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Source-resolved atmospheric metal emissions, concentrations, and deposition fluxes into the East Asian seas

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Section S1. Technical details of the modified CMAQ.

For the revisions of modules for simulate metals in CMAQ, we modified desid_vars.F to add metals emission information. To have tracers undergo the same diffusion, advection, and deposition as PM, we modified AERO_DATA.F, CMAQ_Control_DESID_cb6r5_ae7_aq.nml, AE_cb6r5_ae7_aq.nml files.

Section S2. Technical details of the WRF/CMAQ.

The WRF physics scheme configuration included the Pleim-Xiu land surface model (Xiu and Pleim, 2001), the Rapid Radiative Transfer Model (RRTMG) for shortwave and longwave schemes (Clough et al., 2005), the Asymmetric Convective Model version 2 for the PBL scheme (Pleim, 2007), the Kain–Fritsch cumulus scheme (Kain, 2004) for cumulus parameterization, and the MODIS land cover data (20 categories) employed in this study. In the CMAQ model, two grids on WRF lateral boundary were removed, and thus there were 257×117 grids in d01.

Section S3. Establishment of emission inventory of pollutants other than metals.

For SO_2, NO_x, CO, nonmethane volatile organic compounds (NMVOCs), PM_{10}, $PM_{2.5}$ and NH_3, we categorized them into land anthropogenic and ship sources. For anthropogenic sources, we used the Multiresolution Emission Inventory for China (MEIC) in 2020 (Li et al., 2017a; Zheng et al., 2018; Zheng et al., 2021) in mainland China and the MIX emission data in 2010 (Li et al., 2017b) in Asia excluding mainland China. For ship sources, we used A bottom-up ship emission model based on the Automatic Identification System (AIS) data to calculate the emission inventories of these species. Detailed information on the establishment of the ship emission model can be found in the previous studies (Chen et al., 2017; Fan et al., 2016).

Figure S1. The simulation area.

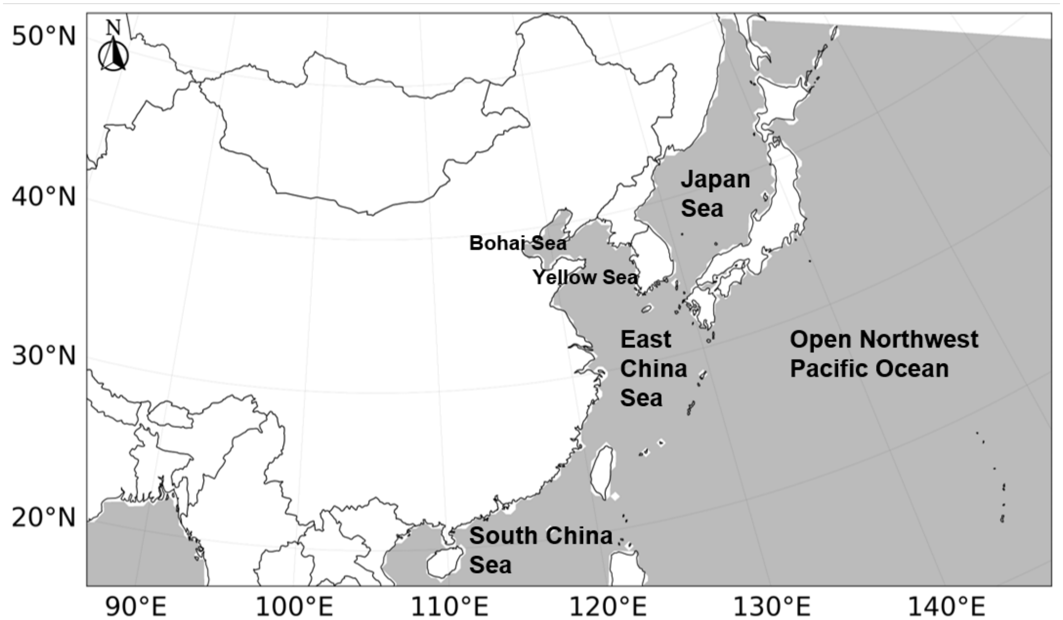

Figure S2. Relative contributions of land anthropogenic, ship, and dust sources to fine mode (a), coarse mode (b) emissions of the six metals (Fe, Al, V, Ni, Zn, Cu) in spring (March-April-May).

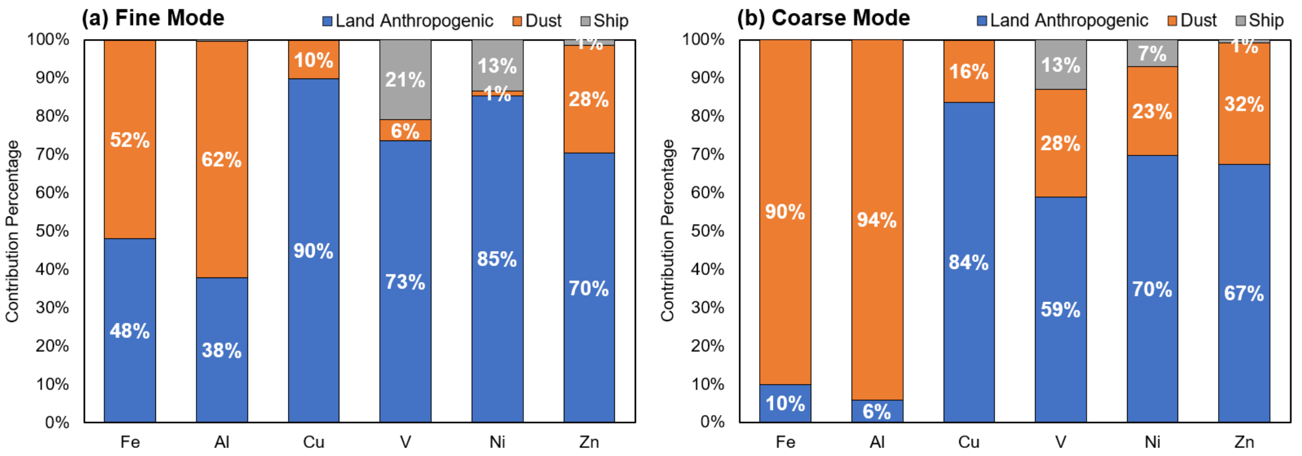

Figure S3. Absolute and relative contributions of seasonal mean concentrations of Fe (a) and Al (b) in different sea areas from land anthropogenic, ship, and dust sources (units: $ng \cdot m^{-3}$), the numbers on top of the stacked bar graphs represent total seasonal mean concentrations from three sources.

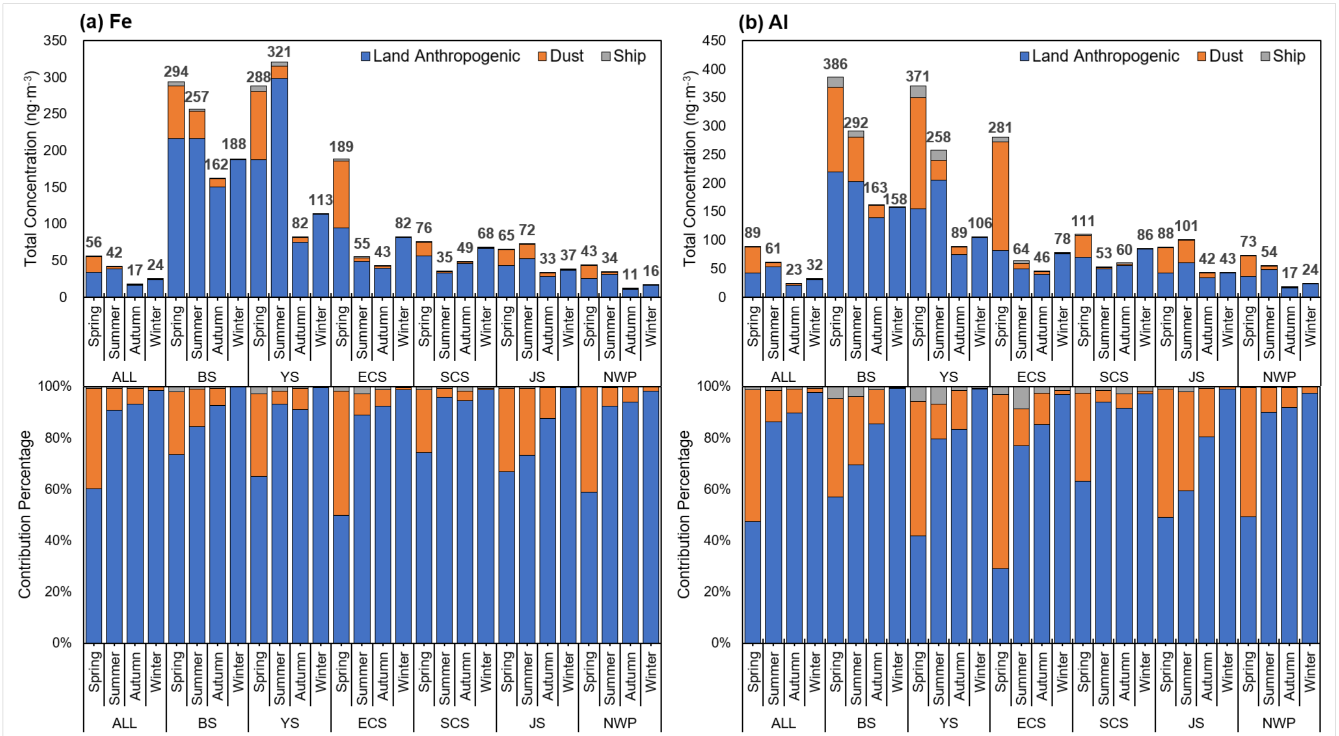

Figure S4. Comparison of simulated daily concentrations of V (a) and Ni (b) with observations at the Pudong site (31.2331° N, 121.5447° E, Shanghai, China).

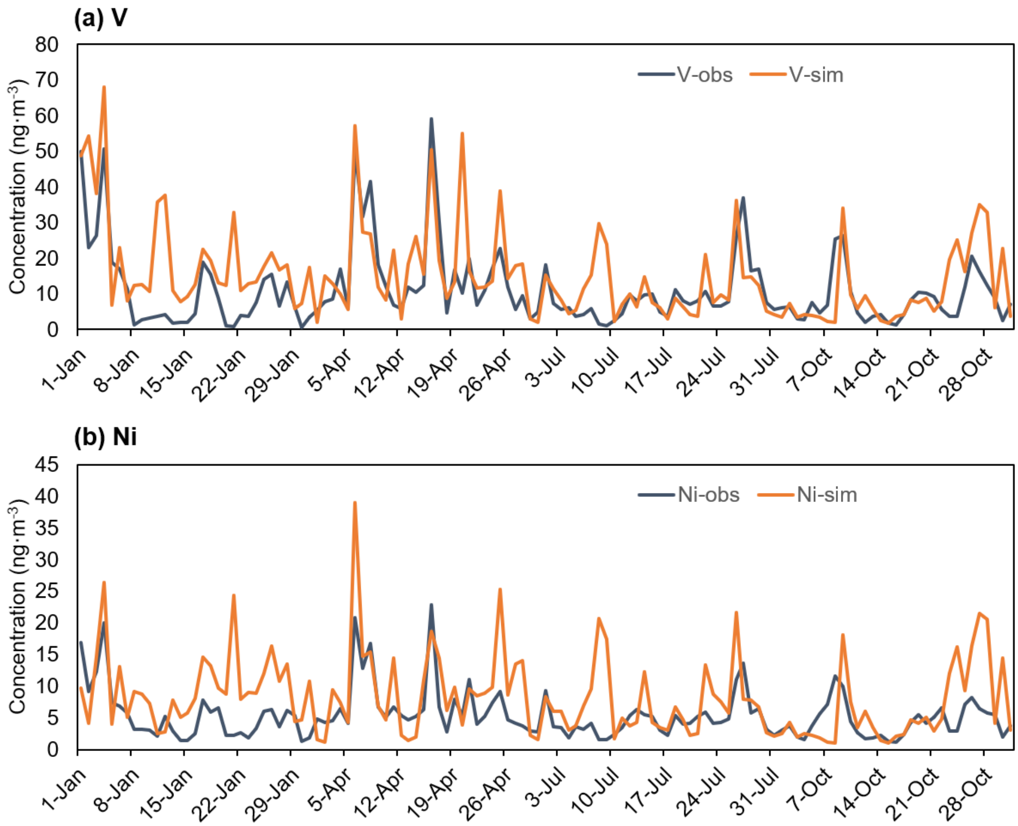

Figure S5. Spatial distribution of the contributions of land anthropogenic, dust sources and ship sources to the seasonal average concentrations of Fe (a-c), Al (d-f), V (g-i), Ni (j-l), Zn (m-o), and Cu (p-r) in the sea area, respectively (36 km × 36 km resolution) in the year of 2017.

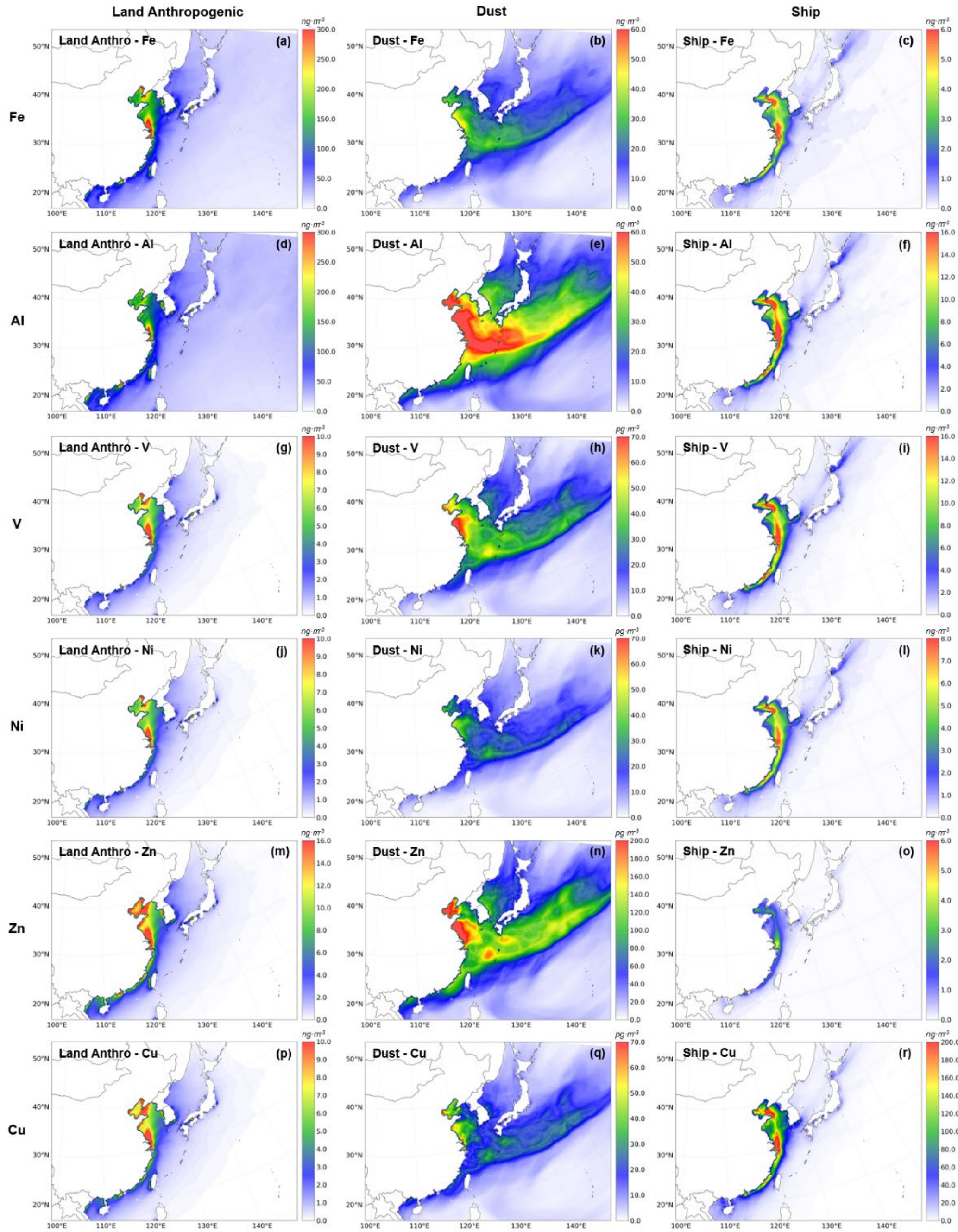

Figure S6. Spatial distribution of depositional fluxes of Fe (a), V (b), Zn (c), Al (d), Ni (e), Cu (f) in the sea area (36 km × 36 km resolution, considering all emission sources) in the year of 2017.

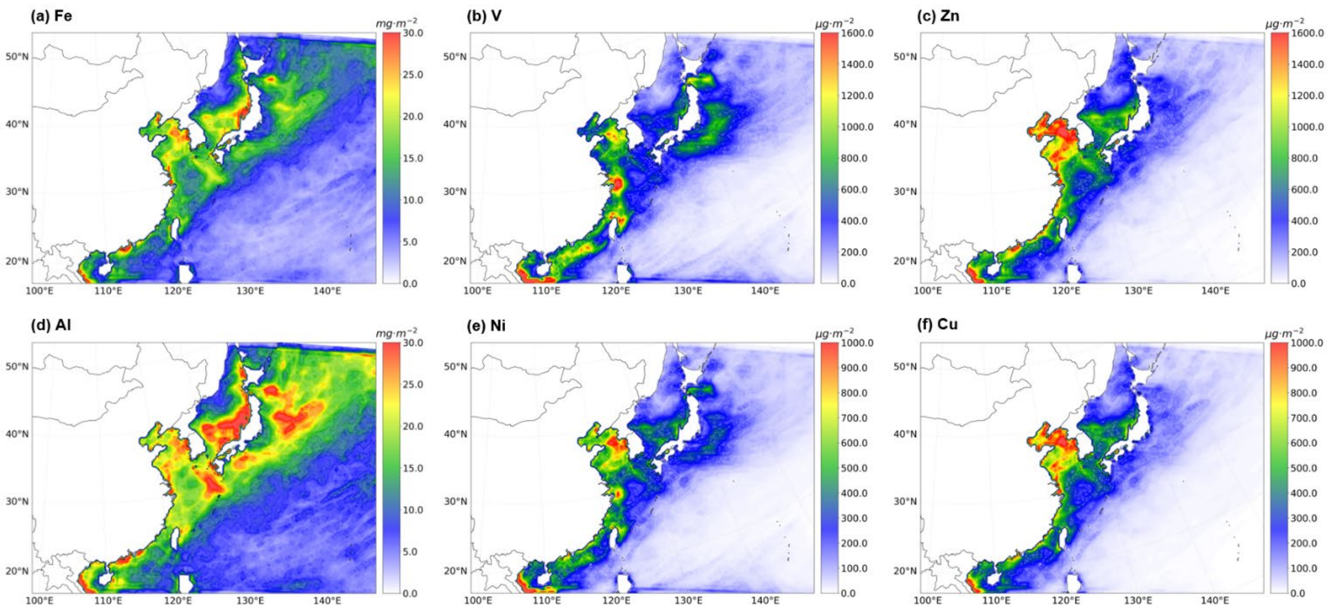

Figure S7. Absolute and relative contributions of soluble iron deposition fluxes from land anthropogenic, ship, and dust sources in different sea areas, fine mode (a), coarse mode (b) (units: $\mu g \cdot m^{-2} \cdot year^{-1}$), the numbers on top of the stacked bar graphs represent total deposition fluxes from three sources.

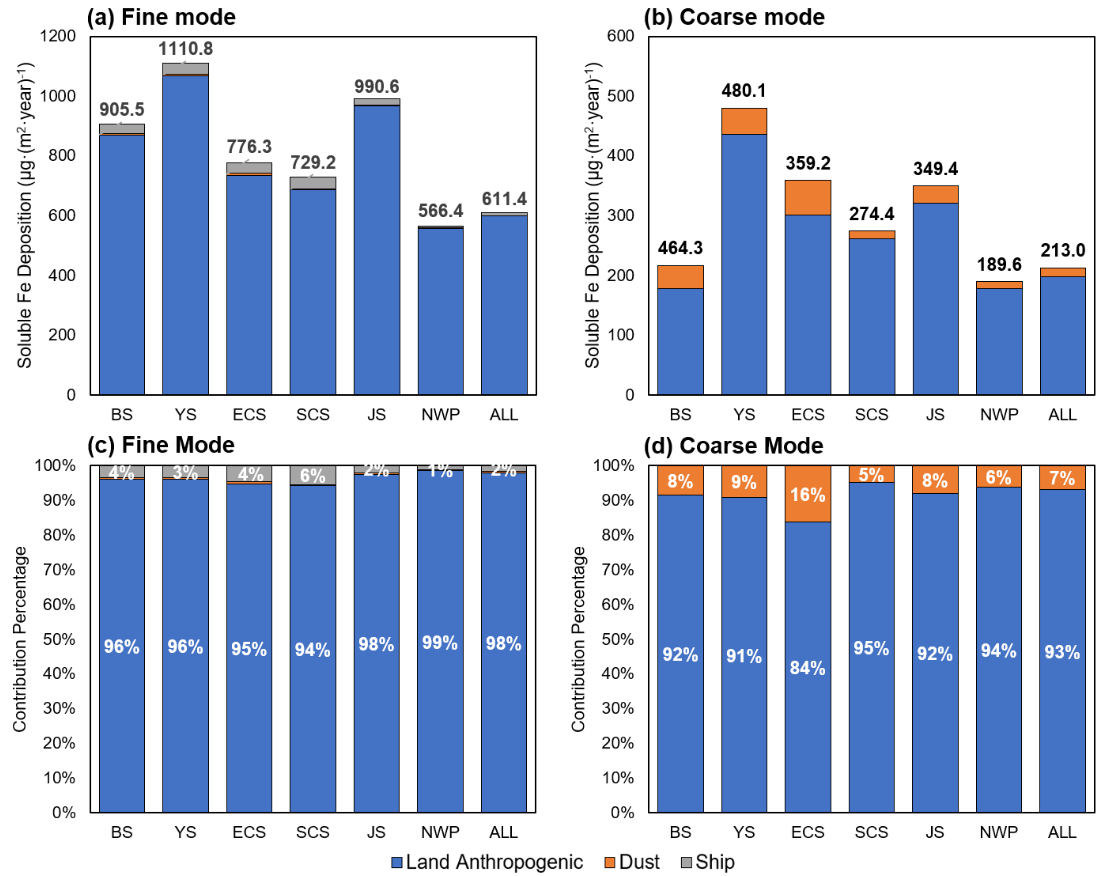

Figure S8. Evolution of the relative contributions of the land anthropogenic, ship, and dust sources to emissions, atmospheric concentrations, and deposition fluxes of Fe (a), V (b), Zn (c), Al (d), Ni (e), Cu (f) for the month of April (Concentrations and depositional fluxes labelled "Ocean" in the figure were for the oceans only, and concentrations and depositional fluxes labelled "Land" were for land only).

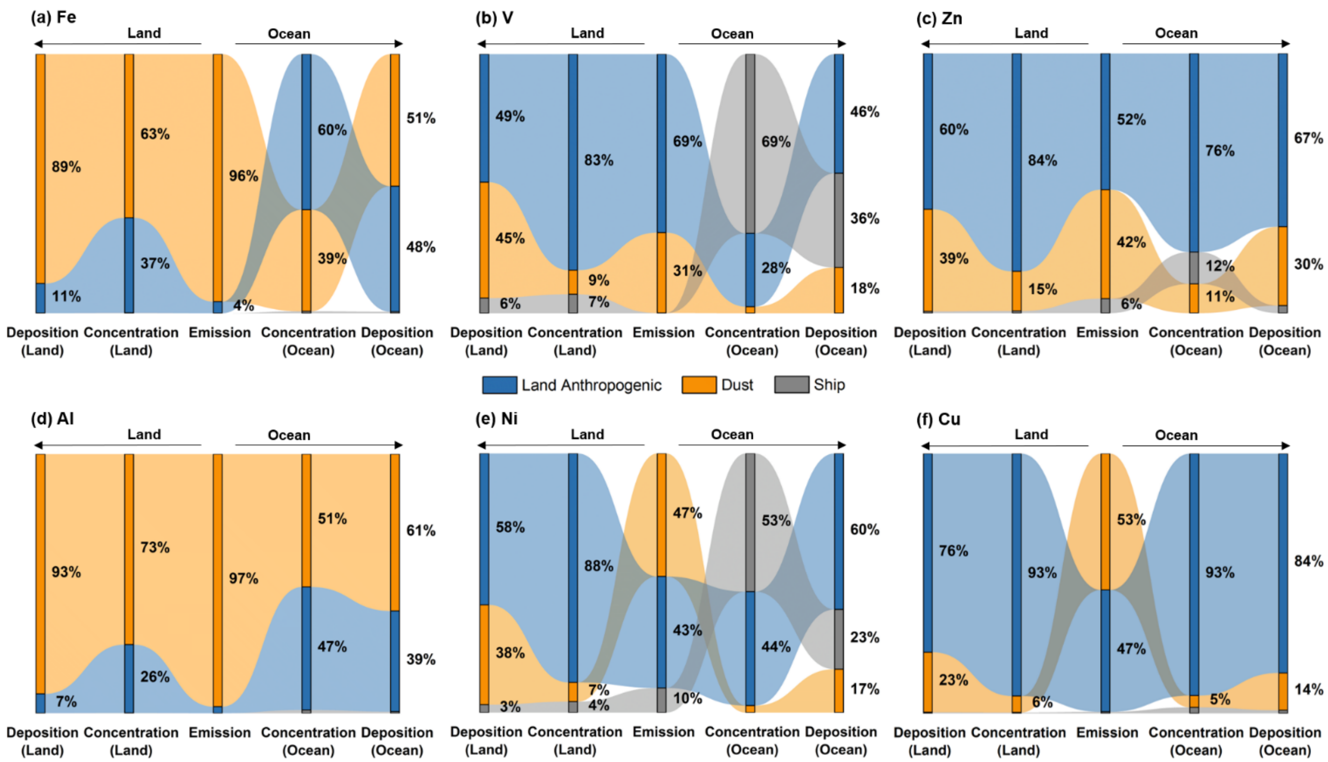

Table S1. Emission factors for ships in previous studies (Unit: g·kWh^{-1})

Engine type	Fuel Type	Sulfur content (%)	Cu	Fe	V	Ni	Zn	Al	Source
ME SSD	HFO	2.70	1.80E-05	4.90E-03	1.94E-02	9.50E-03	1.30E-04	6.10E-04	(Celo et al., 2015)
ME SSD	HFO	2.85	5.00E-05	6.75E-03	1.99E-02	8.69E-03	2.00E-04	1.87E-02	(Agrawal et al., 2008a)
ME SSD	HFO	2.05	1.01E-04	3.89E-03	5.60E-02	1.27E-02	1.43E-04	1.72E-02	(Agrawal et al., 2008b)
ME MSD	HFO	1.48	3.00E-06	4.10E-04	6.80E-03	2.43E-03	1.00E-04	2.30E-03	(Celo et al., 2015)
ME MSD	HFO	2.21	3.00E-06	2.40E-03	1.23E-02	5.50E-03	8.00E-05	4.90E-04	
ME MSD	HFO	2.33	1.35E-05	2.22E-04	1.54E-03	2.91E-04	5.96E-05	1.37E-04	(Corbin et al., 2018)
ME MSD	HFO	0.58		1.21E-04	4.27E-04	1.06E-03	6.50E-05	8.36E-05	(Moldanová et al., 2013)
ME MSD	HFO	0.96		2.46E-04	1.69E-03	1.50E-03	6.63E-05	2.08E-05	
ME MSD	HFO	2.70	1.49E-04	1.27E-03	1.43E-02	4.10E-03	5.80E-04	1.79E-04	(Sippula et al., 2014)
ME MSD	HFO	0.68	3.25E-05	1.55E-03	8.55E-06	1.62E-05	1.31E-04		(Zhang et al., 2018a)
ME MSD	HFO	1.60	3.60E-04	5.30E-03	7.90E-03	2.00E-03	1.30E-03	2.60E-04	(Streibel et al., 2017)
ME MSD	HFO	0.48		1.62E-02	3.60E-02	2.75E-01	1.08E-02		(Zetterdahl et al., 2016)
ME MSD	MDO	0.10		8.69E-06	3.80E-05				(Moldanová et al., 2013)
ME MSD	MDO	0.13		2.29E-06	1.25E-04	1.70E-06			(Zhang et al., 2016)
ME HSD	MDO	0.08		5.08E-05	4.25E-05	7.77E-05			
AE MSD	MGO	0.03		3.06E-06	1.43E-06	3.06E-06	2.45E-04	1.98E-04	(Moldanová et al., 2013)
AE MSD	MGO	0.06	2.00E-05	1.40E-04	1.50E-04	4.00E-05	3.10E-04	3.00E-04	(Agrawal et al., 2008a)

Table S2. Emission factors for ships used in this study (Unit: g·kWh^{-1})

Engine type	Fuel Type	Sulfur content	Cu	Fe	V	Ni	Zn	Al
ME SSD	HFO	2.7%	4.74E-05	5.12E-03	1.94E-02	9.50E-03	1.88E-04	1.77E-02
ME MSD	HFO	2.7%	7.27E-05	1.27E-03	1.24E-02	4.21E-03	3.03E-04	3.89E-04
	MDO	0.5%	1.67E-04	3.18E-04	2.66E-04	9.77E-05	3.33E-03	2.90E-03
ME HSD	MDO	0.5%	1.67E-04	6.09E-04	6.37E-04	1.92E-04	3.33E-03	2.90E-03
AE MSD	MGO	0.5%	1.67E-04	6.09E-04	6.37E-04	1.92E-04	3.33E-03	2.90E-03

Table S3. Dust metal emission factors in previous studies and applied in this study (Unit: %)

	Cu	Fe	V	Ni	Zn	Al	Source
PM3	1.54E-04		9.22E-05	4.78E-05	1.35E-04	7.67E-02	(Li et al., 2022)
Coarse	1.59E-05		6.33E-05	2.18E-05	7.16E-05	6.00E-02	
Coarse	4.71E-05	2.44E-02	6.95E-05	5.10E-05	1.20E-04		(Luo et al., 2022)
Coarse	3.00E-05	2.35E-02	6.38E-05	4.20E-05	8.70E-05		
Coarse	4.77E-05	3.78E-02		7.03E-05	1.93E-04	4.09E-02	(Abbasi et al., 2021)
Coarse	3.80E-05	3.80E-02		7.00E-05	1.45E-04	4.10E-02	
Coarse		2.86E-02				6.03E-02	(Baker et al., 2020)
Coarse	3.41E-05	3.84E-02		2.91E-05	9.31E-05	7.58E-02	(Nishikawa et al., 2013)
Coarse	5.00E-05	7.70E-02	5.00E-04	1.00E-04	1.00E-04	8.40E-02	(Desboeufs et al., 2005)
Coarse	1.00E-05	2.44E-02	3.00E-05	6.70E-05	1.60E-04	7.84E-02	
Fine	1.77E-04	3.36E-02	1.16E-04	5.70E-05	1.32E-03	5.70E-02	Applied in this study
Coarse	3.36E-05	3.24E-02	6.36E-05	5.10E-05	9.93E-05	6.86E-02	

Table S4. Solubility of metals in previous studies and applied in this study (Unit: %)

Mode		Cu	Fe	V	Ni	Zn	Al	Source
Fine	Total	0.6146	0.232	0.6842	0.41	0.8608	0.1404	(Jiang et al., 2014)
		0.1643		0.4342	0.1314	0.6678		(Wang et al., 2015)
		0.349	0.046		0.068	0.413		(Liu et al., 2022)
		0.5769	0.4667	0.421	0.568	0.7		(Karthikeyan et al., 2006)
		0.51	/	0.43	0.49	0.73	0.14	Applied in this study
Coarse	Total	0.41	0.027	-	-	0.816	0.0472	(Jiang et al., 2014)
		0.51	0.077	0.55	0.26	0.84	0.051	(Hsu et al., 2010)
		0.47	0.012	0.262		0.463		(Wang et al., 2015)
		0.41	0.274	0.326	0.284	0.423		(Karthikeyan et al., 2006)
		0.14	0.066		0.12	0.57	0.45	(Jickells et al., 2016)
		0.45	/	0.38	0.27	0.62	0.049	Applied in this study
Fine	Dust		0.01					(Kurisu et al., 2021)
	Land		0.08					(Ito, 2015)
	Ship		0.65					
Coarse	Dust		0.017					(Ooki et al., 2009)
	Land		0.025					(Baker et al., 2020)

Table S5. Monthly metal emissions from ship sources in 2017 (Unit: tons·month^{-1})

Month	Cu	Fe	V	Ni	Zn	Al
Jan	1.1825	34.6850	129.0917	61.7265	18.2189	121.8235
Feb	1.1118	31.7941	118.4216	56.1886	17.2285	111.6897
Mar	1.3335	36.7252	136.4392	64.6687	20.8952	129.2200
Apr	1.4518	41.3125	153.5250	72.8817	22.5648	145.2409
May	1.4068	43.3444	162.0243	77.0775	21.3147	151.9547

Jun	1.2169	36.9126	137.9572	65.5797	18.5189	129.4294
July	1.5311	45.1684	169.1841	80.2542	23.4339	158.3524
Aug	1.5608	43.9847	164.3985	77.8443	24.2096	154.3517
Sep	1.2467	34.1564	127.2538	60.1975	19.5106	119.8588
Oct	1.5415	43.1018	161.1650	76.1933	23.9349	150.8402
Nov	1.6353	45.4896	170.3909	80.4884	25.3904	159.1439
Dec	1.6341	46.1274	172.7453	81.6644	25.2818	161.3140
Sum	16.8529	482.8020	1802.5965	854.7649	260.5023	1693.2192

Table S6. Monthly fine mode metal emissions from land anthropogenic sources in 2017 (Unit: tons·month^{-1})

Month	Cu	Fe	V	Ni	Zn	Al
Jan	626.35	11077.53	566.88	517.94	1373.93	13178.09
Feb	538.29	10242.82	523.12	453.34	1112.31	11625.46
Mar	551.13	10499.15	516.35	449.68	1115.10	12020.72
Apr	543.95	10296.52	539.68	457.48	1069.83	11301.25
May	533.11	10285.04	530.53	448.49	1056.53	11501.22
Jun	529.13	10342.16	527.65	446.97	1055.52	11647.78
July	517.53	10288.07	535.65	445.04	1053.97	12082.21
Aug	511.01	10185.41	513.17	429.63	1047.99	12167.32
Sep	527.01	10223.80	505.84	432.51	1051.61	11659.44
Oct	549.53	10486.76	542.89	458.17	1091.11	11977.92
Nov	557.35	10560.46	541.44	464.38	1127.93	12093.87
Dec	594.54	10952.06	551.70	490.91	1251.36	12789.79
Sum	6578.94	125439.79	6394.90	5494.52	13407.19	144045.08

Table S7. Monthly coarse mode metal emissions from land anthropogenic sources in 2017 (Unit: tons·month^{-1})

Month	Cu	Fe	V	Ni	Zn	Al
Jan	461.51	7255.44	139.25	879.44	685.35	8324.76
Feb	493.37	6678.36	147.77	354.98	714.29	8011.84
Mar	534.38	6955.81	150.49	302.30	750.38	8052.39
Apr	564.46	6757.91	154.73	230.91	775.96	8500.91
May	547.67	6644.82	152.76	208.55	768.33	8327.12
Jun	534.57	6585.42	151.23	208.17	766.45	8269.86
July	512.16	6792.39	149.71	209.89	758.49	8023.75
Aug	503.29	6787.56	148.34	208.26	743.85	7694.13
Sep	540.66	6842.74	151.52	208.03	766.84	7932.16
Oct	565.57	7131.48	154.42	235.22	784.64	8390.68
Nov	539.02	7079.40	151.42	312.63	757.12	8270.60
Dec	499.86	7126.84	145.26	563.84	713.47	8339.15
Sum	6296.51	82638.16	1796.88	3922.21	8985.18	98137.34

Table S8. Monthly fine mode metal emissions from dust sources in 2017 (Unit: tons·month^{-1})

Month	Cu	Fe	V	Ni	Zn	Al
Jan	3.65	680.75	2.43	0.41	26.38	1155.55
Feb	21.75	4054.75	14.50	2.42	157.11	6882.81
Mar	2.27	423.72	1.52	0.25	16.42	719.25
Apr	142.17	26499.65	94.78	15.80	1026.81	44982.26
May	36.11	6730.91	24.07	4.01	260.81	11425.49
Jun	5.84	1088.05	3.89	0.65	42.16	1846.92
July	43.86	8174.67	29.24	4.87	316.75	13876.23
Aug	10.20	1901.40	6.80	1.13	73.68	3227.55
Sep	18.53	3453.22	12.35	2.06	133.81	5861.73

Oct	15.09	2811.89	10.06	1.68	108.96	4773.09
Nov	0.88	164.37	0.59	0.10	6.37	279.00
Dec	15.00	2796.45	10.00	1.67	108.36	4746.88
Sum	315.36	58779.82	210.24	35.04	2277.61	99776.76

Table S9. Monthly coarse mode metal emissions from dust sources in 2017 (Unit: tons·month^{-1})

Month	Cu	Fe	V	Ni	Zn	Al
Jan	9.17	8734.20	17.25	13.75	26.69	18492.78
Feb	54.59	52023.79	102.76	81.89	158.96	110149.13
Mar	5.70	5436.48	10.74	8.56	16.61	11510.56
Apr	356.79	339998.86	671.60	535.18	1038.89	719874.11
May	90.62	86359.67	170.59	135.94	263.88	182847.93
Jun	14.65	13959.97	27.58	21.97	42.66	29557.21
July	110.06	104883.59	207.18	165.09	320.48	222068.34
Aug	25.60	24395.50	48.19	38.40	74.54	51652.20
Sep	46.49	44305.93	87.52	69.74	135.38	93808.24
Oct	37.86	36077.48	71.26	56.79	110.24	76386.26
Nov	2.21	2108.86	4.17	3.32	6.44	4465.05
Dec	37.65	35879.35	70.87	56.48	109.63	75966.76
Sum	791.41	754163.67	1489.71	1187.11	2304.39	1596778.58

Table S10. Comparison of metals emission inventories with other studies (Units: tons·year^{-1})

Area	Emission Source	Period	V	V-this study[a]	Ni	Ni-this study	Zn	Zn-this study	Cu	Cu-this study	Source
China	Solid Waste	2013			43.5	27.9	1790.7	2194.0	382.4	185.6	(Wang et al., 2017)[b]
	solid waste incineration		0.27	0.67							
	No-road transport		1247.0	1803.7							
China	Iron and steel production	2017	79.6	109.2							(Bai et al., 2021)[c]
	Domestic coal combustion		10.5	75.6							
	Total		11505.0	16394.8							
China	iron and steel industry	2011			105.0	196.1			448.8	528.5	(Wang et al., 2016)
East Asia	Ship	2015	1329.8	1802.6	580.4	854.8					(Zhao et al., 2021)[e]
China	primary anthropogenic sources	2012			3395.5	5458.0	22319.6	22526.9	9547.6	12793.1	(Tian et al., 2015)
China	primary anthropogenic sources	2017					19473.1	22526.9	9813.1	12793.1	(Liu et al., 2023)

Table S11. Comparison of mean annual concentration of metals in PM2.5 from other studies (Units: ng·m^{-3})

Area	Periods	Longitude	Latitude	Cu	Fe	V	Ni	Zn	Al	Source

East Asia	2017			3.55	112.56	4.38	3.25	33.02	135.78	This study
Shanghai	2018-2019	120.97°E	31.09°N	3.85	194.5	5.54	3.18	40.84	400.1	(Zou et al., 2020)
Beijing	2010	116.30°E	39.99°N	34	1696	4	4	270	1823	(Tao et al., 2016)
Chengdu	2011	104.下·03°E	30.65°N	23	693	1.7	2.5	350	560	(Tao et al., 2014)
Guangzhou	2014	113.35°E	23.12°N	37	353	9	4	225	305	(Tao et al., 2017)
Gunshan	2012-2013	126.71°E	35.98°N	29	237	-	13	113	166	(Jeon et al., 2012)
Handan	2013	114.50°E	36.69°N	18.8	-	5.4	3.4	300	-	(Wei et al., 2014)
Hangzhou	2005	120.15°E	30.28°N	71	-	-	3	130	-	(Liu et al., 2015)
Jinan	2007	116.98°E	36.67°N	30	1300	7.35	9.05	700	890	(Zhang et al., 2018b)
Lanzhou	2012-2013	103.81°E	36.03°N	47.06	-	7.97	9.4	322	-	(Tan et al., 2017)
Nanjing	2013	118.76°E	32.05°N	71.5	902	5.19	16	497	811.5	(Li et al., 2016)
Niigata	2016	138.86°E	37.80°N	2.14	82.4	1.13	6.06	10.9	144	(Li et al., 2018)
Qingdao	2007	120.38°E	36.07°N	20	630	9.99	6.24	270	470	(Zhang et al., 2018b)
Shanghai	2010	121.53°E	31.22°N	15	1328	-	9	236	1542	(Wang et al., 2013)
Taian	2014	117.11°E	36.18°N	40	1055	-	10	220	350	(Liu et al., 2016)
Tianjin	2008	117.20°E	39.12°N	38.9	145.1	2.2	-	339.4	69.8	(Gu et al., 2011)
Tuoji-island	2012	120.76°E	38.17°N	12.6	466	6	4	94.3	-	(Zhang et al., 2014)
Wuhan	2013	114.21°E	30.30°N	25.27	1680	-	4.8	290.78	-	(Zhang et al., 2015)
Wuhan	2014	114.17°E	30.36°N	30.13	1820.76	6.35	3.57	419.21	-	(Acciai et al., 2017)
Xiamen	2015	118.03°E	24.30°N	26	250	10.95	5.37	220	280	(Zhuang, 2016)

Zaozhuang	2007	117.32°E	34.82°N	40	1000	6.11	9.85	1220	630	(Zhang et al., 2018b)
Zhengzhou	2010	113.51°E	34.80°N	24.1	1248.8	3.7	3.3	444.1	579.6	(Geng et al., 2013)
Zhuhai	2014	113.53°E	22.36°N	20	212	12	7	149	309	(Tao et al., 2017)
the East China Sea				58	410	3.8	1.5	51	615	(Hsu et al., 2010)

Table S12. Comparison of mean annual deposition flux of metals from other studies (Units: $mg \cdot m^{-2} \cdot year^{-1}$)

Area	Periods	Cu	Fe	V	Ni	Zn	Al	Source
East Asia	2017	0.55	62.20	0.42	0.47	1.05	119.43	This study
China	2000-2018	12.77			6.61	96.75		(Chen et al., 2022)
North China	2008-2018	16.30			16.40	129.80		(Peng et al., 2019)
South China	2008-2019	18.80			4.89	88.60		
Tokyo Bay	2004-2005	16.00		6.90	6.80			(Sakata et al., 2008)
Kushiro	2008	0.57	70.81		0.72	4.02	146.00	(Okubo et al., 2013)
		0.11	20.55	0.23	0.14	0.02	35.97	
Otsuchi	2008	2.21	204.77		0.74	14.60	356.61	
		0.24	47.39	0.52	0.29	0.05	90.17	
Hedo	2008	0.35	62.05		0.80	5.48	105.12	
		0.33	20.20	0.62	0.47	0.06	33.29	
Pearl River Delta	2001-2002	18.60	555.00	2.09	8.35	104.00		(Wong et al., 2003)
China	2006-2015	11.56			8.08	72.90		(Ni and Ma, 2018)

Spain	2004-2006	0.79	347.00		0.65	6.26	457.00	(Bacardit and Camarero, 2009)
Jiangsu	2019	11.00			3.30	157.00		(Chen et al., 2019)
Northern China	2007-2010	15.10	3957.10	5.35	7.87	106.50	5046.50	(Pan and Wang, 2015)
Guizhou	2018	1.22				30.27		(Lin et al., 2022)
		0.41	30.46	0.80	0.54	0.11	49.08	
Beijing	2016-2020			0.16	0.53	6.18		(Pan et al., 2021)
		0.46	118.69	1.00	0.55	0.11	240.99	
Hunan	2016-2018	6.41			1.57	73.09		(Feng et al., 2019)
		0.41	28.60	0.77	0.55	0.10	47.03	
Mount Emei	2017-2022	1.70	6.93			41.40	6.38	(Fu et al., 2023)
Daya Bay, China	2015-2017	4.67	209.37	2.41	1.91	93.96	477.40	(Wu et al., 2018)
		0.47	29.78	0.95	0.66	0.20	45.07	
Lushan	2011-2012	6.29	0.00	9.99	2.94	112.87	854.40	(Nie et al., 2017)
		0.41	18.24	0.68	0.56	0.06	30.44	

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