Supporting Information

Use of mercury isotopes to quantify mercury exposure sources in inland populations, China

Buyun Du,[†, ‡] Xinbin Feng,[*,†] Ping Li,[*, †] Runsheng Yin,[§] Ben Yu,[†] Jeroen E. Sonke,[#] Benjamin Guinot,[∇] Christopher W. N. Anderson,[∂] and Laurence Maurice[#]

[†]State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China

[‡]University of Chinese Academy of Sciences, Beijing 100049, China

[§]State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China

[#]Observatoire Midi-Pyrénées, Géosciences Environment Toulouse (GET), CNRS, IRD, Université Paul Sabatier, 14 Avenue Edouard-Belin, 31400 Toulouse, France

[∇]Observatoire Midi-Pyrénées, Laboratoire d'Aérologie (LA), Université de Toulouse, CNRS, UPS, 14 Avenue Edouard-Belin, 31400 Toulouse, France

[∂]Soil and Earth Sciences, Institute of Natural Resources, Massey University, Palmerston North, 4442, New Zealand

Co-corresponding Authors:

Xinbin Feng, Phone: +86 851 85891356; Fax: +86 851 85891609; E-mail: fengxinbin@vip.skleg.cn.

Ping Li, Phone: +86 851 84391375; Fax: +86 851 85891609; E-mail: liping@mail.gyig.ac.cn.

The supporting information contains:

13 Pages

3 Tables

4 Figures

This supporting information contains pages.

Table S1 Gender, age, height, weight and dietary information of participants.

Table S2 THg and MeHg concentrations, %MeHg and Hg isotopic composition in human hair and dietary samples.

Table S3 Parameters used in the diet calculation model.

Figure S1 Locations of the three studied sites.

Figure S2 Correlations between hair THg and MeHg concentrations and ages, heights and weights.

Figure S3 Correlation between Hg isotopic composition and %MeHg of hair, rice and fish from GY and CS

Figure S4 Relative contribution of different THg exposure sources for populations from WS, GY, and CS.

Table S1 Gender, age, height, weight and dietary information of participants.

ID	Gender	Age (year)	Height (cm)	Weight (Kg)	Smoking	Alcohol drinking	Dental Amalgam	Rice consumption (g/day)	Fish consumption (times/month)
Participants in WS									
41-A	Male	9	122	23				350	1.5
42-A	Male	9	119	20				270	2.5
43-A	Female	8	115	19				250	0.75
44-A	Female	9	117	18				250	1
46-A	Female	9	120	19				300	1.5
47-A	Male	9	131	25				400	0.85
48-A	Female	8	118	20				270	0.67
49-A	Female	5	110	40				380	2
50-A	Male	7	114	18				300	1.8
54-A	Male	7	114	19				300	0.5
55-A	Female	6	106	18				270	0.95
56-A	Male	7	112	35				370	1.6
57-A	Male	7	115	18				300	1.2
41-B	Male	64	155	50	Yes	Yes		500	1.5
42-B	Female	41	150	53				550	2.5
43-B	Female	60	155	58				500	0.75
44-B	Female	40	150	55				650	1
46-B	Male	61	165	49	Yes			550	1.5
47-B	Female	75	150	55				400	0.85
48-B	Female	27	158	60				600	0.67
49-B	Female	70	157	43				500	2
50-B	Female	63	154	52				500	1.8
54-B	Male	34	165	70		Yes		600	0.5
55-B	Male	37	169	70				650	0.95
56-B	Male	58	165	60				450	1.6
Participants in GY									
H63-A	Male	8	145	25				300	7.6

H64-A	Female	9	140	28				250	8.9
65-A	Female	8	129	25				150	4.4
67-A	Female	8	146	32				300	3.7
68-A	Female	9	145	41				220	1.8
69-A	Female	8	130	22				170	1
70-A	Female	8	135	30				250	4
71-A	Male	9	155	40				300	8.5
73-A	Male	8	135	28				200	4.3
74-A	Male	8	135	35				270	12
76-A	Male	8	130	30				320	1
77-A	Female	36	165	57				350	6.8
61-B	Female	56	150	50				200	6.3
62-B	Male	39	170	72	Yes	Yes		300	6.3
63-B	Male	39	156	57				370	7.6
64-B	Female	37	163	50				170	8.9
65-B	Female	33	148	50			Yes	200	4.4
66-B	Male	43	183	80	Yes	Yes		220	7.6
68-B	Male	39	170	75				300	3.6
69-B	Male	38	178	80	Yes			270	1
76-B	Female	40	163	50				200	8.5
Participants in CS									
81-A	Male	11	140	45				300	1
82-A	Male	7	100	23				200	0.25
83-A	Female	12	158	98				250	2
89-A	Female	11	142	29				220	0.75
90-A	Female	7	90	20				200	1.2
81-B	Female	68	152	45				400	0.85
82-B	Female	30	155	49				650	1.5
83-B	Male	33	170	60	Yes			750	1.7
84-B	Male	28	167	60	Yes	Yes		700	0.85

Table S2 THg and MeHg concentrations, %MeHg and Hg isotopic composition in human hair and dietary samples.

ID	THg ng/g	MeHg ng/g	% MeHg	δ^{202}Hg ‰	Δ^{199}Hg ‰	Δ^{200}Hg ‰	Δ^{201}Hg ‰
Hair from WS							
H41-A	2680	1333	50	0.03	0.05	0.03	0.01
H42-A	3850	2226	58	0.57	0.10	0.02	0.00
H43-A	2730	1087	40	-0.37	0.00	0.03	-0.05
H44-A	6650	1651	25	0.11	0.02	0.01	0.02
H46-A	3660	2803	77	-0.71	0.08	-0.03	0.06
H47-A	7490	4344	58	-1.43	-0.02	0.01	-0.08
H48-A	2270	1098	48	-0.09	0.01	0.01	0.02
H49-A	6370	3117	50	-0.4	0.08	0.02	0.06
H50-A	3060	1578	52	0.28	0.02	0.02	-0.03
H54-A	2990	1381	46	-0.57	0.00	0.00	-0.05
H55-A	6580	3085	47	-0.52	0.07	0.00	-0.01
H56-A	5220	2574	49	-0.34	0.06	0.00	0.05
H57-A	3500	2954	84	-0.48	0.03	0.01	0.00
H41-B	2430	902	37	-0.05	0.10	0.01	0.05
H42-B	6140	2681	44	-0.86	0.02	0.00	-0.02
H43-B	5370	2188	41	-0.48	0.03	0.00	-0.03
H44-B	2740	1598	58	0.18	0.14	0.00	0.13
H46-B	3600	1619	45	-0.43	0.14	0.01	0.08
H47-B	7280	4057	56	-0.27	-0.27	-0.04	-0.06
H48-B	6150	3571	58	-0.26	0.09	0.01	0.05
H49-B	4260	1642	38	0.65	0.09	0.02	0.08
H50-B	2870	1760	61	0.52	0.06	0.03	0.01
H54-B	4220	2279	54	-0.32	-0.21	0.00	-0.27
H55-B	4780	2950	62	-0.07	-0.06	-0.03	-0.09
H56-B	5070	1585	31	-0.59	-0.24	-0.02	-0.36
Rice from WS							
R41	48	31.7	66	-1.50	0.07	0.03	-0.01
R42	46	17.6	38	-1.89	0.08	0.06	-0.01
R43	120	11.5	9	-3.04	-0.03	0.01	-0.05
R44	89	15.6	18	-2.79	-0.02	0.00	-0.05
R45	47	14.6	31	-2.28	0.04	0.04	-0.02
R46	79	18.8	24	-2.97	0.03	0.00	0.01
R47	166	39	23	-2.43	0.00	0.00	0.02
R48	67	15.3	23	-2.48	0.05	0.00	-0.02
R49	45	14.4	32	-2.6	0.04	-0.02	-0.02
R51	78	10.9	14	-2.29	0.00	0.00	-0.01
R52	56	11.9	21	-2.70	0.04	0.01	-0.01
R55	154	15.8	10	-3.50	0.04	-0.01	0.01
R56	52	16.3	31	-2.78	0.08	0.02	0.01
R28	82	11.6	14	-2.96	0.02	-0.02	0.02

R11	53	9.22	17	-2.59	0.05	0.03	0.01
Vegetables from WS*							
V43	74	__	__	-3.66	-0.01	0.00	-0.05
V44	45	__	__	-2.07	-0.09	-0.03	-0.08
V47	39	__	__	-1.63	-0.08	-0.01	-0.06
V48	70	__	__	-2.18	-0.10	-0.01	-0.09
V49	70	__	__	-2.14	-0.09	0.00	-0.08
V52	440	__	__	-2.61	0.08	0.01	0.01
V55	254	__	__	-2.17	0.02	0.01	0.00
V57	124	__	__	-2.26	-0.09	-0.01	-0.1
Hair from GY							
H61-B	583	175	30	1.65	0.35	-0.02	0.27
H62-B	598	355	59	2.36	0.51	0.04	0.41
H63-A	178	148	83	1.51	0.38	-0.01	0.31
H63-B	299	184	62	1.77	0.3	0.03	0.26
H64-A	640	351	55	0.91	0.54	-0.01	0.44
H64-B	477	210	44	2.14	0.56	0.04	0.4
H65-A	312	191	61	1.66	0.34	0.03	0.26
H65-B	361	224	62	1.67	0.47	0.03	0.37
H66-B	368	267	73	1.84	0.73	0.04	0.55
H67-A	558	210	38	1.65	0.25	0.05	0.21
H68-A	520	374	72	1.15	0.01	0.02	-0.03
H68-B	992	399	40	1.91	0.86	0.05	0.75
H69-A	216	143	66	1.84	0.17	0.02	0.19
H69-B	152	124	82	1.77	0.35	0.02	0.30
H70-A	239	134	56	2.19	0.28	0.01	0.25
H71-A	388	355	90	1.79	0.43	0.03	0.33
H73-A	192	165	86	1.02	0.24	0.02	0.16
H74-A	319	229	72	0.86	0.56	0.01	0.42
H76-A	439	118	27	1.36	0.11	0.02	0.05
H76-B	414	387	93	0.82	0.83	0.03	0.60
H77-A	201	193	96	2.03	0.46	0.04	0.41
Rice from GY							
R61	4.2	1.9	45	-1.5	-0.02	-0.03	-0.12
R62	3.2	2.4	76	-1.22	0.03	-0.03	0.00
R63	3.2	1.7	53	-1.44	0.00	-0.01	-0.08
R64	4	2	49	-1.04	-0.01	0.03	0.04
R65	3.3	1.4	43	-1.74	0.00	0.01	-0.08
R66	3.4	1.7	50	-1.43	0.00	0.05	-0.07
R68	4.4	1.5	34	-1.45	0.02	0.01	0.02
R71	3.1	0.9	28	-1.22	-0.03	0.01	0.01
R73	2.7	1.3	47	-1.38	0.05	0.00	0.06
R74	3.7	2.7	73	-1.09	0.08	0.00	-0.02
R76	3.8	2.5	66	-1.3	0.07	-0.01	0.05
Fish from GY*							
F67-10	70	36	51	-0.63	0.36	0.11	0.43

F61-3	133	89	67	-1.03	0.87	0.19	0.84
F65-4	330	124	38	-1.03	0.28	0.07	0.26
F68-2	64	52	83	0.23	1.80	0.22	1.76
F71-7	37	28	75	0.1	1.88	0.25	1.81
F72-3	63	53	84	-0.06	1.34	0.18	1.52
F74-7	51	34	66	-1.28	0.13	0.12	0.26
F75-5	45	43	97	-0.51	0.66	0.21	0.94
F76-2	160	145	91	-0.14	0.99	0.12	0.81
F78-9	134	72	54	-0.43	0.86	0.07	0.71
Hair from CS							
H81-B	420	252	60	0.8	-0.01	0.02	-0.04
H82-A	278	157	57	1.12	-0.04	0.01	-0.06
H83-A	204	184	90	0.84	0.03	0.02	-0.03
H89-A	285	219	77	0.8	0.01	0.00	0.01
H90-A	547	282	51	0.9	-0.01	0.03	-0.04
H84-B	472	252	53	0.76	0.02	-0.01	-0.02
H81-A	313	185	59	1.14	-0.07	-0.01	-0.08
H82-B	230	184	80	1.35	0.01	-0.01	-0.02
H83-B	330	220	67	1.1	0.01	0.00	0.01
Rice from CS							
R81	5.6	2.89	51	-1.78	-0.13	-0.03	-0.18
R82	4.3	2.74	64	-1.99	-0.11	0.00	-0.05
R83	4.8	2.66	55	-1.72	-0.08	0.03	-0.1
R84	4.7	2.75	59	-1.95	-0.14	0.03	-0.08
R85	4.9	2.9	59	-1.86	-0.05	0.01	-0.07
R86	5	3.02	61	-1.97	-0.12	0.03	-0.04
R87	3.7	2.5	68	-1.59	-0.06	0.03	-0.10
R90	5.7	2.8	50	-1.8	-0.04	0.00	-0.05
R91	5.1	3.1	61	-1.81	-0.03	0.01	-0.04
R92	4.3	2.9	68	-1.91	-0.06	0.01	-0.12
R93	4.9	2.5	50	-1.65	-0.08	0.03	0.00
R94	4.3	2.1	50	-1.72	-0.04	0.01	0.01
R95	5	2.4	47	-1.93	-0.06	0.00	-0.07
R88	4.3	2.7	62	-1.91	-0.03	0.01	-0.02

*Wet weight

Table S3 Parameters used in the diet calculation model

Site	Sample	C THg	SD	C MeHg	SD	C IHg	SD	IR	SD	AE (MeHg)	AE (IHg)
		$ng\ g^{-1}$		$ng\ g^{-1}$		$ng\ g^{-1}$		$g\ day^{-1}$			
WS	Rice	71.6[a]	39	17[a]	8	62[a]	37	418[a]	113	0.95[c]	0.07[d]
	fish	290[e]	157	58[e]	26	235[e]	161	2.2[b]		0.95[c]	0.07[d]
	vegetables	98[a]	140	0[f]	0	98[a]	140	378[b]		0.95[c]	0.07[d]
	meat	220[g]	230	0.8[g]	1.2	215[g]	231	65[b]		0.95[c]	0.07[d]
	air	93[f]*	194			93[f]*	194	20[b]**			0.80[d]
GY	Rice	3.6[a]	0.5	1.8[a]	0.6	1.7[a]	0.6	253[a]	62	0.95[c]	0.07[d]
	fish	43[a]	35	27[a]	16	16[a]	24	9.1[b]		0.95[c]	0.07[d]
	vegetables	2.5[f]	1.2	0.02[f]	0.01	2.5[f]	1.2	378[b]		0.95[c]	0.07[d]
	meat	7.5[h]				7.5 [h]		92[b]		0.95[c]	0.07[d]
	air	9.7[i]*	10.2			9.7*	10	20[b]**			0.80[d]
CS	Rice	4.8[a]	0.5	2.7[a]	0.3	2.1[a]	0.5	408[a]	229	0.95[c]	0.07[d]
	fish	43[a,]	35	27[a]	16	16[a]	24	2.2[b]		0.95[c]	0.07[d]
	vegetables	2.5[f]	1.2	0.02[f]	0.01	2.5[f]	1.2	382[b]		0.95[c]	0.07[d]
	meat	7.5[h]				7.5[h]		65[b]		0.95[c]	0.07[d]
	air	7.9[j]*	2.4			7.9 [j]*	2.4	20[b]**			0.80[d]

*： $ng\ m^{-3}$； **： $m^3\ day^{-1}$

Our team has done lots of work including Hg concentrations of food and air in the three sampling sites. So here in this study, we just cited their results in the particular site. [a]This study; [b]Guizhou Statistical Yearbook (2012); [c]World Health Orgnazation (1991); [d]World Health Orgnazation (1990); [e]Qiu et al. (2010); [f] Zhang et al. (2010); [g]Feng et al. (2008); [h]Wu et al.(2016); [i]Fu et al.(2011); [j]Unpublished data.

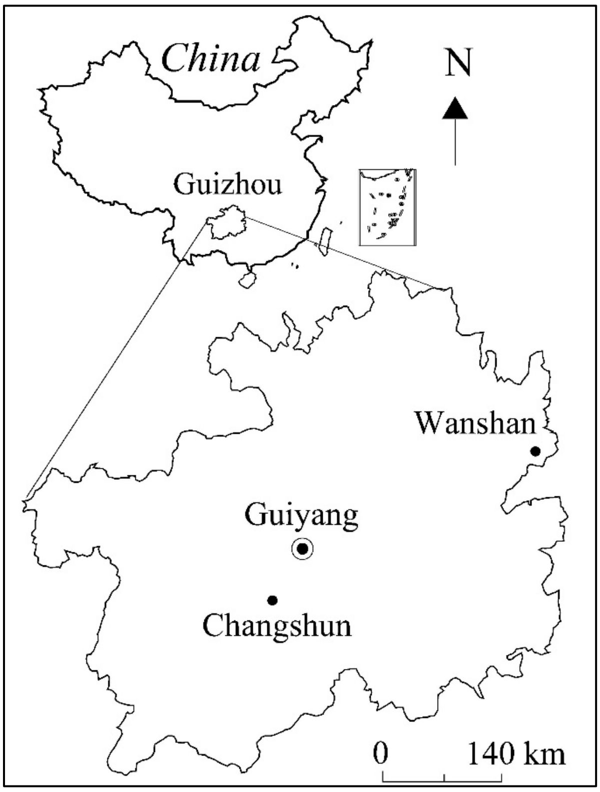

Figure S1 Locations of studied sites

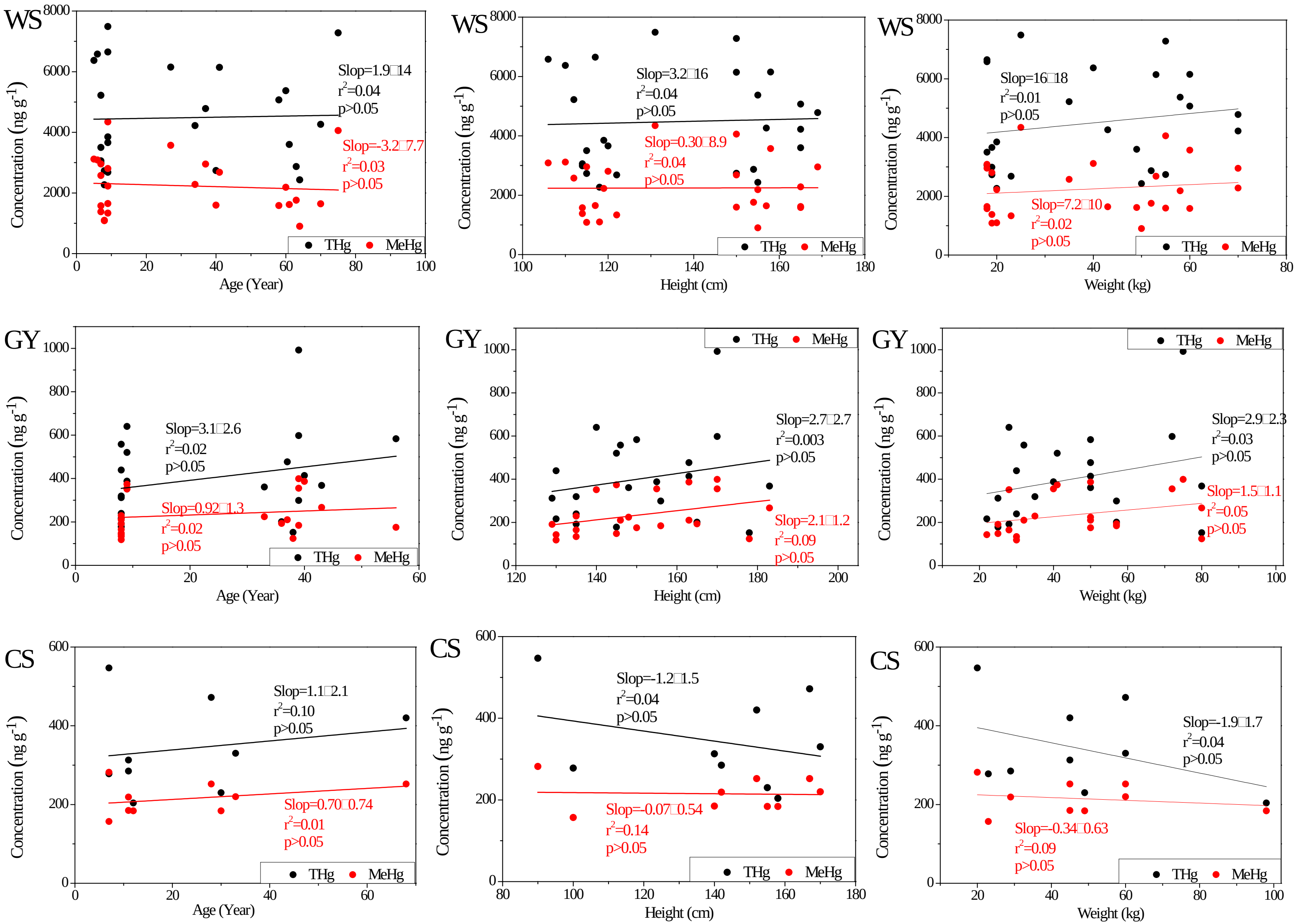

Figure S2 Correlations between hair THg and MeHg concentrations and ages, heights and weights.

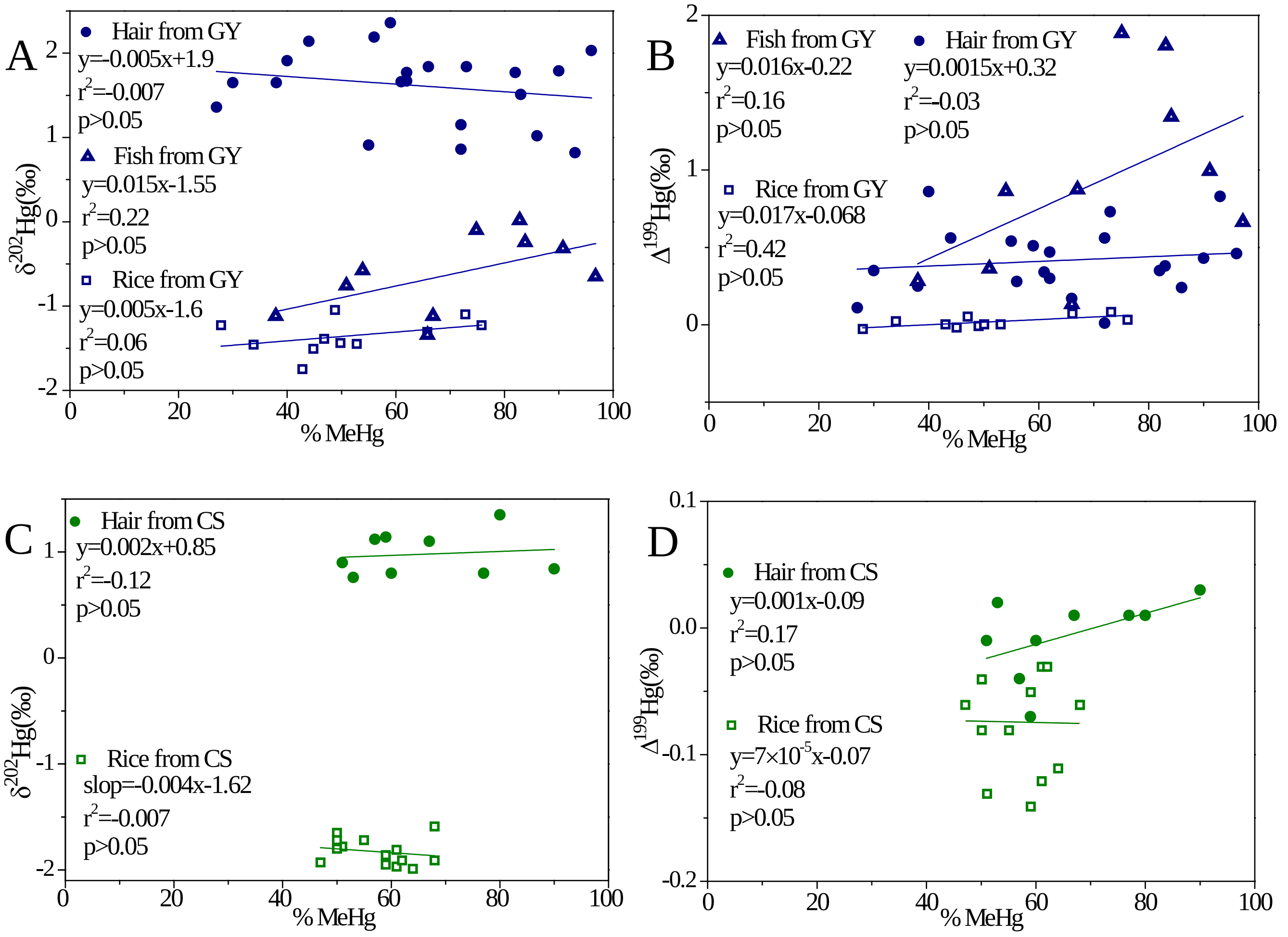

Figure S3 Correlation between Hg isotopic composition and %MeHg of hair, rice and fish from GY and CS.

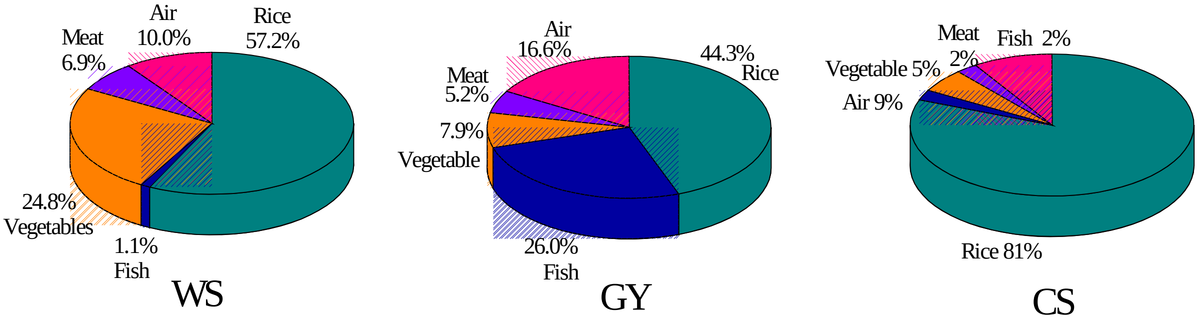

Figure S4 Relative contribution of different THg exposure sources for populations from WS, GY, and CS.

Reference

Feng, X. B.; Li, P.; Qiu, G. L.; Wang, S. F.; Li, G. H.; Shang, L. H.; Meng, B.; Jiang, H. M.; Bai, W. Y.; Li, Z. G.; Fu, X. W. Human exposure to methylmercury through rice intake in mercury mining areas, Guizhou Province, China. *Environ. Sci. Technol.* **2008,** *42,* 326-332.

Fu, X.; Feng, X.; Qiu, G.; Shang, L.; Zhang, H. Speciated atmospheric mercury and its potential source in Guiyang, China. *Atmos. Environ.* **2011,** *45(25),* 4205-4212.

Guizhou Statistical Yearbook, *Guizhou Bureau of Statistics.* China Statistics Press: Beijing, **2012**.

Qiu, G. L.; Feng, X. B.; Wang, S. F.; Fu, X. W.; Shang, L. H. Mercury distribution and speciation in water and fish from abandoned Hg mines in Wanshan, Guizhou province, China. *Sci. Total. Environ*. **2009,** *407,* 5162–5168.

WHO. Environmental health criteria 101: Methylmercury. World Health Organization, Geneva, **1990**.

WHO. Inorganic Mercury Environmental Health Criteria. Vol. 118. World Health Organization, Geneva, **1991**.

Wu, Y.; Zhang, H.; Liu, G.; Zhang, J.; Wang, J.; Yu, Y.; Lu, S. Concentrations and health risk assessment of trace elements in animal-derived food in southern china. *Chemosphere.* **2016,** *144,* 564-570.

Zhang, H.; Feng, X. B.; Larssen, T.; Qiu, G. L.; Vogt, R. D. In inland China, rice, rather than fish is the major pathway for methylmercury exposure. *Environ. Health. Perspect.* **2010,** *118,* 1183-1188.