

An Isotopic Dilution Approach for Quantifying Mercury Lability in Soils

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30 **Supporting Information:**

31 **S.1. Materials and Methods:**

32 **S.1.1. Soil sampling and chemical characterisation**

33 Soil samples were collected from nine different sites (located in some residential areas of
34 Visp (46° 17' 32'' N, 7° 52' 58'' E) and Raron (46° 18' 36'' N, 7° 48'' E) towns, Canton
35 Valais, Switzerland, where a past industrial contamination with Hg is suspected. Three
36 topsoil cores (collected at 0-10 cm depth) were taken from each plot within an area of 10
37 x 10 m or along a line-transect. One of these locations (VS2) was taken as a background
38 reference based on a previous investigation performed by Arcadis AG at this site (mixed
39 samples n=16) which showed THg concentrations of < 0.1 mg kg⁻¹. Samples were
40 collected using a previously cleaned stainless steel auger and sealed in polyethylene bags
41 for transport. Soils were air-dried then sieved to < 2mm particle size for analysis.

42 Soil pH was measured using a glass electrode after 5 g of 2 mm sieved soils were
43 equilibrated (end over end shaker) for 30 min in 12.5 mL of Milli-Q water. Soil organic
44 carbon content (%SOC) and organic nitrogen were determined by a Lyco CN 628 analyser.
45 About 0.25 g of ball milled soils were weighted into tin capsules and combusted at 1000°C.
46 Carbon was measured by an infrared detector in oxygen rich environment while nitrogen
47 was measured by thermal conductivity in helium. Reactive Fe, Al and Mn oxides in soils
48 were extracted by the dithionite procedure developed by Kostka and Luther ¹. Finely
49 ground soils (agate ball mill) were suspended in 25 mL of a solution containing 0.22 M tri-
50 sodium citrate, 0.11 M sodium hydrogen carbonate and 0.1 M sodium dithionite and
51 shaken for 16 hours in a water bath at 40°C. Centrifuged and filtered samples were
52 retained for analysis of Fe, Al, and Mn by ICP-OES and concentrations (mg kg⁻¹) of Fe₂O₃,
53 MnO₂, and Al(OH)₃ were calculated. Concentrations of metals in the dithionite and acid
54 extracts (diluted in 5% v/v HNO₃) were assayed by an Agilent ICP-OES 5100. Sample
55 processing was undertaken using ICP Expert software (version 7.2). Total sulfur in soil
56 samples was extracted by the Swiss standard regulations on soil pollution protocol
57 (Verordnung über Belastungen des Bodens; VBBo) ². In triplicates, 10 g of 2mm sieved

58 soils were extracted with 100 mL 2M analytical grade HNO₃ in a boiling water bath for 120
59 min. Total sulfur in the acid extracts was also assayed by ICP-OES. Total soil mercury
60 (THg) was measured using a Perkin Elmer SMS100 atomic absorption spectrometer
61 according to the EPA standard method 7473. Soil samples were thermally decomposed at
62 750°C followed by amalgamation on a gold-trap and then analysis of Hg cold vapors.
63 Certified reference lake sediment material (IAEA SL-1) was used for calibration. Replicate
64 samples and reference material were analyzed regularly (10% of the sampling sequence)
65 and the precision was under 10% relative standard deviation (SD).

66 **S.1.2. Determination of isotopically exchangeable Hg (Hg_E) in soil**

67 Hamon, et al. ³ and Marzouk, et al. ⁴ demonstrated that the spiked isotopes may disturb
68 the natural isotopic equilibrium leading to a systematic variation in the measured E-values
69 at higher spike levels. However, in practice, the amount of the added spike varied from
70 c. 5% to 400% of the metal soil concentration and the 'appropriate' spike concentrations
71 were usually selected based on the investigated metal, its concentration, soil properties
72 and the robustness and accuracy of the isotopic ratio measurement ⁴⁻¹⁵.

73 In this work, spike levels were chosen based on a preliminary experiment involving 3 soils
74 with high, medium and low THg contents which showed that at least 50% increase in the
75 soil ¹⁹⁹Hg and ¹⁹⁶Hg content was required for robust isotopic ratio (IR) measurements. This
76 was mainly due to our carryover elimination strategy (SI, S.1.3), which included diluting
77 all solutions to < 2 μg L⁻¹ Hg. Therefore, a careful balance was required between
78 maintaining analytical precision while keeping spike concentrations to the minimum to
79 avoid possible perturbation of the natural equilibrium ^{3, 7}. The original plan was not to
80 exceed 100% of soil ¹⁹⁶Hg or ¹⁹⁹Hg; however, this was impractical for ¹⁹⁶Hg in all soils due
81 to its very low natural abundance. The authenticity of the estimated values of Hg_E was
82 determined by checking their consistency against different spike levels ^{3, 4, 16}. The possible
83 precipitation of Hg isotopes at high spike concentrations was modelled with Visual MINTEQ
84 3.1 for the highest spike level in our range (4.1 μg mL⁻¹ ¹⁹⁹Hg). Results showed that, under

85 our experimental conditions, both $\text{Hg}(\text{OH})_2$ and HgCl_2 displayed negative saturation
86 indexes (-7.4 and -1.2, respectively) indicating negligible precipitation.

87 **S.1.3. Measuring equilibrium Hg isotopic abundances**

88 Isotopic abundances of ^{196}Hg , ^{199}Hg , ^{200}Hg , ^{201}Hg and ^{202}Hg in both spiked and un-spiked
89 samples were assayed by an Agilent 7700x ICP-MS in helium collision mode. A three steps
90 washing sequence between samples was used to eliminate memory effect and cross
91 contamination; this included (i) a solution of 1 g L^{-1} EDTA, 0.08 g L^{-1} Triton X-100 and 6
92 g L^{-1} NH_4OH , (ii) technical grade 5% HNO_3/HCl and (iii) the matrix solution (ultra-pure 1%
93 $\text{HNO}_3/ 0.5\%$ HCl). In addition, all samples and standards were diluted to a $0.05 - 2 \mu\text{g L}^{-1}$
94 range of total Hg.

95 For internal mass bias correction, Tl solution ($10 \mu\text{g L}^{-1}$), in a matrix of ultra-pure 1% HNO_3
96 and 0.5% HCl, was introduced in the internal standard line via a T-piece and the variation
97 in the $^{203}\text{Tl}/^{205}\text{Tl}$ ratios was used to correct shifts in Hg isotopic ratios ^{6, 17, 18}. Moreover,
98 external mass discrimination correction was applied by repeatedly analyzing a $2 \mu\text{g L}^{-1}$ Hg
99 ICP-MS standard every three samples ¹⁹. Changes in the measured count rate (CPS) ratios
100 of the bracketing Hg standard were applied by linear extrapolation to the intervening
101 samples ^{5, 11}.

102 The abundance of ^{195}Pt was measured to monitor and correct for possible isobaric
103 interference between ^{196}Pt and ^{196}Hg although in practice Pt concentrations were below
104 detection limits in all samples.

105 To ensure high level of measurement precision, repeated runs of very short quadrupole
106 dwell times were applied. Five runs, each composed of 1000 sweeps, were applied to each
107 sample and the dwell times were 5.01 ms for ^{195}Pt , ^{196}Hg , ^{199}Hg , ^{200}Hg , ^{201}Hg , ^{202}Hg , ^{203}Tl
108 and ^{205}Tl . RSDs between the measured counts per second (CPS) of the five runs, for each
109 isotope, were always kept below 1%; data were rejected and repeated if RSD exceeded
110 1% for any single isotope in the sequence. To allow for the fast jumping quadrupole to

111 settle locally before sweeping through the main isotopes ⁴, ¹⁹⁴Pt (0.3 ms) was also added
112 to the sequence to work as a resting mass.

113 **S.1.4. E-value comparison with extraction methods**

114 Despite the popularity of extraction methods to estimate the available fraction of soil Hg,
115 unfortunately there is no universal protocol available such as the sequential extraction
116 procedure developed by Tessier, et al. ²⁰ for classic hard metals ²¹. The type and sequence
117 of the selected extractants have always been an empirical decision depending on the
118 nature of soil or the targeted Hg species ^{21, 22}. Moreover, when compared to Hg uptake by
119 flora or fauna, good correlations have been observed between bioavailable Hg and Hg from
120 all soil fractions indicating that labile Hg is not exclusively bound to specific soil phases ^{23,}
121 ²⁴. However, the first step of extraction using ammonium acetate or magnesium chloride
122 to extract 'exchangeable Hg' was common in the available literature ^{21, 22, 25-27}.

123 For comparison with Hg E-value, 'Exchangeable' mercury in all soil samples was extracted
124 by ammonium acetate or magnesium chloride as described by Han, et al. ²⁵ and
125 Panyametheekul ²⁶. In duplicates, 2 g of <2mm sieved soils were suspended in 30 mL 1
126 M ammonium acetate or 0.5 M magnesium chloride and shaken for 1 h and 2 h,
127 respectively, at room temperature. Centrifuged and filtered samples were diluted with 5
128 % HCl as necessary and Hg concentrations were measured by an Agilent ICP-OES 5100
129 fitted with Multimode Sample Introduction System (MSIS) for vapour generation.

130 **S.2. Results and Discussion:**

131 **S.2.1. Soil Properties**

132 Key soil parameters including total soil Hg (THg), pH, soil organic carbon (Org-C), total
133 nitrogen, total S, reactive Fe, Mn and Al oxides are summarized by sampling site in Table
134 S1. All soils were alkaline or slightly acidic with a mean pH range of 6.2 to 8.2. In general,
135 soils showed average organic carbon content (2.6 to 4.5 %). Total soil Hg ranged in
136 average from 0.37 to 310 mg kg⁻¹. All locations showed THg concentrations higher than
137 the 0.37 mg kg⁻¹ measured in the VS2 background (reference) site. However, THg

138 concentrations were noticeably elevated (230 - 390 mg kg⁻¹) in location VS2 followed by
139 location TT2 indicating high levels of mercury contamination.

Table S1. Soil properties by sampling locations including total Hg (THg), pH, organic carbon (org-C), N, total S and reactive metal oxides in soils in Visp and Raron, Switzerland differently contaminated by Hg. SD= standard deviation.

Site	THg (mg kg ⁻¹)		pH		Org-C (%)		N (%)		S (%)		Al(OH) ₃ (g kg ⁻¹)		MnO ₂ (g kg ⁻¹)		Fe ₂ O ₃ (g kg ⁻¹)	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
HW1	0.95	0.95	7.29	0.05	2.63	0.31	0.21	0.02	0.36	0.03	0.72	0.16	0.23	0.04	4.00	0.67
HK1	2.80	1.21	6.82	0.52	3.43	0.22	0.31	0.05	0.47	0.06	0.47	0.12	0.13	0.05	3.40	0.74
HK2	2.73	0.25	7.61	0.15	2.95	0.54	0.21	0.05	0.35	0.08	0.43	0.03	0.18	0.02	3.84	0.31
XX1	7.54	3.45	7.36	0.14	4.07	0.30	0.40	0.03	0.63	0.05	0.45	0.05	0.14	0.04	2.91	0.40
XX2	16.9	7.86	7.33	0.38	4.48	0.81	0.47	0.07	0.71	0.05	0.55	0.01	0.19	0.01	4.12	0.26
TT1	2.68	1.63	7.48	0.27	3.89	0.67	0.31	0.03	0.56	0.09	0.42	0.10	0.21	0.16	3.96	0.73
TT2	38.1	8.62	8.20	0.13	2.95	0.12	0.23	0.01	0.78	0.16	0.62	0.01	0.16	0.01	3.82	0.20
VS1	310	80	7.26	0.22	3.49	1.33	0.24	0.15	0.54	0.22	0.70	0.20	0.16	0.06	3.44	0.88
VS2	0.37	0.29	6.20	0.51	3.03	0.96	0.29	0.10	0.42	0.16	0.40	0.04	0.14	0.01	2.85	0.29

Table S2. Mass bias correction factors in the ratios of spike isotopes (^{196}Hg and ^{199}Hg) to reference isotopes (^{200}Hg , ^{201}Hg and ^{202}Hg). The expected drift due to mass discrimination was corrected for by periodically running a $2\ \mu\text{g L}^{-1}$ certified Hg standard and applying the changes in its isotopic ratios to the intervening samples. A, B and C are different soil sampled from the same location while 1 and 2 are replicates of the same soil. Isotopic ratios of the bracketing Hg standard used for mass bias correction are displayed in bold italic font in their exact position in the analysis sequence.

Location	Soil	Spiked Sample	Isotopic ratios mass bias correction factors					
			$^{196}\text{Hg}/^{200}\text{Hg}$	$^{196}\text{Hg}/^{201}\text{Hg}$	$^{196}\text{Hg}/^{202}\text{Hg}$	$^{199}\text{Hg}/^{200}\text{Hg}$	$^{199}\text{Hg}/^{201}\text{Hg}$	$^{199}\text{Hg}/^{202}\text{Hg}$
		<i>Natural Ratio</i>	<i>0.006494</i>	<i>0.011381</i>	<i>0.005023</i>	<i>0.730303</i>	<i>1.279970</i>	<i>0.564970</i>
		<i>Hg std.</i>	<i>0.006748</i>	<i>0.011737</i>	<i>0.00518</i>	<i>0.727679</i>	<i>1.265749</i>	<i>0.558636</i>
HW1	A	1	0.928	0.930	0.926	1.000	1.002	0.998
		2	0.910	0.911	0.907	1.001	1.002	0.997
	B	1	0.892	0.892	0.888	1.001	1.002	0.997
		<i>Hg std.</i>	<i>0.007287</i>	<i>0.01271</i>	<i>0.005611</i>	<i>0.725363</i>	<i>1.265201</i>	<i>0.558526</i>
	C	2	0.882	0.882	0.877	1.001	1.001	0.996
		1	0.891	0.891	0.885	1.000	1.000	0.994
HK1	A	2	0.899	0.899	0.894	0.999	0.999	0.993
		<i>Hg std.</i>	<i>0.00697</i>	<i>0.012133</i>	<i>0.005354</i>	<i>0.726552</i>	<i>1.264826</i>	<i>0.55813</i>
	B	1	0.886	0.886	0.880	0.999	0.999	0.993
		2	0.864	0.864	0.859	0.999	0.998	0.993
	C	1	0.843	0.842	0.838	0.999	0.998	0.994
		<i>Hg std.</i>	<i>0.007735</i>	<i>0.013497</i>	<i>0.00595</i>	<i>0.726829</i>	<i>1.26825</i>	<i>0.559124</i>
HK2	A	2	0.825	0.825	0.821	0.999	0.999	0.994
		1	0.829	0.830	0.825	0.999	1.000	0.995
	B	2	0.833	0.834	0.829	1.000	1.001	0.995
		<i>Hg std.</i>	<i>0.00759</i>	<i>0.013194</i>	<i>0.005839</i>	<i>0.727214</i>	<i>1.264063</i>	<i>0.55947</i>
	C	1	0.839	0.842	0.836	1.000	1.003	0.997
		2	0.838	0.841	0.835	1.000	1.003	0.997
XX1	A	1	0.837	0.839	0.835	1.000	1.003	0.998
		<i>Hg std.</i>	<i>0.007637</i>	<i>0.013286</i>	<i>0.005867</i>	<i>0.727007</i>	<i>1.264733</i>	<i>0.558483</i>
	B	2	0.834	0.836	0.832	1.000	1.002	0.997
		1	0.833	0.834	0.830	1.000	1.001	0.996
	C	2	0.832	0.832	0.829	0.999	1.000	0.996
		<i>Hg std.</i>	<i>0.007662</i>	<i>0.013364</i>	<i>0.005892</i>	<i>0.727356</i>	<i>1.268739</i>	<i>0.55934</i>
XX2	A	1	0.818	0.818	0.815	0.999	0.999	0.995
		2	0.806	0.807	0.803	1.000	1.000	0.995
	B	1	0.794	0.795	0.790	1.000	1.000	0.995
		<i>Hg std.</i>	<i>0.008135</i>	<i>0.014177</i>	<i>0.006264</i>	<i>0.726427</i>	<i>1.266029</i>	<i>0.559366</i>
	C	2	0.764	0.765	0.761	0.999	1.000	0.994
		1	0.746	0.747	0.743	0.998	0.999	0.994
XX2	A	2	0.728	0.729	0.726	0.997	0.998	0.993
		<i>Hg std.</i>	<i>0.007726</i>	<i>0.013443</i>	<i>0.005942</i>	<i>0.727105</i>	<i>1.265086</i>	<i>0.559147</i>
	B	1	0.818	0.819	0.816	1.002	1.003	0.998
		2	0.781	0.782	0.778	1.001	1.003	0.997
	C	1	0.743	0.745	0.740	1.000	1.002	0.996
		<i>Hg std.</i>	<i>0.007802</i>	<i>0.013552</i>	<i>0.00599</i>	<i>0.728428</i>	<i>1.265264</i>	<i>0.559249</i>
		2	0.713	0.715	0.710	0.999	1.001	0.994

TT1	C	1	0.722	0.722	0.718	0.999	1.000	0.994
		2	0.730	0.730	0.726	0.999	0.999	0.993
		<i>Hg std.</i>	<i>0.00754</i>	<i>0.013134</i>	<i>0.005797</i>	<i>0.726265</i>	<i>1.265067</i>	<i>0.558402</i>
	A	1	0.833	0.835	0.831	1.001	1.002	0.998
		2	0.819	0.819	0.816	1.000	1.000	0.996
	B	1	0.804	0.804	0.801	0.999	0.998	0.994
<i>Hg std.</i>		<i>0.008032</i>	<i>0.014019</i>	<i>0.006174</i>	<i>0.727854</i>	<i>1.27036</i>	<i>0.559462</i>	
TT2	C	1	0.799	0.800	0.796	0.998	0.999	0.995
		2	0.804	0.806	0.802	0.999	1.001	0.996
	<i>Hg std.</i>	<i>0.007456</i>	<i>0.013006</i>	<i>0.005741</i>	<i>0.725003</i>	<i>1.264739</i>	<i>0.558205</i>	
	A	1	0.817	0.818	0.812	0.997	0.999	0.991
		2	0.816	0.817	0.811	0.997	0.999	0.991
	B	1	0.814	0.816	0.810	0.997	0.999	0.992
<i>Hg std.</i>		<i>0.009027</i>	<i>0.015686</i>	<i>0.006937</i>	<i>0.727826</i>	<i>1.264719</i>	<i>0.559305</i>	
VS1	C	1	0.889	0.890	0.887	1.001	1.002	0.999
		2	0.869	0.870	0.867	1.001	1.003	0.999
	<i>Hg std.</i>	<i>0.008595</i>	<i>0.015009</i>	<i>0.006621</i>	<i>0.72658</i>	<i>1.268736</i>	<i>0.559692</i>	
	A	1	0.751	0.750	0.746	0.999	0.998	0.993
		2	0.763	0.762	0.758	0.999	0.999	0.993
	B	1	0.775	0.775	0.770	0.999	0.999	0.993
<i>Hg std.</i>		<i>0.008069</i>	<i>0.01405</i>	<i>0.006207</i>	<i>0.726986</i>	<i>1.265819</i>	<i>0.559237</i>	
C	1	0.671	0.672	0.667	0.995	0.997	0.989	
	2	0.614	0.615	0.610	0.994	0.995	0.987	
	<i>Hg std.</i>	<i>0.011371</i>	<i>0.01976</i>	<i>0.008739</i>	<i>0.731391</i>	<i>1.270962</i>	<i>0.562087</i>	

Table S3. E-values of Hg (mg kg⁻¹), for all collected samples, as calculated by equation 1 using ¹⁹⁹Hg as spike isotope and ²⁰⁰Hg, ²⁰¹Hg or ²⁰²Hg as reference isotopes. A, B and C represent different samples collected from the same location. Standard errors are displayed between brackets (two spiked and two un-spiked replicates).

Location	Sample	E-Value (¹⁹⁹ Hg/ ²⁰⁰ Hg)	E-Value (¹⁹⁹ Hg/ ²⁰¹ Hg)	E-Value (¹⁹⁹ Hg/ ²⁰² Hg)
HW1	A	0.16 (0.00)	0.15 (0.00)	0.16 (0.00)
	B	0.07 (0.02)	0.06 (0.01)	0.06 (0.01)
	C	0.08 (0.01)	0.07 (0.00)	0.08 (0.01)
HK1	A	0.41 (0.02)	0.36 (0.03)	0.42 (0.01)
	B	0.54 (0.06)	0.52 (0.09)	0.55 (0.08)
	C	0.14 (0.00)	0.13 (0.00)	0.14 (0.00)
HK2	A	0.62 (0.05)	0.57 (0.05)	0.61 (0.05)
	B	0.72 (0.03)	0.71 (0.25)	0.72 (0.14)
	C	0.64 (0.03)	0.61 (0.02)	0.65 (0.01)
XX1	A	0.56 (0.03)	0.54 (0.04)	0.59 (0.04)
	B	1.29 (0.02)	1.18 (0.04)	1.29 (0.01)
	C	1.08 (0.05)	1.01 (0.04)	1.09 (0.02)
XX2	A	1.69 (0.04)	1.59 (0.05)	1.69 (0.04)
	B	1.66 (0.23)	1.54 (0.15)	1.67 (0.05)
	C	4.91 (0.25)	4.70 (0.08)	4.95 (0.16)
TT1	A	0.78 (0.17)	0.75 (0.18)	0.78 (0.17)
	B	0.12 (0.01)	0.11 (0.02)	0.12 (0.01)
	C	0.35 (0.03)	0.32 (0.15)	0.36 (0.05)
TT2	A	5.99 (0.13)	5.80 (0.11)	5.99 (0.17)
	B	5.42 (0.17)	5.36 (0.48)	5.46 (0.25)
	C	6.83 (0.04)	6.69 (0.14)	6.85 (0.06)
VS1	A	131(16.8)	131 (16.7)	132 (15.9)
	B	74.9 (0.28)	73.7 (1.09)	75.9 (0.65)
	C	43.7 (2.69)	43.3 (3.01)	44.0 (1.89)

Table S4. Mercury lability (%HgE), for all samples, calculated by equation 1, against different ^{199}Hg spike to natural ratios. A, B and C are different samples from the same location. Standard errors are displayed between brackets for two spiked (for each spike level) and two un-spiked replicates.

Location	Sample	Hg Lability (%HgE)		
		<i>Spike / Native ^{199}Hg</i> 50 %	100 %	200 %
HW1	A	7.82 (0.01)	7.76 (0.14)	7.81 (0.12)
	B	16.5 (3.65)	16.2 (3.45)	16.2 (3.48)
	C	19.8 (2.34)	20.2 (2.23)	20.1 (2.2)
HK1	A	11.7 (0.42)	12 (0.18)	12 (0.18)
	B	15.5 (1.78)	15.7 (2.24)	15.7 (2.24)
	C	10.1 (0.25)	10.2 (0.13)	10.1 (0.16)
HK2	A	22.8 (1.82)	22.5 (2.01)	22.6 (1.92)
	B	24.1 (4.32)	24.1 (4.54)	24.2 (4.5)
	C	25.5 (1.29)	26 (1.17)	26 (1.11)
XX1	A	16.5 (0.88)	16.3 (1.02)	16.4 (1)
	B	14.6 (0.43)	14.7 (0.57)	14.7 (0.57)
	C	10.6 (0.26)	10.7 (0.15)	10.7 (0.13)
XX2	A	14.1 (0.28)	14.1 (0.3)	14.2 (0.29)
	B	13 (0.26)	13.1 (0.42)	13.1 (0.41)
	C	18.9 (0.59)	19 (0.61)	19.1 (0.6)
TT1	A	18 (3.80)	18 (3.89)	17.9 (3.84)
	B	10.7 (1.26)	10.8 (1.23)	10.7 (1.29)
	C	13.5 (1.63)	13.7 (1.73)	13.7 (1.79)
TT2	A	15.5 (0.33)	15.4 (0.43)	15.6 (0.43)
	B	18.6 (0.93)	18.8 (0.85)	18.7 (0.83)
	C	14.8 (0.99)	14.8 (0.09)	14.8 (0.13)
VS1	A	33.6 (4.23)	33.8 (4.07)	33.6 (3.84)
	B	24.2 (0.08)	24.5 (0.21)	24.5 (0.19)
	C	19 (1.15)	19.1 (1.17)	19.1 (1.12)
VS2	A	27.5 (2.16)	27.1 (1.78)	27 (1.85)
	B	46 (8.31)	45.9 (8.07)	45.8 (8.15)
	C	15.3 (0.64)	15.1 (0.61)	15.1 (0.64)

Table S5. Pearson correlation coefficients of Hg labile pool (HgE; mg kg⁻¹) and Hg lability (%HgE) with soil parameters in all soils (n=27).

Soil Parameter	Pearson Correlation (r) with HgE (mg kg ⁻¹)	Pearson Correlation (r) with %HgE
THg	0.98	0.63
pH	-0.07	0.14
N	-0.13	-0.26
Org-C	0.11	-0.03
S	0.11	-0.04
Al(OH) ₃	0.47	0.08
MnO ₂	-0.06	-0.17
Fe ₂ O ₃	-0.03	0.00

Table S6. Linear stepwise regression (Minitab 17) coefficients and P-values. Labile Hg (HgE; mg kg⁻¹) and Hg lability (%HgE) parameterised by all other soil parameters including THg, pH, N, Org-C, S and Al, Mn and Fe oxides.

Regression	HgE (mg kg ⁻¹)		%HgE	
	R ² = 0.96		R ² = 0.63	
	Coefficients	P-value	Coefficients	P-value
Intercept	-4.98	0.90	-4.70	0.83
THg	0.27	0.00	0.05	0.01
pH	-1.92	0.73	2.70	0.39
N	-46.2	0.47	1.85	0.96
Org-C	7.57	0.32	-0.52	0.90
S	0.00	0.85	0.00	0.45
Al(OH) ₃	2.89	0.88	-20.5	0.08
MnO ₂	-1.44	0.98	-34.4	0.20
Fe ₂ O ₃	1.58	0.77	5.85	0.07

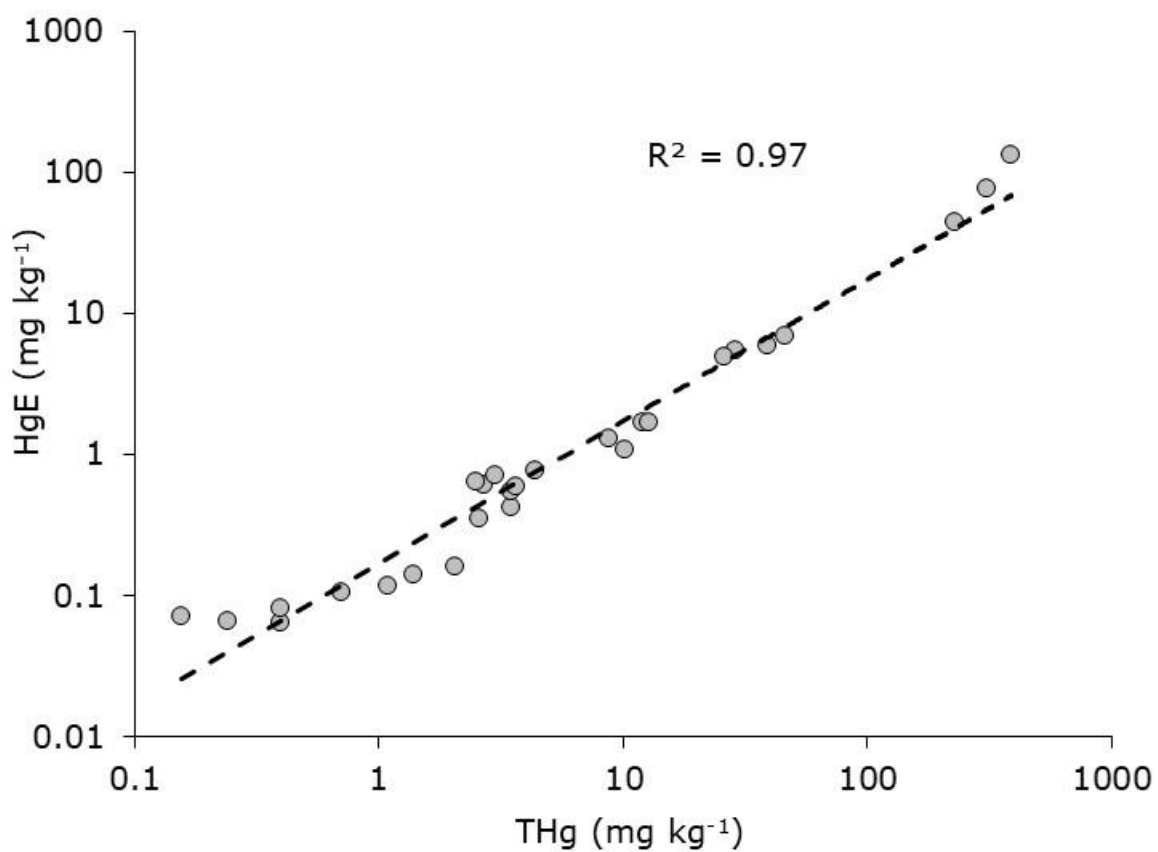


Figure S1. Total soil mercury (THg) (mg kg⁻¹) plotted against corresponding labile Hg (HgE; mg kg⁻¹) for all data points (n=27). The dashed line represents a 'power' relationship between x and y parameters.

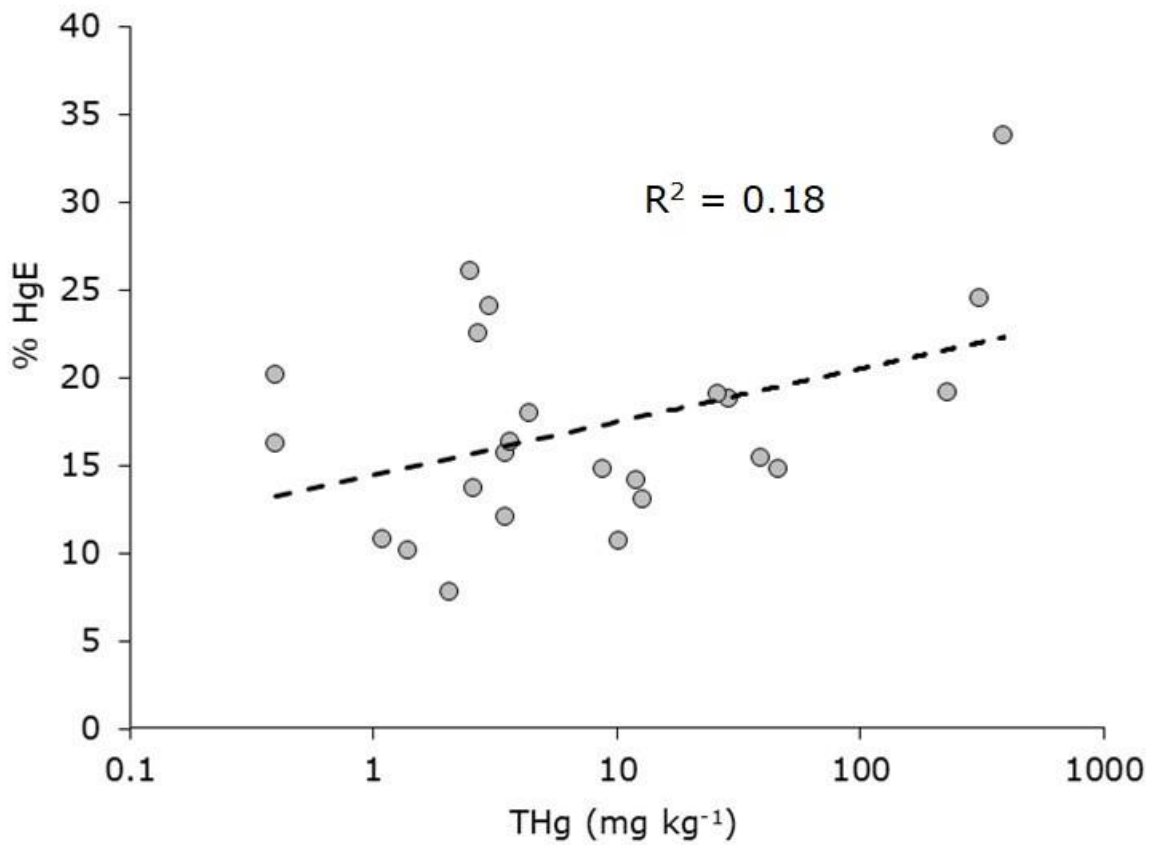


Figure S2. Total soil mercury (THg) (mg kg⁻¹) plotted against Hg lability (%HgE) for all data points (n=27). The dashed line represents a 'logarithmic' relationship between x and y parameters.

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