Source apportionment, criteria derivation, and health risk assessment of soil

heavy metals in the urban green spaces

Zirui Meng ^{a, b}, Qing Xue ^a, Ziyi Wang ^a, Limin Liang ^a, Xijie Ji ^{a, d}, Xueqiang Lu ^c, Xunqiang Mo ^{a*}, Mengxuan

He ^{a*}

^a School of Geographic and Environmental Science, Tianjin Normal University, Tianjin 300382, China

^b State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

^c College of Environmental Science and Engineering, Nankai University, Tianjin 300071, China

^d Key Laboratory of Industrial Ecology and Environmental Engineering, Ministry of Education, School of Environmental Science and Technology, Dalian University of Technology, Dalian 116024, China

* Authors for correspondence:

Xunqiang Mo

Address: School of Geographic and Environmental Science, Tianjin Normal University, Tianjin 300382, China E-mail: <u>421973@163.com</u>

Mengxuan He

Address: School of Geographic and Environmental Science, Tianjin Normal University, Tianjin 300382, China E-mail: <u>hemengxuannku@126.com</u>

Pages: 19

Supplementary materials: 3 Tables: 12 Figures: 1

USEPA PMF 5.0 was used to quantify the source apportionment of soil heavy metals in this study. The original matrix was decomposed into a contribution matrix and a factor profile matrix based on the reliable factorization algorithms under the non-negative constraint. The calculation of PMF receptor model as follows (USEPA, 2011b):

$$x_{ij} = \sum_{k=1}^{p} g_{ik} \times f_{kj} + e_{ij} \#(1)$$

Where x_{ij} is the concentration of *j*-th soil heavy metals in *i*-th sample (mg kg⁻¹), g_{ik} is the contribution matrix of *k*-th source in *i*-th sample, f_{kj} is the source profile of *j*-th soil heavy metals from *k*-th source factor, e_{ij} is the residual for *j*-th soil heavy metals in *i* number of samples. The optimal g_{ik} and f_{kj} are obtained via minimizing the objective function Q:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} \times f_{kj}}{u_{ij}} \right]^2 = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{e_{ij}}{u_{ij}} \right]^2 \#(2)$$

Where u_{ij} is the uncertainty of *j*-th soil heavy metal in the *i*-th sample. The uncertainty can be calculated as follows:

$$u_{ij} = \begin{cases} \frac{5}{6} \times MDL & x_{ij} \le MDL \\ \sqrt{(\delta \times x_{ij})^2 + (0.5 \times MDL)^2} & x_{ij} > MDL \end{cases}$$
(3)

Where δ is the relative standard deviation of the concentration of soil heavy metals, MDL is the species-specific method detection limit value.

In this study, concentration-specific human health risk for adults and Children were calculated via human health risk assessment recommended by. Soil heavy metals could enter human body through direct ingestion, inhalation, and dermal contract. The algorithm of average daily intakes (AAD) through ingestion, inhalation, and dermal contract were calculated as follows:

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \# (1)$$
$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \# (2)$$
$$ADD_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} 10^{-6} \# (3)$$

Where $^{ADD}_{ing}$, $^{ADD}_{inh}$, and $^{ADD}_{dermal}$ (mg kg⁻¹ day⁻¹) are average daily intakes through direct ingestion, inhalation, and dermal contract absorption, respectively, C is the concentration of soil heavy metals, the mean and values of other parameters were listed in the Table S3.

The hazard index (HI) represented the accumulation non-carcinogenic risk, and it could be assessed as follows (USEPA, 2011b; USEPA, 2009):

$$HI = \sum HQ = \sum \frac{ADD_p}{RfD_p} \#(4)$$

Where HI is the sum of the HQ of all the soil heavy metals, HQ is the non-carcinogenic risk of different pathway, RfD is the reference dose (mg kg⁻¹ day⁻¹), p is the type of heavy metals (Table S4).

The algorithm of total carcinogenic risk (TCR) can be assessed as follows (USEPA, 2011b; USEPA, 2009):

$$TCR = \sum CR = \sum ADD_p \times SF_p \#(5)$$

Where TCR is the sum of the CR of all the soil heavy metals, CR is the carcinogenic risk via different pathway, SF is the carcinogenic slope factor, p is the type of heavy metals (Table S4).

Source-specific human health risk in this study was developed combing with PMF model to assess the health risks contributed by each source. Soil heavy metals could enter human body through direct ingestion, inhalation, and dermal contract. The algorithm of AAD through ingestion, inhalation, and dermal contract combing with PMF were calculated as follows (USEPA, 2016; USEPA, 2011b):

$$ADD_{ijing}^{\ \ k} = \frac{C_{ij}^{k} \times IngR \times EF \times ED}{BW \times AT} \times CF\#(1)$$
$$ADD_{ijinh}^{\ \ k} = \frac{C_{ij}^{k} \times InhR \times EF \times ED}{PEF \times BW \times AT} \#(2)$$
$$ADD_{ijdermal}^{\ \ k} = \frac{C_{ij}^{k} \times SA \times AF \times ABF \times EF \times ED}{BW \times AT} \times CF\#(3)$$

Where ADD_{ijing}^{k} , ADD_{ijinh}^{k} , and $ADD_{ijdermal}^{k}$ are average daily intakes via oral ingestion, inhalation, and dermal contact for the *j*-th heavy metals in the *i*-th sampling site from *k*-th source (mg kd⁻¹ day⁻¹), C_{ij}^{k} is the mass contributions of the *j*-th heavy metals from *k*-th source in the *i*-th sample (mg kg⁻¹), the definition and values of

 C_{ij}^k is estimated as follows:

other parameters are listed in Table S3.

$$C_{ij}^k = C_{ij}^{k*} \times C_i \#(4)$$

Where C_{ij}^k is the mass contribution of *j*-th heavy metals from *k*-th source in the *i*-th sample (mg kg⁻¹), C_{ij}^{k*} is the contribution of *j*-th heavy metals from *k*-th source in the *i*-th sample (mg kg⁻¹), C_i is the concentrations of soil heavy metals in the *i*-th sample (mg kg⁻¹).

The ${}^{HQ}_{ij,P}^{k}$ represents the hazard quotient on the *p*-th exposure pathway from *k*-th source of *j*-th heavy metals in *i*-th samples, and ${}^{HI}_{ij}^{k}$ is the sum of ${}^{HQ}_{ij,P}^{k}$. They are calculated as follows (USEPA, 2011b; USEPA, 2009):

$$HI_{ij}^{k} = \sum HQ_{ij}^{k} = \sum \frac{ADD_{ij,p}^{k}}{RfD_{p}} \#(5)$$

Where RfD is the reference dose of soil heavy metals and listed in Table S3.

The carcinogenic risk (CR_{ij}^k) represented the carcinogenic risk of the *j*-th heavy metal in the *i*-th samples from *k*-th source, and the total carcinogenic risk (TCR_{ij}^k) could be calculated by the sum of CR_{ij}^k . They were calculated as follows:

$$CR_{ij}^{k} = \sum CR_{ij,p}^{k} = \sum ADD_{ij,p}^{k} \times SF_{i,p} \#(6)$$

$$TCR_{ij}^k = \sum CR_{ij}^k \#(7)$$

Where SF is the carcinogenic slope factor of soil heavy metals and listed in Table S4.

 Table S1. Data introduction and sources.

Basic data	Data Source	Туре
Digital Elevation Model (30 m)	Geospatial Data Cloud (https://www.gscloud.cn)	Raster
Land use (30 m)	Globel land 30 (http://globeland30.org)	Raster

Environmental indices	Categories									
Geo-accumulation index (I _{geo}) ^a										
Not to weakly contaminated	≤ 0									
Weakly to moderately contaminated	$0 < I_{ m geo} \le 1$									
Moderate contaminated	$1 < I_{ m geo} \le 2$									
Moderate to strongly contaminated	$2 < I_{ m geo} \leq 3$									
Strongly contaminated	$3 < I_{ m geo} \leq 4$									
Strongly to extremely contaminated	$4 < I_{ m geo} \le 5$									
Extremely contaminated	$I_{\rm geo} > 5$									
Pollution factor	(PF) ^b									
Low pollution	≤ 1									
Moderate pollution	$1 < PF \leq 3$									
Considerable pollution	$3 < PF \le 6$									
Very high pollution	PF > 6									

 Table S2. Environmental indices with different classifications.

^a (Huang et al., 2021)

^b (Rehman et al., 2018)

Abbreviate for	Parameters	Unit	Probabilisti c	Adults	Children	Reference
parameters			distribution			
IngR	Ingestion rate	mg d ⁻¹	lognormal	50th:74.5; 95th:257.5	50th:45; 95th:202	(Duan et al., 2014; Duan et al., 2016)
InhR	Inhalation rate	m ³ d ⁻¹	lognormal	50th:14; 95th:17.9	50th:6.4; 95th:7.9	(Duan et al., 2014; Duan et al., 2016)
EF	Exposure frequency	d yr-1	point	350	350	(Wu, 2022)
ED	Exposure duration	yr	uniform	24	12	(Wu, 2022)
BW	Average body weight	kg	point	64.3	21.8	(CCDC, 2015; CCDC, 2020)
۸T	Non-carcinogenic risks	ł	naint	70 imes 365	70×365	(Duan et al., 2014; Duan
AI	Average time Carcinogenic risks	u	point	24×365	6×365	et al., 2016)
PEF	Particle emission factor	m ³ kg ⁻¹	point	1.36E+09	1.36E+09	(USEPA, 2001)
SA	Skin area exposed to soils	cm ²	lognormal	50th:5039; 95th:5938	50th:2074; 95th:2493	(Duan et al., 2014; USEPA, 2001)
AF	Skin adherence factor	mg cm ⁻²	Beta	0.07(0, 0.3)	0.2(0, 3.3)	(USEPA, 2011c)
ABF	Dermal adsorption factor	-	point	0.001(As:0.03)	0.001(As:0.03)	(USEPA, 2011c)

Table S3. Description and probability density functions (PDFs) of parameters in human health risk assessment to assess the health risks with Monte Carlo simulation.

Table S4. Reference dose (*RfD*) and slope factor (SF) of soil heavy metals in different pathways.

Parameters	As	Со	Cr	Cu	Ni	Pb	Zn
RfD _{ing}	3.00E-04 a	3.00E-04 °	3.00E-03 ^f	4.00E-02 a	2.00E-02 a	3.50E-03 a	3.00E-01 a
RfD_{inh}	3.01E-04 ^a	2.80E-05 °	2.86E-05 ^f	4.02E-02 ^a	2.06E-02 a	3.52E-03 a	6.00E-02 ^b
RfD_{dermal}	1.23E-04 °	6.00E-05 °	6.00E-05 ^f	1.20E-02 °	5.40E-03 °	5.25E-04 ^b	6.00E-02 °
SF_{ing}	1.50E+00 ª	-	5.00E-01 ^g	-	1.70E+00 °	-	-
SF_{inh}	1.51E+01 ª	9.8 °	4.20E+01 h	-	9.01E+00 a	8.50E-03 ^b	-
SF_{dermal}	3.66E+00 a	-	2.00E+01 °	-	4.25E+01 °	-	-

a (USEPA, 1999)

b (Men et al., 2021)

c (Cao et al., 2016)

d (Ran et al., 2021)

e (USEPA, 2011c)

f (Rahman et al., 2019)

g (Chen et al., 2019)

h (Gong et al., 2019)

Elements	Category	R ²	Intercept	Intercept-SE	Slope	Slope-SE	Scaled-residuals	Normal-residual
As	Strong	0.999	0.000	0.002	1.000	0.000	-2.052 ~ 1.187	Yes
Co	Strong	0.999	0.017	0.017	0.999	0.001	$-1.376 \sim 3.476$	Yes
Cr	Strong	0.287	42.150	1.815	0.207	0.029	$-0.814 \sim 0.554$	No
Cu	Strong	0.693	7.450	0.846	0.633	0.037	-2.751 ~ 3.221	Yes
Ni	Strong	0.765	6.490	0.714	0.677	0.033	$-1.999 \sim 4.621$	Yes
Pb	Strong	0.998	0.130	0.078	0.992	0.003	$-0.368 \sim 0.839$	Yes
Zn	Strong	0.257	41.180	1.795	0.200	0.030	$-0.733 \sim 0.690$	No

 Table S5. Residual analysis of PMF base model results.

Heavy metals	Factor 1	Factor 2	Factor 3	Factor 4	R ²
As	0.51	3.18	10.36	85.94	0.999
Co	6.77	52.68	31.28	9.27	0.999
Cr	15.00	32.83	46.90	5.27	0.287
Cu	24.21	16.16	54.91	4.73	0.693
Ni	15.18	29.83	52.66	2.33	0.765
Pb	66.51	33.49	0.00	0.00	0.998
Zn	13.38	35.73	45.00	5.88	0.257

Table S6. Factor analysis and contribution percentage (%) for heavy metals using PMF model.

	V	alue	Unit
	Adults	Children	- Omt
Land use			
EF (soil and dust ingestion) ^a	243	243	day yr-1
EF (consumption of homegrown produce)	0	0	day yr-1
EF (skin contact, indoor)	0	0	day yr-1
EF (skin contact, outdoor) ^a	243	243	day yr ⁻¹
EF (inhalation of dust and vapor, indoor)	0	0	day yr-1
EF (inhalation of dust and vapor, outdoor) ^a	243	243	day yr-1
Occupancy Period (indoor)	0	0	hr day-1
Occupancy Period (outdoor) ^a	3	3	hr day-1
Soil to skin adherence factor (indoor)	0	0	mg cm ⁻² day ⁻¹
Soil to skin adherence factor (outdoor) ^b	0.3	0.3	mg cm ⁻² day ⁻¹
Soil and dust ingestion rate ^c	0.2	0.1	g day ⁻¹
Receptor			
Body weight ^d	64.3	21.83	kg
Body height ^d	1.64	1.22	m
Inhalation rate ^a	14.1	14.1	m ³ day ⁻¹
Max exposed skin fraction (indoor)	0	0	m ² m ⁻²
Max exposed skin fraction (outdoor) °	0.48	0.48	m ² m ⁻²

Table S7. Land use and receptor parameters of study area input into the CLEA model.

a (Wu, 2022)

b (Yang et al., 2022)

c (MEPRC, 2019)

d (CCDC, 2020)

Table S8.	Soil and	building	properties	of study	area	input into	o the	CLEA 1	nodel.
		U	1 1	2		1			

	Value	Unit
Soil properties for	S	ilty clay
Porosity, total ^a	0.46	cm ³ cm ⁻³
Porosity, air-filled ^a	0.14	cm ³ cm ⁻³
Porosity, water-filled ^a	0.32	cm ³ cm ⁻³
Residual soil water Content ^b	0.1	cm ³ cm ⁻³
Saturated hydraulic conductivity ^c	3.56E-03	cm s ⁻¹
van Genuchten shape parameter (m) ^c	0.32	dimensionless
Bulk density ^a	1.46	g cm ⁻³
Threshold value of wind speed at 10 m ^d	17.2	m s ⁻¹
Empirical function (Fx) for dust model ^c	1.22	dimensionless
Ambient soil temperature ^c	283	Κ
Building properties for	No	building
Building footprint	0	m ²
Living space air exchange rate	0	hr-1
Living space height (above ground)	0	m
Living space height (below ground)	0	m
Pressure difference (soil to enclosed space)	0	Pa
Foundation thickness	0	m
Floor crack area	0	cm^2
Dust loading factor	0	μg m ⁻³
Air dispersion model		
Mean annual windspeed (10 m) ^c	3	m s ⁻¹
Air dispersion factor at height of 0.8 m $^\circ$	120	g m ⁻² s ⁻¹ per kg m ⁻³
Air dispersion factor at height of 1.6 m ^c	280	g m ⁻² s ⁻¹ per kg m ⁻³
Fraction of site with hard or vegetative cover ^c	0.5	m ² m ⁻²
Vapor model		
Default soil gas ingress rate ^c	25	cm ³ s ⁻¹
Depth to top of source (beneath building) ^c	0	cm
Depth to top of source (no building) ^c	50	cm
Thickness of contaminant layer ^c	0	cm
Time average period for surface emissions ^c	75	year
User defined effective air permeability ^d	7.18E-09	cm ²

a (Li et al., 2018)

b (MEPRC, 2019)

c (EA, 2009)

d (Yang et al., 2022)

		Park green spaces								Pı	rotection are	ea green spac	es				
Risk	Heavy		Ad	ults		Children					Ad	ults			Chil	dren	
	metals	25th	50th	75th	95th	25th	50th	75th	95th	25th	50th	75th	95th	25th	50th	75th	95th
	As	1.81E-01	2.01E-01	2.24E-01	2.61E-01	4.58E-01	5.10E-01	5.67E-01	6.56E-01	1.20E+00	1.33E+00	1.48E+00	1.72E+00	3.03E+00	3.37E+00	3.75E+00	4.36E+00
	Co	1.74E-01	1.95E-01	2.18E-01	2.53E-01	4.34E-01	4.84E-01	5.38E-01	6.27E-01	1.16E+00	1.29E+00	1.43E+00	1.66E+00	2.88E+00	3.21E+00	3.58E+00	4.14E+00
	Cr	4.69E-01	5.16E-01	5.72E-01	6.60E-01	8.76E-02	9.68E-02	1.07E-01	1.23E-01	1.95E-01	2.15E-01	2.38E-01	2.74E-01	5.77E-01	6.38E-01	7.06E-01	8.15E-01
HI	Cu	1.27E-03	1.42E-03	1.58E-03	1.85E-03	3.13E-03	3.50E-03	3.90E-03	4.54E-03	8.43E-03	9.40E-03	1.05E-02	1.21E-02	2.08E-02	2.31E-02	2.57E-02	2.99E-02
	Ni	1.50E-02	1.67E-02	1.86E-02	2.16E-02	6.33E-03	7.05E-03	7.87E-03	9.12E-03	1.70E-02	1.89E-02	2.10E-02	2.45E-02	4.19E-02	4.67E-02	5.20E-02	6.03E-02
	Pb	1.53E-02	1.71E-02	1.90E-02	2.21E-02	3.85E-02	4.28E-02	4.74E-02	5.51E-02	1.01E-01	1.13E-01	1.25E-01	1.47E-01	2.56E-01	2.84E-01	3.16E-01	3.68E-01
	Zn	1.75E-04	1.95E-04	2.17E-04	2.53E-04	4.35E-04	4.84E-04	5.38E-04	6.25E-04	1.15E-03	1.28E-03	1.43E-03	1.67E-03	2.87E-03	3.19E-03	3.55E-03	4.12E-03
THI	Total	8.97E-01	9.54E-01	1.02E+00	1.12E+00	1.08E+00	1.15E+00	1.23E+00	1.35E+00	2.80E+00	2.99E+00	3.20E+00	3.52E+00	7.14E+00	7.63E+00	8.16E+00	8.96E+00
	As	1.21E-05	1.34E-05	1.48E-05	1.71E-05	3.49E-05	3.87E-05	4.28E-05	4.92E-05	7.98E-04	8.81E-04	9.76E-04	1.13E-03	2.31E-03	2.56E-03	2.83E-03	3.26E-03
	Co	2.43E-08	2.71E-08	3.03E-08	3.52E-08	3.25E-08	3.63E-08	4.07E-08	4.78E-08	1.60E-08	1.79E-08	1.99E-08	2.33E-08	2.16E-08	2.40E-08	2.68E-08	3.13E-08
CR	Cr	5.17E-05	5.29E-05	5.43E-05	5.65E-05	7.39E-05	7.05E-05	7.82E-05	9.03E-05	2.66E-04	2.93E-04	3.25E-04	3.76E-04	7.70E-04	8.51E-04	9.42E-04	1.09E-03
	Ni	3.21E-05	3.56E-05	3.95E-05	4.55E-05	6.80E-05	7.52E-05	8.31E-05	9.61E-05	7.72E-06	8.54E-06	9.43E-06	1.09E-05	2.13E-05	2.36E-05	2.61E-05	3.01E-05
	Pb	2.10E-11	2.35E-11	2.61E-11	3.05E-11	2.81E-11	3.14E-11	3.51E-11	4.10E-11	1.39E-11	1.55E-11	1.72E-11	2.02E-11	1.86E-11	2.08E-11	2.32E-11	2.72E-11
TCR	Total	9.59E-05	1.02E-04	1.09E-04	1.19E-04	1.77E-04	1.84E-04	2.04E-04	2.36E-04	1.07E-03	1.18E-03	1.31E-03	1.52E-03	3.10E-03	3.43E-03	3.80E-03	4.38E-03

Table S9. Concentration-specific health risk of soil heavy metals in park green spaces and protection area green spaces for adults and children.

Diale Haarry		Courses	Adults						Children				
ISK	пеачу	Source	5th	25th	50th	75th	95th	5th	25th	50th	75th	95th	
		Factor 1	7.58E-03	8.79E-03	9.81E-03	1.09E-02	1.27E-02	1.91E-02	2.21E-02	2.46E-02	2.74E-02	3.18E-02	
	A a	Factor 2	4.69E-02	5.45E-02	6.05E-02	6.74E-02	7.84E-02	1.19E-01	1.38E-01	1.53E-01	1.70E-01	1.97E-01	
Co	AS	Factor 3	1.52E-01	1.77E-01	1.97E-01	2.19E-01	2.56E-01	3.85E-01	4.47E-01	4.98E-01	5.53E-01	6.41E-01	
		Factor 4	1.27E+00	1.47E+00	1.64E+00	1.82E+00	2.13E+00	3.20E+00	3.72E+00	4.13E+00	4.60E+00	5.31E+00	
		Factor 1	7.30E-03	8.52E-03	9.46E-03	1.06E-02	1.23E-02	1.82E-02	2.12E-02	2.35E-02	2.61E-02	3.04E-02	
	Ca	Factor 2	1.21E+00	1.42E+00	1.58E+00	1.75E+00	2.05E+00	3.03E+00	3.52E+00	3.92E+00	4.36E+00	5.08E+00	
	Co	Factor 3	1.47E-01	1.71E-01	1.91E-01	2.12E-01	2.47E-01	3.66E-01	4.26E-01	4.74E-01	5.28E-01	6.16E-01	
		Factor 4	4.52E-02	5.26E-02	5.85E-02	6.51E-02	7.59E-02	1.11E-01	1.31E-01	1.46E-01	1.62E-01	1.88E-01	
		Factor 1	7.71E-03	8.85E-03	9.79E-03	1.08E-02	1.25E-02	2.26E-02	2.63E-02	2.91E-02	3.22E-02	3.70E-02	
	C.	Factor 2	2.50E-02	2.89E-02	3.19E-02	3.52E-02	4.08E-02	7.43E-02	8.55E-02	9.43E-02	1.05E-01	1.21E-01	
	Cr	Factor 3	2.07E-01	2.38E-01	2.64E-01	2.93E-01	3.38E-01	6.15E-01	7.10E-01	7.87E-01	8.68E-01	1.00E+00	
HI		Factor 4	1.24E-03	1.43E-03	1.59E-03	1.75E-03	2.02E-03	3.64E-03	4.22E-03	4.68E-03	5.18E-03	5.97E-03	
		Factor 1	1.08E-03	1.25E-03	1.40E-03	1.55E-03	1.81E-03	2.65E-03	3.09E-03	3.43E-03	3.81E-03	4.44E-03	
	C	Factor 2	3.29E-04	3.84E-04	4.28E-04	4.76E-04	5.55E-04	8.10E-04	9.44E-04	1.05E-03	1.17E-03	1.37E-03	
	Cu	Factor 3	8.87E-03	1.04E-02	1.15E-02	1.29E-02	1.51E-02	2.19E-02	2.55E-02	2.84E-02	3.16E-02	3.70E-02	
		Factor 4	5.32E-05	6.23E-05	6.94E-05	7.72E-05	9.01E-05	1.31E-04	1.53E-04	1.70E-04	1.89E-04	2.20E-04	
		Factor 1	6.62E-04	7.71E-04	8.58E-04	9.57E-04	1.12E-03	1.64E-03	1.90E-03	2.11E-03	2.35E-03	2.74E-03	
	NI:	Factor 2	2.16E-03	2.51E-03	2.81E-03	3.13E-03	3.66E-03	5.33E-03	6.21E-03	6.91E-03	7.72E-03	8.95E-03	
	111	Factor 3	1.78E-02	2.08E-02	2.31E-02	2.58E-02	3.02E-02	4.43E-02	5.16E-02	5.73E-02	6.37E-02	7.44E-02	
		Factor 4	1.06E-04	1.25E-04	1.39E-04	1.55E-04	1.82E-04	2.65E-04	3.09E-04	3.43E-04	3.82E-04	4.47E-04	
		Factor 1	1.08E-01	1.25E-01	1.39E-01	1.54E-01	1.79E-01	2.70E-01	3.14E-01	3.49E-01	3.88E-01	4.52E-01	
	Pb	Factor 2	1.29E-02	1.50E-02	1.67E-02	1.86E-02	2.16E-02	3.26E-02	3.79E-02	4.21E-02	4.69E-02	5.43E-02	
		Factor 3	4.00E-03	4.64E-03	5.16E-03	5.71E-03	6.65E-03	1.00E-02	1.16E-02	1.29E-02	1.43E-02	1.67E-02	

	-	Factor 4	6.40E-04	7.45E-04	8.29E-04	9.21E-04	1.08E-03	1.63E-03	1.88E-03	2.08E-03	2.31E-03	2.69E-03
		Factor 1	4.48E-05	5.26E-05	5.86E-05	6.51E-05	7.61E-05	1.13E-04	1.31E-04	1.46E-04	1.63E-04	1.88E-04
	7	Factor 2	1.46E-04	1.71E-04	1.90E-04	2.12E-04	2.48E-04	3.66E-04	4.27E-04	4.74E-04	5.28E-04	6.15E-04
	Zn	Factor 3	1.22E-03	1.42E-03	1.58E-03	1.76E-03	2.04E-03	3.04E-03	3.54E-03	3.92E-03	4.36E-03	5.07E-03
		Factor 4	7.29E-06	8.48E-06	9.42E-06	1.05E-05	1.23E-05	1.81E-05	2.11E-05	2.35E-05	2.62E-05	3.04E-05
		Factor 1	1.32E-01	1.53E-01	1.70E-01	1.89E-01	2.20E-01	3.34E-01	3.89E-01	4.32E-01	4.80E-01	5.58E-01
TII	T-4-1	Factor 2	1.30E+00	1.52E+00	1.69E+00	1.88E+00	2.20E+00	3.26E+00	3.79E+00	4.21E+00	4.69E+00	5.47E+00
IHI	Total	Factor 3	5.38E-01	6.23E-01	6.93E-01	7.70E-01	8.95E-01	1.45E+00	1.68E+00	1.86E+00	2.06E+00	2.39E+00
		Factor 4	1.32E+00	1.52E+00	1.70E+00	1.89E+00	2.21E+00	3.32E+00	3.86E+00	4.29E+00	4.77E+00	5.51E+00
		Factor 1	2.92E-07	3.39E-07	3.78E-07	4.20E-07	4.89E-07	6.06E-07	7.01E-07	7.75E-07	8.60E-07	9.92E-07
		Factor 2	1.81E-06	2.10E-06	2.34E-06	2.60E-06	3.03E-06	3.79E-06	4.35E-06	4.81E-06	5.31E-06	6.15E-06
	As	Factor 3	5.87E-06	6.83E-06	7.61E-06	8.45E-06	9.87E-06	1.23E-05	1.42E-05	1.57E-05	1.73E-05	2.00E-05
		Factor 4	4.89E-05	5.68E-05	6.31E-05	7.04E-05	8.24E-05	1.02E-04	1.18E-04	1.30E-04	1.44E-04	1.66E-04
		Factor 1	8.61E-12	1.01E-11	1.13E-11	1.26E-11	1.47E-11	5.62E-12	6.57E-12	7.33E-12	8.16E-12	9.60E-12
	C	Factor 2	1.44E-09	1.68E-09	1.88E-09	2.10E-09	2.47E-09	9.33E-10	1.09E-09	1.22E-09	1.36E-09	1.60E-09
	Co	Factor 3	1.74E-10	2.03E-10	2.27E-10	2.53E-10	2.96E-10	1.12E-10	1.32E-10	1.47E-10	1.65E-10	1.93E-10
		Factor 4	5.35E-11	6.25E-11	6.98E-11	7.78E-11	9.09E-11	3.47E-11	4.07E-11	4.55E-11	5.07E-11	5.93E-11
CR		Factor 1	9.01E-07	1.04E-06	1.15E-06	1.27E-06	1.47E-06	1.25E-06	1.45E-06	1.61E-06	1.78E-06	2.05E-06
	C	Factor 2	2.93E-06	3.38E-06	3.74E-06	4.13E-06	4.79E-06	4.11E-06	4.73E-06	5.22E-06	5.79E-06	6.68E-06
	Cr	Factor 3	2.42E-05	2.79E-05	3.09E-05	3.43E-05	3.96E-05	3.41E-05	3.93E-05	4.35E-05	4.80E-05	5.54E-05
		Factor 4	1.45E-07	1.68E-07	1.86E-07	2.05E-07	2.37E-07	2.01E-07	2.34E-07	2.59E-07	2.86E-07	3.30E-07
		Factor 1	2.60E-06	3.00E-06	3.32E-06	3.67E-06	4.26E-06	3.48E-07	4.00E-07	4.43E-07	4.90E-07	5.68E-07
	NT.	Factor 2	8.49E-06	9.80E-06	1.08E-05	1.20E-05	1.40E-05	1.13E-06	1.31E-06	1.44E-06	1.60E-06	1.85E-06
	N1	Factor 3	7.02E-05	8.10E-05	8.95E-05	9.93E-05	1.15E-04	9.41E-06	1.08E-05	1.20E-05	1.33E-05	1.53E-05
		Factor 4	4.19E-07	4.85E-07	5.37E-07	5.96E-07	6.91E-07	5.62E-08	6.49E-08	7.18E-08	7.94E-08	9.22E-08
	Pb	Factor 1	1.25E-12	1.46E-12	1.63E-12	1.82E-12	2.13E-12	8.10E-13	9.51E-13	1.06E-12	1.19E-12	1.39E-12

		Factor 2	1.50E-13	1.76E-13	1.97E-13	2.19E-13	2.56E-13	9.82E-14	1.14E-13	1.28E-13	1.43E-13	1.67E-13
		Factor 3	4.62E-14	5.42E-14	6.07E-14	6.75E-14	7.88E-14	3.00E-14	3.52E-14	3.93E-14	4.40E-14	5.13E-14
		Factor 4	7.43E-15	8.73E-15	9.76E-15	1.09E-14	1.27E-14	4.88E-15	5.69E-15	6.34E-15	7.09E-15	8.29E-15
TCR		Factor 1	3.79E-06	4.38E-06	4.85E-06	5.36E-06	6.22E-06	2.21E-06	2.55E-06	2.83E-06	3.13E-06	3.61E-06
	Total	Factor 2	1.32E-05	1.53E-05	1.69E-05	1.87E-05	2.18E-05	9.03E-06	1.04E-05	1.15E-05	1.27E-05	1.47E-05
	TOtal	Factor 3	1.00E-05	1.16E-05	1.28E-05	1.42E-05	1.64E-05	5.57E-05	6.43E-05	7.12E-05	7.86E-05	9.08E-05
		Factor 4	4.95E-05	5.75E-05	6.38E-05	7.12E-05	8.33E-05	1.02E-04	1.18E-04	1.30E-04	1.44E-04	1.66E-04

Heavy metals	PDFs for concentration	PDFs for Factor 1	PDFs for Factor 2	PDFs for Factor 3	PDFs for F4
As	LN (3.12, 2.98)	LN (0.02, 0.02)	LN (0.10, 0.09)	LN (0.32, 0.31)	LN (2.68, 2.56)
Со	LN (12.08, 1.50)	LN (0.82, 0.10)	LN (6.36, 0.79)	LN (3.78, 0.47)	LN (1.12, 0.14)
Cr	LN (57.96, 24.63)	LN (8.69, 3.69)	LN (19.03, 8.09)	LN (27.19, 11.55)	LN (3.06, 1.30)
Cu	LN (21.65, 6.85)	LN (5.24, 1.66)	LN (3.50, 1.11)	LN (11.89, 3.76)	LN (1.02, 0.32)
Ni	LN (20.96, 4.94)	LN (3.18, 0.75)	LN (6.25, 1.47)	LN (11.04, 2.60)	LN (0.49, 0.12)
Pb	LN (18.24, 7.10)	LN (12.13, 4.72)	LN (6.11, 2.38)	LN (2.38E-09, 9.28E-10)	LN (3.99E-08, 1.55E-08)
Zn	LN (55.65, 22.07)	LN (7.45, 2.95)	LN (19.88, 7.89)	LN (25.04, 9.93)	LN (3.27, 1.30)

Table S11. Probability density functions (PDFs) of soil heavy metals concentrations and heavy metals concentrations from each source.

Items	PC1	PC2	PC3	
As	0.144	-0.003	0.960	
Со	0.241	0.750	0.188	
Cr	0.973	0.120	0.133	
Cu	0.680	0.157	0.631	
Ni	0.763	0.524	0.135	
Pb	0.089	0.819	-0.046	
Zn	0.976	0.114	0.115	

Table S12. Results of principal component analysis of soil heavy metal concentration.

Table S	13. Limited	values o	f heavy metal	elements ir	n the soil	quality	standards/crit	teria (mg	(kg ⁻¹).
			2			1 2		$\langle U \rangle$	

Elemen	ts Canada ^a	Netherlands (Target value /Intervene value) ^b	Australia (Health/Ecological investigation value) °	France (Target value /Intervene value) ^d	Briain ^e	USA (Eco-screening value) ^f
As	12	29/55	100/50	19/73	20	18
Со	-	9/240	-	-	-	-
Cr	64	100/380	400/75 (Cr ³⁺)	-	130	-
Cu	63	36/190	230/30	95/190	-	70
Ni	-	-	560/10	-	50	38
Pb	70	85/530	1000/270	200/400	-	120
Zn	200	140/720	1300/25	4500/9000	-	160

a (CCME, 2011)

b (NIHE, 2015)

c (NEPC, 2013)

d (Stephen, 2011)

e (EA, 2011)

f (USEPA, 2011a)





Fig. S1. The derivation principle and procedure for GAC in CLEA model.

Reference

- Cao, S., Duan, X., Zhao, X., Chen, Y., Wang, B., Sun, C., Zheng, B., Wei, F. 2016. Health risks of children's cumulative and aggregative exposure to metals and metalloids in a typical urban environment in China. *Chemosphere*, 147, 404-411.
- CCDC. 2015. Report on nutrition and chronic diseases in China (2015). People's Medical Publishing House (In Chinese).
- CCDC. 2020. *Report on nutrition and chronic diseases in China (2020)*. People's Medical Publishing House (In Chinese). CCME. 2011. Canadian Environmental Quality Guidelines Summary Table.
- Chen, G., Wang, X., Wang, R., Liu, G. 2019. Health risk assessment of potentially harmful elements in subsidence water bodies using a Monte Carlo approach: An example from the Huainan coal mining area, China. *Ecotoxicol. Environ. Saf*, **171**, 737-745.
- Duan, X.L., Zhao, X.G., Wang, B.B., Chen, Y.T., Cao, S.Z. 2014. *Highlight of the chinese exposure factors handbook (adults)*. China Environmental Science Press (In Chinese).
- Duan, X.L., Zhao, X.G., Wang, B.B., Chen, Y.T., Cao, S.Z. 2016. *Highlight of the chinese exposure factors handbook* (children). China Environmental Science Press (In Chinese).
- EA. 2009. CLEA Software (Version 1.04) Handbook.
- EA. 2011. Soil Guideline Values.
- Gong, Q., Chen, P.Z., Shi, R.G., Gao, Y., Zheng, S.A., Xu, Y., Shao, C.F., Zheng, X.Q. 2019. Health Assessment of Trace Metal Concentrations in Organic Fertilizer in Northern China. *Int. J. Environ. Res. Public. Health*, 16(6), 1031.
- Huang, J., Wu, Y., Sun, J., Li, X., Geng, X., Zhao, M., Sun, T., Fan, Z. 2021. Health risk assessment of heavy metal(loid)s in park soils of the largest megacity in China by using Monte Carlo simulation coupled with Positive matrix factorization model. J. Hazard. Mater, 415, 125629.
- Li, Y., Yang, J., Li, Y. 2018. Physical Properties of Soil from Different Functional Areas in Tianjin City. Bullet Soil Water Conserv (In Chinese), **38**(4), 331-336.
- Men, C., Liu, R., Wang, Q., Miao, Y., Wang, Y., Jiao, L., Li, L., Cao, L., Shen, Z., Li, Y., Crawford-Brown, D. 2021. Spatial-temporal characteristics, source-specific variation and uncertainty analysis of health risks associated with heavy metals in road dust in Beijing, China. *Environ. Pollut*, **278**, 116866.
- MEPRC. 2019. Technical guidelines for risk assessment of soil contamination of land forconstruction (HJ 25.3-2019) (In Chinese).
- NEPC. 2013. Schedule B5a, guideline on ecological risk assessment.
- NIHE. 2015. Health and safety in the environmental act:ratio and substantiation of current environmental quality standards (in Dutch).
- Rahman, M.S., Khan, M.D.H., Jolly, Y.N., Kabir, J., Akter, S., Salam, A. 2019. Assessing risk to human health for heavy metal contamination through street dust in the Southeast Asian Megacity: Dhaka, Bangladesh. Sci. Total. Environ, 660, 1610-1622.
- Ran, H., Guo, Z., Yi, L., Xiao, X., Zhang, L., Hu, Z., Li, C., Zhang, Y. 2021. Pollution characteristics and source identification of soil metal(loid)s at an abandoned arsenic-containing mine, China. J. Hazard. Mater, 413, 125382.
- Rehman, I.U., Ishaq, M., Ali, L., Khan, S., Ahmad, I., Din, I.U., Ullah, H. 2018. Enrichment, spatial distribution of potential ecological and human health risk assessment via toxic metals in soil and surface water ingestion in the vicinity of Sewakht mines, district Chitral, Northern Pakistan. *Ecotoxicol Environ Saf*, 154, 127-136.
- Stephen, J. 2011. Guidelines for screen contaminated soils.
- USEPA. 2011a. Ecological Soil Screening Levels.
- USEPA. 2016. EPA ExpoBox: a Toolbox for Exposure Assessors. Exposure Assessment Tools by Routes.
- USEPA. 2011b. Exposure Factors Handbook 2011 Edition (Final).
- USEPA. 1999. Risk Assessment Guidance for Superfund. in: Volume I (Part A: Human Health Evaluation Manual; Part E, Supplemental Guidance for Dermal Risk Assessment; Part F, Supplemental Guidance for Inhalation Risk Assessment).
- USEPA. 2009. Risk Assessment Guidance for Superfund (RAGS), Volume I, Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment).
- USEPA. 2011c. The screening level (RSL) tables (Last updated June 2011).
- USEPA. 2001. Supplemental Guidance for Developing Soil Screening Levels for Superfund Sites. in: OSWER 9355.4-24 Office of Solid Waste and Emergency Response.
- Wu, Y. 2022. Research on the occurrence characteristics and nvironmental criteria of beryllium, cobalt, antimony andvanadium in Beijing urban park soil, Vol. Master, Chinese Research Academy of Environmental Sciences.
- Yang, S., Wu, Y., Ma, J., Liu, Q., Qu, Y., Zhao, W. 2022. Human health risk-based Generic Assessment Criteria for agricultural soil in Jiangsu and Zhejiang provinces, China. *Environ. Res*, 206, 112277.