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Insights from mercury stable isotopes on terrestrial–atmosphere exchange of Hg(0) in the Arctic tundra

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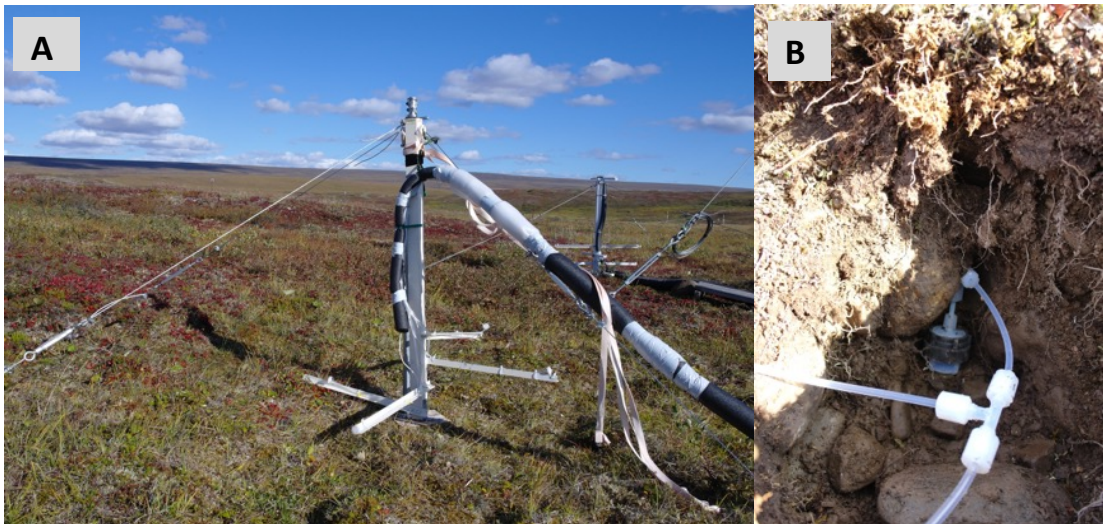

Figure S1: Setup of the sampling inlets for Hg(0) isotope measurements in interstitial snow (A) and soil pore (B) air. Snowpack inlets at 0 cm and 10 cm height were covered by snow as the wintertime snowpack developed allowing for undisturbed interstitial snow air sampling. For soil pore air sampling, soil pits were backfilled with excavated soil after placement of the inlet filters to reconstruct the original soil environment as best as possible.

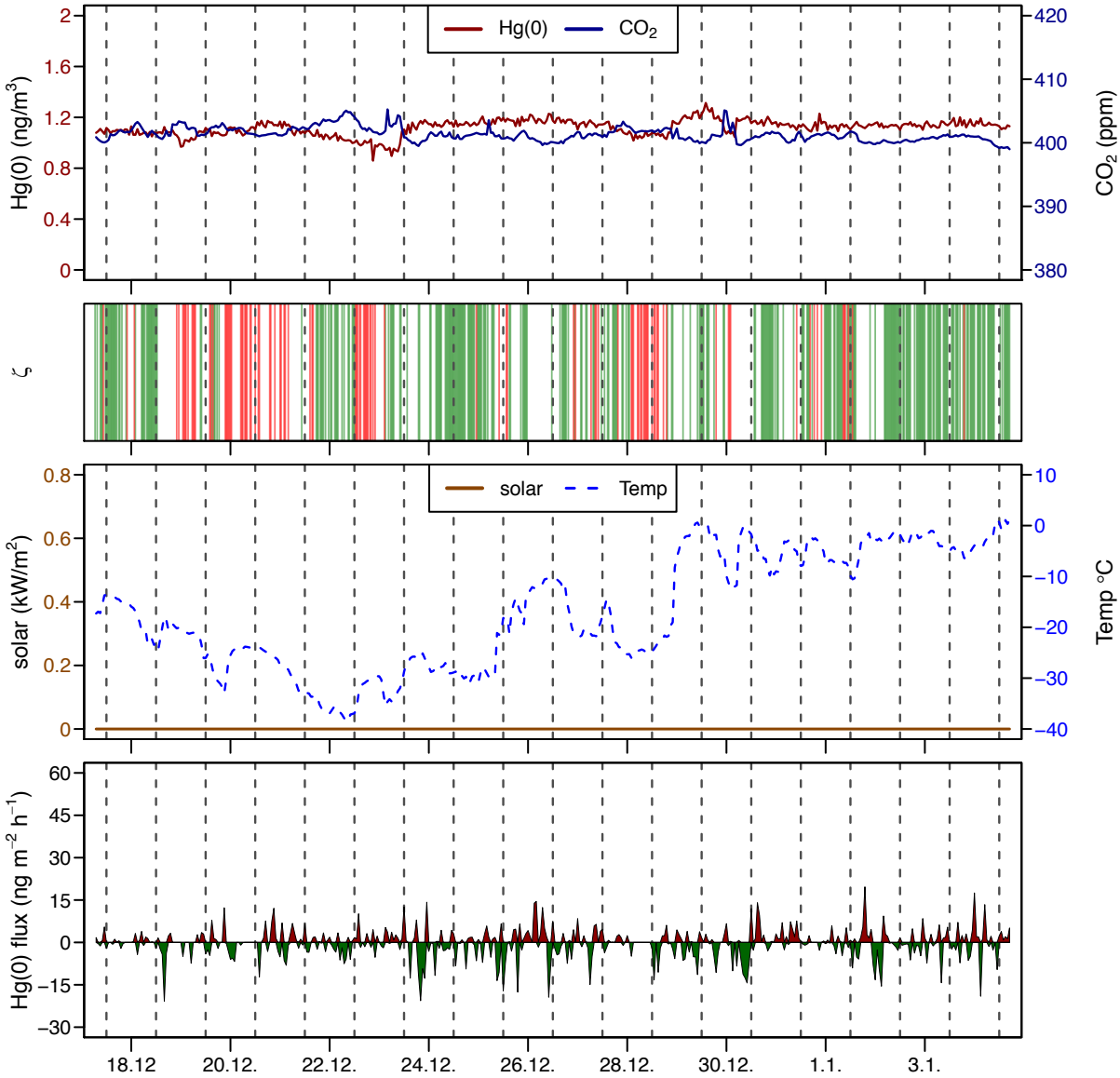

Figure S2: Time series during a winter period (18.12.2015 – 05.01.2016), A: atmospheric Hg(0) concentration and CO_2 mixing ratio, B: median planetary boundary layer stability (ζ), where the shaded areas in green represents stable conditions ($\zeta > 0.1$) and shaded areas in red represent turbulent conditions ($\zeta < -0.1$), C: solar radiation and air temperature, and D: Hg(0) flux, where Hg(0) deposition is in green and net Hg(0) re-emission is in red. Midnight is indicated by dashed lines.

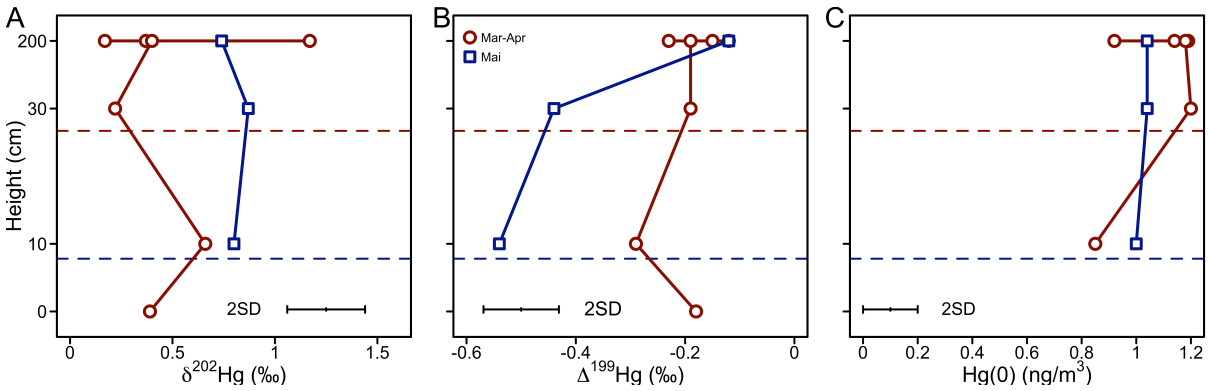

Figure S3: Hg(0) measurements in interstitial snow air profiles **A:** mass dependent fractionation stable isotope signature of Hg(0) (δ^{202}Hg), **B:** mass-independent fractionation stable isotope signature of Hg(0) (Δ^{199}Hg), and **C:** mean Hg(0) concentration. The dashed horizontal lines represent the average snow height during the respective period.

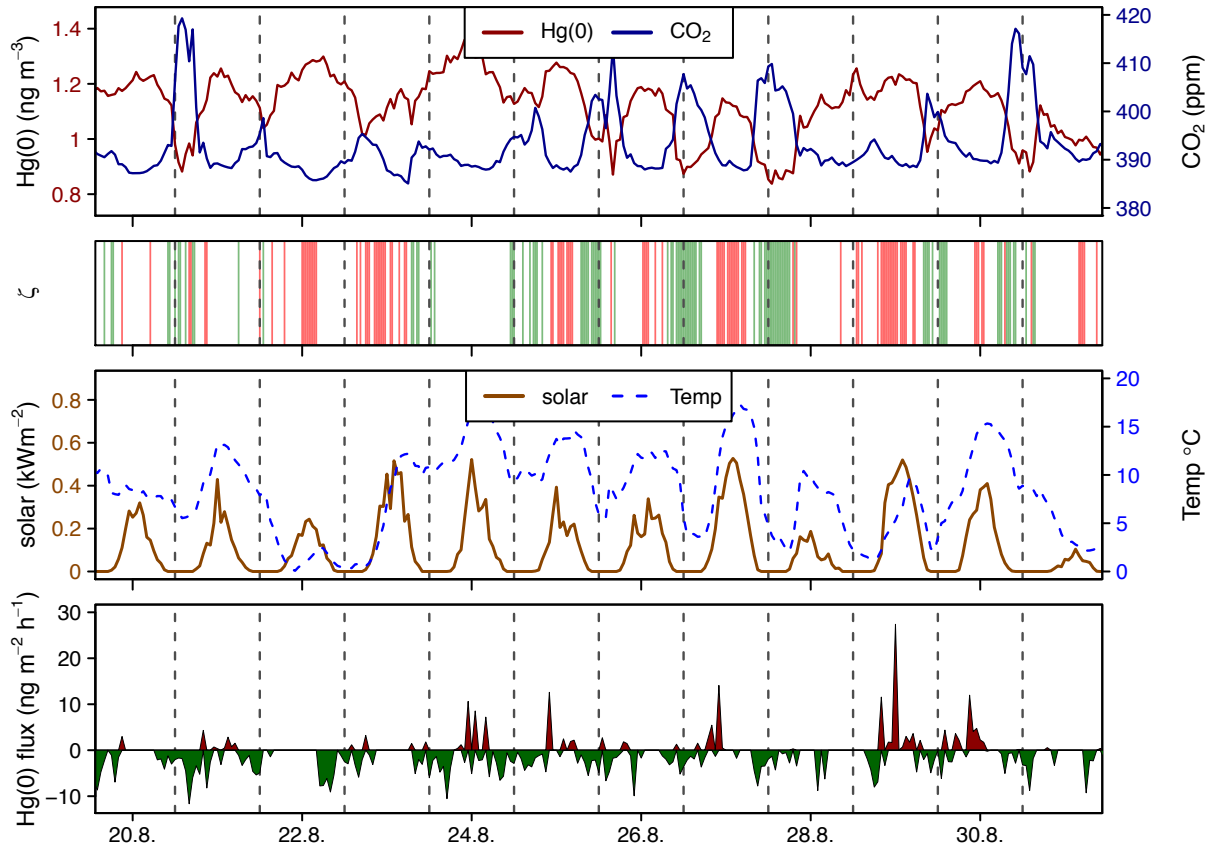

Figure S4: Time series during later summer (20.8.2016-31.8.2016), A: atmospheric Hg(0) concentration and CO_2 mixing ratio, B: median planetary boundary layer stability (ζ), where the shaded areas in green represents stable conditions ($\zeta > 0.1$) and shaded areas in red represent turbulent conditions ($\zeta < -0.1$), C: solar radiation and temperature, and D: Hg(0) flux, where Hg(0) deposition is in green and Hg(0) re-emission is in red. Midnight is indicated by dashed lines.

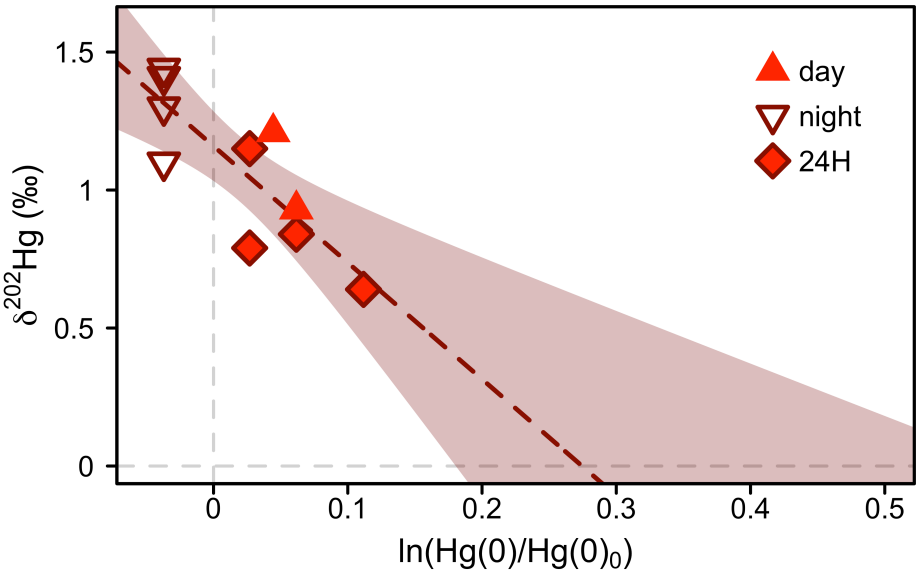

Figure S5: Mass dependent Hg isotope signature (δ^{202}Hg) of atmospheric Hg(0) versus logarithm of Hg(0) concentration (normalized to the initial concentration $Hg(0)_0$) during the snow-free vegetation period (11.6.2016 – 10.9.2016) for the determination of the Hg isotope enrichment factor ε^{202}Hg. The best fit linear regression for δ^{202}Hg using the *lm* function of R = −4.22 ± 1.01 ‰ ln $(Hg(0)/Hg(0)_0)$ + 1.15 ± 0.05‰ (mean ± 1se, R^2= 0.68, p=0.003).

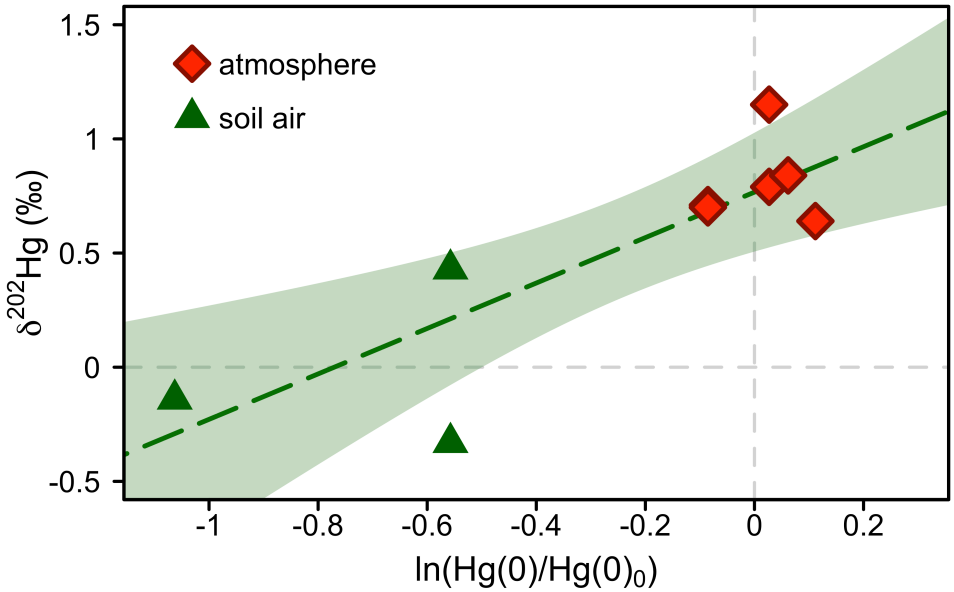

Figure S6: Mass dependent Hg isotope signature (δ^{202}Hg) of atmospheric and soil pore air Hg(0) versus logarithm of Hg(0) concentration (normalized to the initial concentration $Hg(0)_0$) for the determination of the Hg isotope enrichment factor ε^{202}Hg. The best fit linear regression for δ^{202}Hg using the *lm* function of R = 1.00 ± 0.25‰ ln $(Hg(0)/Hg(0)_0)$ + 0.72 ± 0.08‰) (mean ± 1se, R^2 = 0.70, p = 0.005).

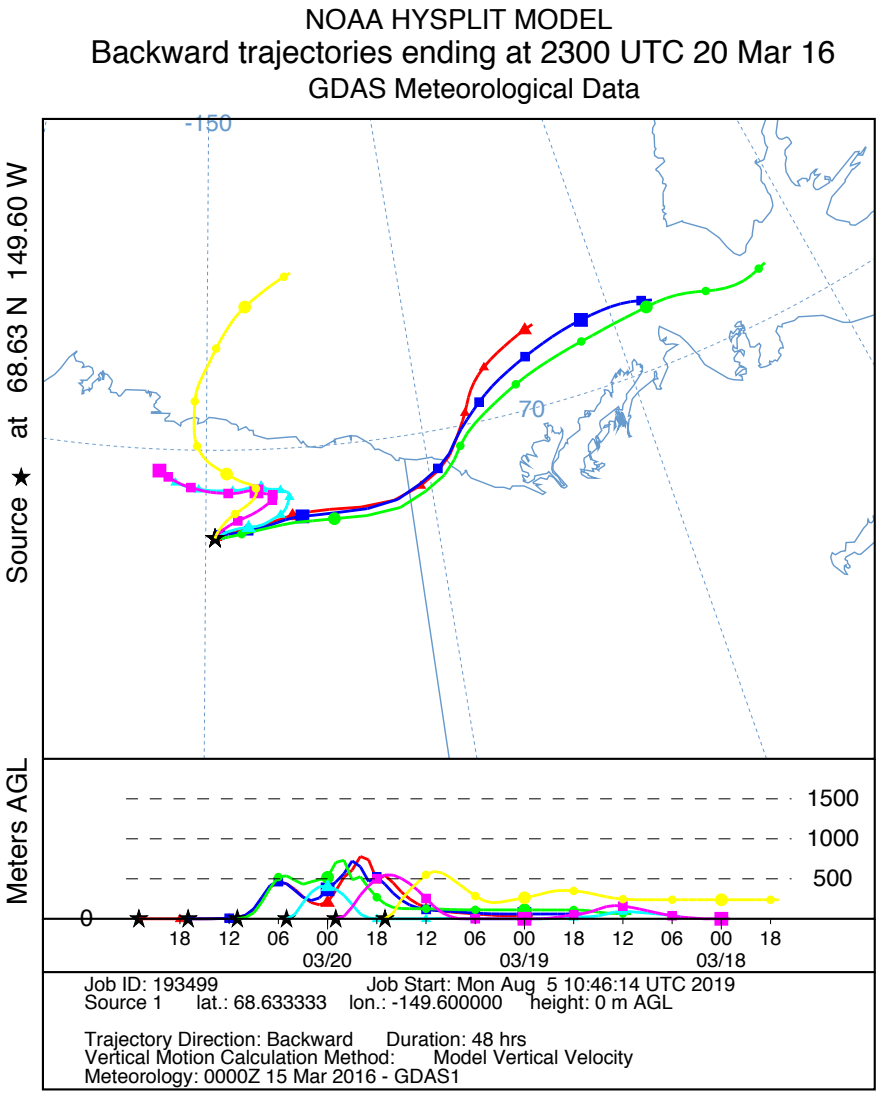

Figure S7: 48 h backward trajectory analysis using the NOAA HYSPLIT model of AMDE Event 1 from 19.3. to 20.2. 2016.

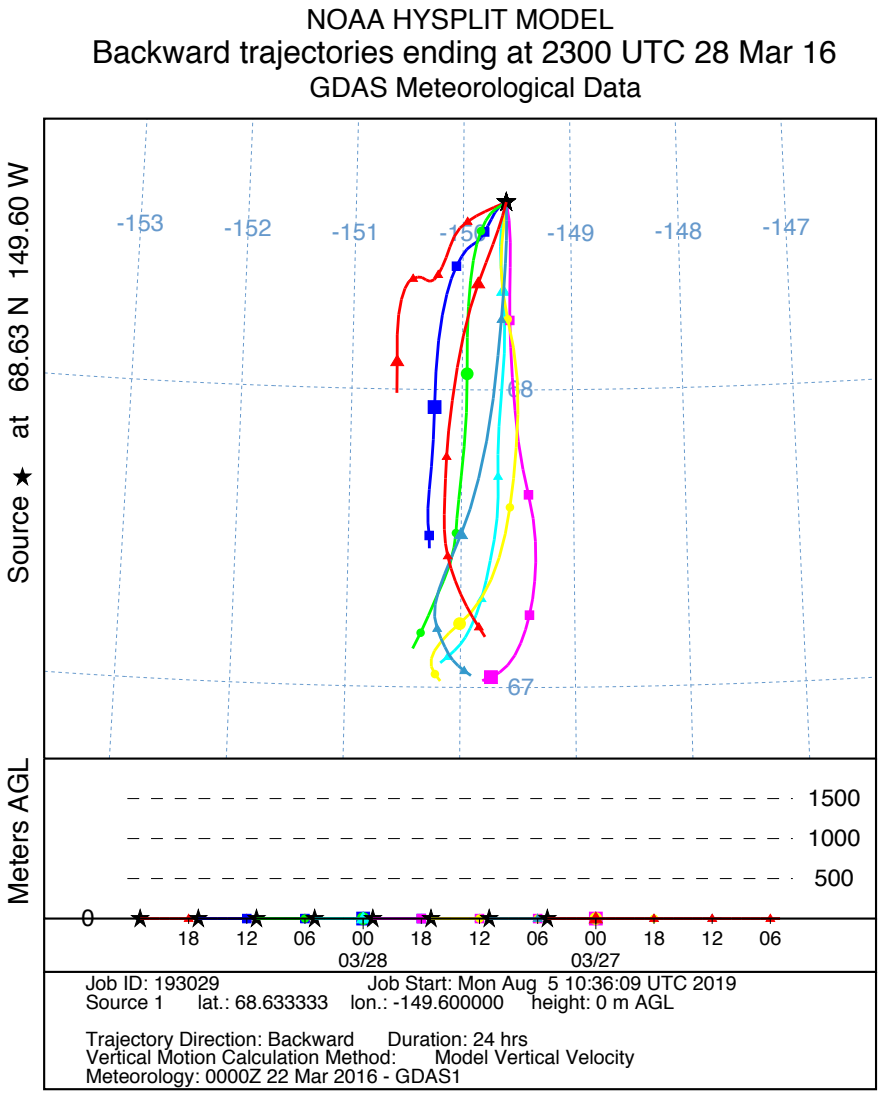

Figure S8: 24 h backward trajectory analysis using the NOAA HYSPLIT model of Event 2 from 27.3. to 28.2. 2016.

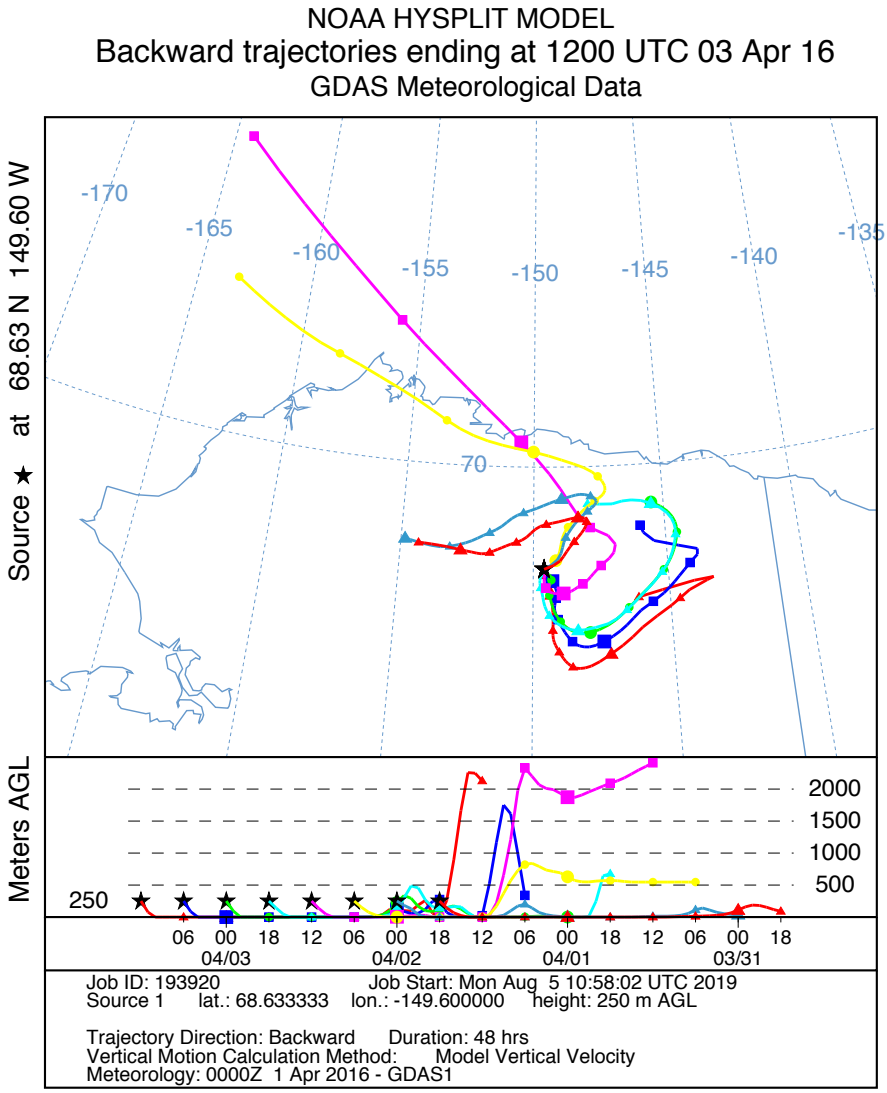

Figure S9: 24 h backward trajectory analysis using the NOAA HYSPLIT model of AMDE Event 3 from 2.4. to 3.4. 2016.

Table S1: Details on atmospheric Hg(0) isotope samples: sampling height, period of day, start date and end date, sampling time, volume sample, average flow, atmospheric Hg(0), Hg recovered from trap (ng) and sampling yield. nm = not measured. Samples marked with * are from Obrist et al. (2017).

name	Height (cm)	time	Start date	End date	time (h)	volume (m^3)	flow (lpm)	GEM (ng m^{-3})	Hg (ng)	yield (%)
GEM_P1-E	-40	24H	17.08.15 20:30	14.10.15 11:00	1383	13.2	0.16	0.38	10.1	201.1
GEM_P1-A	0	24H	17.08.15 20:30	14.10.15 11:00	1383	15.0	0.18	0.49	10.0	136.8
GEM_P1-C *	30	24H	17.08.15 20:30	14.10.15 11:00	1383	16.4	0.20	1.01	22.3	135.1
GEM_P1-D *	200	24H	17.08.15 20:30	14.10.15 11:00	1383	15.7	0.19	1.01	13.1	82.2
GEM_P2-B	10	24H	20.10.15 12:40	04.12.15 10:40	1078	12.6	0.20	0.89	14.8	131.1
GEM_P2-D *	200	24H	20.10.15 12:40	04.12.15 10:40	1078	13.0	0.20	1.06	nm	
GEM_P3-A	0	24H	08.12.15 11:00	27.01.16 08:40	1198	14.4	0.20	0.44	9.6	153.0
GEM_P3-B	10	24H	08.12.15 11:00	27.01.16 08:40	1198	13.3	0.18	0.91	19.2	159.9
GEM_P3-C *	30	24H	08.12.15 11:00	27.01.16 08:40	1198	14.5	0.20	1.10	18.3	114.5
GEM_P3-D	200	24H	08.12.15 11:00	27.01.16 08:40	1198	14.2	0.20	1.10	12.2	77.8
GEM_P4-A	0	24H	02.02.16 12:00	23.03.16 11:00	1199	11.0	0.15	0.73	8.0	
GEM_P4-C *	30	24H	02.02.16 12:00	23.03.16 11:00	1199	11.2	0.16	1.09	13.7	112.2
GEM_P4-D *	200	24H	02.02.16 12:00	23.03.16 11:00	1199	12.3	0.17	1.09	15.9	117.9
GEM_P4b-D	200	24H	23.03.16 16:45	27.03.16 18:30	98	9.3	1.58	1.14	11.7	110.4
GEM_X4	200	24H	27.03.16 18:30	31.03.16 16:20	94	8.8	1.56	1.19	8.2	78.9
GEM_X5-D	200	24H	31.03.16 16:30	05.04.16 08:00	112	10.1	1.51	0.92	11.0	118.7
GEM_P5-A	0	24H	05.04.16 17:00	29.04.16 12:50	572	7.1	0.21		nm	
GEM_P5-B	10	24H	05.04.16 17:00	29.04.16 12:50	572	7.2	0.21	0.85	6.1	99.3
GEM_P5-C	30	24H	05.04.16 17:00	29.04.16 12:50	572	7.2	0.21	1.20	8.9	103.2
GEM_P5-D	200	24H	05.04.16 17:00	29.04.16 12:50	572	7.0	0.21	1.18	nm	
GEM_P6-A	0	24H	03.05.16 17:30	25.05.16 06:35	517	6.4	0.21			
GEM_P6-B	10	24H	03.05.16 17:30	25.05.16 06:35	517	6.4	0.21	1.00	7.0	108.2
GEM_P6-C	30	24H	03.05.16 17:30	25.05.16 06:35	517	6.0	0.19	1.04	7.9	126.5
GEM_P6-D	200	24H	03.05.16 17:30	25.05.16 06:35	517	6.3	0.20	1.04	13.6	206.4
GEM_P7-D	200	24H	11.06.16 11:00	13.07.16 10:30	767	10.7	0.23	1.13	10.8	89.3
GEM_P7-A	200	night	11.06.16 11:00	13.07.16 10:30	767	9.5	0.21	1.06	13.1	130.4
GEM_P7-B	200	night	11.06.16 11:00	13.07.16 10:30	767	8.6	0.19	1.06	7.6	82.9
GEM_P7-C	200	day	11.06.16 11:00	13.07.16 10:30	767	11.7	0.25	1.17	14.6	106.6
GEM_J1	200	24H	24.06.16 17:30	26.06.16 19:15	50	6.1	2.03	1.23	8.9	119.7
GEM_J2	200	24H	26.06.16 19:15	28.06.16 21:30	50	5.9	1.94	1.17	7.0	102.7
GEM_P8-D	200	24H	13.07.16 11:15	10.09.16 10:30	1415	17.7	0.21	1.13	16.1	81.2
GEM_P8-A	200	night	13.07.16 11:15	10.09.16 10:30	1415	17.5	0.21	1.06	21.7	117.1
GEM_P8-B	200	night	13.07.16 11:15	10.09.16 10:30	1415	16.0	0.19	1.06	27.7	164.1
GEM_P8-C	200	day	13.07.16 11:15	10.09.16 10:30	1415	11.5	0.14	1.15	9.8	74.6
GEM_P8-E	-40	24H	18.08.16 11:00	09.09.16 08:30	525	7.2	0.23	0.63	3.3	72.4
GEM_P8-F	-40	24H	18.08.16 11:00	09.09.16 08:30	525	5.7	0.18	0.63	2.4	67.9

Table S2: Hg stable isotope values of Hg(0) samples measured in the atmosphere and in interstitial snow and soil air. n refers to the number of re-analysis of the same sample. Samples marked with * are from (2017).

name	n	δ^{204}Hg (‰)	δ^{202}Hg (‰)	δ^{201}Hg (‰)	δ^{200}Hg (‰)	δ^{199}Hg (‰)	Δ^{204}Hg (‰)	Δ^{201}Hg (‰)	Δ^{200}Hg (‰)	Δ^{199}Hg (‰)
GEM_P1-E	1	0.06	-0.14	-0.20	-0.12	-0.15	0.26	-0.10	-0.06	-0.11
GEM_P1-A	2	2.00	1.34	0.71	0.55	-0.01	0.00	-0.30	-0.12	-0.35
GEM_P1-C *	3	1.09	0.71	0.31	0.30	-0.05	0.03	-0.22	-0.05	-0.23
GEM_P1-D *	2	1.01	0.70	0.32	0.32	-0.01	-0.04	-0.21	-0.04	-0.19
GEM_P2-B	5	1.42	0.89	0.42	0.39	-0.14	0.09	-0.25	-0.06	-0.36
GEM_P2-D *	1	0.96	0.55	0.20	0.20	-0.10	0.15	-0.21	-0.08	-0.23
GEM_P3-A	3	1.80	1.15	0.61	0.52	0.02	0.10	-0.25	-0.05	-0.27
GEM_P3-B	5	1.45	0.85	0.35	0.35	-0.09	0.12	-0.29	-0.07	-0.31
GEM_P3-C *	2	1.13	0.64	0.26	0.22	-0.13	0.18	-0.22	-0.11	-0.29
GEM_P3-D	3	1.45	0.95	0.54	0.44	0.14	0.03	-0.17	-0.03	-0.09
GEM_P4-A	2	1.75	1.15	0.68	0.60	0.05	0.03	-0.19	0.02	-0.24
GEM_P4-C *	3	1.30	0.78	0.41	0.31	-0.07	0.13	-0.18	-0.08	-0.27
GEM_P4-D *	2	1.43	0.88	0.55	0.36	-0.03	0.13	-0.11	-0.08	-0.26
GEM_P4b-D	3	0.29	0.17	-0.04	0.00	-0.18	0.03	-0.17	-0.09	-0.23
GEM_X4	3	1.53	1.05	0.62	0.53	0.15	-0.04	-0.17	0.00	-0.11
GEM_X5-D	3	0.68	0.37	0.17	0.13	-0.03	0.12	-0.11	-0.06	-0.12
GEM_P5-A	1	1.14	0.39	0.24	0.23	-0.09	0.55	-0.05	0.03	-0.18
GEM_P5-B	2	1.23	0.66	0.33	0.26	-0.12	0.24	-0.17	-0.07	-0.29
GEM_P5-C	3	0.61	0.29	0.07	0.12	-0.14	0.18	-0.15	-0.02	-0.21
GEM_P5-D	3	0.71	0.40	0.16	0.12	-0.09	0.12	-0.14	-0.08	-0.19
GEM_P6-A	3	1.99	1.00	0.62	0.44	-0.02	0.33	-0.13	-0.06	-0.27
GEM_P6-B	3	1.02	0.61	0.06	0.22	-0.47	0.11	-0.39	-0.09	-0.62
GEM_P6-C	1	1.03	0.87	0.24	0.38	-0.22	-0.26	-0.41	-0.05	-0.44
GEM_P6-D	3	1.22	0.74	0.43	0.34	0.06	0.11	-0.13	-0.03	-0.12
GEM_P7-D	4	1.38	0.79	0.41	0.31	-0.03	0.20	-0.18	-0.09	-0.23
GEM_P7-A	4	1.91	1.10	0.66	0.40	-0.04	0.27	-0.16	-0.15	-0.32
GEM_P7-B	1	2.46	1.44	0.99	0.65	0.20	0.31	-0.09	-0.07	-0.16
GEM_P7-C	4	1.33	0.93	0.53	0.42	0.05	-0.07	-0.17	-0.05	-0.18
GEM_J1	2	1.08	0.64	0.26	0.24	-0.08	0.12	-0.23	-0.08	-0.24
GEM_J2	2	1.55	0.84	0.53	0.32	-0.03	0.30	-0.10	-0.10	-0.24
GEM_P8-D	4	1.80	1.15	0.62	0.49	-0.05	0.08	-0.25	-0.09	-0.34
GEM_P8-A	4	2.33	1.41	0.78	0.64	0.04	0.23	-0.28	-0.07	-0.31
GEM_P8-B	3	2.08	1.30	0.75	0.58	0.01	0.14	-0.22	-0.07	-0.32
GEM_P8-C	3	1.91	1.21	0.62	0.48	0.01	0.11	-0.28	-0.13	-0.30
GEM_P8-E	2	-0.09	-0.33	-0.41	-0.10	-0.33	0.40	-0.17	0.06	-0.25
GEM_P8-F	2	0.47	0.43	-0.08	0.05	-0.08	-0.17	-0.41	-0.16	-0.19