

Supplementary Table 1. Rainwater sampling location, date and time of collection, measured surface air temperature (T), noble gas concentrations and measured He isotopic ratios (R) normalized to atmospheric He ratio (Ra). Samples from Milan and Ann Arbor were collected at altitudes of 215m and 290m a.s.l respectively. Surface weather station records used to derive appropriate precipitation characteristics for corresponding rainfall event is also indicated

Sample	Location	Date	Time	T	He	Ne	Ar	Kr	Xe	R/R _a ^b +/- 1σ	Surface weather station
			(UTC ^a)	(°C)	(x 10 ⁻⁸) (ccSTPg ⁻¹)	(x10 ⁻⁷) (ccSTPg ⁻¹)	(x 10 ⁻⁴) (ccSTPg ⁻¹)	(x 10 ⁻⁸) (ccSTPg ⁻¹)	(x 10 ⁻⁹) (ccSTPg ⁻¹)		
nr1	Milan	15/5/11	15:40	8.2	4.67	1.78	3.66	8.70	12.43	0.95	0.02 Ann Arbor
n2a	Milan	4/7/11	0:27	25.0	5.56	1.94	3.03	6.50	8.34	0.97	0.02 -
nr2b	Milan	4/7/11	0:27	25.0	4.57	1.67	2.57	5.20	6.48	0.93	0.02 -
nr3-1 ^c	Milan	23/7/11	15:00	27.0	4.67	1.55	2.13	4.21	4.87	-	- Custer
nr3-2 ^c	Milan	23/7/11	15:30	21.4	4.63	1.74	2.63	5.29	6.55	0.98	0.03 "
nr4-1 ^c	Ann Arbor	23/7/11	21:35	26.0	4.62	1.70	2.69	5.28	6.42	0.98	0.03 Ann Arbor
nr4-2 ^c	Ann Arbor	23/7/11	21:35	26.0	4.99	1.55	1.99	3.91	5.10	0.93	0.03 "
nr5-1 ^c	Milan	27/7/11	23:40	23.5	4.51	1.71	2.68	5.51	7.30	1.05	0.02 Ann Arbor
nr5-2 ^c	Milan	28/7/11	5:00	22.5	5.28	1.74	2.80	5.72	7.43	1.03	0.04 "
nr6	Milan	29/7/11	8:25	22.6	4.37	1.64	2.60	5.30	7.26	0.98	0.02 Ann Arbor
nr7	Milan	3/8/11	7:00	24.0	5.44	1.80	2.92	6.05	7.78	0.96	0.12 Ann Arbor
nr8	Milan	6/8/11	18:40	28.6	4.39	1.68	2.72	5.49	7.17	0.99	0.14 Custer
nr9	Milan	9/8/11	9:35	20.8	4.72	1.72	2.87	5.94	7.56	1.18	0.16 Ann Arbor
nr10-1 ^c	Milan	9/8/11	21:05	23.1	5.22	1.68	2.74	5.48	6.94	1.15	0.07 Willow Run
nr10-2 ^c	Milan	9/8/11	21:55	20.3	4.44	1.66	2.65	5.41	6.75	1.02	0.03 "
nr11	Milan	14/8/11	11:40	18.4	4.96	1.79	2.99	6.55	8.88	1.05	0.03 Ann Arbor
nr12	Milan	24/8/11	7:50	19.6	4.52	1.78	3.23	7.03	9.61	1.05	0.04 Custer
nr13	Milan	4/9/11	0:47	23.9	5.32	1.82	3.02	6.75	8.84	0.92	0.04 Ann Arbor
nr14	Milan	9/9/11	15:10	18.5	5.06	1.78	3.14	6.91	9.38	1.04	0.03 Ann Arbor

^aUTC is Coordinated Universal Time. UTC=Eastern Standard time+4 hours during daylight savings in USA.

^bR is the measured $^3\text{He}/^4\text{He}$ ratio and Ra is the atmospheric $^3\text{He}/^4\text{He}$ ratio of 1.384×10^{-6} [Clarke *et al.*, 1976]

^cExcept for nr5-1 and nr5-2, all other samples were collected on the same day and almost at the same time (± 1 hour) indicating that they are likely from the same storm system