SCIENTIFIC REPORTS

OPEN

SUBJECT AREAS:
POLLUTION REMEDIATION
GEOCHEMISTRY
ATMOSPHERIC CHEMISTRY
ENVIRONMENTAL MONITORING

Received
10 December 2012

Accepted
8 November 2013

Published
25 November 2013

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Atmospheric mercury inputs in montane soils increase with elevation: evidence from mercury isotope signatures

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The influence of topography on the biogeochemical cycle of mercury (Hg) has received relatively little attention. Here, we report the measurement of Hg species and their corresponding isotope composition in soil sampled along an elevational gradient transect on Mt. Leigong in subtropical southwestern China. The data are used to explain orography-related effects on the fate and behaviour of Hg species in montane environments. The total- and methyl-Hg concentrations in topsoil samples show a positive correlation with elevation. However, a negative elevation dependence was observed in the mass-dependent fractionation (MDF) and mass-independent fractionation (MIF) signatures of Hg isotopes. Both a MIF (Δ^{199}Hg) binary mixing approach and the traditional inert element method indicate that the content of Hg derived from the atmosphere distinctly increases with altitude.

M [illegible]

[illegible]

a ac ca a a a b c c -
K c H (' a a. c').
N K , K a H aK b c c
a. a a a a a - Ka a a -
c [9,10,13]. C , c, b -
c ca c c H a c a
. I c ca , a b a a-
ba / c a H b a
a a c - c a Ka -
(. ., CMAQ-H [14] a GEOS-C [15]), a -
a a. a aK b K a c
c . I b a a a. c H
a , K , a Ka . B a
, 27% Ea ' a a ca b ca a a -
, c a a a , a ab b 22%
' a [16]. I C a, a acc -
a a a a, >50% c a a a-
a Ka > 1,000 a. . [16].. T ,
H a a a c C a
a a c a a c Ka a
a a ba H b a c c .
S c a aK ca b a -
c c a a b ca a -
a c a a , c ca c
a [8,17]. A c a a H a c c
14 ac U.S. a a b a
H (90%), b (8%) a ab K
b a (<1%)[18]. H K , a H c c -
a a a c a c a b ca
a H c c a b ca a c -
, a a a a bac , a a c a a
a a c [9].
O a b a H c a b
a c , a ca , c K -
a H c c H . T a c a K b a. -
a. ac ac c H a a
c c ca c a a. c H c c . R c aK
a a H a Ka a
c a a a a H ca b a ca ac -
a c c K a c [19 21]. I a
a - ac a (MDF), a - ac a-
(MIF) - a H ([199]H a [201]H) a cc
a a c c c a c a a c c
(MIE)[22,23] a c a K c (NVE)[20,24]. A c b a a
MDF a MIF H a a ca b a a
c K ac a c c [25].
M . L (' ') a (2,179 a. . .,
26.39°N, 108.20°E) M a Ra G P K c ,
C a (F . S1) a a c . T a ,
c a a b ca c , ca
a Na a Na R K (473 ^{2}). B ca
a a K c a a
[11], a a c c . T b c M .
L a c - a a c c
P -S a A , a a b [26]. T
a K S C a S a (~750) ca M .
L b a a. c b
ab a a a (a a . c. a a ~1250
a >1600). F a
a a a , a a a a c a b
0.46°C 100 Ka a a . 9.2°C
[12]. T (~300
a $^{-1}$), a c <25% a
a [11,26].
T a c c c a
a a ac a a c K c

a c H a a c a a c -
a [27]. T K H a a Ka a a c
a a a K a c c a
a a H a a H [10]. H
a a ac
a a H c a b
c a ' a a. c' H .

Results

Hg levels and distribution along the elevation gradient. E Ka
a H (TH) K b K a c a
bac aK a C a (0.052 · $^{-1}$ (28)),
a a (a) 0.18 (0.07 0.34) · $^{-1}$ a 0.20 (0.08
0.38) · $^{-1}$ a c c S b 2009 a 2010,
c K (F . 1). T a (a) M H c
c a 2010 a 2.16 (0.26 5.05) µ . $^{-1}$.
N ca c (ANOVA, > 0.05) TH K
a b K b a c c a a a
2009 a 2010; , a a b c b
c . T TH K (a 0.19 · $^{-1}$)
M . L c a aK ab b
a a U.S. (0.15 · $^{-1}$), N a
(0.19 · $^{-1}$) a S (0.25 · $^{-1}$)[9].
T TH a M H c c a a
c a a (^{2} = 0.68 0.71, < 0.01 b)
(F . 1). T a 0.12 µ . $^{-1}$ $^{-1}$ a
3.1 . $^{-1}$ $^{-1}$ TH a M H , c K . B TH a
M H c c a a c a
a. a a a a .
F , TH c c a a a a -
b a Ka (^{2} = 0.33 0.39, < 0.01
b) (F . S2 S3). T a ca a a a
a ca c (. a a. c) a c
H M . L . S K a -a aK

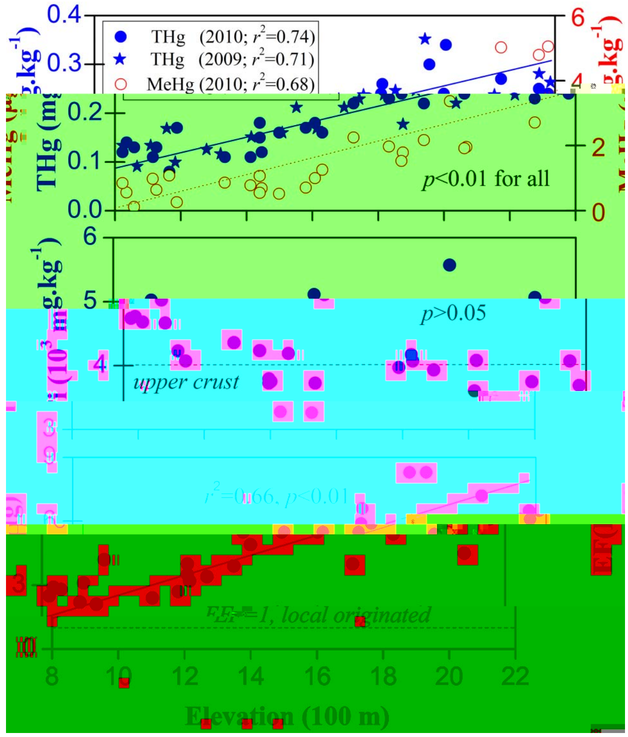

Figure 1 | **Scatter plots of soil THg and MeHg contents (upper panel), soil Ti content (centre panel) and calculated enrichment factors (EF(Hg) = $(Hg/Ti)_{soil}/(Hg/Ti)_{crust}$) (lower panel) versus elevation.**

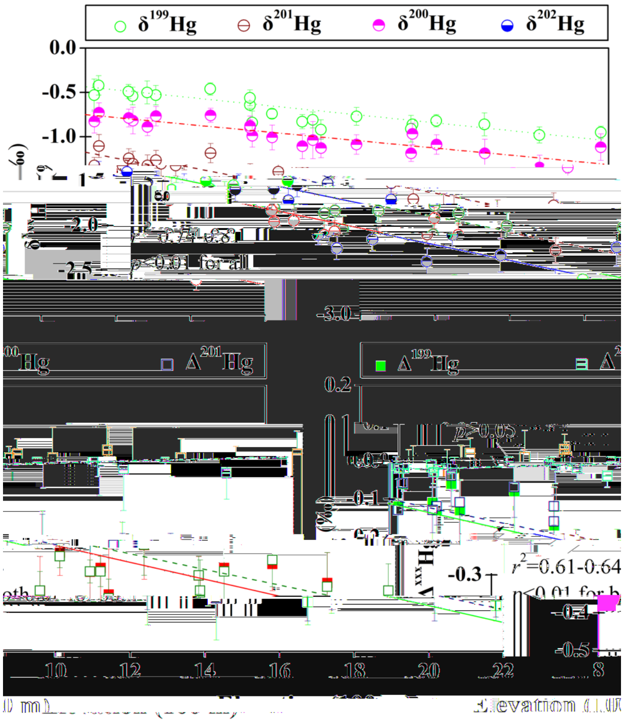

Figure 2 | Scatter plots of mean δxxxHg (upper panel) and mean ΔxxxHg (MIF, lower panel) isotope ratios in surface soil versus elevation. All error bars represent ± 2 s.d.

Ka a c c c a a a H [10,17,18]. H c , a b c c a a c K c a c H a .

Hg isotope ratios distribution along the elevation gradient. S ca MDF (a 1.2‰ a δ^{202}H) a MIF (a

0.3‰ a b Δ^{201}H a Δ^{199}H) a b K a a (F . 2). A c a K Ka a b K TH a Ka (^{2} = 0.61 0.82, < 0.01) (F . 2), c (‰ 100 Ka a a) b a b a −0.039, −0.040, −0.083 a −0.083 δ H (= 199, 200, 201 a 202, c K) a −0.020 a −0.021 Δ^{199}H a Δ^{201}H , c K . F , Δ^{201}H Ka c a Δ^{199}H Ka (^{2} = 0.98, < 0.01) (F . 3). N ca MIF K (. ., ^{200}H a ^{204}H) a b K a K a a (c , a c a).

Tracing and quantifying the atmospheric Hg inputs in soil samples. (1). I . T a a b H c M . L a . T a (T), a c Ka K c ca a . c , a c a c ca c a H c ac (EF) (a EF[H]) ba ac a c c c a [28] acc a :

$$EF(H\ ) = (H\ /T\ )\ \ /(H\ /T\ ) \quad (1)$$

$$H\ = H\ - T\ (H\ /T\ ) \quad (2)$$

A F . 1 a , EF(H) a K c a Ka (^{2} = 0.66, < 0.01). A EF(H) Ka c 1 ca a H c . T c a EF(H) Ka c a Ka a - c c H (a b a H , a c) c a Ka . T a a c H TH K c a a . a 90% a a 50% a ba a (^{2} = 0.64, < 0.01) (F . 4), a ca c b - c c (a a c) ac H M . L , a c a a Ka .

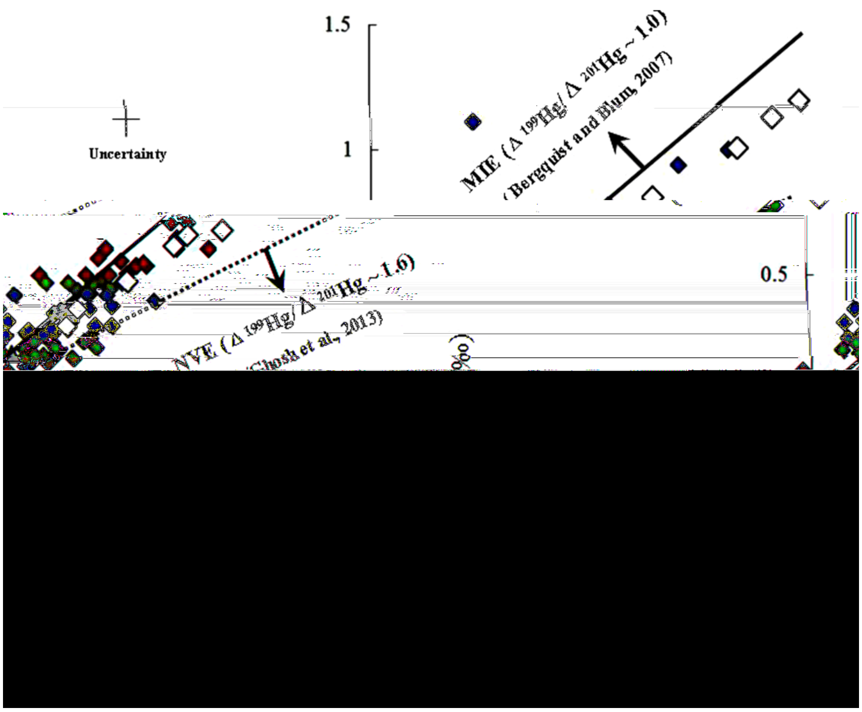

Figure 3 | A comparison of the relationship between Δ199Hg and Δ201Hg from various studies (MIE = magnetic isotope effect; NVE = nuclear volume effect).

(2). H a . A b B a B[19], a. ca ab H a a c a c a a a H ca b c a , ac a MDF a ca c . C a P MDF, MIF a a c c c a c ca c , a b ab b Δ^{199}H /Δ^{201}H a . Acc c P , MIF c b a b MDF c , b ca b c a b MIF c y y y H P y MIF y a . S K a aK b K ca a K Δ^{199}H Ka c c a c H [29 31]. H P K , a P F . 3, c y c a a a c H c aK P K Δ^{199}

S MDF (δ^{202}H) b a aK b cc -
a H c a c [40,41].
I , δ^{202}H a 1/TH a a -
K a c a (^{2} = 0.48, < 0.001) (F . S5), ca
b a a c H a c H . A ca
c a (^{2} = 0.68, < 0.001) b δ^{202}H a X
a b K (F . S6). Ba c a , MDF -
a - b b a (F . S6). T δ^{202}H
c a a −1.38‰, a a
a c a a −2.50‰. T δ^{202}H
a c H MDF c a b K δ^{202}H
a (δ^{202}H −2.21 ± 0.14‰, 2 . . , = 3) a a
a MDF −0.29 ± 0.14‰ a aK cc ab -
H b . M a c a a c a
a c H a a. I ac -
a aK a (. , C , Z a F) aK b a
cc , a a a ca H
a . R c a a a a (. , c [31]
a c [36]) ca a c a H
. T c c H a K
a b c . T b c M . L -
, a aK a δ^{202}H c a −0.89 ± 0.10‰
(2 . . , = 2), c ca a a MDF −0.49 ± 0.10‰
δ^{202}H a cc a . M . L a a b- ca
c a a a a c a 1250 1700 [11]. I c a
c a , a a K K ac . R c ac -
a H a c a a
b H ac a a c aK H
[42]. T δ^{202}H a c H (−2.50‰) c a -
ab K a a a (. , c [31], a [34] a
c [36]), ca a a ca a c a
H .

Discussion

Potential mechanisms for Hg isotope signatures in montane soils. V ca K a H cc c
ca b a b ac a H
c c c a / H
c (. , a ca c). I a
a a c a M . L , K a a (. ,
a , c a a a a a) K c
b c ca b aK H a a H
ac a . T ac a H M . L
K a a b a c c -c ca
c , c a K a H ^{0} ,
a c H (. , c a , a a)
a - - H . T b -
, K a H a K K c c
a - c [22], K a a [24,43] a c b a c
H [37]. G a , a c c ca c MDF
K a H a c H ^{0} ca
δ^{202}H K a a H $^{2+}$. T - c H
a a MIF H [20,22,44], a -
ca MIF c b c K a a a
c b a c c [24,37,43]. A c b D
a [34]. a ca a - c , K a a a c -
b a c c b a c K a H
a a . I c a a M . L , H
b a c a -
c a a c a [9]. S K a a a
a a b ca a c a
c , b a c K , a ca a [34].
O M . L , ac aK c K H ca
c (. , a) a a c c (a
). I , H K c a (0.10
± 0.02 · $^{-1}$, 2 . . , = 2) a

a . T δ^{202}H (−0.89‰ ± 0.04, 2 . . , = 2) a
Δ^{199}H (−0.02‰ ± 0.04, 2 . . , = 2) c a a
c a a K , a H c
a a (. , a [45], a [45] a K -
ca [46]) a aK δ^{202}H K a a −0.60‰,
K c a ca MIF (Δ^{199}H < 0.2‰)[19].
D c c H , a c c
H c a aK b c a a c
(. , c a) a (. , a c a H a
a) a c H . F aK c
H c c a a c a c
H c [25,32,33]. I a a c b
aK a a ca MIF K (. , ^{200}H a
^{204}H) c a a c a c H a [25,32,34].
G a , H ^{0} c a ac b a K Δ^{200}H K a , a -
c a (c c a a H $^{2+}$ a H) a K
Δ^{200}H K a . I , ab c a MIF K
H ac c b a b
a c H c a c a . A a K ,
C a [33]. a MIF K H
- a H ^{0} a , b c b a-
c , c a , a , a
a a a a a K . S K a aK
a - Δ^{200}H K a a b a H G a La
[32], A c c [25] a a Wa a M c M [31], -
ca a - a H ^{0} a cc c a a a
a c a ca MIF K H . T c -
a MIF K H c a [33], a
a .
I , ca a K MIF H -
ab a a a a K a a -
(. , c a ac). T a b c a a
a -N MIF H c (1)
a c c (MIE)[47] a (2) c a K c
(NVE)[48]. T Δ^{199}H /Δ^{201}H a MIF c b
c a a b a c. Acc K a c ,
MIF cc NVE (. , H ^{0} K a a , ab c a
c H $^{2+}$ a b H $^{2+}$- c a) a
a a Δ^{199}H /Δ^{201}H a 1.5 2.0[20,23,24]. T
MIE a b c c ca ac a -
H c (. , M H a H $^{2+}$). W Δ^{199}H a Δ^{201}H
K a a ac c ca c ,
CH_3H^+ a H $^{2+}$ - c aK 1.36 a 1.00,
c K [22]. T MIF c b MIE
a c a K K [20,22,44]. A F . 3,
a K Δ^{199}H K a ac M . L ca
a c , c c a c /
c a a a ac
[23]. T a a 0.98 a a b a
Δ^{201}H /Δ^{199}H (c c a b NVE) ca
a a H a aK
- c c b b c -
a .

Potential mechanisms for Hg magnification in montane soils. W K H , a ca K a a , a c a
a a K a c
a c a b ' c '
a ' ' H a c c ca c
a a c b K a c . A c a
a ca a acc a H a a
K a a a a a c F . 5 a a
a c .
(1). L a . L a a c ca H a
c a , c a c a
aK c [17]. H ^{0} a K c

c aK ca P a a a -
ca P a Pa
a a [49,50]. T a c a a-
H 0 b a Ka a a a c c K .
F , H b c c c a
P ab K a (50% 800%)[17,18]. D (-
a) ca acc 40% a 80% a H a a -
P a , c K [17,18]. I a
K c ca [10], P a H a
M . L P , a a ba , b a -

b H a a G (2012)[7] a ca K H
a Ka .

O M . L , c a c a a a - ca a (a S I a)[11,12]. S ca K c a b H K a c a K a (^{2} = 0.67 0.69, < 0.01 b) (F . S7) b K , a a a b ca K a c / H - Ka a - a c a- - a ca c b ab K . H K , c a - a a c b a a c . F a , a a a a b a a ca ac c H b a a [59]. S a a a ca Ka M . L [26] a a (c a c c K a a c a - b a a)[16], b c c a a [1], c , c H a ac a .

O M . L , a a a a a a c a a c a , /c a a c a c a- Ka [11,12,26]. H c , - (' a c'). - c a a K c a a c H a c a H c c a , ca a Ka . I c a , caK a c H b c a a a c a K c a H c c a a a . T c a b a a a a c a H c c a a Ka .

Implications for regional or global Hg cycling. A a K Ka - a c a b K MDF a MIF a H . T a ca a MIF (Δ[199]H) b a a ac a a a a ca a ac H K a c c a a . O , a a a ' a a c' -K a H ca cc - a K a K a ac c b ca . M c a ca - Ka a K b a a c a , c a , a a ac (. ., a a a), O ac a a b c ca . O b Ka a K a ba H c c / b a aK ca a H a a b a , a a acc a ca ba a a a. O a H ab a ca b ac a c H a . T c a b a a K a c a a c .

Methods

T TH c c a a a c a c - Ka a c ab c (CVAAS), a TH c c a a a a a - a a a- a a a c -Ka a c c c c (CVFAS) c USEPA 1631. T M H c c a a a , a a GC CVAFS c USEPA 1630. T H c a MC-ICP-MS a N -Pa a a c 12 Fa a a c . T a - c H c c a a a c ba , ba , a , c c a a b ca . T - c b c a a a a a a ca a (ca = 2). W a a a UM-A a a a c a a a (c K 10 a) a b ac a a NIST 3133. T H c - c a UM-A a a a a a ac a a ca .

D a a a c , a c c a - a a , a a H c c a a H a , a a a c a c , a c a c a a a K S a I a .

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Acknowledgments

T P Pa b "973" P a (2013CB430003), Y I Ka P A ca C Aca Sc c , Na a Sc c F a C a (40825011 a 41203092) a S -N P a c a c 'Ca ac b c c C a - a ca G K c ', P c Pa b N P a G K .

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H.Z., R.S.Y., X.B.F. a T.L. c c K c. H.Z., R.S.Y., X.W.F. a A.S. a a . H.Z. a R.S.Y. a H c c c a a H a . H.Z., R.S.Y., X.B.F., T.L., C.W.N.A. a J.S. a a a a a. H.Z. a R.S.Y. P a. P c a a .

Additional information

Supplementary information acc a a. a ://PPP. a .c / c c .

Competing financial interests: T a ca c a c a .

How to cite this article: Z a , H. . A c c a c a P Ka : K c c a . . . 3, 3322; DOI:10.1038/ . 03322 (2013).