHP	0.11-1.41×10 ³	-	

-not

found

Table S8 Comparison of PAE concentrations and compositions in this study and previous studies.

Matrix	Predominant Chemicals	Region	Range (ng/L)	Mean	Median	Target	Reference
TW	DIBP, DBP, DEP, DMP, DEHP	China	0-447	97	121±104	22	This study
TW	DMP, DEP, DEHP, DBP	Tehran, Iran	450-1080	-	730	12	Abtahi et al., 2019
TW	DEP, DMP, DnBP	Delhi, India	N.D.-5765	-	-	13	Chakraborty et al., 2022
TW	DEHP, DBP	Tianjin, China	2940-1.39×10 ⁴	1.08×10 ⁴	-	6	Ji et al., 2014
TW	DEHP	Tianjin, China	-	-	2409±391	6	Wang et al., 2021
TW		Shanghai, China	-	-	1037	8	Xue et al., 2021
TW	DBP, DIBP	French	DBP: 1300, DIBP: 1650	-	-	9	Bach et al, 2020
TW	DEHP, DIBP, DBP	Vietnam	2100-1.8×10 ⁴	-	9270	10	Le et al., 2021
TW	DBP, DEHP, nBiBP	Moscow, Russian	51.1-84.5	-	-	5	Eremina et al., 2016
TW	DMP, DEP, BBP	Nanjing, China	N.D.-527.8	-	-	4	Li et al., 2018
TW	DBP	Spain	430-909	-	-	5	Morueco et al., 2014
TW	DEP, DMP	Hangzhou, China	25-80	55	-	4	Jin et al., 2019
BW	DEHP, DBP	Egypt	N.D.-469	-	-	6	Zaki et al., 2018
WVM	DBP, DEHP, BBP	Tianjin, China	-	-	1960±160	6	Wang et al., 2021
BW	DMEP, DEP, DBP	Pakistan	9×10 ⁵ -1.13×10 ⁵	-	-	9	Surhio et al., 2017
BW	DBP	Malaysia	680-1110	-	-	5	Hazira et al., 2021
BW	DEHP, DIBP, DBP	Dalian, China	770-4.8×10 ⁴	-	-	15	Xue et al., 2021
BW	DBP	Beijing, China	10-510	-	-	21	Li et al., 2018

Table S9 The estimated daily intakes of PAEs (ng/kg/day) via ingestion of TW, WVM, and WBM water in two exposure scenarios.

		DBP		DIBP		DEP		DMP		DEHP		DMEP		DHP		Σ_{22} PAEs	
		N ^a	H ^b	N	H	N	H	N	H	N	H	N	H	N	H	N	H
TW	<1 m	0.73	1.16	1.16	4.50	0.18	0.35	0.55	1.25	0.00	1.71	0.00	0.94	0.00	2.53	3.98	10.81
	1-3 m	0.68	1.07	1.07	4.15	0.17	0.32	0.50	1.16	0.00	1.58	0.00	0.87	0.00	2.34	3.67	9.98
	3-6 m	0.73	1.16	1.16	4.50	0.18	0.35	0.55	1.25	0.00	1.71	0.00	0.94	0.00	2.53	3.98	10.81
	6-12 m	0.58	0.91	0.91	3.55	0.14	0.28	0.43	0.99	0.00	1.35	0.00	0.74	0.00	2.00	3.14	8.53
	1-2 y	0.32	0.51	0.51	1.99	0.08	0.16	0.24	0.55	0.00	0.76	0.00	0.42	0.00	1.12	1.76	4.78
	2-3 y	0.32	0.51	0.51	1.99	0.08	0.16	0.24	0.55	0.00	0.76	0.00	0.42	0.00	1.12	1.76	4.78
	3-6 y	0.25	0.40	0.40	1.56	0.06	0.12	0.19	0.43	0.00	0.59	0.00	0.33	0.00	0.88	1.38	3.74
	6-11 y	0.20	0.31	0.31	1.21	0.05	0.09	0.15	0.34	0.00	0.46	0.00	0.25	0.00	0.68	1.07	2.91
	11-16 y	0.14	0.22	0.22	0.87	0.03	0.07	0.10	0.24	0.00	0.33	0.00	0.18	0.00	0.49	0.77	2.08
	16-18 y	0.13	0.20	0.20	0.78	0.03	0.06	0.09	0.22	0.00	0.30	0.00	0.16	0.00	0.44	0.69	1.87
WVM	18-21 y	0.13	0.20	0.20	0.78	0.03	0.06	0.09	0.22	0.00	0.30	0.00	0.16	0.00	0.44	0.69	1.87
	<21 y	0.18	0.29	0.29	1.12	0.05	0.09	0.14	0.31	0.00	0.43	0.00	0.23	0.00	0.63	0.99	2.70
	21-65 y	0.20	0.31	0.31	1.21	0.05	0.09	0.15	0.34	0.00	0.46	0.00	0.25	0.00	0.68	1.07	2.91
	All y	0.20	0.31	0.31	1.21	0.05	0.09	0.15	0.34	0.00	0.46	0.00	0.25	0.00	0.68	1.07	2.91
	<1 m	0.74	5.41	1.38	17.03	0.02	0.08	0.05	0.26	0.40	2.70	0.51	2.39	1.21	2.34	6.84	34.30
	1-3 m	0.68	5.00	1.28	15.72	0.02	0.07	0.05	0.24	0.37	2.50	0.48	2.21	1.12	2.16	6.31	31.66
	3-6 m	0.74	5.41	1.38	17.03	0.02	0.08	0.05	0.26	0.40	2.70	0.51	2.39	1.21	2.34	6.84	34.30
	6-12 m	0.58	4.27	1.09	13.43	0.01	0.06	0.04	0.21	0.31	2.13	0.41	1.88	0.96	1.84	5.39	27.05
	1-2 y	0.33	2.39	0.61	7.53	0.01	0.03	0.02	0.12	0.18	1.20	0.23	1.06	0.54	1.03	3.02	15.17
	2-3 y	0.33	2.39	0.61	7.53	0.01	0.03	0.02	0.12	0.18	1.20	0.23	1.06	0.54	1.03	3.02	15.17
WBM	3-6 y	0.26	1.87	0.48	5.89	0.01	0.03	0.02	0.09	0.14	0.94	0.18	0.83	0.42	0.81	2.37	11.87
	6-11 y	0.20	1.46	0.37	4.58	0.00	0.02	0.01	0.07	0.11	0.73	0.14	0.64	0.33	0.63	1.84	9.24

11-16 y	0.14	1.04	0.27	3.27	0.00	0.01	0.01	0.05	0.08	0.52	0.10	0.46	0.23	0.45	1.32	6.60	
16-18 y	0.13	0.94	0.24	2.95	0.00	0.01	0.01	0.05	0.07	0.47	0.09	0.41	0.21	0.40	1.18	5.94	
18-21 y	0.13	0.94	0.24	2.95	0.00	0.01	0.01	0.05	0.07	0.47	0.09	0.41	0.21	0.40	1.18	5.94	
<21 y	0.18	1.35	0.35	4.26	0.00	0.02	0.01	0.07	0.10	0.68	0.13	0.60	0.30	0.58	1.71	8.58	
21-65 y	0.20	1.46	0.37	4.58	0.00	0.02	0.01	0.07	0.11	0.73	0.14	0.64	0.33	0.63	1.84	9.24	
All y	0.20	1.46	0.37	4.58	0.00	0.02	0.01	0.07	0.11	0.73	0.14	0.64	0.33	0.63	1.84	9.24	
<1 m	0.74	6.73	1.20	3.26	0.09	0.37	0.30	1.15	0.15	0.83	0.26	1.56	1.05	2.74	4.24	22.44	
1-3 m	0.69	6.21	1.11	3.01	0.09	0.34	0.28	1.06	0.14	0.77	0.24	1.44	0.97	2.53	3.92	20.71	
3-6 m	0.74	6.73	1.20	3.26	0.09	0.37	0.30	1.15	0.15	0.83	0.26	1.56	1.05	2.74	4.24	22.44	
6-12 m	0.59	5.31	0.94	2.57	0.07	0.29	0.24	0.91	0.12	0.66	0.21	1.23	0.83	2.16	3.34	17.69	
WBM	1-2 y	0.33	2.98	0.53	1.44	0.04	0.16	0.13	0.51	0.06	0.37	0.12	0.69	0.46	1.21	1.88	9.92
	2-3 y	0.33	2.98	0.53	1.44	0.04	0.16	0.13	0.51	0.06	0.37	0.12	0.69	0.46	1.21	1.88	9.92
	3-6 y	0.26	2.33	0.41	1.13	0.03	0.13	0.11	0.40	0.05	0.29	0.09	0.54	0.36	0.95	1.47	7.77
	6-11 y	0.20	1.81	0.32	0.88	0.03	0.10	0.08	0.31	0.04	0.22	0.07	0.42	0.28	0.74	1.14	6.04
	11-16 y	0.14	1.29	0.23	0.63	0.02	0.07	0.06	0.22	0.03	0.16	0.05	0.30	0.20	0.53	0.82	4.31
	16-18 y	0.13	1.16	0.21	0.56	0.02	0.06	0.05	0.20	0.03	0.14	0.05	0.27	0.18	0.47	0.73	3.88
	18-21 y	0.13	1.16	0.21	0.56	0.02	0.06	0.05	0.20	0.03	0.14	0.05	0.27	0.18	0.47	0.73	3.88
	<21 y	0.19	1.68	0.30	0.81	0.02	0.09	0.08	0.29	0.04	0.21	0.07	0.39	0.26	0.68	1.06	5.61
	21-65 y	0.20	1.81	0.32	0.88	0.03	0.10	0.08	0.31	0.04	0.22	0.07	0.42	0.28	0.74	1.14	6.04
	All y	0.20	1.81	0.32	0.88	0.03	0.10	0.08	0.31	0.04	0.22	0.07	0.42	0.28	0.74	1.14	6.04

^aN: Normal exposure scenario, median concentrations of PAEs were used.

^bH: High exposure scenario, 90 percentiles of PAE concentrations were used.

M:

month;

y:

year.

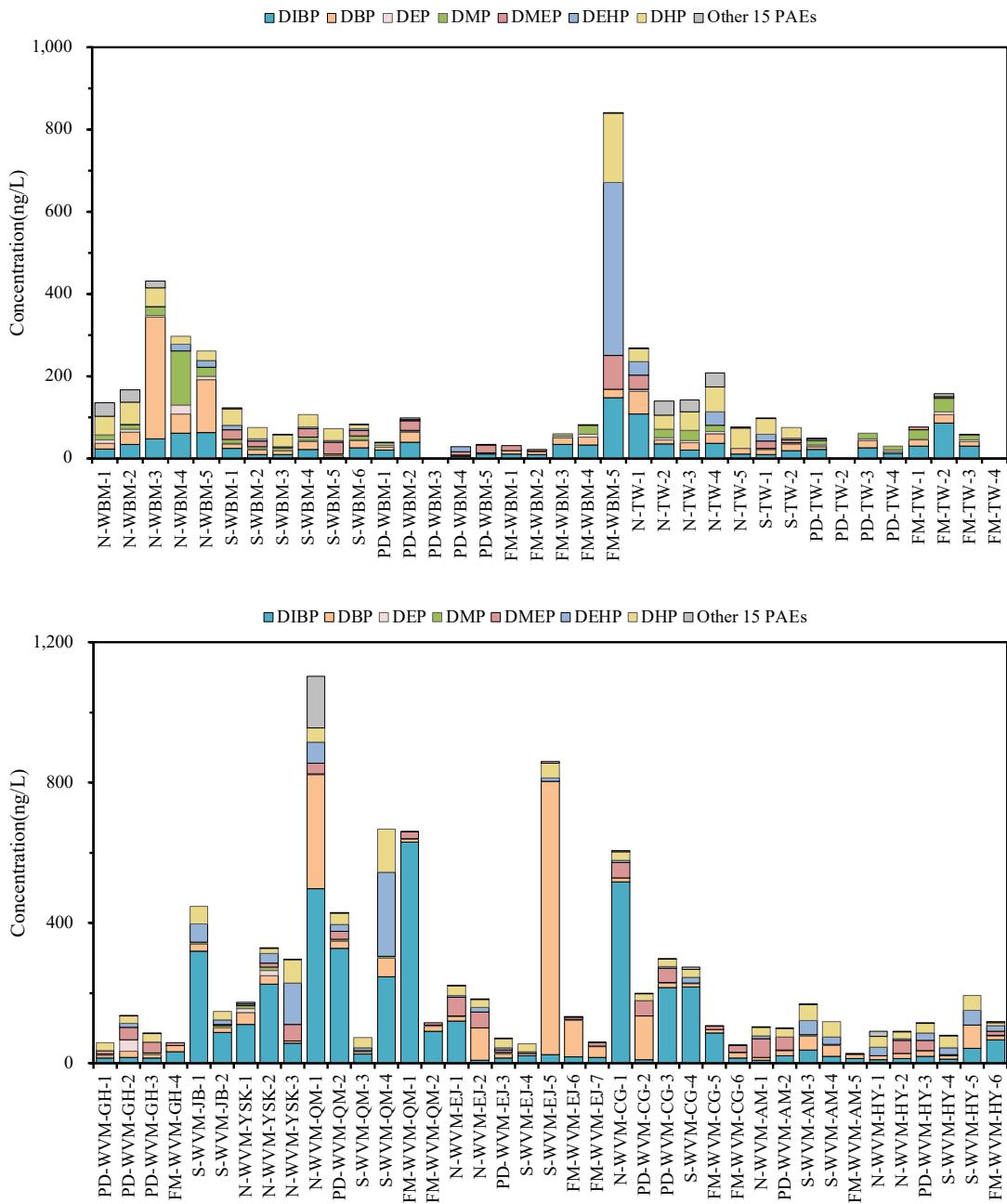


Fig. S1. Concentrations of PAEs in three types of samples.

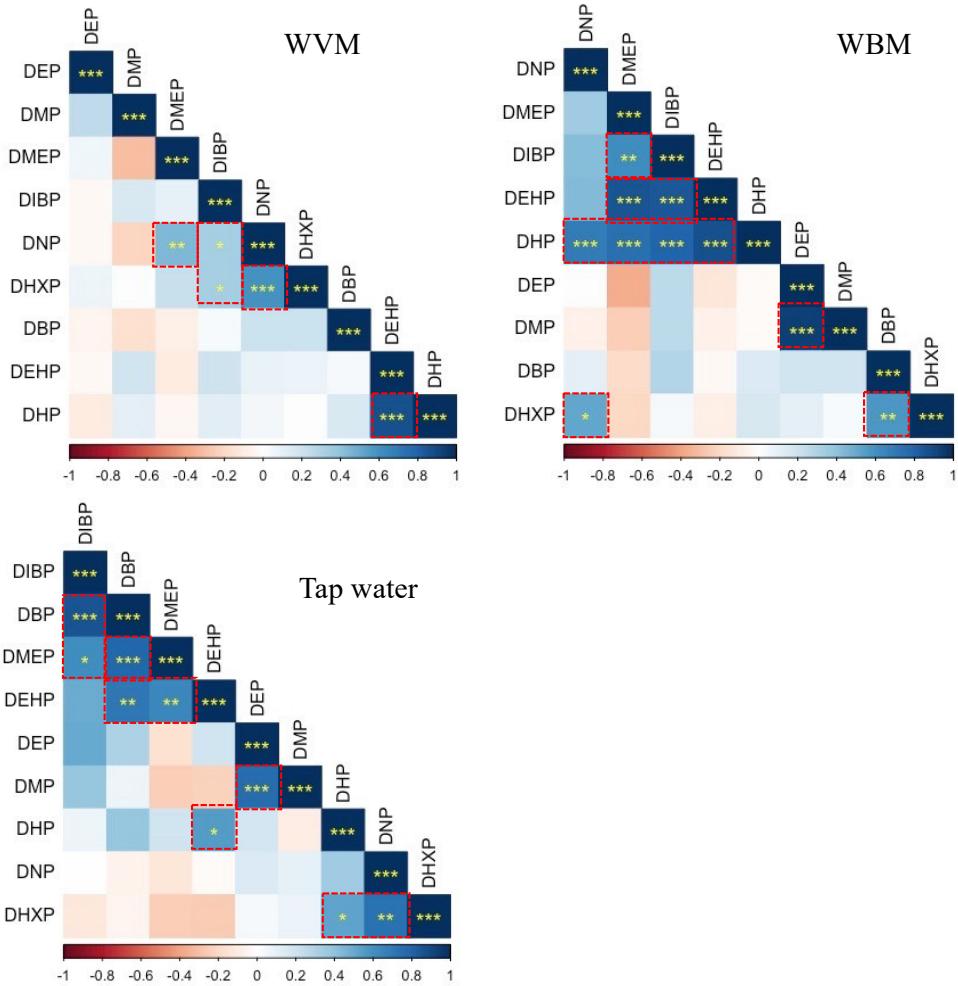


Fig. S2. Heatmap of the correlation matrix of PAEs in TW, WVM, and WBM samples (hollow square represent significant correlations).

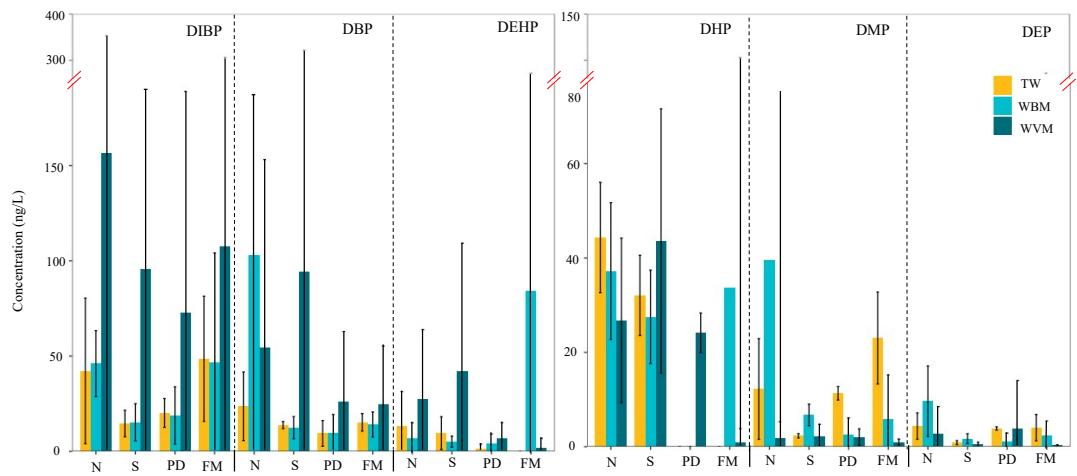


Fig. S3. Comparison of the individual PAE mean levels in TW, WBM, and WVM.

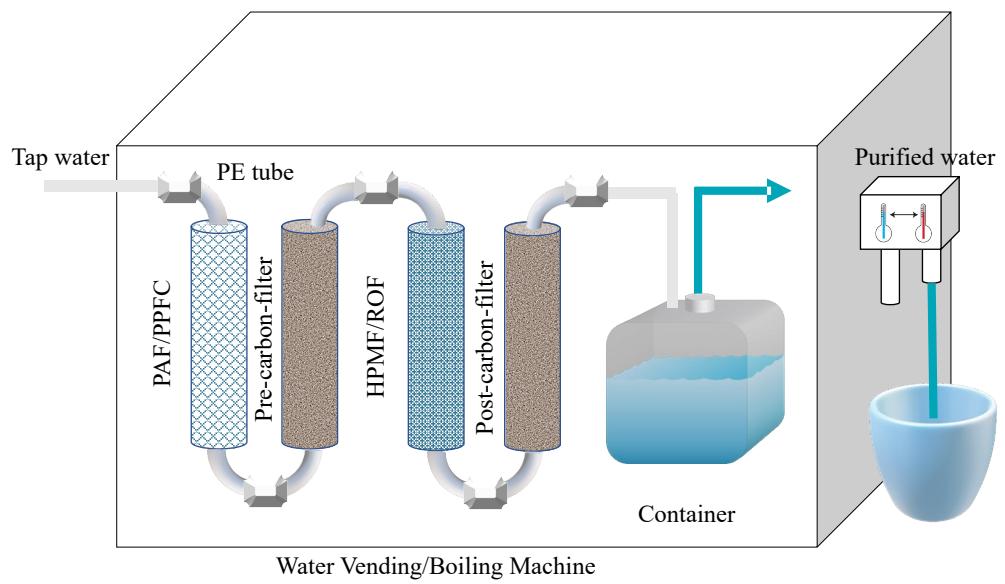


Fig. S4. Schematic diagram of purification processes inside water purifier.

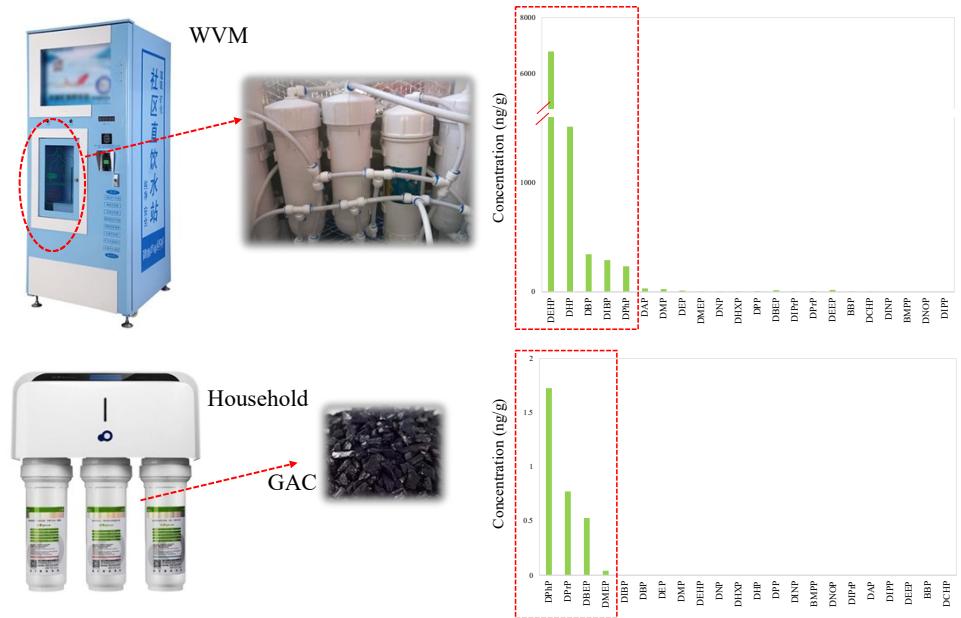


Fig. S5. PAE levels in materials (HPMF and GAC)

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