

SUPPORT INFORMATION
for
**Development of Lead Source-specific Exposure Standards Based on Aggregate Exposure
Assessment: Bayesian Inversion from Biomonitoring Information to Multipathway
Exposure**

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Materials and Methods

Integrated Exposure Uptake Biokinetic model. In this study, IEUBK model was used to link the external exposure and the biomonitoring information, and the pharmacokinetics procedure is detailed in technical support document (1). The model consists of three components (Support Information Figure S1): 1) exposure components, in which average daily lead intake is determined from multiple environmental media, including air, soil, food, drinking water, paint and others; 2) uptake component, which converts the media-specific intake in exposure component into media-specific uptake for the blood plasma; 3) biokinetic component, which simulates the absorption, transportation, metabolism and elimination in the body, and estimates BLLs finally. In this research, the model was coded in the Matlab programming language, and the distribution families for the daily intake amounts of air, soil, grain, vegetable, paint can be seen in the Support Information Tables S1 and S2.

Markov chain Monte Carlo (MCMC) Sampling Algorithm. In this study, the Gibbs and Metropolis Hastings (MH) samplers were used to update the object parameters. We iteratively update the parameters in the following sequence: σ^2 , Σ , μ , and C_{ext} . On the basis of Bayesian analysis of hierarchical linear model, the population parameters, σ^2 , Σ , μ , were randomly draw from inverse gamma, inverse gamma and normal distribution, respectively, using the Gibbs sampler. However, since the toxicokinetic model is nonlinear, the conditional distributions for C_{ext} have no closed form. Therefore, we sampled the C_{ext} using the steps of the Metropolis algorithm as described previously (2), and the following Gaussian distribution was adopted to execute the M-H sampling:

$$C_{ext}^{new} = C_{ext}^{k-1} + N(0, cov(C_{ext})/7) \quad (1)$$

where $cov(C_{ext})$ in the Gaussian distribution is a diagonal matrix with its diagonal entry being set to the variances of prior C_{ext} and zeros elsewhere.

Relationships among Lognormal Distributional Descriptors. Given a lognormal

distribution of a parameter X for which $\hat{\mu}$ and $\hat{\sigma}$ are the mean and the standard deviation of the distribution $Ln(X)$, respectively. GM, AM, GSD, SD represent the geometric mean, arithmetic mean, geometric standard deviation, standard deviation of the distribution X , respectively. α_i , β_i are the cumulative probabilities of the distribution X , and p_i is the corresponding cumulative probability of the standard normal distribution. The following relationships hold:

$$GM = \exp(\hat{\mu})$$

$$GSD = \exp(\hat{\sigma})$$

$$AM = \exp(\hat{\mu} + \hat{\sigma}^2 / 2)$$

$$SD = \sqrt{\exp(2 \times \hat{\mu} + \hat{\sigma}^2) \exp(\hat{\sigma}^2 - 1)}$$

$$\alpha_i = \exp(\hat{\mu} + \hat{\sigma} \times \beta_i)$$

$$p_i = \int_{-\infty}^{\beta_i} \frac{1}{\sqrt{2 \times \pi}} \exp\left(-\frac{t^2}{2}\right) dt$$

Data combined for biomonitoring data from each city. The data for each city, which was obtained from a separate study, was combined to create one sample size weighted geometric mean BLL according to the method described previously (3) as follows:

$$m_i = \frac{N_i}{\sum_{i=1}^n N_i} \quad (2)$$

$$Ln(BLLs) = \frac{\sum_{i=1}^n m_i \frac{Ln(BLL_i)}{SD[Ln(BLL_i)]^2}}{\sum_{i=1}^n \frac{m_i}{SD[Ln(BLL_i)]^2}} \quad (3)$$

$$SD[Ln(BLL)] = \sqrt{\frac{1}{\sum_{i=1}^n \frac{m_i}{SD[Ln(BLL_i)]^2}}} \quad (4)$$

where m_i is the weight factor, and N_i is the sample size of each city.

Analytical Method for Lead in Drinking Water. Drinking water samples were collected

from the 34 cities' waterworks in China, and were frozen at the -4°C temperature refrigerator in the plastic bottle. 1.5 mL sample were mixed with 1.5 mL 6% HNO₃ (A.R) and inductively coupled plasma mass spectroscopy was used to determine the lead content in the mixture. The sensitivity was monitored daily and optimized when required. The elements measured were lead (*m/z* 208) and Rh (*m/z* 103). Concentration was determined using a six-point calibration of 0, 0.1, 0.5, 1, 2 and 10 µg/L (R²=0.99). Standards were run after every set of 10 samples.

Sensitivity Analysis for the Parameters in IEUBK Model. The sensitivity coefficient (*s*) was calculated as following Equation (5):

$$s = \frac{f(x + \Delta \times x, \Phi) - f(x, \Phi)}{\Delta} \quad (5)$$

where *x* is the objective parameter; Δ is the changing scale of the objective parameter, and Δ was set as 1% as the study by Yang (4); Φ is the model default parameter family that excluded the objective parameter; *f* is the IEUBK model. In this study, the 43 parameters listed in the Supporting Information TABLE S3 were performed for sensitivity analysis in turn. The parameter with a sensitivity coefficient over 0.1 is usually considered a sensitivity parameter (4), which means varying the sensitivity parameter value by 1% has a 0.1% impact on the response. The default uptake for each month is an arithmetic sequence with initial value of 9.2 µg/day and common difference of 0.2 µg/day for children aged 13~84 months, thus BLLs for children aged 1~6 are calculated to be 4.47 µg/dL, 4.71 µg/dL, 5.26 µg/dL, 5.78 µg/dL, 6.15 µg/dL, 6.35 µg/dL, respectively, which are close to the observed blood lead concentration in chinese children. The output of function *f* is a vector with six elements which represented the BLLs of children aged 1~6. Thus, the sensitivity coefficient listed in the Supporting Information TABLE S3 is the average of the sensitivity coefficient for children aged 1~6. Also, the pseudocode used to calculate the BLLs in children was listed as follows:

```
function BLLs=ass(uptake,timestep);
%the input parameters including uptake(each month),BLLs in children aged 1 and timestep
%% basic parameters setting
```

```

a=1/30*timestep;
b=floor(1/a);
t=0:a:85-a;
%% physiological parameter
wtbody=(8.375./(1+exp(-(t-3.8)/3.6)))+(17.261./(1+exp(-(t-48.76)/20.63)));
tblur=20.*(wtbody/12.3).^0.33;
tblliv=10.*(wtbody/12.3).^0.33;
tblloth=10.*(wtbody/12.3).^0.33;
tblkid=10.*(wtbody/12.3).^0.33;
tblbone=(wtbody/12.3).^0.33;
RATOUTFEC=0.75;
tblfec=0.75.*tblur;
tblout=RATOUTFEC.*tblfec;
crbonebl=6+(215.*(1-exp(-0.000942.*t)));
for i=1:(85*b);
    if t(i)<12 | t(i)==12;
        wtbone(i)=0.111*wtbody(i);
    else
        wtbone(i)=0.838+0.02*wtbody(i);
    end
end
end
wttrab=0.2.*wtbone;
wtcort=0.8.*wtbone;
volblood=(10.67./(1+exp(-(t-6.87)/7.09)))+(21.86./(1+exp(-(t-88.15)/26.73)));
tbonebl=crbonebl.*tblbone.*((wttrab+wtcort)./(volblood/10));
tplrbc=0.1;
ratblpl=100;
trbcpl=tplrbc*(ratblpl-0.55/(0.55+0.73));
tplur=tblur./ratblpl;
tplliv=tblliv./ratblpl;
crkidbl=0.777+(2.35.*(1-exp(-0.0468.*t)));
crlivbl=1.1+(3.5.*(1-exp(-0.0462.*t)));
crothbl=0.931+(0.437.*(1-exp(-0.00749.*t)));
wtliver=(0.261./(1+exp(-(t-9.82)/3.62)))+(0.584./(1+exp(-(t-55.76)/37.64)));
tlivpl=crlivbl.*(tblliv./(1-tblliv./tblfec)).*(wtliver./(volblood./10));
tlivfec=crlivbl.*tblfec.*(wtliver./(volblood./10));
tplkid=tblkid./ratblpl;
wtkidney=(0.05./(1+exp(-(t-5.24)/4.24)))+(0.106./(1+exp(-(t-65.67)/34.11)));
tkidpl=crkidbl.*tblkid.*(wtkidney./(volblood./10));
tpltrab=tblbone./(0.2*ratblpl);
ttrabpl=tbonebl;
tplcort=tblbone./(0.8*ratblpl);
tcortpl=tbonebl;
tploth=tblloth./ratblpl;
wtblood=1.056.*volblood./10;
wtecf=0.73*volblood./10;
wtother=wtbody-wtkidney-wtliver-wttrab-wtcort-wtblood-wtecf;
tothpl=crothbl.*(tblloth./(1-tblloth./tblout)).*(wtother./(volblood./10));
tothout=crothbl.*tblout.*(wtother./(volblood./10));
volrbc=(4.31./(1+exp(-(t-6.45)/10)))+(26.47./(1+exp(-(t-129.61)/25.98)));

```

```

for i=2:85*b;
    volrbc1(1,i-1)=volrbc(1,i);
end
volplasm=(6.46./(1+exp(-(t-6.81)/5.74)))+(8.83./(1+exp(-(t-65.66)/23.62)));
volecf=0.73.*volblood;
conrbc=1200;
    pbldmat=2.5;
pbldo=0.85*pbldmat;
volplasm0=2.0269;
volrbc0=1.6623;
hct0=0.45;
trbcpl0=trbcpl;
%% initial value
mplecf(1)=pbldo*(volplasm0+volrbc0)*(tplrbc/timestep)*(1.7-hct0)/(trbcpl0/timestep+tlrbc/timestep);
mrbc(1)=pbldo*(volplasm0+volrbc0)*(trbcpl0/timestep)/(trbcpl0/timestep+tlrbc/timestep);
mplasm(1)=mplecf(1)/(1.7-hct0);
wtcort0=wtcort(1);
mcort(1)=78.9*pbldo*wtcort0;
mkidney(1)=10.6*pbldo*wtkidney(1);
mliver(1)=13*pbldo*wtliver(1);
mother(1)=16*pbldo*wtmother(1);
mtrab(1)=51.2*pbldo*wttrab(1);
tplrbc2(1)=tplrbc./(1-mrbc(1)./(conrbc.*volrbc1(1)));
tothall=1./(1./tothpl+1./tothout);
tlivall=1./(1./tlivpl+1./tlivfec);
%% compartmental lead masses
    for pp=1:84;
        sss=0;
for ii=(pp*b-b+2-sign(pp-1)):(pp*b);
    tplrbc2(ii)=tplrbc./(1-mrbc(ii-1)./(conrbc.*volrbc1(ii-b*sign(pp-1))));
sum1(ii)=1./tplur(ii)+1./tlliv(ii)+1./tplkid(ii)+1./tplot(ii)+1./tpltrab(ii)+1./tpltort(ii)+1./tplrbc2(ii);
sum2(ii)=1/(tplrbc2(ii)*(trbcpl/timestep+1))+1/(tlliv(ii)*(tlivpl(ii)/timestep+1+tlivpl(ii)/tlivall(ii)))+1/(tothpl(ii)/timestep+1+tothpl(ii)/tothall(ii))+1/(tplkid(ii)*(tkidpl(ii)/timestep+1))+1/(tpltrab(ii)*(ttrabpl(ii)/timestep+1))+1/(tpltort(ii)*(tcortpl(ii)/timestep+1));
sum3(ii)=mrbc(ii-1)/(trbcpl/timestep+1)+mliver(ii-1)/(tlivpl(ii)/timestep+1+tlivpl(ii)/tlivall(ii))+mother(ii-1)/(tothpl(ii)/timestep+1+tothpl(ii)/tothall(ii))+mkidney(ii-1)/(tkidpl(ii)/timestep+1)+mtrab(ii-1)/(ttrabpl(ii)/timestep+1)+mcort(ii-1)/(tcortpl(ii)/timestep+1);
mplecf(ii)=(mplecf(ii-1)+(uptake(pp)*timestep/30)+sum3(ii))/(1+(timestep*sum1(ii))-timestep*sum2(ii));
    mrbc(ii)=(mrbc(ii-1)+(mplecf(ii)*(timestep/tplrbc2(ii))))/(1+timestep/trbcpl);
    mliver(ii)=(mliver(ii-1)+(mplecf(ii)*(timestep/tlliv(ii))))/(1+timestep/tlivall(ii));
    mother(ii)=(mother(ii-1)+(mplecf(ii)*(timestep/tplot(ii))))/(1+timestep/tothall(ii));
    mcort(ii)=(mcort(ii-1)+(mplecf(ii)*(timestep/tpltort(ii))))/(1+timestep/tcortpl(ii));
    mkidney(ii)=(mkidney(ii-1)+(mplecf(ii)*(timestep/tplkid(ii))))/(1+timestep/tkidpl(ii));
    mtrab(ii)=(mtrab(ii-1)+(mplecf(ii)*(timestep/tpltrab(ii))))/(1+timestep/ttrabpl(ii));
    mplasm(ii)=(mplecf(ii)*volplasm(ii))/(volecf(ii)+volplasm(ii));
    sss=sss+(mrbc(ii)+mplasm(ii))/volblood(ii);    pbb(pp)=sss*a;
end

```

end

BLLs=pbb(13:84); %BLLs for children aged 13-84 months

Sensitivity analysis of the prior C_{ext} . To illustrate the sensitivity of the prior C_{ext} for source allocation, the variation in source allocation was investigated by changing the pPDF of diet which shows the highest source allocation. GM of dietary lead changed from GM/GSD to $GM \times GSD$ where prior GSD was set as an average value of 2.5, and GSD changed from 2 to 4. Generally, GM is relatively sensitive to source allocation compared with GSD since the uncertainty from its prior distribution has been reduced during Bayesian inversion as described above. When the GM for dietary lead varied from 0.4 to 2.5 folds (Support Information Figure S2), the source allocation from diet changed from 60.46% to 66.91%. Following the change of GM and GSD for dietary lead, the source allocations for other sources are also changed as exemplified in Figure 2a~2d. Their source allocation changes limited to the range of 1.89%~3.56% for air, 0.07%~0.51% for water, 12.38%~14.42% for soil, and 16.41%~23.40% for paint, respectively.

SUPPORTING INFORMATION TABLE S1. Distributions and Parameters for the Daily Amounts of Soil, Grain, Vegetables, Drinking Water and Paint

Age		1	2	3	4	5	6	Distribution-type	Ref.
Soil and dust (mg/day)	GM	0.11	0.11	0.11	0.090	0.088	0.087	Lognormal	5
	GSD	1.23	1.23	1.23	1.16	1.14	1.14		
Grain (mg)	GM	45.64*	97.80	111.80	136.80	146.30	171.27*	Lognormal	6, 7
	GSD	1.50	1.50	1.50	1.50	1.50	1.50		
Vegetable (mg)	GM	48.72*	104.4	140.3	144	182.7	213.89*	Lognormal	6, 7
	GSD	1.50	1.50	1.50	1.50	1.50	1.50		
	Min	0.19	0.19	0.19	0.19	0.19	0.19		
Water (L)	Max	1.9	1.9	1.9	1.9	1.9	1.9	Triangular	8
	Mode	0.60	0.60	0.60	0.60	0.60	0.60		
Paint (mg)	GM	0.622	0.622	0.622	0.46	0.417	0.392	Lognormal	8
	GSD	2.23	2.23	2.23	2.10	2.05	2.03		

* No the daily amounts of vegetable and grain for children aged 1 and 6 can be obtained. Fortunately, the IEUBK model adopt the defaulted lead intake rate for game animal meat, fish, home-grown vegetables, or home-grown fruit as 0.14, 0.3, 0.33, 0.35, 0.41, 0.48 μg for children aged 1, 2, 3, 4, 5, 6 (1), respectively. So, the daily amount of children aged 1 and aged 6 were estimated by the rate of the daily amount between children aged 1 and 2 (0.14/0.3), aged 5 and 6 (0.48/0.41).

SUPPORTING INFORMATION TABLE S2. Distribution of Body Weight for Children Aged 1~6 (9).

Age	Mean (kg)	SD(kg)	Age	Mean (kg)	SD(kg)
1	10	1.6	4	16	3.0
2	12	2.2	5	18	2.6
3	14	1.8	6	24	4.2

SUPPORTING INFORMATION TABLE S3 Sensitivity Analysis of IEUBK Model Parameters

Parameters	Description	Sensitivity	Parameters	Description	Sensitivity
Vol _{blood}	Volume of blood	-0.25	T _{bonebl}	Lead transfer time from bone to blood	-0.057
W _{t_{kidney}}	Weight of kidney	-0.00034	W _{t_{trab}}	Weight of trabecular bone	-0.0109
Hct ₀	Hematocrit at birth	0.00	Vol _{ecf}	Volume of extra-cellular fluid (ECF)	-0.0025
W _{t_{bone}}	Weight of bone	-0.053	W _{t_{blood}}	Weight of blood	0.00035
T _{cortpl}	Lead transfer time from cortical bone to plasma-ECF	-0.046	Vol _{plasm}	Volume of plase	0.00073
W _{t_{ecf}}	Weight of extra-cellular fluid (ECF)	0.00024	Ra _{t_{blpl}}	Ratio of lead mass in blood to lead mass in plasma-ECF	-0.0104
T _{othpl}	Lead transfer time from soft tissues to plasma-ECF	-0.0399	T _{pl_{rbc}}	Lead transfer time from plasma-ECF to red blood cells	-0.0099
T _{trabpl}	Lead transfer time from trabecular bone to plasma-ECF	-0.0115	T _{bl_{liv}}	Lead transfer time from blood to liver	0.0238
W _{t_{liver}}	Weight of liver	0.00074	W _{t_{body}}	Weight of body	0.12
T _{kidpl}	Lead transfer time from kidney to plasma-ECF	-0.00074	Cr _{oth_{bl}}	Ratio of lead concentration in other soft tissue to blood lead concentration	-0.0035
P _{bbld_{mat}}	Maternal blood lead concentration	0.0067	P _{bbld₀}	Lead concentration in blood	0.0067
T _{bl_{kid}}	Lead transfer time from blood to kidney	0.00	T _{liv_{pl}}	Lead transfer time from liver to plasma-ECF	-0.091
T _{bl_{bone}}	Lead transfer time from blood to bone	0.00058	Ra _{t_{out_{fec}}}	Ratio of elimination rate via soft tissues to endogenous fecal lead elimination rate	0.36
Vol _{r_{bc}}	Volume of red blood cells	0.0028	T _{oth_{out}}	Lead transfer time from soft tissues to elimination pool	0.0367
W _{t_{cort}}	Weight of cortical bone	-0.043	T _{pl_{kid}}	Lead transfer time from plasma-ECF to	0.00074

T_{blur}	Lead transfer time from blood to urine	0.86	T_{pltrab}	kidney Lead transfer time from plasma-ECF to trabecular bone	0.0115
T_{livfec}	Lead transfer time from liver to feces	0.093	$Ra_{toutfec}$	Ratio of elimination rate via soft tissues to endogenous fecal lead elimination rate	0.36
Cr_{livbl}	Ratio of lead concentration ($\mu\text{g}/\text{kg}$) in liver to blood lead concentration ($\mu\text{g}/\text{L}$)	0.0012	T_{othout}	Lead transfer time from soft tissues to elimination pool	0.0367
Con_{rbc}	Maximum lead concentration capacity of red blood cells	0.0128	T_{plcort}	Lead transfer time from plasma-ECF to cortical bone	0.046
T_{bloth}	Lead transfer time from blood to other soft tissue	0.0365	Cr_{kidbl}	Ratio of lead concentration ($\mu\text{g}/\text{kg}$) in kidney to blood lead concentration ($\mu\text{g}/\text{L}$)	-0.00074
Ra_{tfecur}	Ratio of endogenous fecal lead elimination rate to urinary lead elimination rate	0.63			

SUPPORTING INFORMATION TABLE S4. Prior distributions of the selected IEUBK model parameters for the Bayesian MCMC studies

Parameters	Mean	CV	Parameters	Mean	CV
Volblood	*	0.20	ABSD	0.3	0.20
Ratoutfec	0.75	0.20	ABSF	0.5	0.20
Ratfecur	0.75	0.20	ABSW	0.5	0.20
Tblur	*	0.20	ABSP	0.17	0.20
Air_absorb	0.32	0.20			

Abbreviations: Volblood, Volume of blood; Ratoutfec, Ratio of elimination rate via soft tissues to endogenous fecal lead elimination rate; Ratfecur, Ratio of endogenous fecal lead elimination rate to urinary lead elimination rate; Tblur, Lead transfer time from blood to urine; Air_absorb, Net percentage absorption of air lead; ABSD, ABSF, ABSW, ABSP, Total absorption for dust, food, water, paint at low saturation, respectively. *The default function in the IEUBK model.

SUPPORTING INFORMATION TABLE S5. References of Biomonitoring Information in Different Regions in China

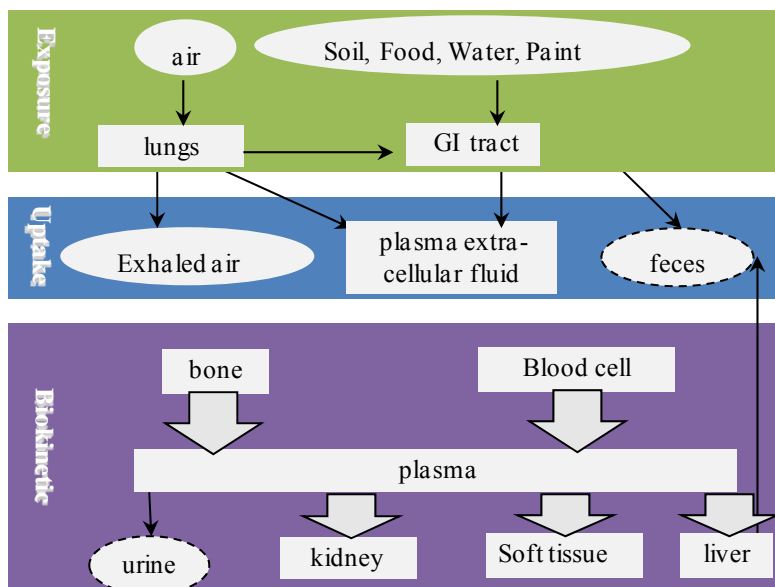
City	Ref.	City	Ref.	City	Ref.	City	Ref.
Chengdu	<i>10</i>	Beijing	<i>15</i>	Kunming	<i>20</i>	Yinchuan	<i>25</i>
Huhhot	<i>11</i>	Wenzhou	<i>16</i>	Xi'ning	<i>21</i>	Harbin	<i>26</i>
Nanchang	<i>12</i>	Shenyang	<i>17</i>	Hefei	<i>22</i>	Haikou	<i>27</i>
Guangzhou	<i>13</i>	Shijiazhuang	<i>18</i>	Zhengzhou	<i>23</i>	Xi'an	<i>28</i>
Tianjin	<i>14</i>	Qingdao	<i>19</i>	Changsha	<i>24</i>		

SUPPORTING INFORMATION TABLE S6. Air lead concentration for different regions in China.

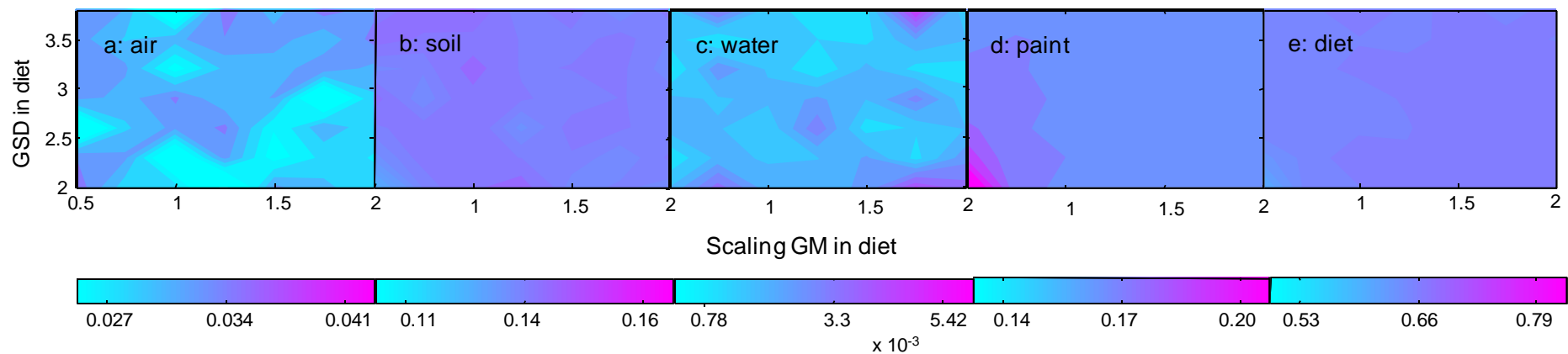
	Air ($\mu\text{g}/\text{m}^3$)	Reference
Chengdou	0.38	29
Huhhot	0.44	30
Nanchang	0.080	31
Guangzhou	0.27	32
Tianjin	0.27	33
Beijing	0.082	34
Shenyang	0.88	35
Hefei	0.072	32
Zhengzhou	0.13	32
Yinchuan	0.047	36
Harbin	0.17	37
Xi'an	0.44	38
Nation	0.34	

SUPPORTING INFORMATION TABLE S7 Prior Distributions, Posterior Distributions, and G-R Test for All Exposure Pathways

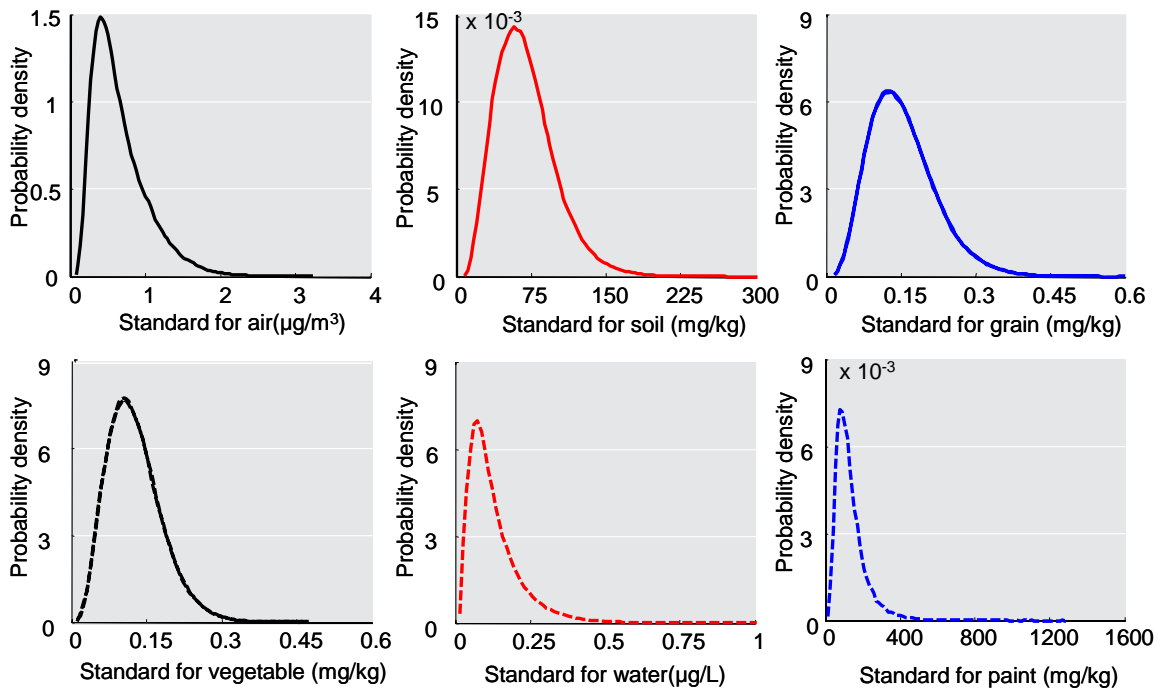
Pathways	Prior distribution			Posterior distribution			G-R
	Population GM	Uncertainty GSD	Interindividual variability	Population GM	Uncertainty GSD	Interindividual variability	
Air ($\mu\text{g}/\text{m}^3$)	0.250	2.28	2.28	0.68	1.28	1.31	1.00
Soil (mg/kg)	25.8	2.67	2.67	82.2	1.50	1.56	1.00
Grains (mg/kg)	0.0800	2.34	2.34	0.0850	1.90	1.91	1.00
Vegetable s(mg/kg)	0.0520	2.76	2.76	0.0700	1.87	1.88	1.00
Water (mg/L)	0.054	2.05	2.05	0.0760	1.55	1.43	1.00
Paint (mg/kg)	553	2.59	2.59	266	2.11	2.12	1.00



SUPPORTING INFORMATION FIGURE S1. IEUBK model structure for predicting lead exposure through the multipathway (I) .



SUPPORTING INFORMATION FIGURE S2. Variation of Source allocations in different media when changing GM and GSD of pPDF for diet exposure. We used colour contour shading with a rainbow scale from blue to rose red to visualize source allocation. a)~e) Source allocation for air, soil, water, paint and diet when scaling GM, GSD of diet lead from 0.5 to 2 and from 2 to 4, respectively.



SUPPORTING INFORMATION FIGURE S3. Standard probability distribution in air, soil, grains, vegetables, drinking water, and paint.

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