



Supplement of

Unprecedented radioactive pollution in Spitsbergen's air – first data of the 21st century

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Table S1: Meteorological parameters measured at the Stanislaw Siedlecki Polish Polar Station in Hornsund.

Variable	Location	Sensor	Height	Start operation	Unit
Air temperature	77°00'1.261" N 15°32'12.267" E	traditional thermometer in a Stevenson screen, Vaisala HMP 45D (since January 2001), HMP155 (since January 2018)	2 m a.g.l.	1979	°C
Relative humidity	77°00'1.261" N 15°32'12.267" E	hygrometer, HMP45D (since January 2001), HMP155 (since January 2018)	2 m a.g.l.	1979	%
Sum of precipitation	77°00'5.734" N 15°32'17.077" E	Hellmann rain gauge D-200	1 m a.g.l.	1979	mm
Atmospheric pressure	77°00'1.261" N 15°32'12.267" E	Mercury barometer, PTB200A (since January 2001), Baro-1QML-AV (since January 2018)	reduced to the sea level	1979	hPa
Wind speed and wind direction	77°00'1.261" N 15°32'12.267" E	Fuess 90z wind meter, Vaisala WAA151 (since January 2001), Vaisala WMT702 (since January 2018)	10 m a.g.l.	1983	m s ⁻¹
Sunshine duration	77°00'5.935" N 15°32'14.3" E	Campbell–Stokes heliograph	2 m a.g.l.	1979	h
Cloudiness	on location	visual observations	-	1983	octa
Visibility	on location	visual observations	-	1983	marine scale

Table S2: Results of quality assurance for blank samples.

Code	^{238}Pu (Bq)	$^{239+240}\text{Pu}$ (Bq)	^{241}Am (Bq)
blank1	< 0.000063	< 0.000068	< 0.000076
blank2	< 0.000043	< 0.000040	< 0.00016
blank3	< 0.000039	< 0.000034	< 0.00015
blank filter	< 0.00021	< 0.00012	< 0.00014

Table S3: Results of quality assurance for reference materials (with 95% confidence intervals for error assessment).

Reference material	^{238}Pu (Bq kg ⁻¹)		$^{239+240}\text{Pu}$ (Bq kg ⁻¹)		^{241}Am (Bq kg ⁻¹)	
	certified	calculated	certified	calculated	certified	calculated
IAEA-447	0.150 ± 0.020	0.148 ± 0.052	5.30 ± 0.40	5.27 ± 0.31	2.30 ± 0.40	1.89 ± 0.13
IAEA-385	0.392 ± 0.021	0.440 ± 0.044	2.98 ± 0.26	3.50 ± 0.18	4.42 ± 0.23	4.59 ± 0.36

Table S4: Meteorological parameters in quarters used for correlation assessment (input dataset available at <https://doi.org/10.48733/no6.25.015>).

year	quarter	Air temperature (°C)	Sum of precipitation (mm)	Relative humidity (%)	Cloudiness (octa)	Sunshine duration (h)	Visibility (marine scale)	Atmospheric pressure (hPa)	N (m s ⁻¹)	NE (m s ⁻¹)	E (m s ⁻¹)	SE (m s ⁻¹)	S (m s ⁻¹)	SW (m s ⁻¹)	W (m s ⁻¹)	NW (m s ⁻¹)
2007	1	-6.78	87.50	75.79	5.31	71.10	6.99	1001.03	0.00	1.04	3.81	0.58	0.47	0.33	0.06	0.05
2007	2	-2.70	48.30	78.23	6.22	543.70	7.34	1009.81	0.00	0.42	3.96	0.75	0.14	0.33	0.13	0.13
2007	3	3.38	130.80	85.50	6.51	382.60	7.85	1008.08	0.00	0.34	2.58	0.55	0.37	0.22	0.29	0.11
2007	4	-3.27	165.10	78.62	6.20	28.70	7.23	999.49	0.00	1.26	2.49	0.78	0.38	0.54	0.61	0.33
2008	1	-8.49	77.10	77.18	5.35	149.80	7.39	1001.15	0.00	0.71	4.88	0.32	0.39	0.35	0.33	0.16
2008	2	-2.82	56.50	78.17	5.47	709.40	7.88	1017.36	0.00	0.33	2.89	0.34	0.32	0.48	0.14	0.43
2008	3	3.69	226.20	88.21	6.62	369.10	6.93	1010.78	0.00	0.21	1.16	0.64	0.62	0.57	0.21	0.26
2008	4	-5.26	80.10	78.75	5.47	19.80	7.35	1001.67	0.00	0.93	3.29	0.54	0.31	0.42	0.46	0.13
2009	1	-9.28	89.60	78.65	5.69	66.00	7.03	1005.44	0.00	0.49	5.41	0.53	0.24	0.11	0.14	0.19
2009	2	-3.27	71.90	79.69	5.52	630.10	7.79	1013.26	0.00	0.02	2.80	0.54	0.25	0.39	0.48	0.34
2009	3	3.85	89.80	83.43	6.42	468.50	7.67	1009.05	0.00	0.24	3.09	1.09	0.19	0.13	0.23	0.05
2009	4	-2.15	228.40	82.94	6.57	34.90	7.14	1007.94	0.00	1.38	3.11	0.43	0.32	0.21	0.35	0.19
2010	1	-8.80	119.70	77.72	5.30	186.40	7.00	1011.58	0.00	1.17	3.79	0.46	0.40	0.48	0.24	0.19
2010	2	-1.77	65.80	83.03	6.51	488.90	7.57	1014.18	0.00	0.64	3.85	0.68	0.42	0.16	0.18	0.00
2010	3	3.32	151.80	87.20	6.51	376.40	7.47	1012.12	0.00	0.06	2.12	0.47	0.56	0.60	0.17	0.04
2010	4	-6.59	111.20	76.90	5.27	14.50	7.64	1009.20	0.00	1.27	3.13	0.53	0.28	0.35	0.11	0.00
2011	1	-10.62	139.10	79.48	5.30	115.40	7.65	1002.26	0.00	0.47	4.56	0.58	0.16	0.51	0.30	0.00
2011	2	-0.72	74.70	82.05	5.71	621.10	7.83	1010.62	0.00	0.50	2.52	0.57	0.43	0.38	0.38	0.06
2011	3	4.06	207.80	88.60	6.92	258.10	7.11	1010.55	0.00	0.14	3.44	0.47	0.41	0.33	0.33	0.00
2011	4	-3.09	195.70	79.07	6.02	42.20	7.07	996.87	0.05	1.40	3.00	0.53	0.28	0.23	0.29	0.19
2012	1	-2.97	172.50	81.35	6.28	90.30	6.92	1002.71	0.00	0.61	3.54	0.72	0.26	0.40	0.86	0.00
2012	2	-2.53	39.90	77.41	5.75	579.40	7.59	1011.17	0.00	1.17	2.39	0.42	0.21	0.40	0.30	0.11
2012	3	4.08	280.60	83.65	6.65	276.10	7.52	1005.71	0.00	0.03	2.82	0.65	0.36	0.30	0.31	0.19
2012	4	-3.07	68.80	75.64	5.26	34.20	7.58	1010.13	0.00	1.47	2.96	0.34	0.43	0.23	0.26	0.00
2013	1	-9.49	61.30	71.26	4.43	121.00	7.68	1013.27	0.00	1.20	4.06	0.40	0.22	0.27	0.26	0.13
2013	2	-2.44	76.80	81.16	5.81	474.90	7.56	1011.46	0.00	0.37	3.07	0.50	0.28	0.60	0.33	0.14
2013	3	5.44	321.40	86.22	6.68	327.00	7.43	1008.64	0.00	0.14	2.96	0.60	0.18	0.38	0.41	0.06
2013	4	-4.91	86.30	73.33	5.36	25.70	7.38	998.76	0.00	1.48	3.10	0.51	0.20	0.28	0.43	0.22

2014	1	-3.64	93.60	78.71	5.97	127.20	7.39	1006.77	0.00	0.55	3.84	0.60	0.25	0.28	0.43	0.07
2014	2	-1.96	56.50	76.39	6.12	566.60	7.84	1012.80	0.00	0.47	1.68	0.57	0.28	0.31	0.61	0.22
2014	3	4.33	151.50	82.19	6.43	437.00	7.48	1010.84	0.00	0.04	1.70	0.41	0.65	0.42	0.49	0.41
2014	4	-4.10	111.90	78.36	5.85	13.70	6.89	1005.38	0.00	0.97	3.19	0.49	0.26	0.30	0.62	0.16
2015	1	-7.34	107.20	78.79	5.66	78.40	6.49	998.53	0.00	0.59	5.09	0.30	0.39	0.27	0.49	0.18
2015	2	-1.41	30.20	81.13	6.27	547.10	7.54	1008.03	0.00	0.50	2.93	0.61	0.26	0.30	0.10	0.00
2015	3	4.34	251.60	85.21	6.30	420.00	7.62	1013.86	0.00	0.05	2.54	0.70	0.55	0.13	0.35	0.12
2015	4	-2.45	178.00	80.22	5.82	13.70	7.01	997.30	0.00	0.60	2.86	0.68	0.61	0.26	0.40	0.11
2016	1	-3.91	104.20	78.17	5.65	132.80	6.92	1003.05	0.01	0.99	3.23	0.61	0.31	0.55	0.11	0.05
2016	2	-0.03	50.60	80.95	6.20	432.10	7.67	1013.49	0.00	0.03	3.13	0.51	0.32	0.33	0.43	0.12
2016	3	5.07	241.80	85.92	6.74	354.30	7.29	1008.25	0.00	0.09	2.34	0.62	0.41	0.50	0.42	0.21
2016	4	0.02	410.10	81.34	6.44	1.40	6.44	1006.74	0.00	0.36	2.67	0.29	0.37	0.53	1.17	0.12
2017	1	-7.61	142.00	75.14	5.52	97.40	7.13	1000.40	0.00	0.81	2.43	0.63	0.28	0.28	0.88	0.24
2017	2	-2.62	49.10	78.17	5.54	657.80	8.21	1015.19	0.00	0.12	2.86	0.54	0.36	0.11	0.56	0.12
2017	3	5.13	223.80	86.60	6.72	327.30	7.33	1011.43	0.00	0.01	2.58	0.53	0.31	0.52	0.77	0.00
2017	4	-1.55	142.30	74.46	5.96	19.60	7.06	1003.29	0.00	1.96	3.99	0.24	0.05	0.16	0.21	0.00
2018	1	-6.36	91.70	72.98	5.05	171.60	7.39	1013.21	0.00	0.50	4.19	0.27	0.33	0.26	0.35	0.00
2018	2	-1.02	163.80	79.63	6.15	497.20	7.78	1008.66	0.00	0.01	2.38	0.58	0.82	0.70	0.33	0.26
2018	3	4.05	286.90	86.62	6.84	279.00	7.20	1006.39	0.00	0.25	2.29	0.30	0.56	0.57	0.57	0.21
2018	4	-2.26	227.80	77.40	6.17	42.00	7.41	1003.14	0.00	0.90	2.56	0.52	0.46	0.39	0.62	0.12
2019	1	-8.96	32.40	70.18	5.30	90.50	7.49	1001.67	0.00	1.58	4.04	0.38	0.33	0.06	0.29	0.08
2019	2	-0.42	59.20	76.42	6.13	598.30	7.87	1015.35	0.00	0.46	2.04	0.48	0.24	0.58	0.78	0.62
2019	3	4.22	116.10	84.99	6.34	354.30	7.67	1011.94	0.00	0.32	2.21	1.02	0.48	0.16	0.30	0.08
2019	4	-5.84	56.80	75.65	4.98	28.70	7.54	1008.33	0.00	1.72	3.11	0.27	0.20	0.24	0.04	0.08
2020	1	-12.81	40.10	71.55	4.27	132.30	7.51	994.36	0.00	0.91	3.72	0.59	0.34	0.43	0.21	0.21
2020	2	-1.72	67.40	81.20	6.12	461.40	7.72	1009.72	0.00	0.12	2.61	0.74	0.70	0.35	0.78	0.39
2020	3	4.81	231.30	88.92	6.46	348.10	7.59	1004.88	0.00	0.10	3.40	0.63	0.39	0.34	0.17	0.00
2020	4	-2.34	183.20	78.69	5.55	26.70	7.76	1007.07	0.00	1.16	3.50	0.56	0.25	0.20	0.58	0.11
2021	1	-6.36	107.70	76.72	5.29	79.10	7.60	1010.40	0.00	0.88	3.64	0.90	0.20	0.35	0.38	0.20
2021	2	-1.83	57.70	78.72	5.89	572.70	7.73	1009.64	0.00	0.70	2.16	0.46	0.22	0.68	0.74	0.23
2021	3	4.27	201.20	91.69	6.67	308.50	7.38	1008.19	0.00	0.00	2.52	0.64	0.53	0.44	0.44	0.00
2021	4	-4.54	49.30	73.32	5.43	17.30	7.27	1008.67	0.00	1.59	3.81	0.25	0.24	0.24	0.20	0.13

Table S5: Activity concentrations of radionuclides and suspended particulate matter (SPM) in quarters used for correlation assessment (input dataset available at <https://doi.org/10.48733/no6.25.015>).

year	quarter	SPM ($\mu\text{g m}^{-3}$)	^7Be ($\mu\text{Bq m}^{-3}$)	^{137}Cs ($\mu\text{Bq m}^{-3}$)	^{210}Pb ($\mu\text{Bq m}^{-3}$)	$^{239+240}\text{Pu}$ (nBq m^{-3})	^{238}Pu (nBq m^{-3})	^{241}Am (nBq m^{-3})
2007	1	6.86	2092.60	0.49	290.60	0.56	0.39	9.61
2007	2	4.75	2254.98	-	224.03	0.55	0.04	37.43
2007	3	3.75	970.43	0.36	79.61	0.63	0.17	8.99
2007	4	12.40	1397.65	0.38	275.79	0.85	0.20	13.67
2008	1	9.46	2614.72	0.30	448.23	0.64	0.03	16.03
2008	2	4.30	2885.33	0.25	358.31	0.84	0.11	11.96
2008	3	7.88	1100.93	0.19	79.60	0.45	0.05	8.01
2008	4	16.07	1655.87	0.27	186.11	2.27	0.43	11.37
2009	1	8.08	3097.29	0.26	479.99	0.86	0.34	24.18
2009	2	3.14	3389.85	0.21	399.13	0.54	0.29	73.72
2009	3	7.59	1594.00	0.37	93.28	2.11	2.04	13.68
2009	4	11.13	3155.70	0.37	254.12	0.62	0.81	21.02
2010	1	8.41	2800.51	0.35	524.72	0.96	0.32	35.04
2010	2	4.32	1772.40	0.44	144.70	1.18	0.49	19.57
2010	3	5.82	1282.44	0.25	139.40	0.81	0.08	41.44
2010	4	11.21	2518.43	0.29	432.70	-	0.31	-
2011	1	9.11	3082.54	0.32	464.99	-	-	-
2011	2	7.77	3027.80	0.48	192.55	-	-	-
2011	3	3.65	-	0.57	81.04	-	-	-
2012	3	7.97	-	0.26	46.76	-	-	-
2012	4	7.14	922.21	0.23	131.42	-	-	-
2013	1	5.40	2613.09	0.30	516.92	-	-	-
2013	2	6.52	1154.99	-	115.75	-	-	-
2013	3	8.33	1739.72	0.62	105.21	-	-	-
2013	4	7.97	1715.32	-	282.60	-	-	-
2014	1	7.36	2042.56	0.78	240.69	0.23	0.08	17.00
2014	2	4.82	1996.95	0.54	202.66	0.29	0.02	13.24

2014	3	8.41	1704.00	-	102.39	0.72	0.09	11.27
2014	4	7.60	2165.69	-	312.11	0.84	0.15	5.03
2015	1	15.32	2582.53	-	472.00	0.70	0.03	9.36
2015	2	5.79	2229.30	-	251.88	0.35	0.04	12.32
2015	3	3.80	1249.22	-	117.68	-	-	38.12
2015	4	11.94	1477.26	0.44	205.09	1.51	0.03	81.54
2016	1	10.41	1990.43	0.55	310.61	0.86	0.06	-
2016	2	7.07	1433.78	-	105.59	0.66	0.10	90.96
2016	3	10.00	1449.87	-	93.89	0.47	0.05	18.29
2016	4	15.87	1534.01	-	232.81	0.84	0.14	7.33
2017	1	5.93	1962.82	-	364.99	0.27	0.05	12.61
2017	2	4.33	2354.54	-	282.05	0.47	0.13	18.67
2017	3	4.62	991.90	-	100.92	0.38	-	4.85
2017	4	7.77	1567.15	-	161.76	1.13	1.82	10.42
2018	1	11.45	2205.41	-	359.75	0.35	1.20	14.85
2018	2	10.20	1656.99	-	179.04	0.32	0.69	34.10
2018	3	10.18	1051.68	0.17	81.86	1.65	0.29	86.74
2018	4	12.41	1247.86	-	159.19	0.91	-	-
2019	1	5.91	2407.05	-	494.42	0.77	-	-
2019	2	5.73	2023.55	-	194.05	0.51	0.22	40.44
2019	3	4.90	1154.00	-	100.46	0.78	0.05	25.53
2019	4	10.26	1900.83	0.55	255.57	-	0.39	22.40
2020	1	10.74	2419.59	0.63	593.17	-	0.75	15.07
2020	2	10.45	2789.05	-	202.10	0.80	0.32	13.12
2020	3	4.98	1302.47	0.11	90.09	0.90	-	10.45
2020	4	10.97	1550.83	0.07	162.02	1.43	0.21	18.03
2021	1	6.74	2549.31	-	307.59	0.44	0.17	2.55
2021	2	6.51	2564.23	-	161.97	0.43	0.09	8.54
2021	3	8.36	-	0.20	66.92	0.34	0.13	4.48
2021	4	12.68	2507.91	-	331.23	-	0.35	11.47

Table S6: Reference signatures (activity ratios) of selected past nuclear events.

Activity ratio	Global fallout ¹ (+ SNAP 9A)	Chernobyl ² (NPP)	Fukushima ³ (NPP)	Thule ⁴ (Weapon grade)	Cap de la Hague ⁵ (NRP)	Sellafield ⁵ (NRP)	Mayak ⁵ (PPP)	Stepovogo Fjord ⁵ (Sunken submarine reactor)	Abrosimov Fjord ⁵ (Sunken submarine reactor)
²³⁸ Pu/ ²³⁹⁺²⁴⁰ Pu	0.028 ^a	0.38 ^a	1.2 ^b	0.0161 ± 0.0005 ^c	0.36 ^d	0.19 ^d	0.005–0.018 ^d	0.12 ^d	0.45 ^d
²⁴¹ Am/ ²³⁹⁺²⁴⁰ Pu	0.38 ^a	2.0 ^a	-	0.169 ± 0.005 ^c	-	-	-	-	-

¹(UNSCEAR, 2000)

²(Salminen et al., 2005)

³(Hirose and Povinec, 2015)

⁴(Eriksson et al., 2008)

⁵(Oughton et al., 2001)

^adecay corrected to 2013

^bdecay corrected to 2011

^cdecay corrected to 2001

^dreference date specified elsewhere (Oughton et al., 2001)

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Table S7: Activity ratios of $^{238}\text{Pu}/^{239+240}\text{Pu}$ and $^{241}\text{Am}/^{239+240}\text{Pu}$ in quarters.

year	quarter	$^{238}\text{Pu}/^{239+240}\text{Pu}$	$^{241}\text{Am}/^{239+240}\text{Pu}$
2007	1	0.70 ± 0.12	17.1 ± 2.0
2007	2	0.068 ± 0.068	67.8 ± 6.7
2007	3	0.271 ± 0.074	14.2 ± 1.6
2007	4	0.231 ± 0.056	16.1 ± 2.0
2008	1	0.042 ± 0.042	25.0 ± 2.4
2008	2	0.126 ± 0.032	14.3 ± 1.3
2008	3	0.11 ± 0.11	17.9 ± 2.1
2008	4	0.191 ± 0.031	5.0 ± 0.4
2009	1	0.39 ± 0.41	28.0 ± 8.5
2009	2	0.537 ± 0.098	136 ± 13
2009	3	0.966 ± 0.066	6.48 ± 0.50
2009	4	1.30 ± 0.15	33.8 ± 3.3
2010	1	0.334 ± 0.046	36.4 ± 2.8
2010	2	0.413 ± 0.056	16.6 ± 1.2
2010	3	0.104 ± 0.059	51.1 ± 3.9
2010	4	0.0507 ± 0.0078	-
2011/2012	1-4	0.86 ± 0.20	13.7 ± 2.0
2013	1, 2	1.99 ± 0.25	35.8 ± 3.9
2013	2, 3	0.203 ± 0.066	62.6 ± 7.7
2014	1	0.33 ± 0.34	74 ± 16
2014	2	0.064 ± 0.065	45.5 ± 5.2
2014	3	0.124 ± 0.037	15.6 ± 1.3
2014	4	0.181 ± 0.042	6.03 ± 0.72
2015	1	0.037 ± 0.037	13.4 ± 1.1
2015	2	0.10 ± 0.10	35.0 ± 3.9
2015	3	0.426 ± 0.017	2.46 ± 0.12
2015	4	0.021 ± 0.021	54.1 ± 4.0
2016	1	0.075 ± 0.028	141 ± 10
2016	2	0.157 ± 0.033	138 ± 10
2016	3	0.10 ± 0.10	39.2 ± 4.2
2016	4	0.165 ± 0.044	8.75 ± 0.92
2017	1	0.19 ± 0.20	47.5 ± 7.6

2017	2	0.286 ± 0.089	39.6 ± 3.7
2017	3	6.29 ± 0.68	12.9 ± 1.5
2017	4	1.62 ± 0.12	9.23 ± 0.69
2018	1	3.41 ± 0.52	42.2 ± 5.9
2018	2	2.14 ± 0.34	105 ± 14
2018	3	0.173 ± 0.029	52.5 ± 3.4
2018	4	3.79 ± 0.30	-
2019	1	8.29 ± 0.55	459 ± 30
2019	2	0.434 ± 0.075	79.0 ± 6.3
2019	3	0.063 ± 0.063	32.7 ± 2.4
2019	4	0.133 ± 0.019	7.62 ± 0.41
2020	1	0.0766 ± 0.0067	1.539 ± 0.078
2020	2	0.406 ± 0.050	16.4 ± 1.3
2020	3	2.68 ± 0.21	11.60 ± 0.90
2020	4	0.149 ± 0.023	12.61 ± 0.80
2021	1	0.380 ± 0.074	5.86 ± 0.65
2021	2	0.213 ± 0.044	19.7 ± 1.8
2021	3	0.39 ± 0.10	13.3 ± 2.0
2021	4	0.115 ± 0.016	3.72 ± 0.29

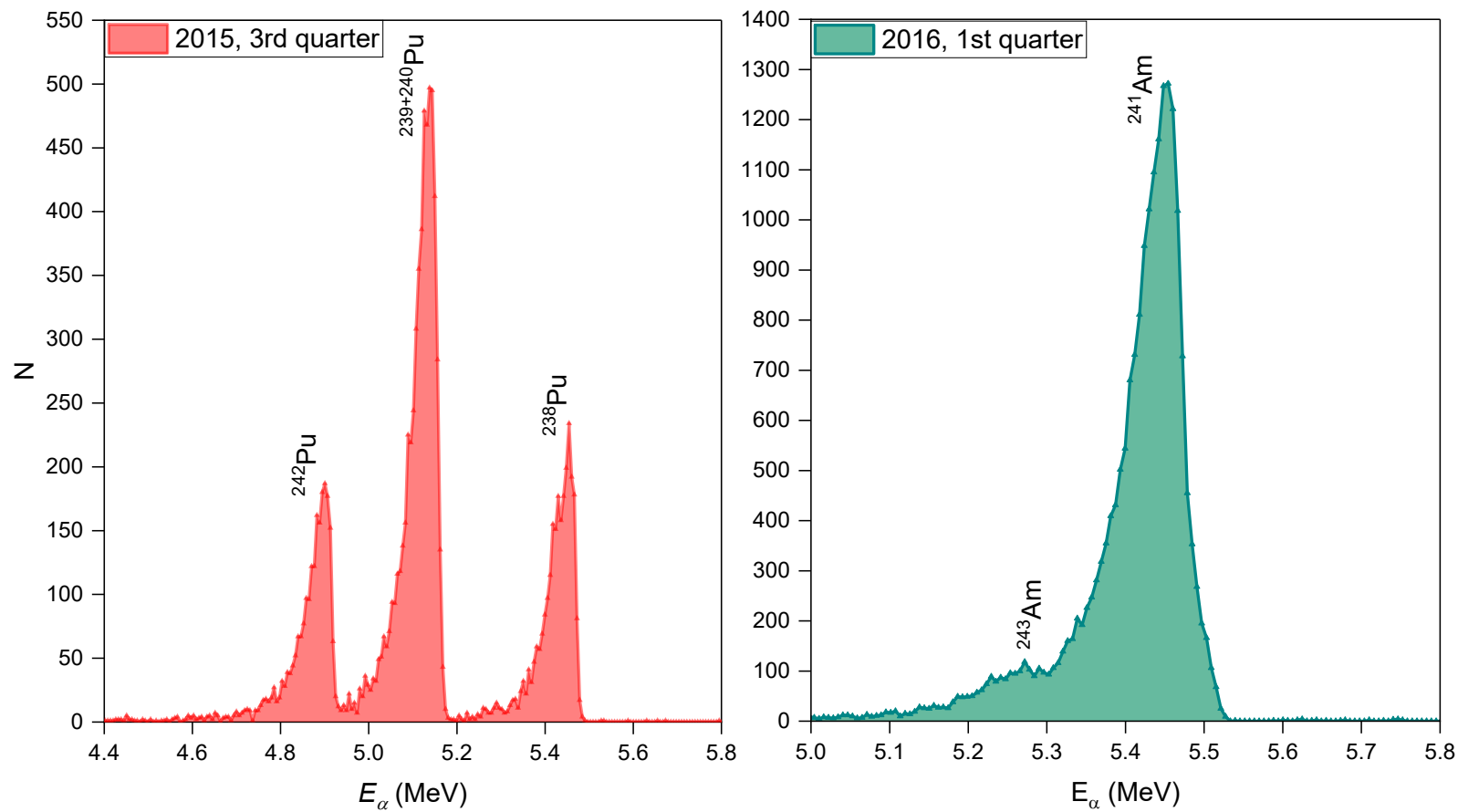


Figure S1: Alpha spectra of Pu and Am isotopes for air filter samples collected in the 3rd quarter of 2015 and 1st quarter of 2016, respectively. The acquisition time was two weeks for Pu and one week for Am.

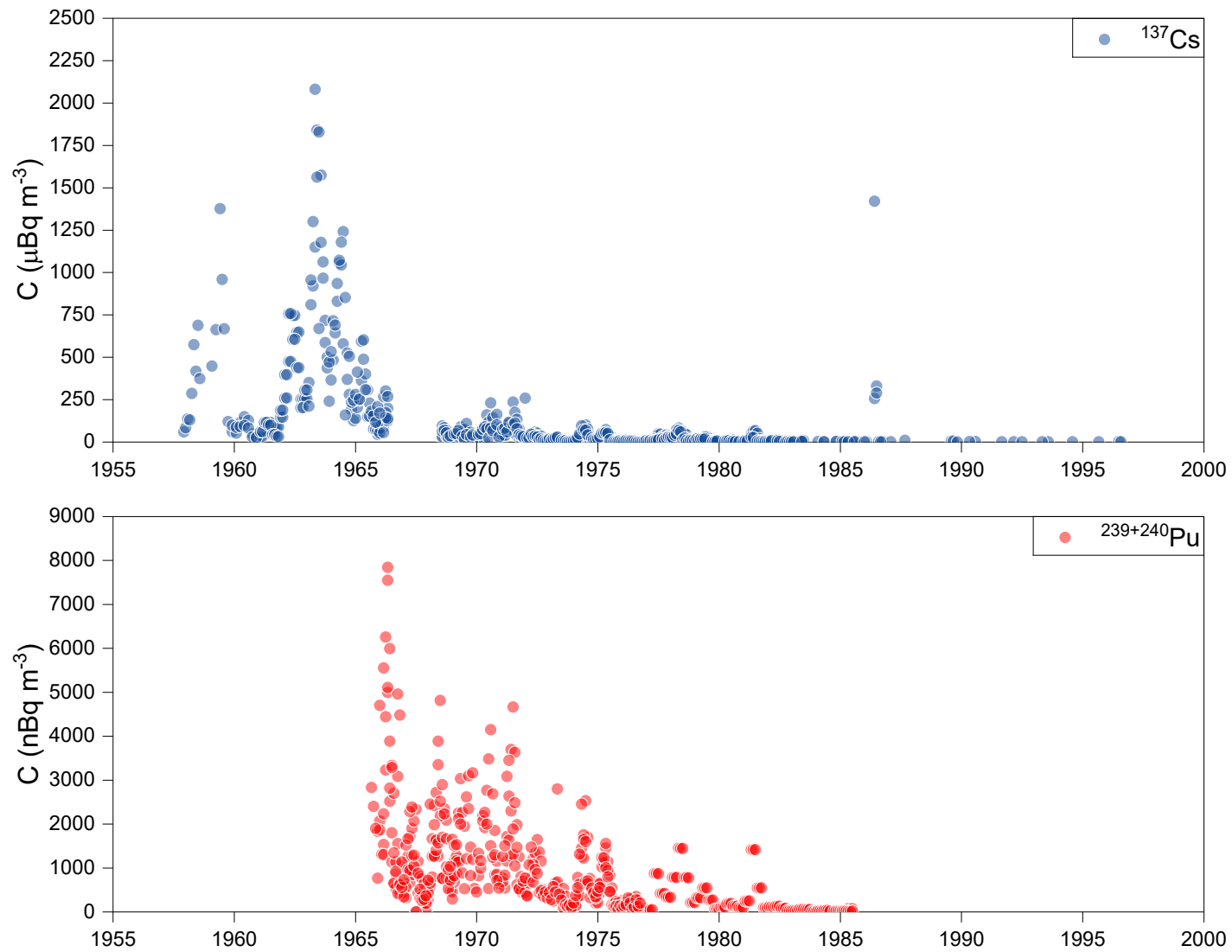


Figure S2: Time series of ^{137}Cs and $^{239+240}\text{Pu}$ activity concentrations in surface air for sites north of 50°N . Data collected by EML within the Surface Air Sampling Program (SASP) in the years 1963–1999 (and NRL during the 1957–1962 period).

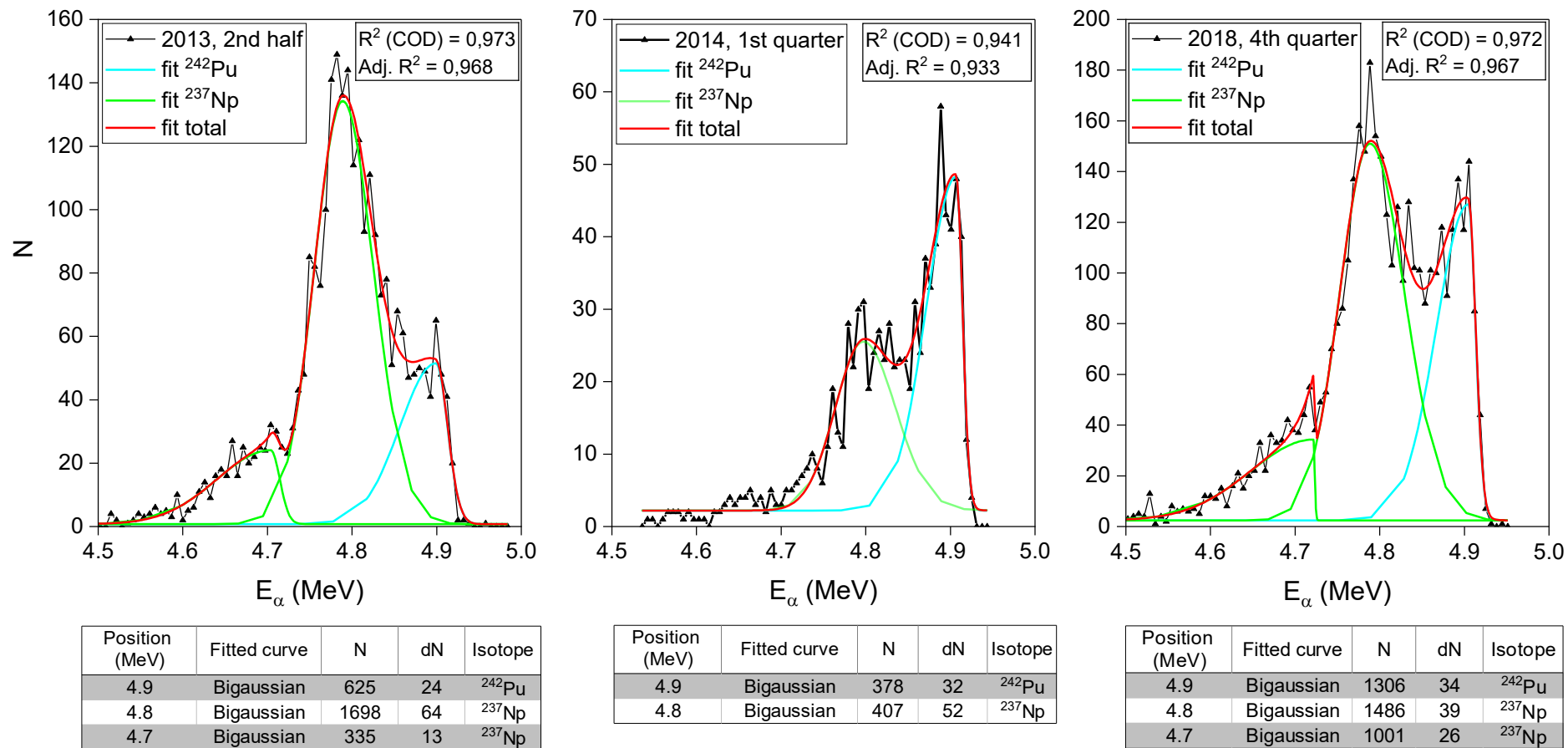


Figure S3: Bigaussian fitting procedure to resolve overlapping ^{242}Pu and ^{237}Np peaks with count integration for each spectral line.

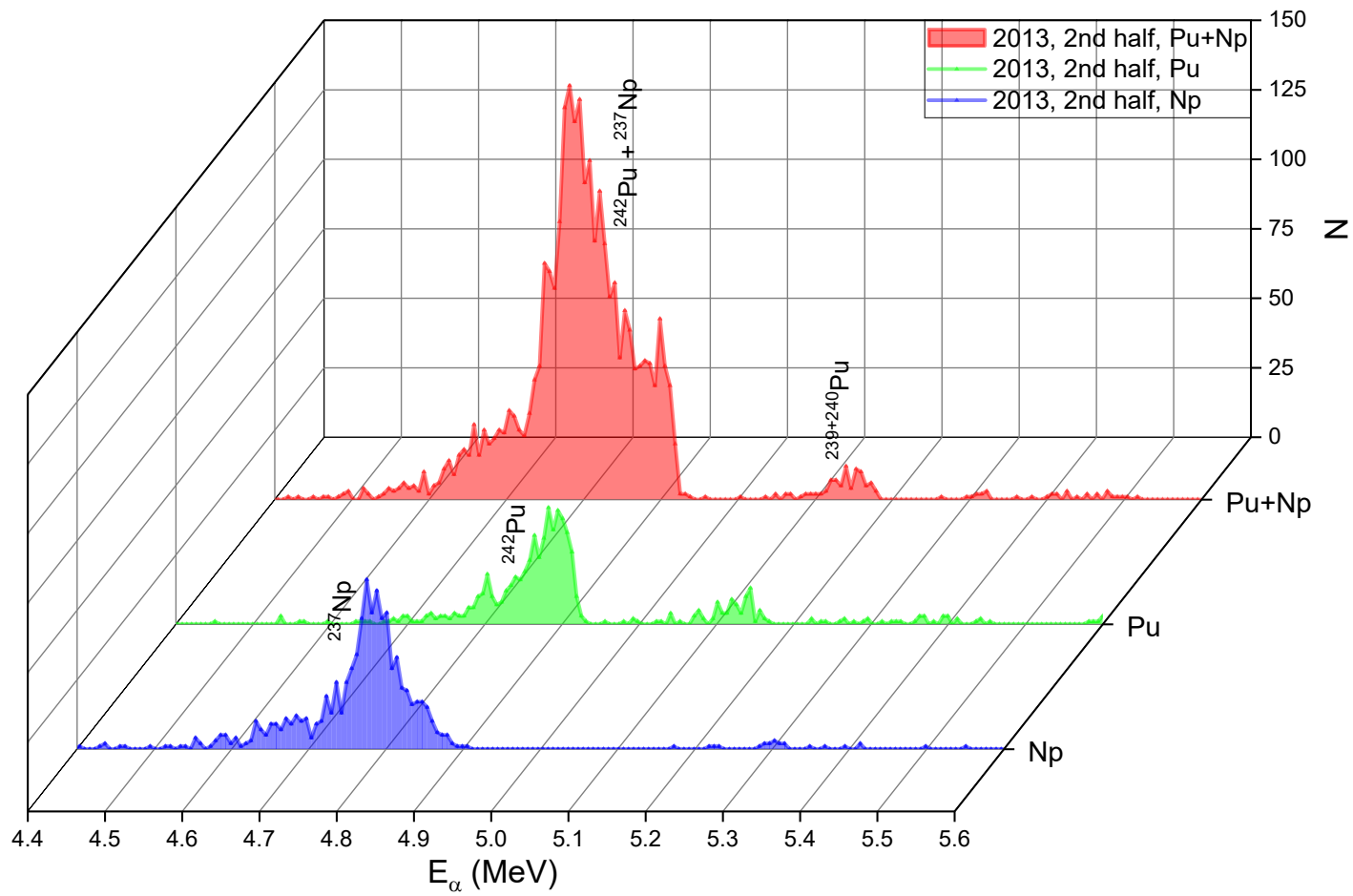


Figure S4: Alpha spectra of Pu+Np isotopes prior to separation, along with Pu isotopes and ^{237}Np after separation, from an air filter sample collected in the second half of 2013. The acquisition time was one week.

