Supplementary Material

Long-term investigation of spatial-temporal variations, risk assessments, and controlling factors of persistent organic pollutants (HCHs, DDTs, and PAHs) and heavy metals in coastal regions along the Chinese Bohai Sea

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Supplementary Table 1. The summary of heavy metals concentrations (μg L^{-1}) in seawater of the coastal regions along the Chinese Bohai Sea and other coastal systems.

Areas	As	Cd	Cu	Hg	Pb	Zn	References
Coastal regions along the Chinese	2.72±1.01	0.25±0.11	3.32±0.80	0.06±0.02	1.09±0.83	19.41±10.78	This study
Yellow River Estuary	0.43-1.40	0.10-3.22	0.10-4.46	0.004-0.028	0.22-1.33	12.00-81.84	(Tang et al., 2010)
Lianyungang Port	ND	0.131	1.93	ND	0.21	12.9	(Ge et al., 2010)
Jiaozhou Bay	ND	1.13	3.48	0.0228±0.0139	22.72	48.93	(Chen et al., 2023; Ling, 2010)
Yalu River Estuary	1.60-2.05	0.83-1.33	1.80-4.70	0.006-0.049	0.40-1.80	9.20-19.60	(Li et al., 2017)
Yangtze River Estuary	3.6	0.387	1.99	0.172	0.9	6.1	(Sun et al., 2009)
Luoyuan Bay	9.31	2.50	5.58	0.04	0.16	4.99	(Qu et al., 2009)
Hangzhou Bay	1.9	0.13	1.76	0.042	ND	ND	(Li et al., 2013)
Belgian coastal region	ND	0.21	3	--	0.14	12	(Gao et al., 2013)
Coast of Saronikos Gulf, Greece	ND	0.32	2.80	--	2.85	4.21	(Ladakis et al., 2007)
Port Jackson, Australia	ND	0.01-0.10	0.93-2.55	--	ND	ND-9.66	(Hatje et al., 2003)
Bohai Sea	1.5±0.3	0.31±0.12	1.9±0.8	0.0077±0.0039	1.1±0.4	ND	(Chen et al., 2022; Wang and Wang, 2007)
Yellow Sea	ND	0.14	0.8	0.0057±0.0031	0.35	3.8	(Chen et al., 2022; Ci et al., 2011; Tian et al., 2009)
East China Sea	ND	0.02	1.40	0.0015	0.13	1.2	(Wang et al., 2016; Wang, 2008)
Background value of natural surface seawater	0.01-0.04	0.01	0.01-0.04	--	0.005-0.015	0.01	(Wu and Zeng, 1983)

Supplementary Table 2. The summary of HCHs, DDTs, and PAHs concentrations (mg kg^{-1}) in sediment of the coastal regions along the Chinese Bohai Sea and other coastal systems.

Areas	HCHs	DDTs	PAHs	References
Coastal regions along the Chinese Bohai Sea	0.023±0.032	0.003±0.002	0.26±0.10	This study
Yellow River estuary, China			0.15	(Hu et al., 2013)
Haihe estuary, China		ND-0.00034		(Zhao et al., 2010)
East China Sea estuaries, China		ND-0.005	0.032-0.17	(Adeleye et al., 2016)
South west coast of India		0.0007	0.18	(Sreedevi and Harikumar, 2023)
Hugli estuary, India	0.0001-0.0004	0.00034-0.0019	0.27	(Guzzella et al., 2005)
Gulf of Batabano, Cuba		0.00014		(Alonso-Hernandez et al., 2014)
Hainan Island, China		0.0028		(Qiu et al., 2019)
Mumbai coast, India		0.00053		(Shete et al., 2009)
Singapore coast		0.0008		(Zhang et al., 2015)
Moorea island		0.00076		(Fey et al., 2019)

Supplementary Table 3. The summary of heavy metals concentrations (mg kg^{-1}) in sediment of the coastal regions along the Chinese Bohai Sea and other coastal systems.

Areas	As	Cd	Cu	Hg	Pb	Zn	References
Coastal regions along the Chinese Bohai Sea	10.38±4.41 3.89-24.60	0.20±0.08 0.06-0.42	14.35±4.37 8.50-27.90	0.02±0.02 0.003-0.12	16.70±6.20 9.24-38.40	39.93±11.51 12.50-59.90	This study
Yellow River Estuary	6.5-12.6	0.11-0.2	21-38	0.03-0.05	11.6-15.0	21-38	(Wu et al., 2007)
Haihe Estuary	ND	0.4-2.1	21-246	ND	21.7-78.9	ND	(Liu et al., 2006)
Jiaozhou Bay	2.04-20.1	0-1	4.45-211	0.005-4.1	12.3-109	10-384	(He et al., 2013)
Qingdao coastal	8.4	0.1	22	0.05	27.6	58	(Wang et al., 2007)
Yalu River Estuary	7.1	0.24	93.15	0.07	29.6	108.4	(Gao et al., 2008)
Lianyungang coastal	8.4	0.1	36	0.05	14.8	ND	(He et al., 2008)
Yangtze River Estuary	ND	0.261	30.7	0.12	27.3	94.3	(Zhang et al., 2009; Zhao et al., 2008).
Luoyuan Bay	9.28	0.075	21.2	0.041	29.5	90.86	(Chen, 2011; Qu et al., 2009)
Hangzhou Bay	8.27	0.112	23.2	0.080	26.3	78.4	(Cai, 2015)
western Gulf of Thailand	3.78	0.09	12.25	0.02	21.35	45.02	(Liu et al., 2016)
Mediterranean coast, Morocco	--	0.12	6.92	0.19	22.47	98.72	(Saddik et al., 2019)
Persian Gulf, Iran	--	0.8	32.1	--	48.3	62.5	(Pejman et al., 2017)
Admiralty Bay, Antarctica	5.6	0.4	64	0.02	4.8	59	(Trevizani et al., 2016)
Masan Bay, Korea	--	1.24	43.40	--	43.97	206.26	(Hyun et al., 2007)

Supplementary Table 4. The summary of heavy metals in the organisms (mg kg^{-1}) of the coastal regions along the Chinese Bohai Sea and other coastal systems.

Areas	Organisms	As	Cd	Cu	Hg	Pb	Zn	References
Coastal regions along the Chinese Bohai Sea	arthropod	2.69±0.0.62	0.15±0.03	10.16±1.41	0.01±0.002	0.30±0.04	18.58±0.77	This study
	mollusk	1.82±0.61	0.19±0.16	4.61±3.61	0.01±0.006	0.33±0.09	12.36±3.20	
	fish	1.52±1.51	0.07±0.003	0.31±0.20	0.02±0.005	0.20±0.03	4.01±1.99	
Jiaozhou Bay	fish	ND	ND	0.13-1.02	ND	0.095-1.81	1.42-10.59	(Sun and Yang, 1992; Zhang and Zhang, 2015)
	molluscs	ND	ND	1.12-14.46	ND	0.18-2.13	10.60-23.79	
	shellfish	ND	ND	1.45-7.57	0.001-1.096	0.36-0.80	11.58-12.86	
Lianyungang coastal	fish	ND	0.01-0.36	0.37-0.71	0.05-0.32	0-0.43	ND	(Yan, 1993)
Zhejiang coastal	fish	ND-7.6	0.001-0.131	0.10-1.95	0.002-0.032	0.008-0.142	3.6-42.4	(Fang et al., 2004)
	shellfish	0.7-2.4	0.067-13	0.87-135	0.009-0.044	0.029-0.348	5.42-244	
	shrimp	1.1-9.2	0.046-1.12	1.43-23.4	0.004-0.028	0.006-0.244	5.49-15.4	
Yangtze River Estuary	fish	ND	0.091	0.17	0.012	0.52	4.75	(Yi and Zhang, 2012)
Luoyuan Bay	fish	0.45	0.01	0.2	0.032	0.19	12.44	(Qu et al., 2009)
	shrimp	0.5	ND	3.1	0.023	0.04	9.07	
	shellfish	0.7	1.28	39.53	0.081	0.06	15.88	(Li et al., 2013)
Hangzhou Bay	oyster	1.2	1.70	123.05	0.032	1.59	ND	(Li et al., 2013)
Nantong coastal	fish	0.46-14.31	0-3.84	0.06-30.7	0.02-0.18	0.01-0.41	0-68.65	(Liu et al., 2017)

Supplementary Table 5. Temporal variations of heavy metals concentrations in seawater and surface sediment of the coastal regions along the Chinese Bohai Sea.

Matrixes	Year	As	Cd	Cu	Hg	Pb	Zn	HCHs	DDTs	PAHs
	2015	2.81±1.11	0.25±0.10	3.40±0.79	0.06±0.02	1.14±0.94	18.55±11.03			
Water	2016	2.40±0.96	0.23±0.08	3.05±0.69	0.06±0.01	1.23±0.85	20.44±8.86			
μg L^{-1}	2017	2.44±0.85	0.23±0.12	3.27±0.88	0.05±0.02	0.98±0.50	27.96±11.78			
	2018	2.89±0.39	0.32±0.10	3.18±0.80	0.05±0.01	0.83±0.13	14.53±2.74			
	2015	16.78±4.57	0.22±0.10	17.26±5.65	0.02±0.01	21.34±5.76	44.84±3.71	0.004±0.003	0.003±0.003	0.15±0.08
Sediment	2016	10.27±4.13	0.18±0.04	13.04±2.69	0.01±0.01	13.91±2.40	35.54±13.67	0.011±0.006	0.003±0.003	0.24±0.10
mg kg^{-1}	2017	8.64±2.60	0.19±0.06	12.80±2.45	0.03±0.03	12.97±1.98	37.84±9.84	0.027±0.026	0.003±0.002	0.30±0.08
	2018	8.26±1.42	0.21±0.11	15.41±5.31	0.02±0.02	20.36±7.98	43.38±12.33	0.042±0.047	0.004±0.001	0.30±0.07

Supplementary Table 6. Summary of I_{geo} and E_i^r of heavy metals in sediment of the coastal regions along the Chinese Bohai Sea.

Heavy metal	I_{geo}	E_i^r
As	2.65 ±11.73 (-30.65-65.43)	16.58 ±7.04 (6.21-39.90)
Cd	0.61 ±0.35 (0.32-2.52)	185.12 ±76.17 (59.25-390.94)
Cu	-3.97 ±18.81 (-144.65-23.46)	4.68 ±1.42 (2.77-9.09)
Hg	-0.24 ±2.82 (-9.35-15.54)	41.79 ±40.53 (6.69-250.53)
Pb	-2.64 ±9.08 (-56.65-14.97)	6.22 ±2.31 (3.44-14.30)
Zn	-0.76 ±0.23 (-1.26--0.33)	0.58 ±0.17 (0.18-0.87)

Supplementary Table 7. Spearman analysis of heavy metals with environmental parameters in seawater of the coastal regions along the Chinese Bohai Sea.

Spearman analysis	As	Cd	Cu	Hg	Pb	Zn
depth	0.13**	-0.09*	-0.06	-0.34**	0.08	0.12**
T	0.12**	0.00	-0.13**	-0.36**	0.14**	0.18**
pH	0.12**	-0.18**	-0.24**	0.03	0.17**	0.16**
S	-0.20**	0.20**	0.27**	-0.11*	-0.22**	-0.14**
DO	-0.04	0.03	0.09*	0.30**	-0.14**	-0.14**
COD	0.27**	-0.21**	-0.01	0.17**	0.24**	0.13**
PO_4^{3-}	-0.32**	0.17**	0.18**	-0.08	-0.16**	-0.13**
NO_2^-	0.03	0.14**	-0.07	-0.05	-0.08	-0.10*
NO_3^-	-0.13**	0.04	0.07	0.10*	0.05	0.01
NH_4^+-N	-0.31**	0.05	-0.01	0.04	-0.10*	-0.03
TN	-0.15**	0.13**	0.10*	0.02	0.07	-0.02
TP	-0.41**	0.34**	0.12**	0.06	-0.26**	-0.31**
SiO_4^{3-}	0.19**	-0.01	-0.20**	-0.08	0.03	-0.04
SPM	-0.21**	0.28**	0.05	0.15**	-0.14**	-0.23**
Chl-*a*	0.33**	-0.30**	-0.20**	-0.06	0.33**	0.26**

Supplementary Table 8. Pearson analysis of HCHs, DDTs, PAHs, and heavy metals with parameters in surface sediment of the coastal regions along the Chinese Bohai Sea.

	TOC	Sulfide	Eh	As	Cd	Cu	Hg	Pb	Zn	HCHs	DDTs	PAHs
TOC	1.00											
Sulfide	-0.16	1.00										
Eh	0.11	0.20	1.00									
As	0.30**	0.41**	0.07	1.00								
Cd	0.41**	-0.22	-0.28	0.04	1.00							
Cu	0.36**	-0.06	-0.30	0.35**	0.37**	1.00						
Hg	0.59**	0.09	-0.04	0.16	0.65**	0.05	1.00					
Pb	0.34**	-0.05	-0.28	0.42**	0.26*	0.59**	0.01	1.00				
Zn	0.39**	0.15	0.02	0.31**	-0.02	0.31**	0.04	0.16	1.00			
HCHs	0.31**	-0.08	-0.03	-0.32*	0.39**	0.04	0.05	-0.00	-0.16	1.00		
DDTs	0.29**	-0.15	0.13	-0.09	0.10	0.11	0.41**	0.11	-0.07	0.49**	1.00	
PAHs	0.39**	-0.41**	0.09	-0.56**	0.37**	-0.08	-0.15	-0.07	-0.26	0.26	0.15	1.00

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