

Supporting Information
for
Quantifying soil accumulation of atmospheric mercury using fallout radionuclide chronometry

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Supporting Information consists of three pages, Figures S1-S3, and Table S1.

S1. **Identifying robust soil Hg chronologies**. Soils are notoriously complex, with pedogenic processes redistributing mass vertically and horizontally, through both physical and chemical processes, operating variously at different depths in different horizons, and possibly changing over decadal to centennial timescales with climate, vegetation, and land use disturbances. To every extent possible we sampled putative 'reference' soils at sites of known land-use histories, undisturbed for >150 years, on flat ground, near local topographic high points or ridgetops to reduce the influence of horizontal flowpaths, and over relatively large areas (30 x 30 cm) to reduce localized heterogeneities. The gold standard of ensuring that a soil site may be trusted to produce an honest record of atmospheric deposition is the concordance of multiple independent chronometers. The LRC age model typically dates the ^{241}Am bomb-pulse to its expected peak of 1963-4 with errors <5 years or +10%. Here our global compilation has geometric mean = 1965.8 +1.9 SE, $k=1$, $n=18$; Table S1). When this is the case, total soil ^{210}Pb and ^{241}Am inventories are tightly correlated based on their common atmospheric source ($R^2=0.88$, $n=19$; Fig. S1). When this is not the case, the ^{210}Pb and ^{241}Am have likely been redistributed differentially with respect to each other by some of the disturbance problems listed above, and due to the different timeframes over which they are deposited; ^{210}Pb is deposited continuously and forms an exponential depth profile, whereas ^{241}Am was deposited in a pulse and appears as a sharp peak in the soil subsurface at depths ranging 5-10 cm.

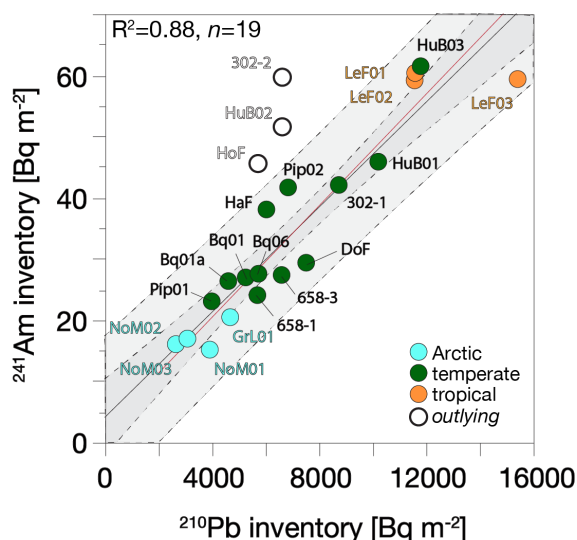


Figure S1: Linear correlation of ^{241}Am and ^{210}Pb soil inventories for Arctic, temperate/boreal and tropical sites. Two-sigma confidence intervals for fit and prediction are shaded. Typical $^{210}\text{Pb} : ^{241}\text{Am} = 233 \pm 9$ (1-sigma SD). Points colored white are considered outlying and possibly impacted by disturbance.

Soils that do not produce accurate ^{241}Am dates, fall outside the Pb-Am correlation confidence intervals (Fig. S1), or have total Hg inventories significantly different than ecosystem means (Table S1), are omitted from analysis of long-term Hg chronologies. For example, in reconstructing Hg accumulation for northeastern soils, we omitted 3 pits likely to have been disturbed (302-2, 658-1, and HoF01), and two that show atypical Hg low-accumulation as described in the main text (658-3 and DoF01). Historical reconstruction with sites omitted in the main text are shown in the alternative Fig. S2 below.

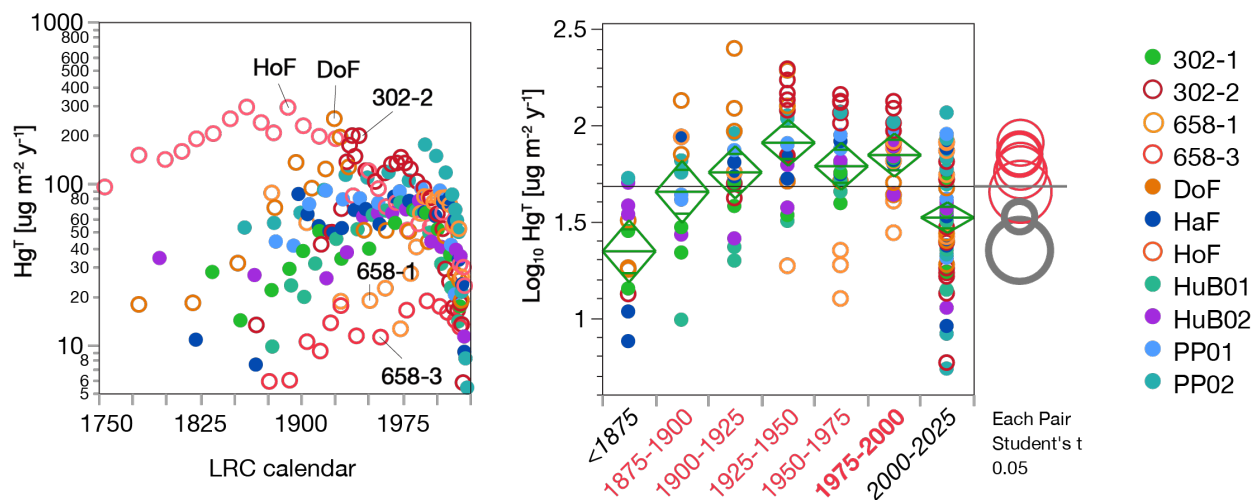


Figure S2. Reconstructed rates of soil Hg accumulation, including outlying sites with evidence of disturbance (HoF, 302-2) or Hg remobilization (658-1, 658-3).

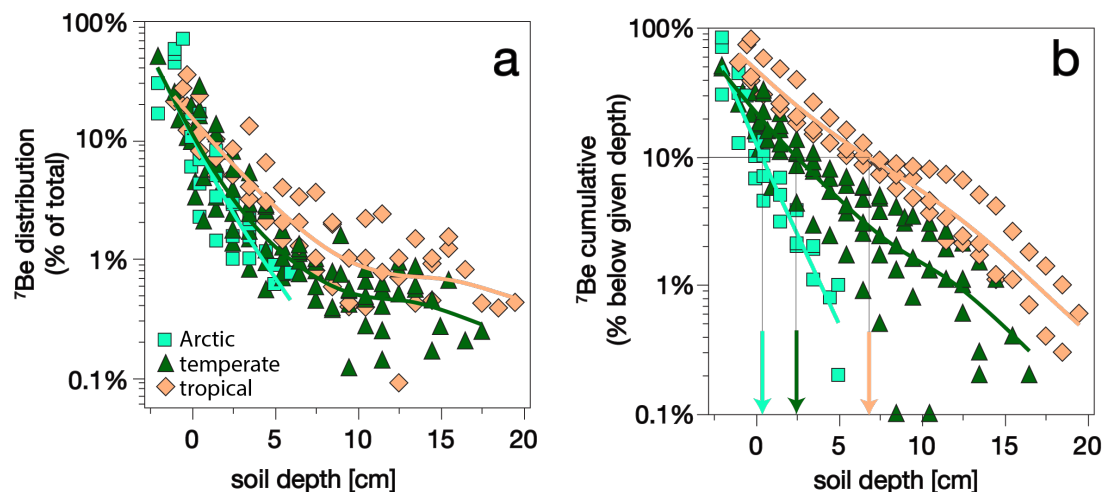


Figure S3: (a) mass-depth distributions of ^7Be (half-life 54 days) in global soils. Lines show spline best fits. (b) cumulative mass distributions for ^7Be below a given depth. In Arctic soils 10% of ^7Be is found below soil depths of 0.5 cm, and corresponding distributions in temperate and tropical soils are 2.5 cm and 7 cm, respectively.

Table S1: site locations and characteristics for reconstruction of Hg accumulation in soils by FRN chronometry

site name	site code	pit code ^a	canopy species	biome	ecotype	elevation m a.s.l.	lat N	long W	MAT C	MAP cm	soil order	parent lithology	experimental condition	Hg ^T mg m ⁻²	²¹⁰ Hg ^T _{atm} mg m ⁻²	²¹⁰ Pb _{soil} Bq m ⁻²	²⁴¹ Am Bq m ⁻²	bomb year**	¹³⁷ Cs Bq m ⁻²	²¹⁰ Pb/Am R	²¹⁰ Pb/Hg R			
Beaver Meadow	Sharon	VT	BeM	FLH	<i>Pinus strobus</i>	temperate	coniferous	464	43.80	72.40	6.8	108	inceptisol	schist, phyllite										
				FLH	<i>Quercus rubra</i>	temperate	deciduous	464	43.80	72.40			inceptisol	schist, phyllite										
Camel's Hump State Forest	Duxbury	VT	CaH	FLH	<i>Sphagnum</i>	temperate	moss	1067	44.32	72.88	2.4	203	spodosol	schist, phyllite										
Downer State Forest	Stratford	VT	DoF	FLH	<i>Pinus resinosa</i>	temperate	coniferous	460	43.80	72.39	6.8	108	inceptisol	schist, phyllite										
				FLH	<i>Tsuga canadensis</i>	temperate	coniferous	460	43.80	72.39			inceptisol	schist, phyllite										
				DoF01	<i>Pinus strobus</i>	temperate	coniferous						inceptisol	schist, phyllite	reference	13.9	6.1	7529	29.3	1963	1689	257	1232	
Experimental Lakes Area	Kenora	ON	ELA302	302-1	<i>Picea mariana</i>	boreal	coniferous	460	49.68	93.77	3.5	80	inceptisol	gneiss	reference	9.0	8.7	8748	42.0	1966	1484	209	1004	
				302-2	<i>Picea mariana</i>	boreal	coniferous						inceptisol	gneiss	reference	10.0	7.6	6638	59.6	1967	1217	111	872	
				658-1	<i>Picea mariana</i>	boreal	coniferous						inceptisol	gneiss	disturbed, fire	3.9	3.2	5707	24.0	n/a	842	237	1763	
				658-3	<i>Picea mariana</i>	boreal	coniferous						inceptisol	gneiss	reference	1.9	1.6	6611	27.3	1966	1376	243	4161	
Harvard Experimental Forest	Petersham	MA	HaF	FLH	<i>Quercus rubra</i>	temperate	deciduous	340	42.32	72.11	8.8	122	inceptisol	glacial till										
				HaF	<i>Quercus rubra</i>	temperate	deciduous						forest regrowth	8.8	5.8	6044	38.0	1964	1139					
Howland Experimental Forest	Howland	ME	HoF	FLH	<i>Tsuga canadensis</i>	temperate	coniferous	60	45.20	68.70	6.2	115	spodosol	granitic till										
				HoF	<i>Tsuga canadensis</i>	temperate	coniferous						woody debris	34.1	31.0	5730	45.5	1947	1499	>	126	185		
Hubbard Brook Experimental Forest	N. Woodstock	NH	HuB	HuB01	<i>Betula alleghaniensis</i>	temperate	deciduous	584	43.96	71.72	6.7	150	spodosol	granitic till	hillslope	5.9	5.8	10223	45.8	1967	841	>	223	1748
				HuB02	<i>Betula alleghaniensis</i>	temperate	deciduous	410					spodosol	granitic till	hillslope	8.7	8.7	6636	51.5	1951	908	>	129	767
				HuB03	<i>Picea rubens</i>	temperate	coniferous	710					spodosol	granitic till	hillslope			11807	61.4	n/a	4263	192		
Berlin chloralkali	Berlin	NH	BqP	BqP1	<i>Betula</i>	temperate	deciduous						spodosol	alluvium	Hg pointsource	n/a		5270	*	26.9	n/a	1700	196	
				BqP1a	<i>Tsuga canadensis</i>	temperate	coniferous	298	44.45	71.18		107	spodosol	alluvium	Hg pointsource	>28	>28	4619	*	26.3	n/a	1101	>	176
				BqP6	<i>Tsuga canadensis</i>	temperate	coniferous						spodosol	alluvium	Hg pointsource	n/a		5747	27.5	1962	1301	>	209	
Mink Brook watershed	Etna	NH	MiB	FLH	<i>Acer saccharum</i>	temperate	deciduous	435	43.68	72.18		110	inceptisol	amphibolite										
Shattuck Observatory	Hanover	NH	ShO	FLH	<i>Quercus rubra</i>	temperate	deciduous	165	43.70	72.29	6.9	109	spodosol	amphibolite										
				PIP01	<i>Pinus strobus</i>	temperate	coniferous						alluvial till	reference	10.5	8.1	3999	23.0	1966	896	>	174	492	
				PIP02	<i>Pinus strobus</i>	temperate	coniferous						alluvial till	forest cut	19.2	13.8	6854	41.6	1957	1752	165	495		
Woody Adams Conservation	Norwich	VT	WoA	FLH	<i>Quercus rubra</i>	temperate	deciduous	425	43.77	72.35	6.8	111	inceptisol	schist, phyllite										
				FLH	<i>Acer saccharum</i>	temperate	deciduous	425	43.77	72.35			inceptisol	schist, phyllite										
				FLH	<i>Pinus strobus</i>	temperate	coniferous	425	43.77	72.35			inceptisol	schist, phyllite										
				FLH	<i>Tsuga canadensis</i>	temperate	coniferous	425	43.77	72.35			inceptisol	schist, phyllite										
Seward Peninsula	Nome	AK	NoM	NoM01	<i>Sphagnum</i>	Arctic	moss	120	64.98	166.21	-6.0	43	gellisol	phyllite	hillslope	2.8	2.5	3928	15.1	1965	925			
				NoM02	<i>Sphagnum</i>	Arctic	moss						gellisol	phyllite	reference	3.3	2.5	3092	16.9	1962	748			
				NoM03	<i>Sphagnum</i>	Arctic	moss						gellisol	phyllite	hillslope	2.3	2.0	2660	16.0	1967	769			
Sisimuit	Sisimuit	GR	GRL	GRL1	<i>Sphagnum</i>	Arctic	moss						inceptisol	gneiss		2.7	2.5	4690	20.4	1967	611			
Luquillo Experimental Forest	Luquillo	PR	LeF	LeF01	<i>Dacryodes</i>	tropical	evergreen	440	18.32	65.82	24.0	260	ultisol	volcaniclastic	reference	30.9	13.4	11601	60.3	1978	2910			
				LeF02	<i>Cyrtilla</i>	tropical	evergreen	773	18.28	65.79	22.0	360	inceptisol	diorite	reference	21.5	11.3	11580	59.1	1982	1910			
				LeF03	<i>Eugenia</i>	tropical	evergreen	1078	18.27	65.76	20.0	420	inceptisol, hydric	diorite	reference	13.6	8.6	15432	59.3	1962	2058			

**peak loading or cumulative deposition interpolated to 48.3% in 1963 (Appleby 1991)

^aFLH denotes collection of foliage, litter and humus only; named pits were quantitative to 50 cm or refusal^batmospheric Hg is distinguished from soil total Hg using Al as an index element

*not at steady state

>value is underestimated due to deep percolation

red text highlights outlying values

temperate median	9.0	7.6	6637	42	1965	1297	192	938
SD	8.8	7.9	2131	13	7	931	51	1140
CV	98%	104%	32%	31%	12.5%	72%	26%	122%
count					11.0		13.0	10.0
SE					3.8%		7.3%	38.4%

global median 1965.8

SD 8.0

CV 14.0%

SE 1.9

count 18.0