

Unprecedented radioactive pollution in Spitsbergen air - first data as of 21st century - supplementary material

Anna Cwanek¹, Agnieszka Burakowska², Ewa Nalichowska¹, Magdalena Długosz-Lisiecka³, Marek Kubicki⁴, Tomasz Wawrzyniak⁴, Edyta Łokas¹, Michał Gryziński², Kamil Brudecki¹

¹Institute of Nuclear Physics, Polish Academy of Sciences, 31-342 Kraków, Poland

²National Centre for Nuclear Research, 05-400 Otwock-Świerk, Poland

³Lodz University of Technology, Institute of Applied Radiation Chemistry, 90-924 Łódź, Poland

⁴Institute of Geophysics, Polish Academy of Science, 01-452 Warszawa, Poland

Correspondence to: Anna Cwanek (anna.cwanek@ifj.edu.pl)

Table S1: Meteorological parameters characteristics.

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	%
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, VaisalaWAA151 (since January 2001), Vaisala WMT702 (since January 2018)	10 m a.g.l.	1983	m/s
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	octas
Visibility	On location	Visual observations	-	1983	marine scale

Table S2: Results of quality assurance for blank samples.

Code	²³⁸ Pu (Bq)	²³⁹⁺²⁴⁰ 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 (95% confidence intervals in error assessment).

Reference material	²³⁸ Pu (Bq/kg)		²³⁹⁺²⁴⁰ Pu (Bq/kg)		²⁴¹ Am (Bq/kg)	
	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 in correlation assessment.

year	quarter	Air temperature (°C)	Sum of precipitation (mm)	Relative humidity (%)	Cloudiness (octas)	Sunshine duration (h)	Visibility (a.u.)	Atmospheric pressure (hPa)	N (m/s)	NE (m/s)	E (m/s)	SE (m/s)	S (m/s)	SW (m/s)	W (m/s)	NW (m/s)
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) concentrations in quarters used in correlation assessment.

year	quarter	SPM ($\mu\text{g}/\text{m}^3$)	^7Be (Bq/kg)	^{137}Cs (Bq/kg)	^{210}Pb (Bq/kg)	$^{239+240}\text{Pu}$ (Bq/kg)	^{238}Pu (Bq/kg)	^{241}Am (Bq/kg)
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 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 $^{241}\text{Am}/^{239+240}\text{Pu}$.

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

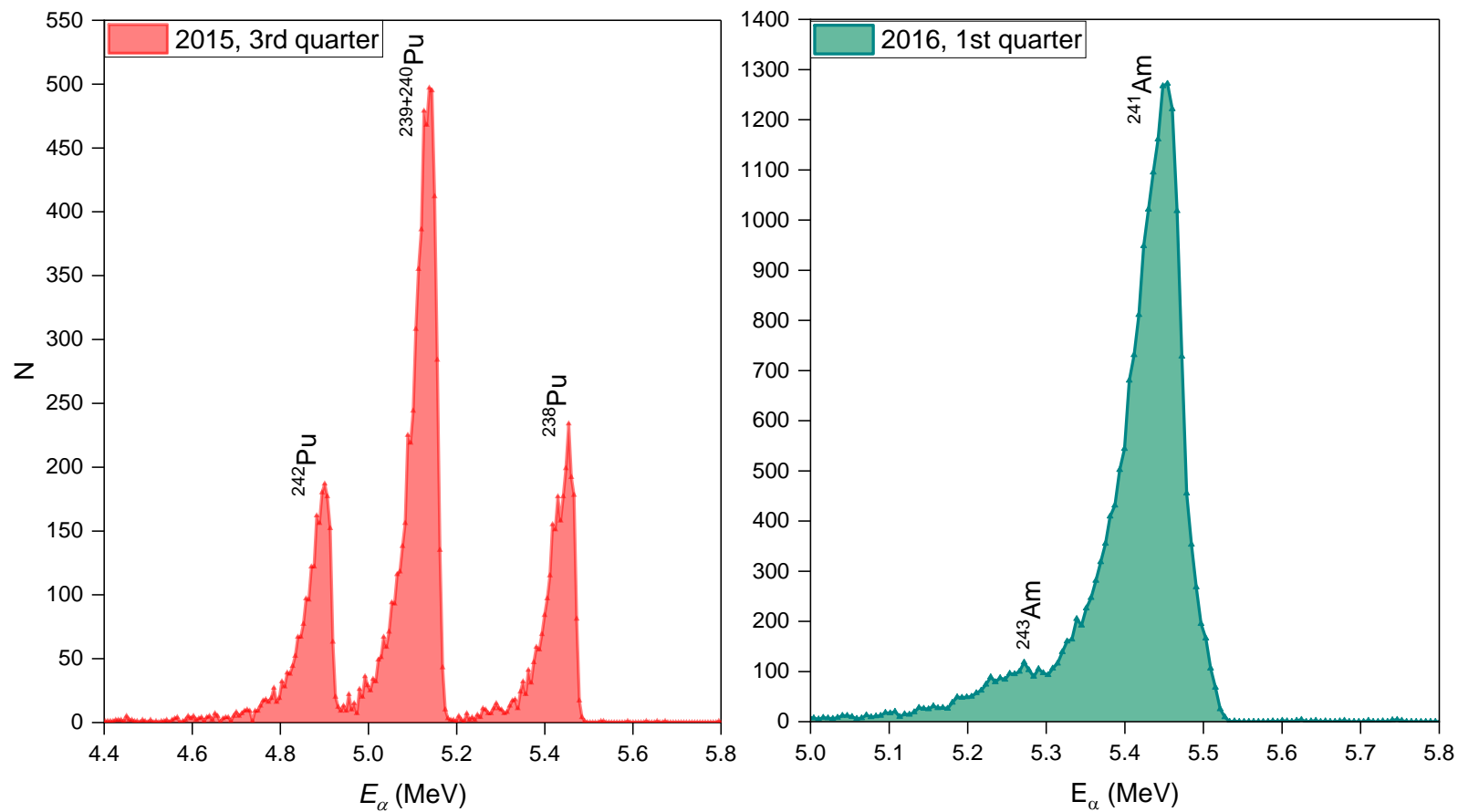


Figure S1: Alpha spectra of Pu and Am isotopes for air filter samples collected in the 3rd quarter of 2015 and 1st quarter of 2016, respectively. The acquisition time was 2 weeks for Pu and 1 week for Am.

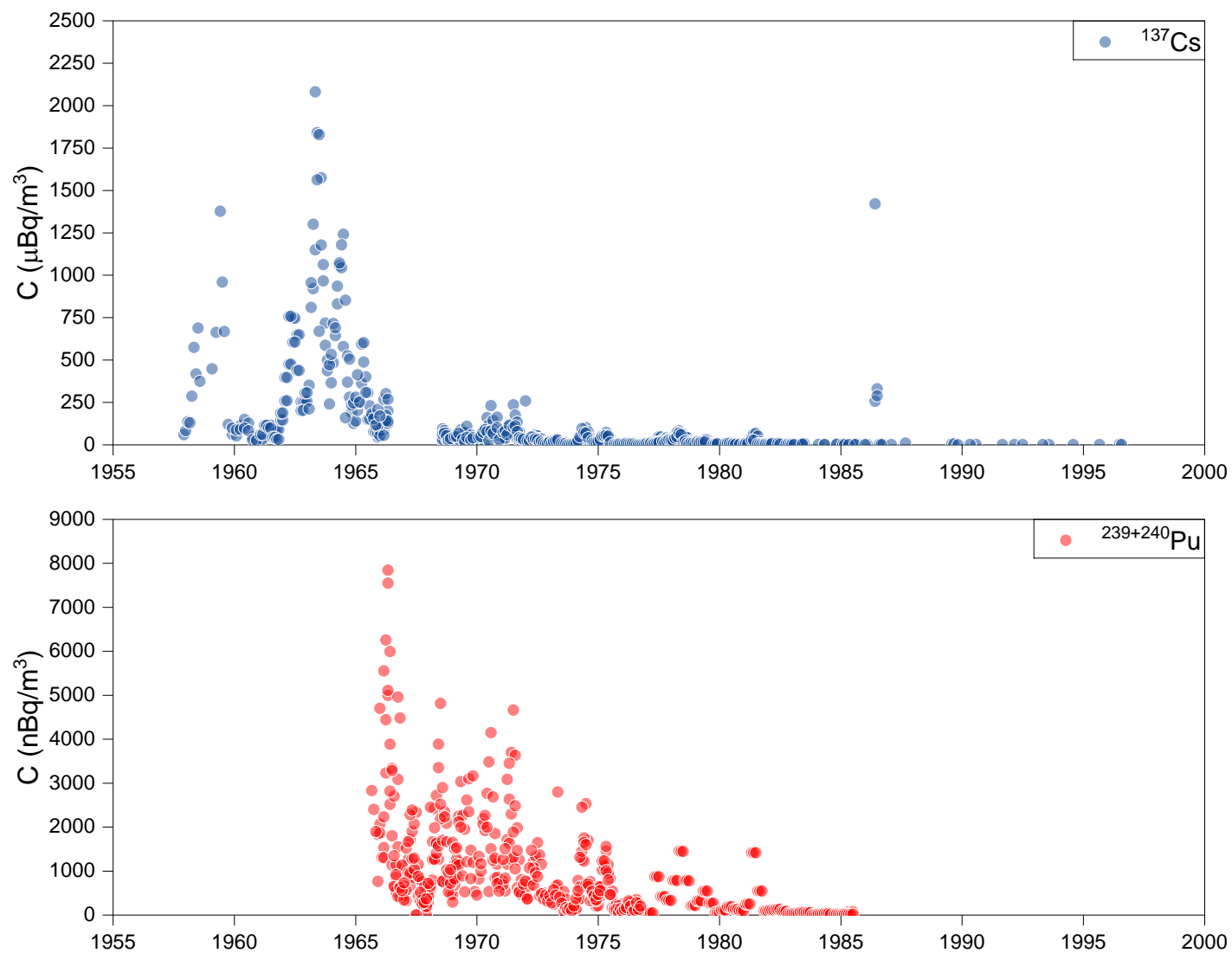


Figure S2: Time series of ^{137}Cs and $^{239+240}\text{Pu}$ activity concentrations in surface air of sites north of 50°N . Data collected by EML within the Surface Air Sampling Program (SASP) in the years 1963-1999 (and NRL during 1957-1962 period).

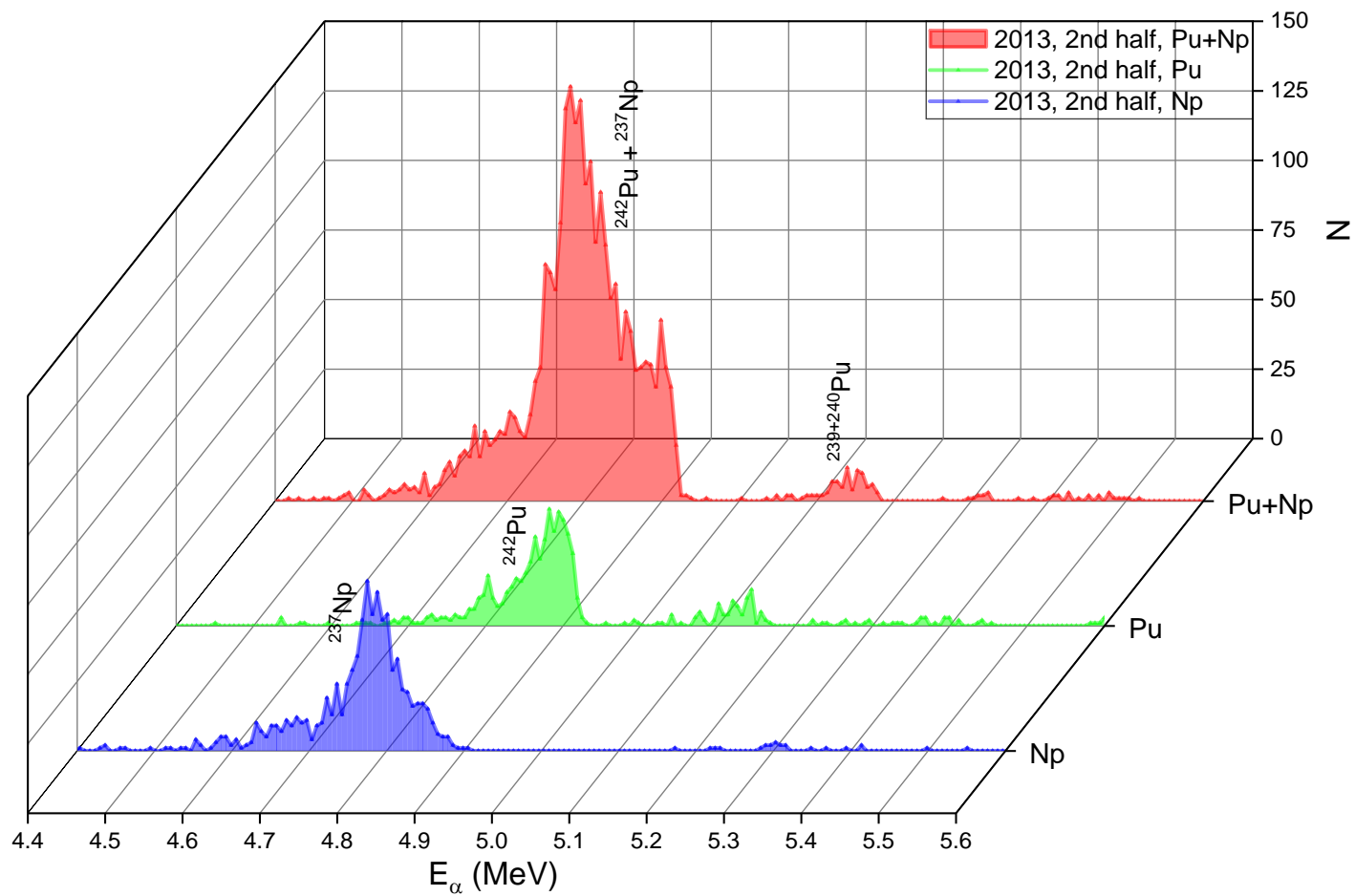


Figure S3: Alpha spectra of Pu+Np isotopes before separation as well as Pu isotopes and ^{237}Np after separation for air filter sample collected in 2nd half of 2013. The acquisition time was 1 week.

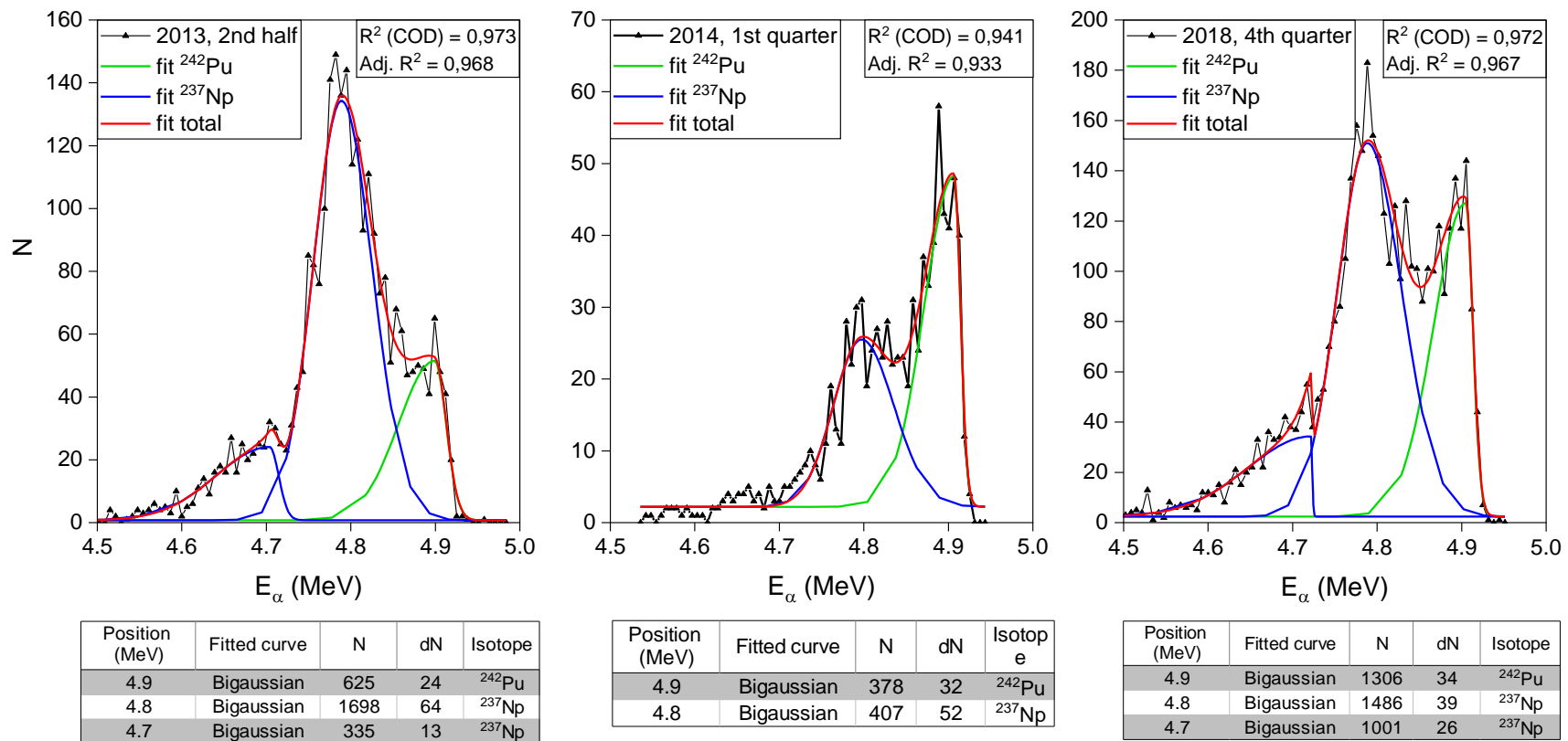
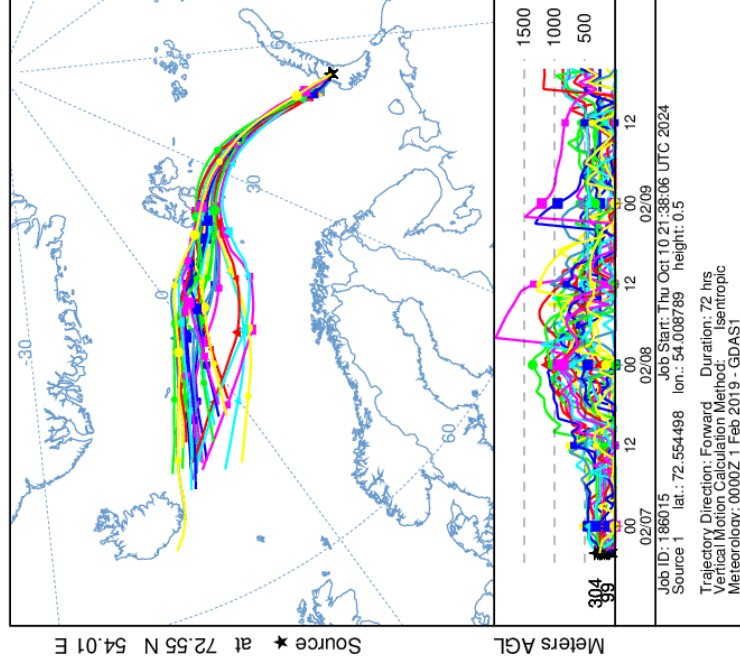
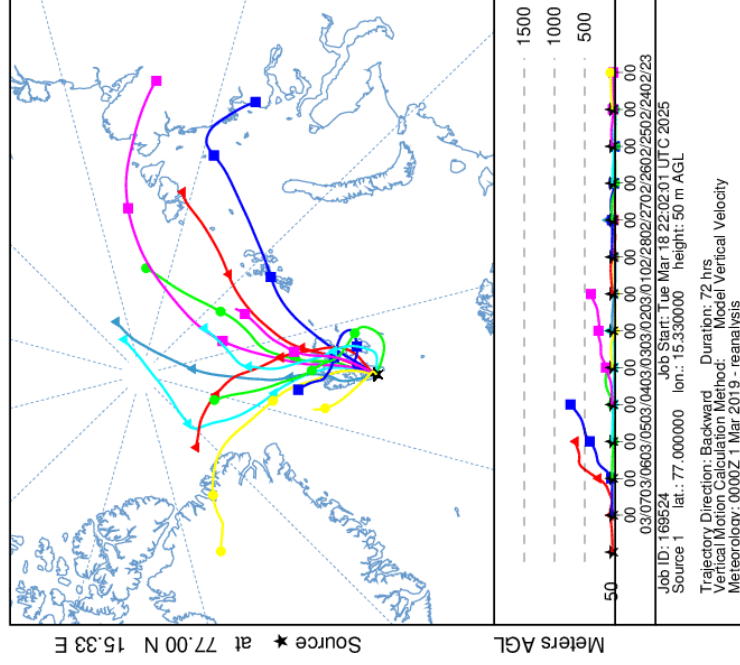


Figure S4: Bigaussian fitting procedure for ^{242}Pu and ^{237}Np peak separation with integrated counts for each spectral line.

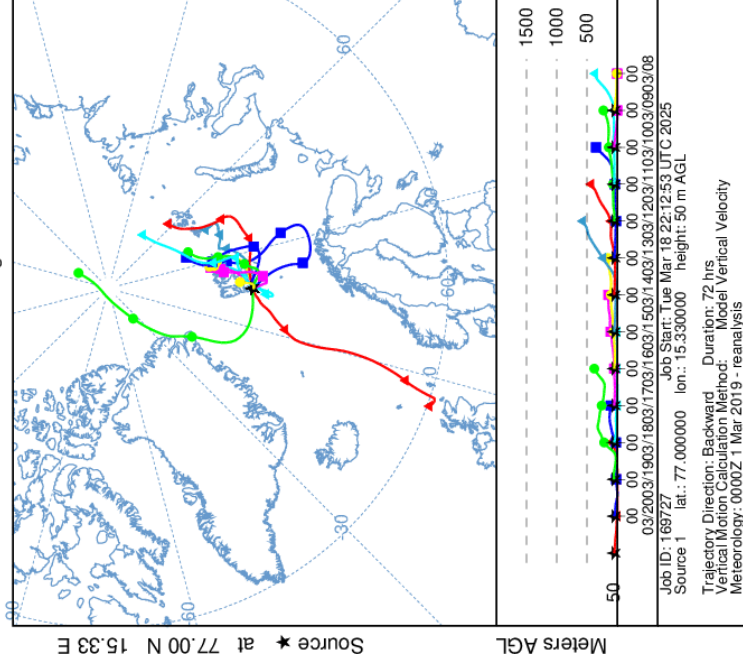
NOAA HYSPLIT MODEL
Forward trajectories starting at 2000 UTC 06 Feb 19
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 0000 UTC 08 Mar 19
CDC1 Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 0000 UTC 21 Mar 19
CDC1 Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 0000 UTC 31 Mar 19
CDC1 Meteorological Data

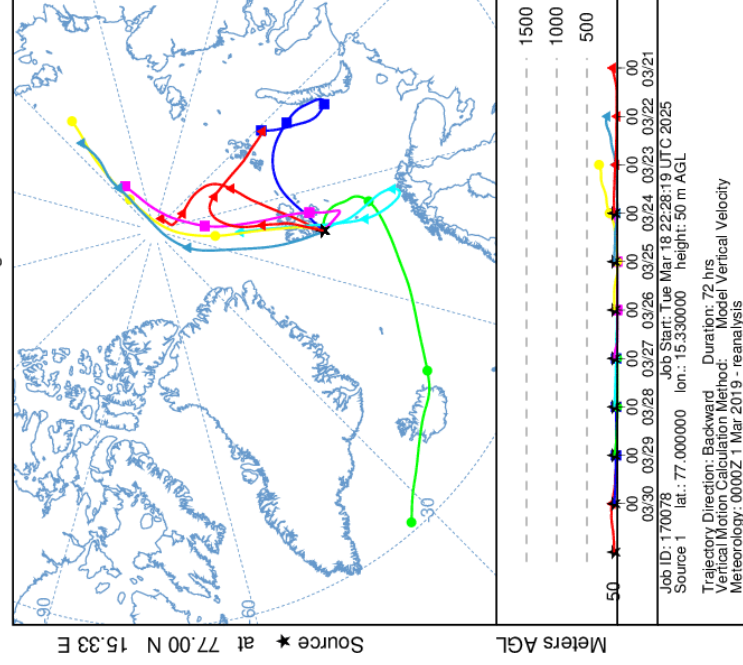


Figure S5: Trajectory simulations of aerosols transported through the troposphere during the 1st quarter of 2019 (forward and backward trajectories) performed for the location of Stanislaw Siedlecki Polish Polar Station in Hornsund, Spitsbergen.