Supporting information

Occurrence and Dietary Intake of Organophosphate Esters via Animal-origin Food Consumption in China: Results of a Chinese Total Diet Study

Xuelei Chen[1], Sai Fan[2], Bing Lyu[3], Lei Zhang[3], Shunying Yao[1], Jiaying Liu[4,], Zhixiong Shi[1,*], Yongning Wu[3]*

[1] School of Public Health and Beijing Key Laboratory of Environmental Toxicology, Capital Medical University, Beijing 100069, China

[2] Beijing Center for Disease Prevention and Control, Beijing Research Center for Preventive Medicine, Beijing 100013, China

[3] NHC Key Laboratory of Food Safety Risk Assessment, Chinese Academy of Medical Science Research Unit (No. 2019RU014), China National Center for Food Safety Risk Assessment, Beijing 100021, China

[4] Department of Nutrition and Health, China Agricultural University, Beijing 100083, China

Number of pages: 29

Number of tables: 5

Number of figures: 9

Corresponding authors

Jiaying liu

Department of Nutrition and Health, China Agricultural University, Beijing 100083, China

liujiaying@cau.edu.cn

Zhixiong Shi

School of Public Health and Beijing Key Laboratory of Environmental Toxicology, Capital Medical University, Beijing 100069, China

szx0127@ccmu.edu.cn

Table of content

Figure S7. EDI of OPEs via various food groups

Figure S8. A comparison between EDI of OPEs via food consumption and other pathways

Figure S9. A comparison between mean EDI of OPEs and BFRs in the 6th TDS.

References

1. Composition of the four food groups

Food items consumed by “standard man” were aggregated into 13 food groups. Four animal-origin food groups of these 13 groups were subjected for OPEs analysis included: (a) eggs and egg products, including chicken and duck eggs, and salted and limed duck eggs; (b) aquatic foods, including fish, shrimp, and oysters; (c) milk and milk products, including cow milk, cow milk powder, yogurt, and sheep milk; (d) meat and meat products, including pork, mutton, beef, chicken, duck, rabbit, pork liver, and swine blood.

2. Chemicals used in sample analysis

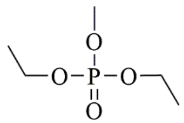
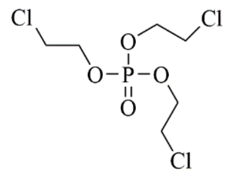
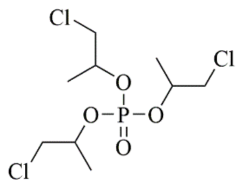
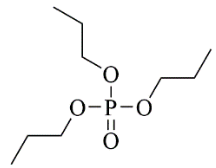
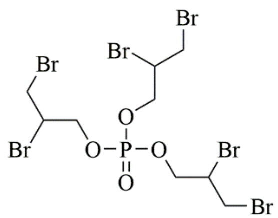
HPLC-grade methanol, acetonitrile and formic acid were from Fisher Scientific (Massachusetts, America). HPLC-grade ammonium acetate was from Aladdin Corporation (Shanghai, China). Reagent grade anhydrous magnesium sulfate ($MgSO_4$) was acquired from Guangfu Fine Chemical Research Institute (Tianjin, China). Primary secondary amine (PSA) and octadecyl-modified silica (C_{18}) were from Agilent Technologies (Palo Alto, CA, USA). Oasis HLB extraction cartridges (6 cc/200 mg) were provided by Waters Corporation (MA, USA).

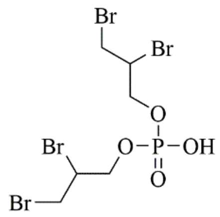
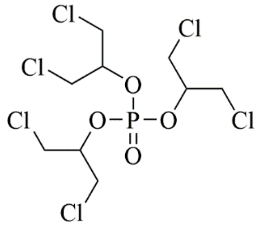
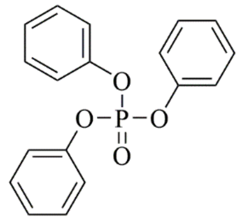
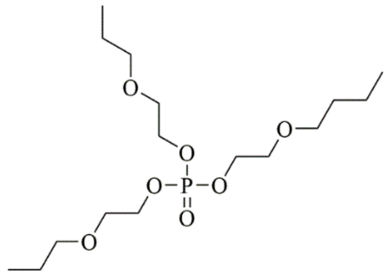
3. Sample pretreatment and instrumental analysis

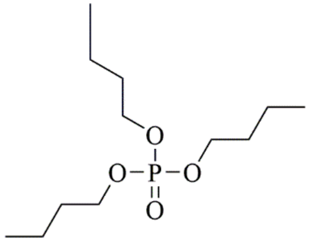
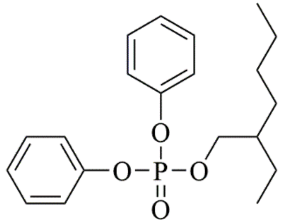
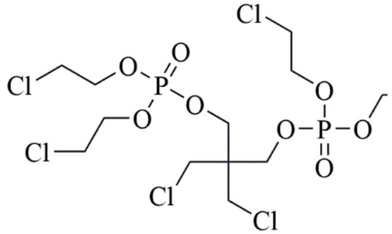
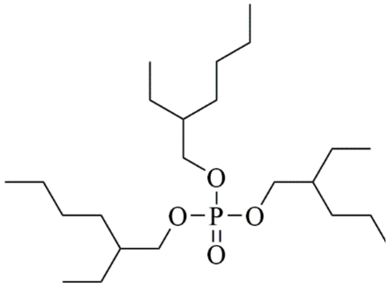
All samples were freeze-dried before extraction. After freeze-drying, 0.5 g±0.01 g dried sample and 5 ng each of the internal standards were added into a 15-mL polypropylene (PP) centrifuge tube and mixed with 5 mL of 0.5% formic acid in acetonitrile. After vortexing for 1 min, the tube was shaken by an orbital shaker for 2 h and ultrasonicated for 10 min. After centrifugation at 5000 rpm for 10 min, the supernatant was transferred into another precleaned PP tube. Then, dispersive SPE sorbent that consisted of 250 mg MgSO4, 150 mg PSA and 150 mg C18 were added. The tube was vortexed for 1 min and centrifuged at 5000 rpm for 5 min. The supernatant was collected and purified by an Oasis HLB (6 cc/200 mg) extraction cartridge. The cartridge was preconditioned with 5 mL of ACN, and after loading of the sample extract (Fraction 1), the cartridge was eluted with 3.5 mL of ACN (Fraction 2). The purified extracts (a mixture of fractions 1 and 2) were evaporated to dryness under N_2. The residues were dissolved in 200 μL of methanol and filtered through a 0.22-μm filter membrane for instrumental analysis.

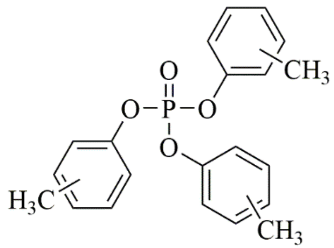
Quantification was accomplished on an Acquity ultra-performance liquid chromatographic system coupled with a Micromass Xevo TQS mass detector (Waters, MA, USA). The analytes were separated by an Acquity UPLC BEH C18 column (100×2.1 mm i.d., 1.7 μm) maintained at 40 °C. Water (with 0.01% formic acid) (A) and methanol (with 10 mmol/L ammonium acetate) (B) were used as mobile phase at a flow rate of 0.2 mL/min, and the inject volume was 5 μL. The gradient program for the mobile phase started with 30% B, and changed linearly to 80% B in 4 min and maintained for 4 min, and then increased to 100% B in 0.1 min, held for 3.5 min and returned to the initial condition (30% B). The MS detector was equipped with an electrospray ionization probe operated in positive mode with multiple reaction monitoring, and the best conditions of all target chemicals were presented in the Supplementary Table S1.

Table S1. The structure, full name, abbreviation, CAS number, retention time and monitoring transition of 14 OPEs and isotopically labeled internal standards

Full name	Abbreviation	Structure	CAS.NO	RT (min)	Monitoring transition (*m/z*)	CE (eV)	ILIS
Trimethyl phosphate	TMP		512-56-1	1.87	141→79 141→109 [a)]	20 16	d_{12}-TCEP
Tris(2-chloroethyl) phosphate	TCEP		115-96-8	4.11	287→99 285→99 [a)]	30 20	d_{12}-TCEP
Tri(3-chloropropyl) phosphate	TCIPP		13674-84-5	5.34	329→99 327→99 [a)]	20 20	d_{15}-TDCIPP
Tri-*n*-propyl phosphate	TPrP		513-08-6	5.36	225→141 225→99 [a)]	10 20	d_{21}.TPrP
Tris(2,3-dibromopropyl) phosphate	TDBPP		126-72-7	6.41	716→99 699→99 [a)]	40 25	d_{15}-TDCIPP

Bis(2,3-dibromopropyl) phosphate	DDBPP		5412-25-9	6.41	698→99 a) 698→298	30 10	d_{15}-TDCIPP
Tris(1,3-dichloro-2-propyl) phosphate	TDCIPP		13674-87-8	5.99	433→99 431→99 a)	20 20	d_{15}-TDCIPP
Triphenyl phosphate	TPHP		115-86-6	6.12	327→77 a) 327→152	40 40	$^{13}C_{18}$-TPhP
Tris(2-butoxyethyl) phosphate	TBOEP		78-51-3	7.27	399→199 399→299 a)	15 10	$^{13}C_{6}$-TBOEP

Tri-*n*-butyl phosphate	TnBP		126-73-8	6.84	267→99 [a)]	20	d_{27}-TnBP
2-ethylhexyl diphenyl phosphate	EHDPP		1241-94-7	9.64	363.1→77.0 363.1→251.0 [a)]	40 20	$^{13}C_{18}$-TPhP
Tetrakis(2-chloroethyl) dichloroisopentyl diphosphate	V6		38051-10-4	5.21	582.9→234.9 [a)] 582.9→360.9	30 20	$^{13}C_6$-TBOEP
Tris(2-ethylhexyl) phosphate	TEHP		78-42-2	11.24	435.4→98.9 [a)] 435.4→323.2	10 10	$^{13}C_6$-TBOEP

Tricresyl phosphate	TMPP		1330-78-5	8.42	369.03→165.20 [a] 369.03→91.03	40 15	$^{13}C_{18}$-TPhP
d_{12}-Tris(2-chloroethyl) phosphate	d_{12}-TCEP	-	-	4.14	299→67 299→102 [a]	30 25	-
$^{13}C_{18}$-Triphenyl phosphate	$^{13}C_{18}$-TPhP	-	-	6.05	342→82 342→162 [a]	25 25	-
$^{13}C_{6}$-Tris(2-butoxyethyl) phosphate	$^{13}C_{6}$-TBOEP	-	-	7.27	405→102 405→303 [a]	10 10	-
d_{27}-Tri-*n*-butyl phosphate	d_{27}-TnBP	-	-	6.72	294→102 [a]	20	-
d_{21}-Tri-*n*-propyl phosphate	d_{21}-TPrP	-	-	5.30	246.2→102 246.2→150.1 [a]	15 30	-
d_{15}-Tris(1,3-dichloro-2-propyl) phosphate	d_{15}-TDCIPP	-	-	5.99	446→102 [a] 445→216	50 30	-

a) Quantitative transition or ion;
RT: retention time;
CE: collision energy.
TMPP: mixture of isomers

Table S2. Average recoveries, relative standard deviation (RSD) and LODs of target OPEs measured in spiked four animal-origin samples.

Compound	Basa catfish			Chicken			Chicken eggs			Cow milk		
	Average recovery (*n*=3)	RSD (%)	LODs (ng/g ww)	Average recovery (*n*=3)	RSD (%)	LODs (ng/g ww)	Average recovery (*n*=3)	RSD (%)	LODs (ng/g ww)	Average recovery (*n*=3)	RSD (%)	LODs (ng/g ww)
TMP	63.7	1.57	0.008	80.7	7.45	0.008	78.1	12.96	0.007	115.7	8.73	0.009
TCEP	103.0	11.24	0.003	82.3	10.46	0.003	92.4	6.19	0.002	104.8	1.57	0.004
TDBPP	115.5	6.72	0.08	113.9	1.89	0.08	95.4	10.14	0.06	115.2	8.14	0.01
DDBPP	112.4	8.61	0.02	106.0	1.50	0.04	85.5	12.20	0.02	108.4	13.30	0.03
V6	111.9	11.15	0.02	96.1	13.19	0.02	106.4	8.10	0.02	107.9	14.87	0.02
TDCIPP	103.3	9.50	0.006	84.8	13.99	0.006	78.7	2.09	0.005	103.2	6.90	0.007
TBOEP	115.7	11.49	0.002	100.0	2.03	0.002	76.8	6.88	0.002	88.1	12.35	0.004
TCIPP	111.0	13.02	0.002	111.4	10.69	0.003	83.0	6.23	0.003	104.2	12.91	0.005
TPhP	99.3	9.09	0.001	109.8	4.63	0.001	76.9	8.08	0.001	91.6	6.75	0.001
TnBP	118.2	8.37	0.004	108.5	1.01	0.004	95.6	4.28	0.004	103.8	12.16	0.004
TPrP	96.5	13.91	0.003	82.4	1.20	0.003	73.4	4.42	0.003	101.3	14.69	0.003
TMPP	117.1	5.98	0.004	95.9	3.07	0.008	91.5	0.31	0.004	106.6	11.73	0.004
TEHP	109.6	8.55	0.004	109.9	11.80	0.004	89.1	9.70	0.006	115.2	14.70	0.008
EHDPP	113.4	7.88	0.004	106.6	13.41	0.005	93.9	6.15	0.008	100.2	15.17	0.002

The spiking level of the recovery test was 10 ng/g.

Table S3. Concentrations of OPEs (ng/g ww) in food composites.

Analyte	Statistics	Total samples (*n*=96)	Aquatic food (*n*=24)	Meat (*n*=24)	Eggs (*n*=24)	Milk (*n*=24)
TMP	%DF	98	100	96	96	100
	Min	<LOD	0.019	<LOD	<LOD	0.010
	25th	0.045	0.08	0.09	0.03	0.038
	Median	0.093	0.137	0.187	0.063	0.062
	75th	0.2	0.211	0.298	0.143	0.077
	Max	1.28	0.426	1.28	0.61	0.407
	AM	0.15	0.16	0.239	0.127	0.075
	SD	0.17	0.12	0.26	0.16	0.08
TCEP	%DF	100	100	100	100	100
	Min	0.107	0.438	0.366	0.22	0.107
	25th	0.411	0.613	0.777	0.788	0.194
	Median	0.812	0.798	0.982	1.05	0.237
	75th	1.13	0.941	1.29	1.7	0.282
	Max	4.13	3.08	3	4.13	0.909
	AM	0.928	0.906	1.11	1.41	0.283
	SD	0.78	0.55	0.60	1.1	0.18
TDBPP	%DF	29	25	17	17	58
	Min	<LOD	<LOD	<LOD	<LOD	<LOD
	25th	<LOD	<LOD	<LOD	<LOD	<LOD
	Median	<LOD	<LOD	<LOD	<LOD	0.014
	75th	0.015	0.022	<LOD	<LOD	0.019
	Max	0.441	0.23	0.441	0.172	0.063
	AM	0.023	0.031	0.031	0.018	0.013
	SD	0.06	0.06	0.09	0.04	0.02
DDBPP	%DF	81	88	79	91	67
	Min	<LOD	<LOD	<LOD	<LOD	<LOD
	25th	0.036	0.076	0.057	0.045	<LOD
	Median	0.089	0.124	0.167	0.111	0.038
	75th	0.189	0.187	0.449	0.192	0.05
	Max	3.43	0.452	3.43	0.528	0.1
	AM	0.205	0.141	0.493	0.152	0.034
	SD	0.45	0.11	0.84	0.14	0.03
V6	%DF	41	33	38	13	79
	Min	<LOD	<LOD	<LOD	<LOD	<LOD
	25th	<LOD	<LOD	<LOD	<LOD	0.028
	Median	<LOD	<LOD	<LOD	<LOD	0.044
	75th	0.036	0.028	0.023	<LOD	0.062
	Max	0.13	0.119	0.037	0.13	0.115
	AM	0.02	0.017	0.01	0.011	0.044
	SD	0.03	0.03	0.01	0.03	0.03
TDCIPP	%DF	98	96	100	100	96

	Min	<LOD	<LOD	0.121	0.076	<LOD
	25th	0.251	0.401	0.37	0.163	0.226
	Median	0.414	0.508	0.558	0.406	0.292
	75th	0.638	0.638	0.892	0.802	0.392
	Max	6.72	1.114	1.43	6.72	0.971
	AM	0.562	0.531	0.63	0.756	0.332
	SD	0.72	0.25	0.38	1.3	0.20
TBOEP	%DF	97	100	100	91	96
	Min	<LOD	0.008	0.003	<LOD	<LOD
	25th	0.027	0.043	0.052	0.018	0.027
	Median	0.136	0.145	0.136	0.105	0.178
	75th	0.496	0.401	1.05	0.458	0.462
	Max	3.53	1.63	3.53	0.733	1.87
	AM	0.383	0.283	0.697	0.235	0.317
	SD	0.59	0.37	0.95	0.26	0.42
TCIPP	%DF	99	96	100	100	100
	Min	<LOD	<LOD	0.065	0.107	0.116
	25th	0.631	0.582	1.09	0.757	0.514
	Median	0.977	0.982	1.75	1.27	0.664
	75th	1.71	1.3	2.32	2.24	0.901
	Max	25.5	25.5	6.15	6.94	1.844
	AM	1.61	1.99	1.96	1.78	0.713
	SD	2.74	5.04	1.35	1.7	0.35
TPHP	%DF	100	100	100	100	100
	Min	0.023	0.034	0.082	0.046	0.023
	25th	0.121	0.176	0.251	0.15	0.059
	Median	0.274	0.318	0.453	0.237	0.109
	75th	0.556	0.472	0.576	0.445	0.303
	Max	20.9	5.51	1.63	20.9	4.719
	AM	0.689	0.626	0.512	1.18	0.436
	SD	2.22	1.11	0.37	4.2	0.98
TnBP	%DF	92	100	92	74	100
	Min	<LOD	0.027	<LOD	<LOD	0.015
	25th	0.048	0.075	0.079	0.003	0.043
	Median	0.105	0.135	0.147	0.05	0.089
	75th	0.174	0.157	0.26	0.128	0.166
	Max	0.545	0.342	0.515	0.545	0.54
	AM	0.134	0.131	0.184	0.093	0.129
	SD	0.12	0.07	0.14	0.13	0.13
TPrP	%DF	97	96	100	91	100
	Min	<LOD	<LOD	0.003	<LOD	0.004
	25th	0.009	0.009	0.028	0.006	0.013
	Median	0.018	0.011	0.06	0.009	0.021
	75th	0.046	0.044	0.093	0.013	0.029

	Max	0.381	0.381	0.272	0.039	0.194
	AM	0.044	0.057	0.075	0.011	0.032
	SD	0.07	0.11	0.07	0.01	0.04
TMPP	%DF	96	100	100	91	92
	Min	<LOD	0.045	0.021	<LOD	<LOD
	25th	0.051	0.164	0.162	0.113	0.02
	Median	0.166	0.239	0.26	0.161	0.026
	75th	0.308	0.327	0.538	0.368	0.047
	Max	7.21	6.542	7.21	3.13	0.572
	AM	0.514	0.727	0.917	0.353	0.06
	SD	1.3	1.49	1.95	0.62	0.12
TEHP	%DF	96	100	96	96	92
	Min	<LOD	0.006	<LOD	<LOD	<LOD
	25th	0.037	0.057	0.068	0.035	0.019
	Median	0.14	0.182	0.178	0.073	0.129
	75th	0.646	0.406	0.943	0.237	1.02
	Max	18.5	18.5	5.56	5.77	4.45
	AM	0.843	1.21	0.799	0.541	0.819
	SD	2.2	3.75	1.30	1.3	1.28
EHDPP	%DF	96	100	96	100	88
	Min	<LOD	0.009	<LOD	0.014	<LOD
	25th	0.284	0.284	0.463	0.494	0.301
	Median	1.63	4.62	4.61	2.3	0.84
	75th	7.45	8.5	11.9	6.06	1.56
	Max	35.5	18.9	35.5	26.2	16.6
	AM	5.13	5.32	8.62	4.56	2.03
	SD	7.2	5.62	10.5	6.2	3.65

The concentrations below LOD were treated as zero during statistical analysis.

Table S4. Spearman's rank correlation coefficient between concentrations of OPEs in food composites.

	TCEP	DDBPP	TDCIPP	TBOEP	TCIPP	TPHP	TNBP	TPRP	TMPP	TEHP	EHDPP
TMP (*r*)	0.173	**0.257**	**0.342**	-0.005	**0.380**	**0.345**	**0.221**	**0.282**	**0.437**	0.162	**0.388**
p	0.099	**0.013**	**0.001**	0.965	**0.000**	**0.001**	**0.034**	**0.007**	**0.000**	0.123	**0.000**
TCEP (*r*)		**0.382**	0.121	-0.016	**0.390**	**0.302**	0.035	-0.029	**0.505**	-0.011	**0.212**
p		**0.000**	0.249	0.879	**0.000**	**0.003**	0.742	0.783	**0.000**	0.921	**0.043**
DDBPP (*r*)			**0.321**	0.175	**0.504**	0.198	0.169	0.042	**0.307**	**0.256**	0.176
p			**0.002**	0.096	**0.000**	0.059	0.108	0.694	**0.003**	**0.014**	0.093
TDCIPP (*r*)				0.001	**0.453**	**0.424**	**0.315**	0.105	**0.353**	-0.058	**0.302**
p				0.991	**0.000**	**0.000**	**0.002**	0.318	**0.001**	0.583	**0.003**
TBOEP (*r*)					-0.036	**0.237**	**0.225**	**0.329**	-0.049	**0.377**	-0.056
p					0.732	**0.023**	**0.031**	**0.001**	0.646	**0.000**	0.597
TCIPP (*r*)						**0.257**	0.193	0.130	**0.310**	0.178	0.115
p						**0.013**	0.066	0.218	**0.003**	0.090	0.276
TPHP (*r*)							0.101	0.188	**0.532**	0.016	**0.433**
p							0.338	0.073	**0.000**	0.880	**0.000**
TNBP (*r*)								**0.234**	0.128	**0.228**	0.052
p								**0.025**	0.222	**0.029**	0.623
TPRP (*r*)									0.151	**0.217**	0.165
p									0.150	**0.038**	0.116
TMPP (*r*)										0.045	**0.421**
p										0.673	**0.000**
TEHP (*r*)											0.065
p											0.541

Note：1) Compounds with DF>50% were included in analysis; 2) *p* and corresponding *r* were marked in bold when $p<0.05$.

Table S5-1. The upper bound dietary intake of OPEs by the "standard Chinese man" (ng/kg bw/day)

Province	TMP	TDBPP	DDBPP	V6	TBOEP	TNBP	TPRP	Σ_4Cl-OPEs	$\Sigma_5$alkyl-OPEs	$\Sigma_3$aryl-OPEs	Σ_2Br-OPEs
Heilongjiang	0.11	0.169	0.258	0.048	0.303	0.42	0.105	6.92	1.86	4.09	0.427
Liaoning	0.467	0.186	0.101	0.127	0.277	0.32	0.433	8.86	1.56	64.7	0.287
Hebei	0.083	0.117	0.106	0.058	1.39	0.197	0.118	7.04	2.31	24.9	0.223
Beijing	0.438	0.173	0.337	0.096	0.455	0.203	0.139	9.99	2.12	28.7	0.509
Jilin	0.283	0.143	0.451	0.075	2.67	0.362	0.196	10.2	9.61	5.13	0.595
Shanxi	0.17	0.244	0.489	0.124	8.69	0.548	0.385	10.2	10.1	1.88	0.732
Shaanxi	0.223	0.085	0.087	0.034	1.19	0.089	0.112	3.27	1.95	1.42	0.172
Henan	0.09	0.111	0.24	0.05	1.72	0.486	0.105	3.18	5.11	7.86	0.351
Ningxia	0.114	0.129	0.309	0.034	1.57	0.504	0.034	3.74	3.98	11.8	0.438
Inner Mongolia	0.205	0.157	0.773	0.05	1.32	0.22	0.056	6.83	2.49	7.85	0.93
Qinghai	0.425	0.122	0.534	0.047	1.1	0.221	0.095	5.54	3.7	43	0.656
Gansu	0.286	0.076	0.167	0.037	0.217	0.226	0.033	5.14	0.944	13.3	0.243
Shanghai	0.235	0.302	4.76	0.204	0.309	0.81	0.186	13	4.04	42.3	5.06
Fujian	0.822	0.227	1.4	0.1	0.159	0.551	0.056	12.3	1.61	14.5	1.63
Jiangxi	1.22	0.204	0.44	0.067	0.125	0.324	0.046	29.3	6.73	6.51	0.644
Jiangsu	0.784	0.255	0.389	0.076	2.03	0.55	0.178	9	6.55	16.1	0.643
Zhejiang	0.579	0.263	7.2	0.104	0.174	0.322	0.061	16.3	1.26	27	7.46
Shangdong	0.558	0.299	0.402	0.127	0.248	0.72	0.152	17.6	7.97	7.8	0.701
Hubei	0.326	0.255	0.932	0.16	2.69	0.252	0.162	6.43	9.91	34.3	1.18
Sichuan	0.404	0.198	0.418	0.065	0.022	0.026	0.362	13.8	1.18	27.5	0.616
Guangxi	0.112	0.351	0.351	0.115	0.399	0.627	0.086	9.91	1.9	39.4	0.702
Hunan	3.56	1.171	2.05	0.094	2.95	1.1	0.107	9.34	9.41	28.5	3.23
Guangdong	0.819	0.264	0.329	0.114	0.174	1.16	0.168	5.78	2.53	49.5	0.593
Guizhou	0.785	0.179	1.66	0.067	0.102	0.449	0.112	14.1	4.42	12.3	1.84

Mean	0.546	0.237	1.01	0.086	1.26	0.445	0.145	9.91	4.3	21.7	1.24
Median	0.365	0.192	0.41	0.076	0.427	0.391	0.112	9.17	3.12	15.3	0.64
RfD	na	na	na	na	1.5×10^{3}[d]	1×10^{4}[b]	na				

The concentrations below LOD were treated as LOD.

na: RfD value is not available.

[a] EU Commission, IUCLID Dataset, 2000.[1]

[b] U.S. Environmental Protection Agency Regional Screening Levels (RSLs) (2017).[2]

[c] From ref.[3]

[d] From ref.[4]

Table S5-2. The lower bound dietary intake of OPEs by the "standard Chinese man" (ng/kg bw/day)

Province	TMP	TDBPP	DDBPP	V6	EHDPP	TCIPP	TCEP	TDCIPP	TPHP	TBOEP	TNBP	TPRP	TMPP	TEHP
Heilongjiang	0.11	0.006	0.258	0	2.35	1.35	4.95	0.574	0.319	0.303	0.42	0.104	1.42	0.918
Liaoning	0.467	0	0.028	0.084	60	3.53	2.3	2.9	3.24	0.277	0.32	0.433	1.47	0.064
Hebei	0.083	0	0.094	0.026	9.68	2.85	2.75	1.39	14.5	1.39	0.194	0.118	0.649	0.52
Beijing	0.438	0	0.337	0.055	25.8	3.93	4.66	1.31	1.36	0.455	0.201	0.139	1.53	0.881
Jilin	0.283	0	0.451	0.072	3.35	2.76	6.89	0.462	1.57	2.67	0.362	0.196	0.212	6.1
Shanxi	0.17	0.06	0.489	0.113	0.657	5.46	2.21	2.43	0.994	8.69	0.548	0.385	0.227	0.269
Shaanxi	0.223	0.006	0.063	0.012	0.471	2.05	0.711	0.473	0.717	1.19	0.089	0.112	0.227	0.329
Henan	0.09	0.004	0.24	0.021	6.9	1.5	0.956	0.67	0.668	1.72	0.486	0.104	0.289	2.71
Ningxia	0.114	0.107	0.309	0.011	4.46	1.79	1.31	0.604	0.79	1.57	0.504	0.034	6.6	1.76
Inner Mongolia	0.205	0.049	0.773	0	6.06	3.36	1.75	1.68	1.4	1.31	0.215	0.056	0.401	0.69
Qinghai	0.425	0	0.509	0	40.4	2.39	1.77	1.34	1.78	1.1	0.221	0.095	0.866	1.86
Gansu	0.286	0.005	0.161	0.028	12.6	2.91	0.65	1.55	0.488	0.217	0.226	0.033	0.152	0.182
Shanghai	0.235	0.175	4.74	0.169	39.9	6.4	4.57	1.85	1.29	0.309	0.81	0.186	1.103	2.49
Fujian	0.822	0	1.36	0.071	13.1	4.45	5.86	1.87	0.755	0.158	0.551	0.056	0.661	0.016
Jiangxi	1.22	0.005	0.429	0.046	4.9	24.8	1.36	3.06	0.966	0.125	0.324	0.045	0.636	5.02
Jiangsu	0.784	0.082	0.33	0.037	9.38	4.83	2.26	1.83	4.57	2.03	0.55	0.178	2.18	3.01
Zhejiang	0.579	0.007	7.18	0.054	21.8	8.23	3.67	4.27	4.39	0.174	0.321	0.061	0.863	0.12
Shangdong	0.558	0.166	0.328	0.127	6.12	6.4	4.69	6.38	0.877	0.248	0.72	0.152	0.817	6.29
Hubei	0.326	0.155	0.686	0.081	32.4	3.74	1.39	0.272	0.129	2.6	0.148	0.158	1.16	5.48
Sichuan	0.404	0.017	0.418	0.056	11.6	10.9	2.18	0.655	1.02	0.022	0.016	0.362	15	0.364
Guangxi	0.112	0.144	0.351	0.079	37.6	3.32	4.59	1.88	0.948	0.399	0.626	0.086	0.866	0.678
Hunan	3.56	1.06	2.05	0.018	27	4.93	2.22	2.1	0.967	2.95	1.1	0.107	0.482	1.7
Guangdong	0.819	0	0.237	0.052	47.2	0.894	3.04	1.74	0.869	0.174	1.16	0.168	1.44	0.211
Guizhou	0.785	0.154	1.64	0.03	10.9	11.3	1.95	0.816	0.812	0.102	0.448	0.112	0.554	2.97
Mean	0.546	0.092	0.978	0.052	18.1	5.17	2.86	1.75	1.89	1.26	0.44	0.145	1.66	1.86
Median	0.365	0.007	0.384	0.049	11.2	3.63	2.24	1.61	0.967	0.427	0.391	0.112	0.84	0.899

RfD	na	na	na	na	1.5×10^{4}[a]	1×10^{4}[b]	7×10^{3}[b]	1.5×10^{4}[c]	7×10^{4}[c]	1.5×10^{3}[d]	1×10^{4}[b]	na	1.3×10^{3}[d]	1×10^{5}[b]

The concentrations below LOD were treated as zero.

na: RfD value is not available.

[a] EU Commission, IUCLID Dataset, 2000.[1]

[b] U.S. Environmental Protection Agency Regional Screening Levels (RSLs) (2017).[2]

[c] From ref.[3]

[d] From ref.[4]

Table S5-3. The lower bound dietary intake of OPEs by the "standard Chinese man" (ng/kg bw/day).

Province	Σ_4Cl-OPEs	$\Sigma_5$alkyl-OPEs	$\Sigma_3$aryl-OPEs	Σ_2Br-OPEs	∑OPEs
Heilongjiang	6.87	1.85	4.09	0.264	8.32
Liaoning	8.82	1.56	64.7	0.028	73
Hebei	7	2.31	24.9	0.094	31.6
Beijing	9.95	2.11	28.7	0.337	36.7
Jilin	10.2	9.61	5.13	0.451	18.6
Shanxi	10.2	10.1	1.88	0.549	20.8
Shaanxi	3.25	1.95	1.42	0.069	6.05
Henan	3.15	5.11	7.86	0.243	15.4
Ningxia	3.72	3.98	11.8	0.417	18.7
Inner Mongolia	6.78	2.48	7.85	0.822	16.3
Qinghai	5.49	3.7	43	0.509	51.2
Gansu	5.13	0.944	13.28	0.165	18.9
Shanghai	13	4.03	42.28	4.92	60.3
Fujian	12.2	1.6	14.5	1.36	24.2
Jiangxi	29.3	6.73	6.51	0.434	41.7
Jiangsu	8.96	6.55	16.1	0.412	29.9
Zhejiang	16.2	1.25	27	7.19	48.1
Shangdong	17.6	7.97	7.82	0.494	29.3
Hubei	5.48	8.71	33.7	0.841	47.3
Sichuan	13.8	1.17	27.5	0.435	40.8
Guangxi	9.88	1.9	39.4	0.495	47.1
Hunan	9.26	9.41	28.5	3.12	48.1
Guangdong	5.72	2.53	49.5	0.237	55.1
Guizhou	14.1	4.42	12.3	1.8	30.7
Mean	9.84	4.25	21.7	1.07	34.1
Median	9.11	3.12	15.3	0.443	31.1

The concentrations below LOD were treated as zero.

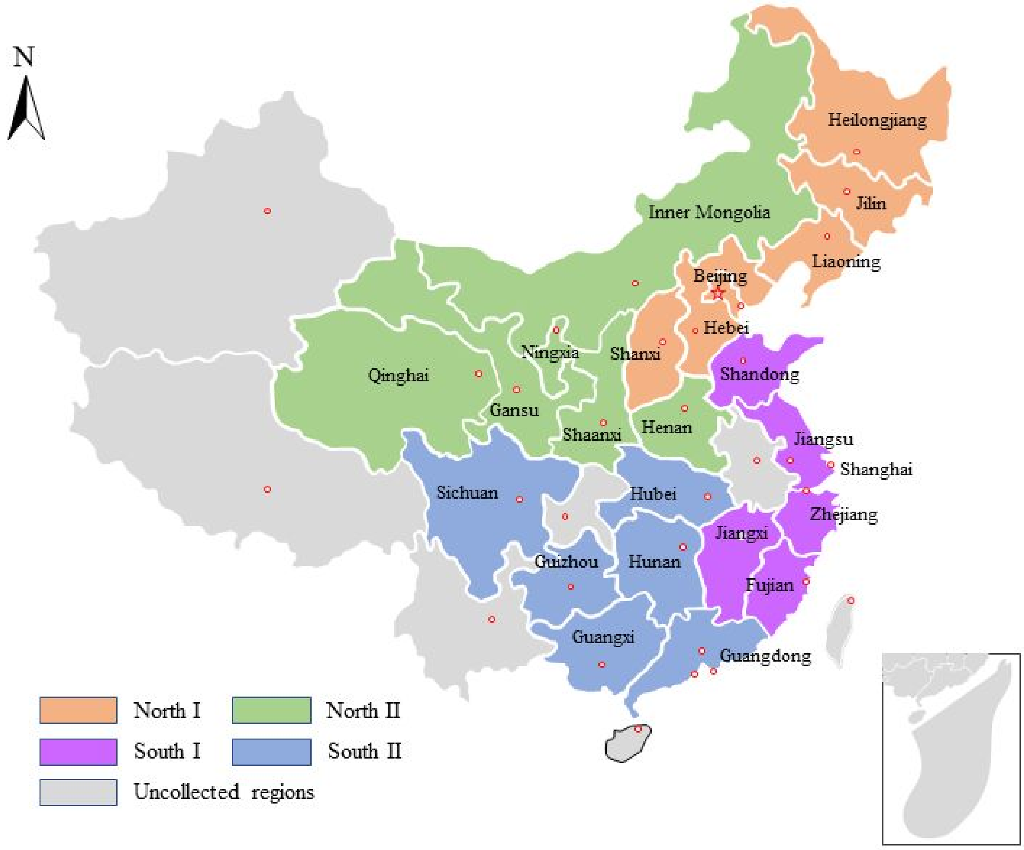


Figure S1. The covered provinces in the 6th Chinese TDS (the 24 provinces included Heilongjian (HLJ), Jilin (JL), Liaoning (LN), Hebei (HeB), Beijing (BJ), Shanxi (SX), Inner Mongolia (IM), Ningxia (NX), Qinghai (QH), Gansu (GS), Shaanxi (SaX), Henan (HN), Jiangxi (JX), Fujian (FJ), Shanghai (SH), Zhejiang (ZJ), Jiangsu (JS), Shandong (SD), Hubei (HuB), Sichuan (SC), Guangxi (GX), Guizhou (GZ), Hunan (HuN), Guangdong (GD))

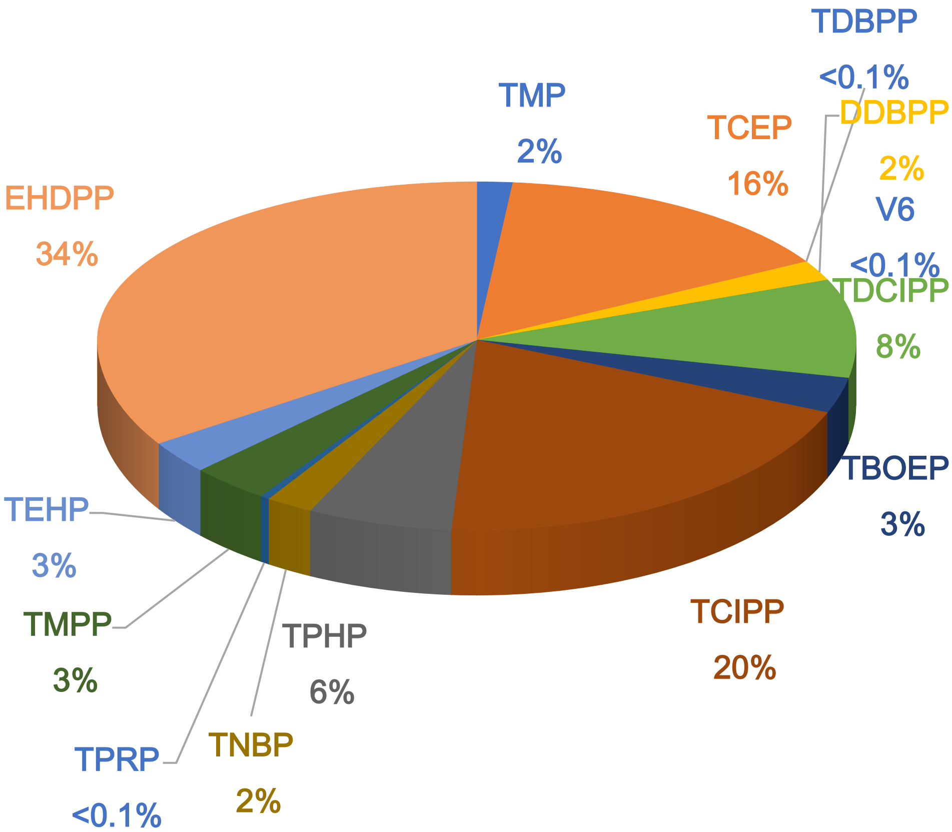


Figure S2. Profile of OPEs in all food composites (the ratio of median level of each analyte in total median levels)

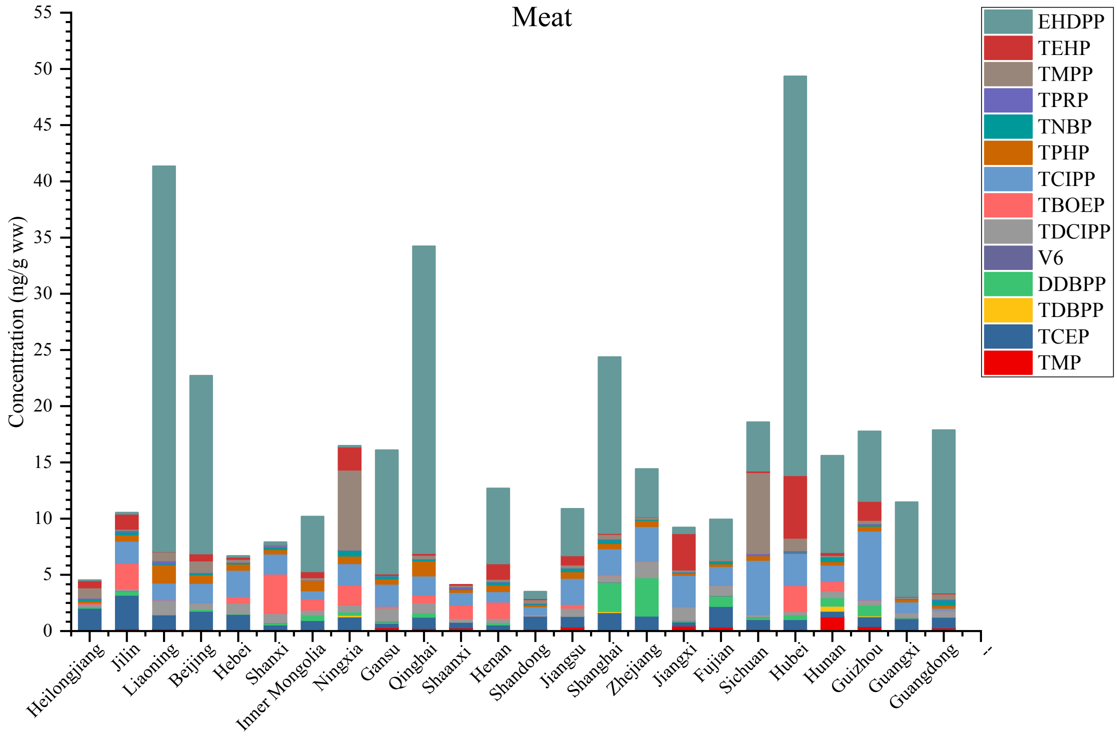


Figure S3-1. Concentrations and profiles of OPEs in meat from various areas

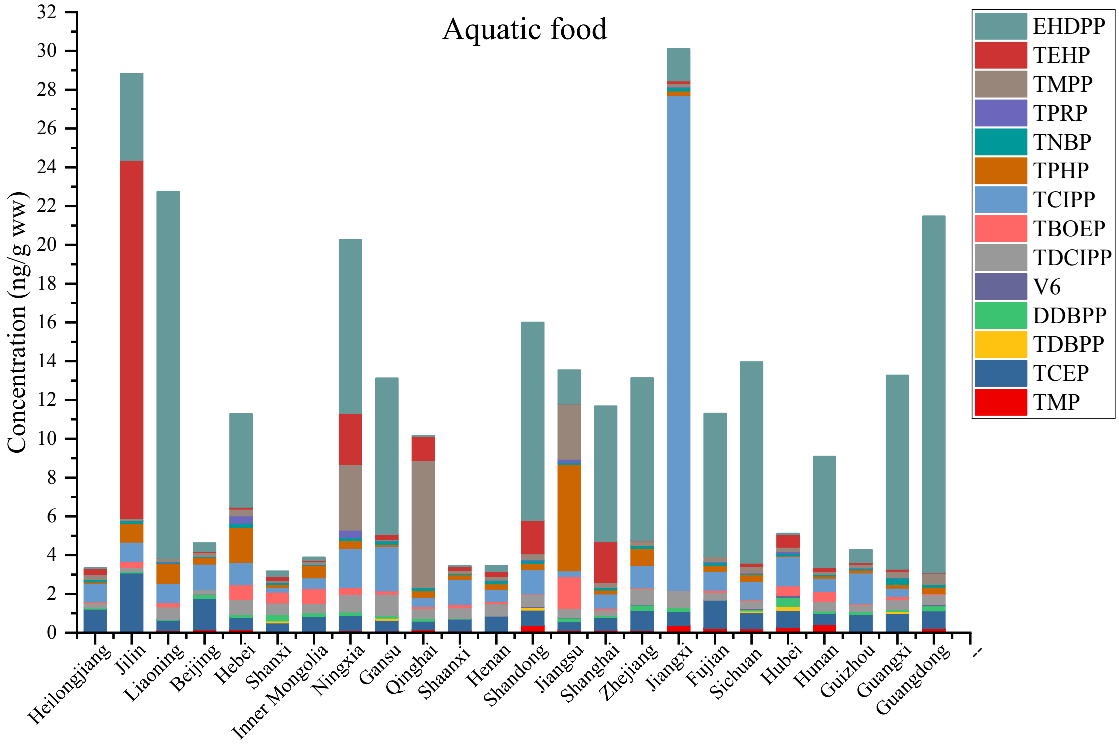


Figure S3-2. Concentrations and profiles of OPEs in aquatic food from various areas

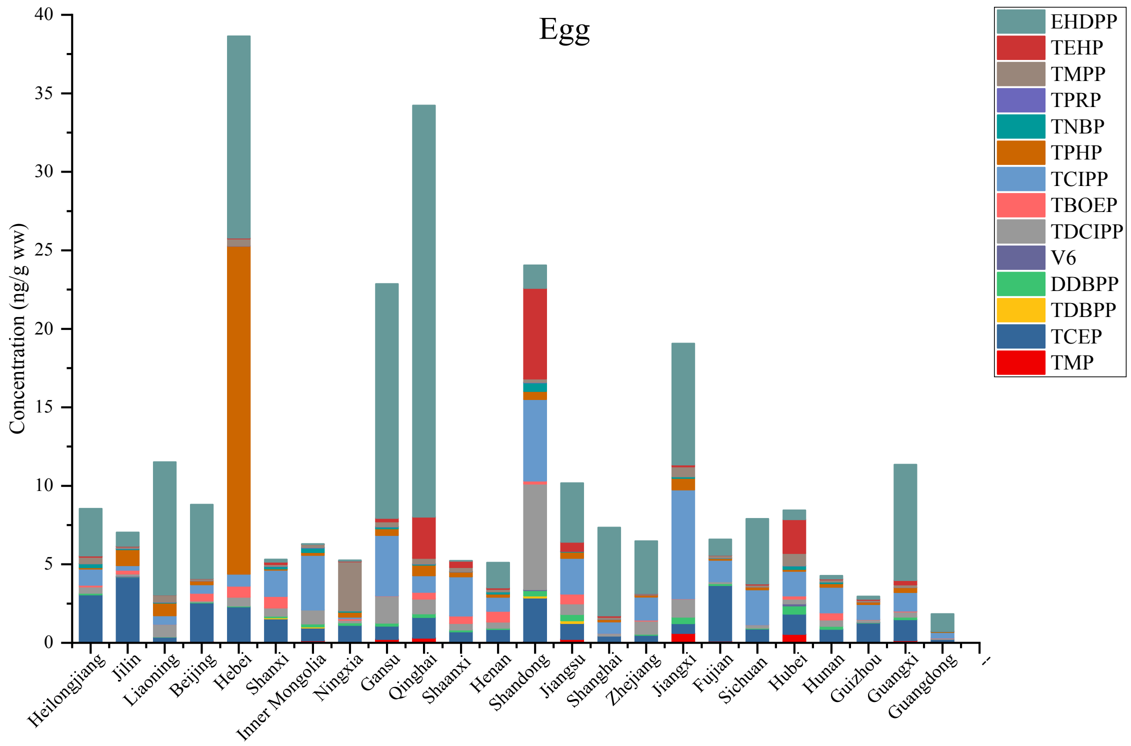


Figure S3-3. Concentrations and profiles of OPEs in egg from various areas

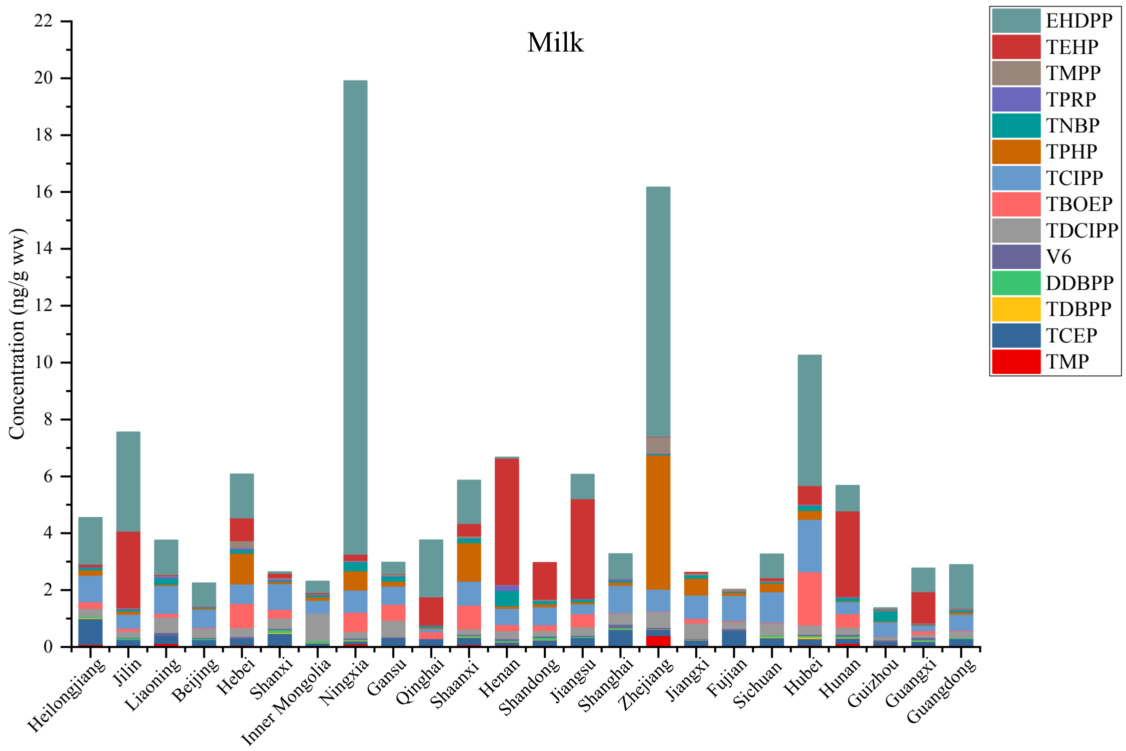


Figure S3-4. Concentrations and profiles of OPEs in milk from various areas

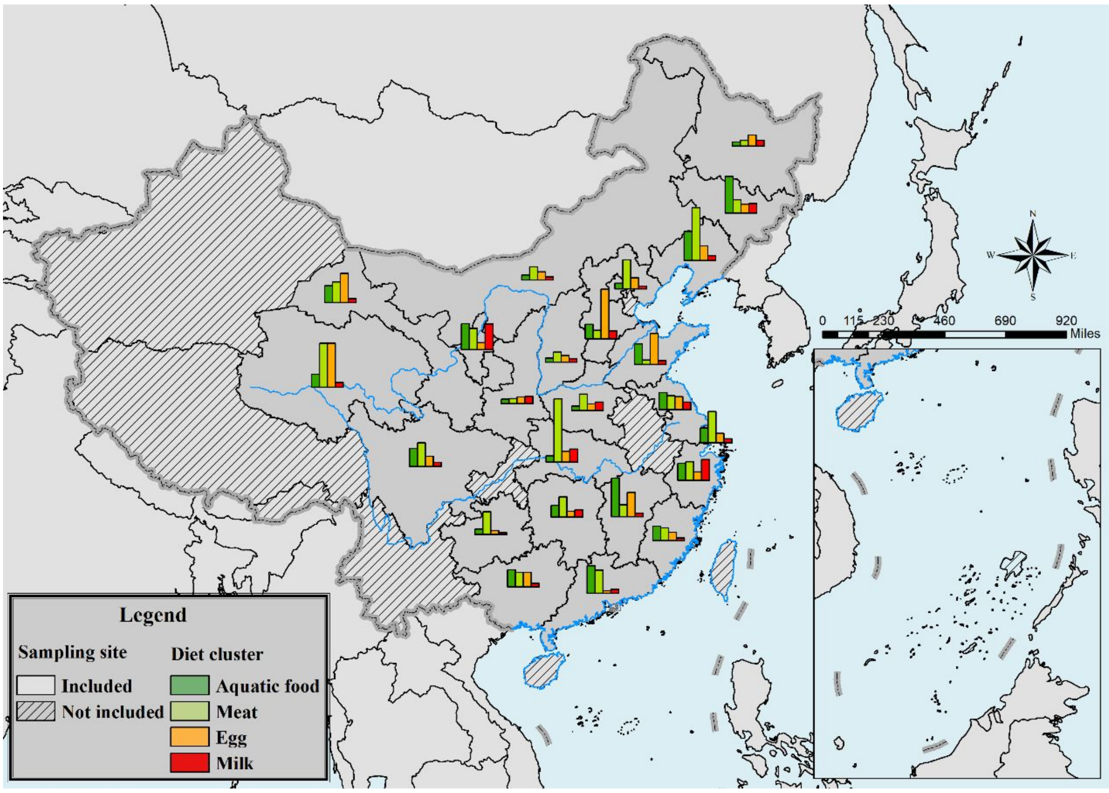


Figure S4. ∑OPEs levels in animal-origin foods from 24 provinces

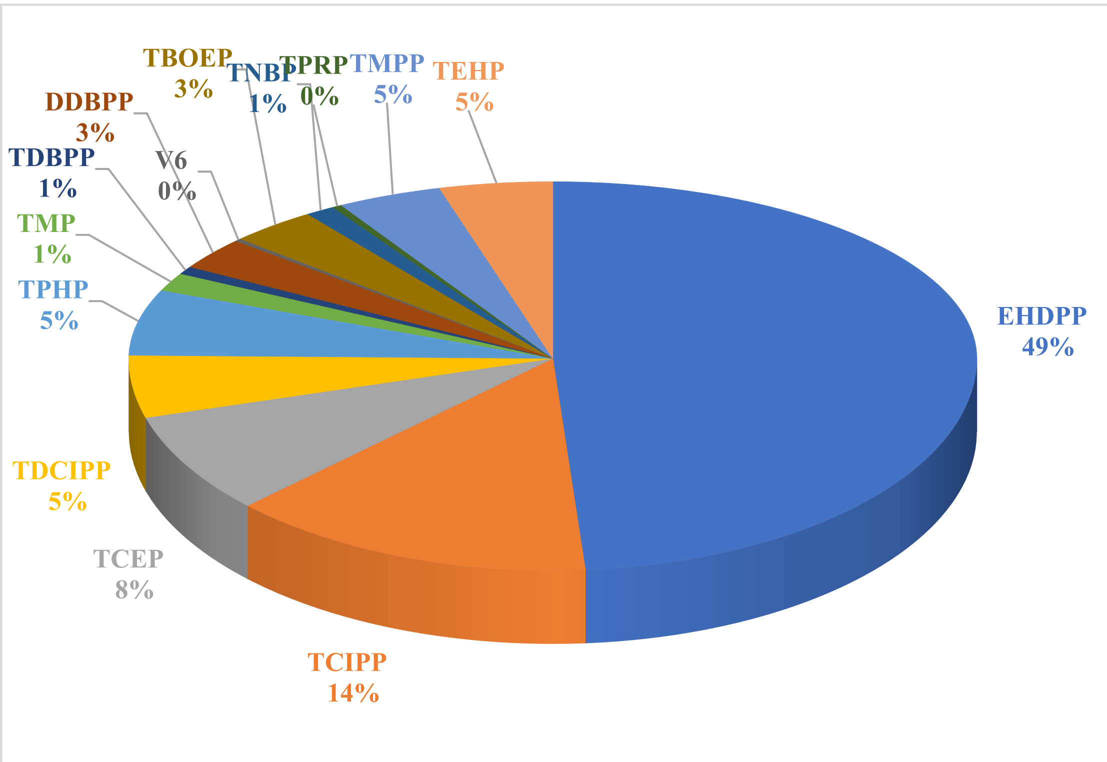


Figure S5. The proportion of EDI of each OPE in total mean upper EDI

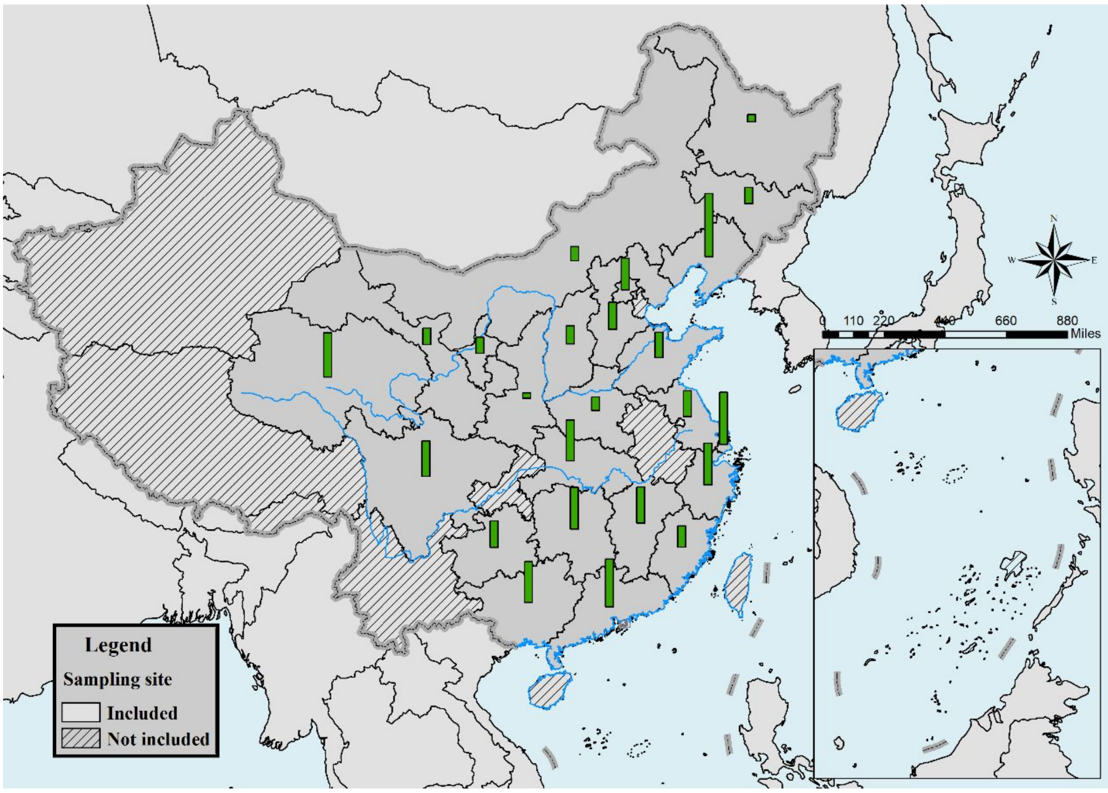


Figure S6. EDI of ∑OPEs in 24 provinces

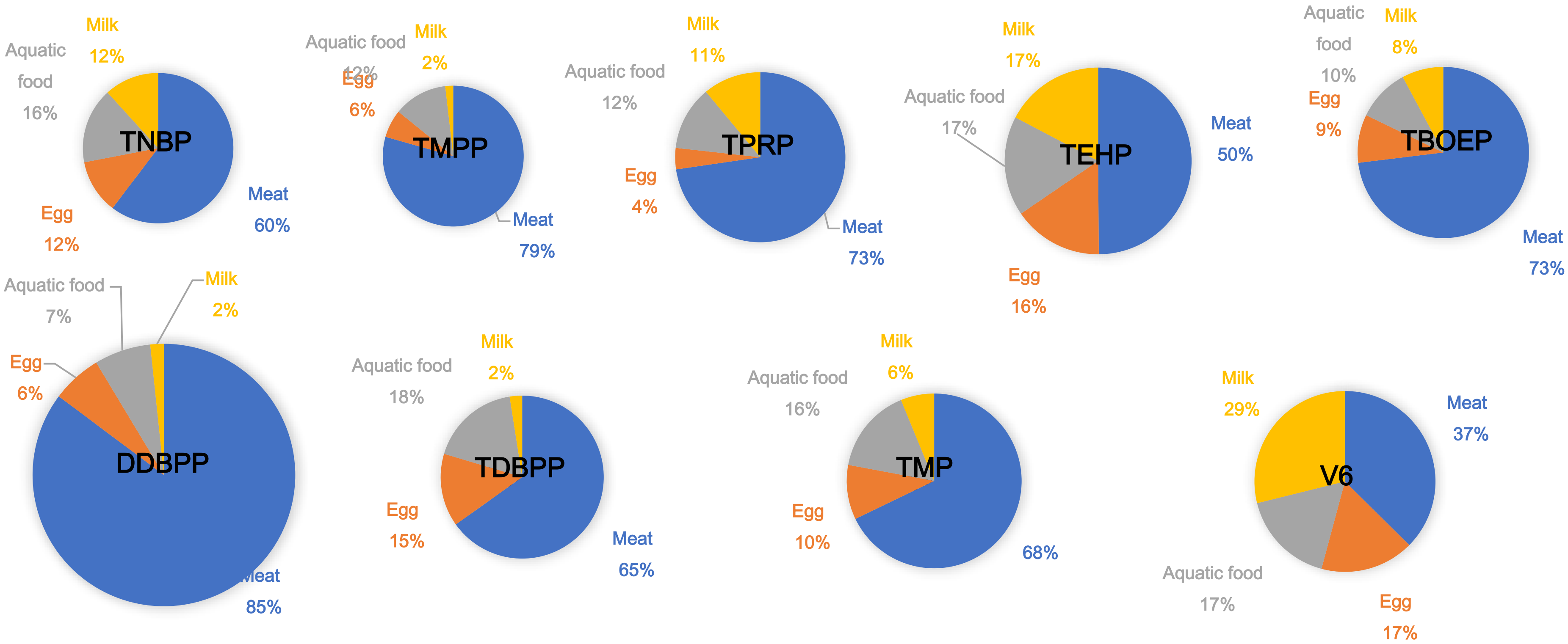


Figure S7. EDI of OPEs via 4 animal-origin food groups

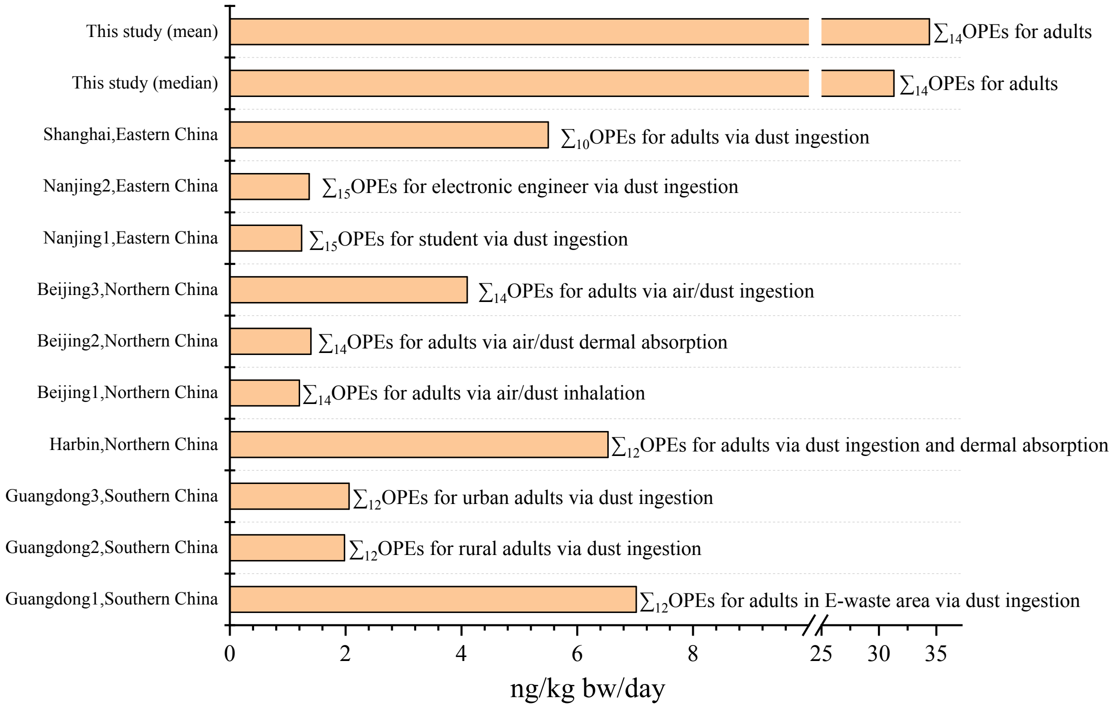


Figure S8. A comparison between EDI of OPEs via food consumption and other pathways (data were from studies conducted in Shanghai[5], Nanjing[6], Beijing[7], Harbin[8] and Guangdong[9])

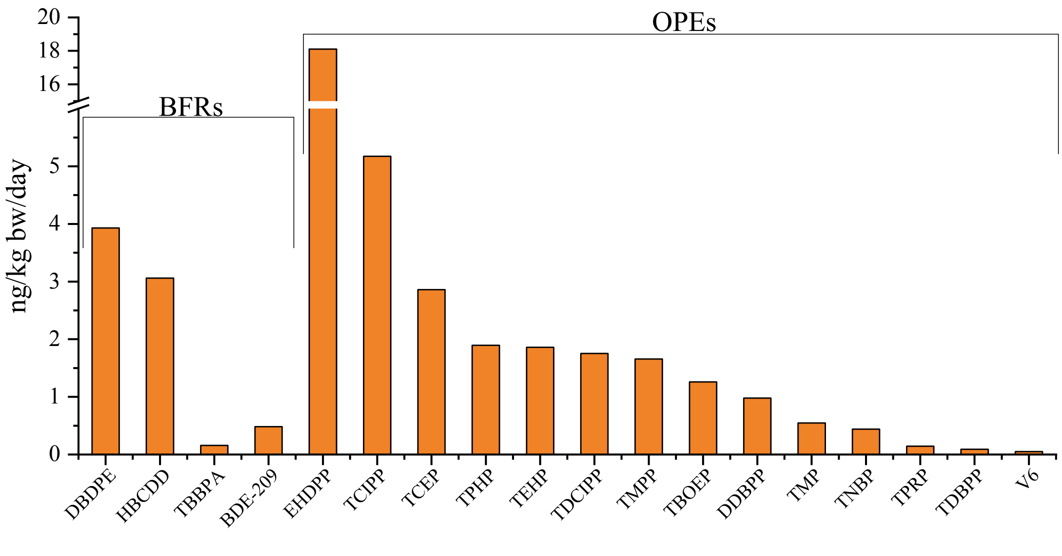


Figure S9. A comparison between mean EDI of OPEs and BFRs in the 6th TDS.

References

1. EC, European Commission/European Chemical Substances Information System (ESIS). IUCLID Dataset, 2-Ethylhexyl diphenyl phosphate (1241-94-7). 2000 (http://esis.jrc.ec.europa.eu).
2. EPA, Risk Assessment: Regional Screening Levels (RSLs). Technical Report; U.S. Environmental Protection Agency: Washington, DC (https://www.epa.gov/risk/regional-screening-levels-rsls-users-guidejune-2017#toxicity).
3. Ali, N.; Dirtu, A. C.; Van den Eede, N.; Goosey, E.; Harrad, S.; Neels, H.; t Mannetje, A.; Coakley, J.; Douwes, J.; Covaci, A., Occurrence of alternative flame retardants in indoor dust from New Zealand: indoor sources and human exposure assessment. *Chemosphere* **2012,** *88*, (11), 1276-82.
4. He, C.; Wang, X.; Tang, S.; Thai, P.; Li, Z.; Baduel, C.; Mueller, J. F., Concentrations of Organophosphate Esters and Their Specific Metabolites in Food in Southeast Queensland, Australia: Is Dietary Exposure an Important Pathway of Organophosphate Esters and Their Metabolites? *Environ. Sci. Tech.* **2018,** *52*, (21), 12765-12773.
5. Peng, C.; Tan, H.; Guo, Y.; Wu, Y.; Chen, D., Emerging and legacy flame retardants in indoor dust from East China. *Chemosphere* **2017,** *186*, 635-643.
6. Zhao, L.; Zhang, Y.; Deng, Y.; Jian, K.; Li, J.; Ya, M.; Su, G., Traditional and emerging organophosphate esters (OPEs) in indoor dust of Nanjing, eastern China: Occurrence, human exposure, and risk assessment. *Sci. Total Environ.* **2020,** *712*, 136494.
7. Cao, D.; Lv, K.; Gao, W.; Fu, J.; Wu, J.; Fu, J.; Wang, Y.; Jiang, G., Presence and human exposure assessment of organophosphate flame retardants (OPEs) in indoor dust

and air in Beijing, China. *Ecotox. Environ. Safe.* **2019,** *169*, 383-391.

8. Sun, Y.; Liu, L. Y.; Sverko, E.; Li, Y. F.; Li, H. L.; Huo, C. Y.; Ma, W. L.; Song, W. W.; Zhang, Z. F., Organophosphate flame retardants in college dormitory dust of northern Chinese cities: Occurrence, human exposure and risk assessment. *Sci. Total Environ.* **2019,** *665*, 731-738.

9. He, C. T.; Zheng, J.; Qiao, L.; Chen, S. J.; Yang, J. Z.; Yuan, J. G.; Yang, Z. Y.; Mai, B. X., Occurrence of organophosphorus flame retardants in indoor dust in multiple microenvironments of southern China and implications for human exposure. *Chemosphere* **2015,** *133*, 47-52.