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Scientific paper

Analysis of ^{7}Be and ^{210}Pb air concentrations in Ny-Ålesund, Svalbard: CHIMERPOL II project, preliminary results

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Abstract: Half-daily high volume aerosol particles samples have been collected onto cellulose and glass fiber filters at Ny-Ålesund, Svalbard, from April 14th to May 7th 2004, in the framework of CHIMERPOL II program. The filters have been analyzed for ^{210}Pb (22.3 y) and ^{7}Be (53 d) by semiconductor gamma spectrometry. The median concentration values were 70 μBq m^{-3} STP (8–264 μBq m^{-3} STP) for ^{210}Pb, and 404 μBq m^{-3} STP (35–824 μBq m^{-3} STP) for ^{7}Be respectively. The ^{7}Be and ^{210}Pb activity concentrations measured are relatively "low" compared to those measured in Svalbard and in other Arctic sites for the same period time (April–May) during previous studies. The ^{210}Pb atmospheric concentration and atmospheric pressure are positively correlated, which may indicate that the ^{210}Pb atmospheric concentration increase as a function of air mass exchange over the Svalbard region. The results of ^{7}Be and ozone concentrations show also the absence of direct ^{7}Be stratospheric injections during our sampling campaign, in the low troposheric air masses. The performed backward trajectory analysis showed that collected aerosols and associated radionuclides concentrations were associated to three different air masses, originating from Atlantic ocean, Artic ocean and Barents Sea, and continental areas (North Finland–Russia) respectively.

key words: Svalbard, arctic aerosols, ^{7}Be, ^{210}Pb

1. Introduction

Recent studies have been showing a net increase of mercury deposits in Arctic lacustrine sediments (Lockhart *et al*., 1998), as well as abnormally elevated or increasing mercury concentrations in Arctic fauna for about a decade (Wagemann *et al*., 1996; Dietz *et al*., 2000). The native populations of Arctic also show worrying mercury levels in their body (AMAP, 2002). Atmospheric mercury deposits through classical

mechanisms (wet deposits) as well as scarce mercury sources in Arctic areas (Pacyna and Keeler, 1995) cannot reasonnably explain these high levels. A part of these observations can be explained by the discovery in Alert (82.5° in North Canada) of an atmospheric depletion in gaseous elementary mercury concentrations: "Mercury Depletion Events" or MDEs (Schroeder *et al*., 1998). MDEs, today called Atmospheric Mercury Depletion Events (AMDEs), are characterized by an abrupt decrease of mercury concentrations occuring at the same time as a decrease of ozone concentration during boreal spring. This depletion can be very fast and leads to atmospheric mercury (Hg^0) levels close to zero during several hours. Since the observations were made at Alert, AMDEs have been registered in other Arctic sites, at Barrow, Alaska (Lindberg *et al*., 2001), at Ny-Ålesund, Svalbard (Berg *et al*., 2001), at Station Nord, Greenland (Skov *et al*., 2004), as well as in a sub-arctic site at Kuujjuarapik/Whapmagoostui in Quebec (Poissant *et al*., 2002). The origin of the AMDEs is not elucidated yet, and there are still many questions about the reaction mechanism, the reaction products and kinetics, as well as the role of the snow mantle. The role played by the atmospheric particles (origin, concentration, size distribution and so on) is not properly known, but it seems that they have a key position in AMDEs formation (Gauchard *et al*., 2003, 2005a,b). One of the aim of the CHIMERPOL II program is to develop the study of aerosol particles during Atmospheric Mercury Depletion Events. The origin and transport of air masses, as well as the local nature or not of the involved phenomenons looks to be of major importance. The use of radiochemical tools, widely used for the monitoring of Arctic pollution clouds (Arctic haze) (Lal and Peters, 1967; Robbins, 1978; Graustein and Turekian, 1996), and for the characterization of air masses and aerosols, represents one of the key point of this program.

Radon-222 (3.8 d), which is the Uranium-238 daughter, is a noble gas, released by the soils at a constant rate. The rate of release of this nuclide is twice weaker from the ocean surface than from the soils, and it is considered as negligeable for the areas covered by glaciers and permafrost (Robbins, 1978). The airborne ^{222}Rn of 99% originates from land and only 1% from the sea (Baskaran *et al*., 1993). Once released in the atmosphere, ^{222}Rn is rapidly desintegrated and produces numerous short-lived daughter nuclide *e.g*. ^{214}Pb (26.8 mn), ^{214}Bi (19.7 mn), as well as longer lived daughter elements like ^{210}Pb (22.3 y). These elements, that are mainly positive ions, are extremely reactive with atmospheric aerosols particles of sub-micronic size (0.05–0.5 μm) (Junge, 1963; Vohra *et al*., 1969). Once they are formed, the ions quickly link with these particles. The cosmogenic ^{7}Be (53 d), radioactive nuclide is formed quickly by spallation processes of light atmospheric nuclei such as carbon ^{12}C, nitrogen ^{14}N and oxygen ^{16}O with protons and neutrons (Papastefanou and Ioannidou, 1995; Gaggeler, 1995) and indiscriminately attaches to available aerosols, as well. As a consequence, the study of natural ^{210}Pb and ^{7}Be both associated to particles in surface air makes it possible to obtain information about the past and present history of the aerosols and their future, as well as the origin of the studied air masses. The analyses of ^{210}Pb together with ^{210}Po allow to estimate the residence time of aerosols as well as their origin (Poet *et al*., 1972; Moore *et al*., 1973; Suzuki *et al*., 1996; Baskaran and Shaw, 2001). The complementary analysis of ^{7}Be also gives further key information about the origin of the air masses and associated aerosols that exclusively come from the troposphere or

are a mixing between troposphere and stratosphere. Indeed, since the ^{7}Be is mainly of stratospheric origin, it has been used in many studies as a tracer for stratosphere-troposphere exchange (Danielsen, 1968; Husain *et al*., 1977; Sanak *et al*., 1985; Tremblay and Servranckx, 1993; Elbern *et al*., 1997). Beryllium-7 have also served as useful tracer for studying aerosol deposition velocities (Turekian *et al*., 1983) and deposition patterns of airborne contaminants (Graustein and Turekian, 1983). In conjunction with other components such as ^{210}Pb, this cosmogenic radionuclide thus provide an excellent examination of the wet scavenging parameterisation in general circulation models, since both tracers are removed from the troposphere primarily by precipitation (Brost *et al*., 1991; Koch *et al*., 1996).

In this paper, we report the preliminary results of daily variation of atmospheric ^{210}Pb and ^{7}Be concentrations in aerosol samples, in Ny-Ålesund, Svalbard from April 14th to May 7th 2004, in the framework of CHIMERPOL (CHIMIE DU MERCURE EN ZONE POLAIRE) II program. We compare the observations to database in other Arctic sites, and analyze together the analytical results and meteorological parameters. The results presented in this paper also allow to insert new values in the radionuclide database in Arctic area, and in particular in Svalbard, to address the issues on the sources and transport of arctic aerosols.

2. Material and methods

2.1. Field measurements

2.1.1. Air sampling

Large volume air samples from Ny-Ålesund (78° 56′N, 11° 52′E; 10 m asl), Svalbard (Fig. 1), were filtered through an air filtration assembly at a typical flow rate of 30–40 $m^3 h^{-1}$. Air was continuously pumped and filtered in a 'protected' area located 2.5 m above snow surface, 300 m far away from first Ny-Ålesund habitations (Fig. 2). We checked that the snow drift was not strong enough to interfere with the air sampling. The filtration system was protected from local precipitations, which could occur at this time of the year. The air-filters used are made of cellulose and glass fibers (C-577 type, Bernard DUMAS Cie), representing discs of 108 mm useful diameter for a sampling surface of 91.6 cm^2. These filters have been shown to be 99.9% efficient for the sub-micron size aerosols carrying ^{7}Be and ^{210}Pb. However, we have to be cautious and keep in mind that concentrations measured during the campaign, and reported later in the paper may be 10–20% lower than they are supposed to be. In any case, ratios will not be affected. The filters were changed every 12 hours, which corresponded to a filtered air volume of about 250–500 m^3 STP. Every 10 samples, one filter is unexposed and used as a field blank sample. Sample collection was carried out from April 14th to May 7th 2004. Then, air filters were sent to the Laboratory of Glaciology and Geophysics of Environment (LGGE) for measurements.

2.1.2. Meteorological parameters

Since 1992 a meteorological observatory monitoring programme is carried out at Koldewey Station, Ny-Ålesund, by the Alfred Wegener Institute (AWI), in close cooperation with the Norsk Polarinstitutt (NP). Synoptic observations are carried out from the Norsk Polarinstitutt. The activities include measurements of several meteor-

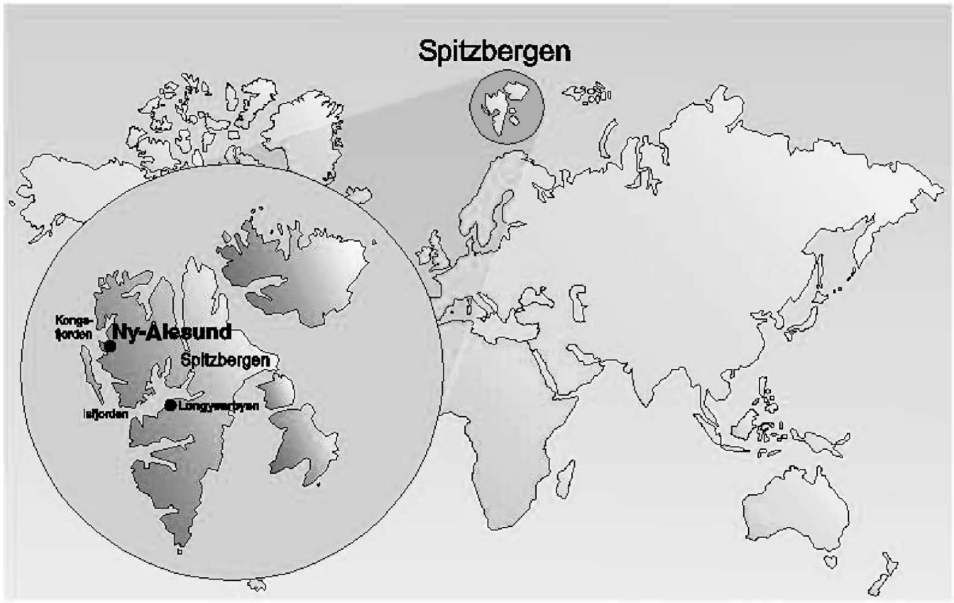

Fig. 1. Location of Ny-Ålesund, in Svalbard region.

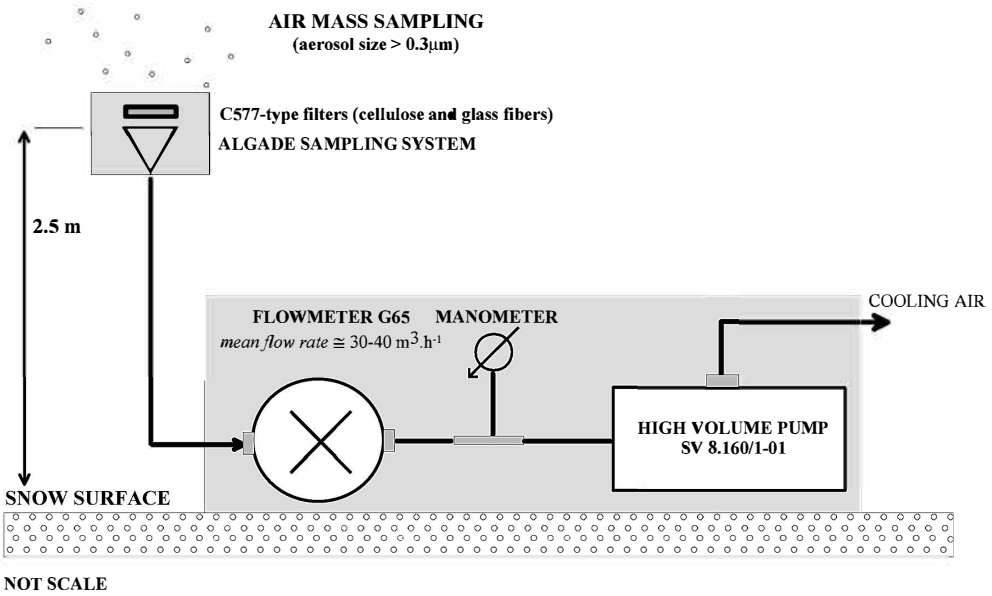

Fig. 2. Aerosol sampling device system.

ological parameters such as air temperature (at 2 m height), air pressure (values are reduced to mean sea level), wind vector (direction and speed, at 10 m height), dew point temperature (at 2 m height), clouds (cloud amount, type and height), horizontal visibility, and present and past weather observations. The full observation programme is carried out at 6, 12, 18 UTC. The data is coded (FM12-SYNOP) and transferred directly to the Global Telecommunication System (GTS) where they contribute to weather forecasting. The meteorological and aerological data measured at the station are sent on a regular basis to the World Meteorological Organisation (WMO).

Quantitative measurements of precipitation at Ny-Ålesund are not available in this study, to investigate the role of wet scavenging for the radionuclides surface level concentrations. This is due to the fact that, during our studied period, snow is generally the vast component of precipitation in this region, and together with strong winds and drifting snow, make such measurements very difficult. In order to try investigating the possible role of wet scavenging, we used the observations carried out by AWI in Koldewey Sation, at 0600, 0900 and 1200 AM and PM GMT. Although these are only qualitative data, they can be used to obtain an estimate of precipitation frequency and intensity.

2.1.3. Ozone measurements

In addition to the above meteorological parameters, total O_3 was used to investigate the role of possible direct stratospheric intrusions on the ^{7}Be activity concentrations at Ny-Ålesund. Ozone has been then monitored at the Norwegian research station on the nearby Zeppelin mountain (474 m a.s.l.), at 300 m far from first Ny-Ålesund habitations. Model 400 ozone analyzer (Advanced Pollution Instrumentation, USA) has been used. This model is based on UV spectrophotometry at 254 nm, with O_3 concentration following the Beer-Lambert law (Gauchard *et al*., 2005b). Method detection limits are ~ 0.6 ppb, precisions of 1 ppb. Both instruments were setup to output 5 min average data.

2.2. ^{7}Be and ^{210}Pb analysis

The filters were directly analyzed by gamma spectrometry using a very low background germanium detector (germanium diode N type) (Pinglot and Pourchet, 1979, 1994). This detector has a relative efficiency of 20%. For high resolution gamma spectrometry, the analyzer was protected against all interfering ambient radioactivity, in particular using an anti-Compton device. This system provides a lower detection threshold, especially for the isotopes of interest such as ^{210}Pb and ^{7}Be (Pinglot and Pourchet, 1994, 1995; Pinglot *et al*., 1999). Standard ^{137}Cs and ^{210}Pb liquids from the CEA or Amersham laboratories (2% uncertainty at 95% of confidence level) were used to calibrate the detector and to correct the calculated activities. The analytical procedures were the same as those used for the air samples. The ^{210}Pb and ^{7}Be measurements were carried out with a time resolution between 24 and 72 h. Quantitative analysis software (Genie 2000 v1.4; Canberra) is used to compute the activity of existing radionuclides and the associated accuracies. The total uncertainties due to both sampling procedures and counting statistics are of the order of 20% for ^{210}Pb and 50% for ^{7}Be. The specific activities have been corrected for decay to the deposition time and counting. This time period was generally near 30–40 days for the majority of filters, but occasionally approached more than 100 days. For this last type of samples,

counting time was increased to estimate ^{7}Be activities and to limit uncertainties. Blank and background values were regularly checked.

3. Results and discussion

3.1. ^{210}Pb and ^{7}Be concentrations

All the measured concentrations data of ^{210}Pb, ^{7}Be and O_3 are listed in Table 1. Daily variations of ^{210}Pb and ^{7}Be atmospheric concentrations are shown in Fig. 3.

Lead-210—Results give a ^{210}Pb mean concentration and a standard deviation ($\pm 1\sigma$) of $82 \pm 59\,\mu$Bq m^{-3} (n=43) ranging from 8 to $264\,\mu$Bq m^{-3}. The median concentration is around $70\,\mu$Bq m^{-3} (n=43), *i.e.*, 15% lower than the mean value. Figure 4 shows that data population has not a gaussian distribution. More than 75% of the ^{210}Pb concentrations values are below $100\,\mu$Bq m^{-3} and remaining data are included between 100 and $270\,\mu$Bq m^{-3}. In this case, the mean value is strongly influenced by the extreme values, thus driving to the observed difference between the mean and the median values. As a consequence, the median value, $70\,\mu$Bq m^{-3}, will be preferentially used as reference for comparison with other datasets.

Concerning Svalbard atmospheric radionuclides database, Paatero *et al.* (2003) measured ^{210}Pb activity concentrations in the air during one year (2001), at Mt Zeppelin Global Atmospheric Laboratory (474 m a.s.l.), close to Ny-Ålesund. The values vary between 11 and $620\,\mu$Bq m^{-3}, with annual arithmetic mean concentrations of $144\,\mu$Bq m^{-3}, and the 50% percentile close to $83\,\mu$Bq m^{-3}. Although the considered temporal scale is different, this last value is close to the 50% percentile observed in our study. Paatero *et al*. (2003) found that the seasonal variation of airborne ^{210}Pb showed lowest concentrations—$<100\,\mu$Bq m^{-3}—during summer (June to October) and highest values —$>100\,\mu$Bq m^{-3}—during wintertime and springtime (November to May), and in particular in March–April. This could differ from what we observed during our measurements period—April 14th to May 7th 2004—characterized by relatively "low" concentrations compared to 2001. However, if we carefully observe the evolution of ^{210}Pb activity concentrations in the air at Mt Zeppelin in 2001, Paatero *et al.* (2003)'s data show two short periods, between April and the beginning of May 2001, during which the ^{210}Pb concentrations sharply drop, reach concentration levels close to what we measure in April–May 2004, and sharply increase again after a few days. Actually, the transition period between winter and summer, is characterized by progressive changes in the atmospheric circulation (Preiss *et al*., 1996), implying changes in the origin of air masses reaching Svalbard. As described by Preiss *et al.* (1996), the artic winter is characterized by the extend of the polar front down to mid-latitudes including large areas of the North American and Eurasian continents. At this time of the year, these regions are often subjected to large anticyclones. The Artic troposphere constitutes also a mixed reservoir which includes emissions from mid latitudinal continental areas, source of ^{222}Rn and then, atmospheric ^{210}Pb. This leads to high ^{210}Pb concentrations during winter in Artic area such as Svalbard (Larsen and Sanderson, 1991). The small amount of precipitation in the northern regions in winter and low temperatures limit the aerosol condensation rate and increases the mean residence time of the aerosol particles,

Table 1. Concentrations of ^{7}Be, ^{210}Pb and $^{7}Be/^{210}Pb$ ratio in aerosol filter samples, and total ozone data from Ny-Ålesund Arctic site, Svalbard, from 14th of April to 7th of May 2004.

No	Sampling date (every 12 hours) (local time)	Activity concentrations * ($\mu Bq\ m^{-3}$ STP) ^{7}Be	^{210}Pb	^{7}Be / ^{210}Pb	Total Ozone £ (mean ± 1σ) (ppbv)
1	22h00 – 04/14/04	247 ± 27	115 ± 20	2.1	51 ± 2
2	22h00 – 04/15/04	258 ± 21	96 ± 15	2.7	50 ± 0.3
3	10h00 – 04/16/04	521 ± 42	161 ± 24	3.2	50 ± 0.8
4	22h00 – 04/16/04	416 ± 25	94 ± 11	4.4	47 ± 0.8
5	10h00 – 04/17/04	337 ± 34	70 ± 24	4.8	41 ± 1.5
6	22h00 – 04/17/04	366 ± 33	95 ± 18	3.8	39 ± 1.0
7	10h00 – 04/18/04	181 ± 18	73 ± 15	2.5	38 ± 0.5
8	22h00 – 04/18/04	280 ± 28	93 ± 19	3.0	39 ± 0.6
9	10h00 – 04/19/04	341 ± 31	82 ± 19	4.1	40 ± 0.4
10	22h00 – 04/19/04	370 ± 37	167 ± 23	2.2	42 ± 0.7
11	08h30 – 04/20/04	598 ± 54	214 ± 24	2.8	42 ± 0.6
12	22h00 – 04/20/04	824 ± 66	264 ± 24	3.1	46 ± 0.3
13	10h00 – 04/21/04	610 ± 61	125 ± 26	4.9	46 ± 0.3
14	22h00 – 04/21/04	546 ± 71	172 ± 29	3.2	48 ± 0.7
15	10h00 – 04/22/04	556 ± 67	137 ± 29	4.1	49 ± 0.5
16	22h00 – 04/22/04	739 ± 74	210 ± 31	3.5	50 ± 2.1
17	10h00 – 04/23/04	433 ± 56	94 ± 25	4.6	44 ± 2.1
18	22h00 – 04/23/04	439 ± 53	66 ± 26	6.6	45 ± 2.6
19	10h00 – 04/24/04	672 ± 67	117 ± 23	5.7	51 ± 2.5
20	22h00 – 04/24/04	773 ± 69	121 ± 25	6.4	49 ± 4.8
21	10h00 – 04/25/04	745 ± 67	54 ± 22	13.8	48 ± 3.0
22	22h00 – 04/25/04	522 ± 52	72 ± 21	7.2	53 ± 1.9
23	10h00 – 04/26/04	35 ± 16	n.d.	/	49 ± 1.1
24	22h00 – 04/26/04	n.d.	26 ± 16	/	48 ± 1.0
25	10h00 – 04/27/04	57 ± 25	22 ± 17	2.6	47 ± 0.7
26	22h00 – 04/27/04	n.d.	61 ± 9	/	45 ± 0.4
27	10h00 – 04/28/04	n.d.	8 ± 11	/	44 ± 1.9
28	22h00 – 04/28/04	n.d.	35 ± 14	/	26 ± 2.9
29	10h00 – 04/29/04	n.d.	55 ± 17	/	27 ± 1.4
30	22h00 – 04/29/04	n.d.	33 ± 15	/	24 ± 2.7
31	10h00 – 04/30/04	n.d.	n.d.	/	23 ± 1.6
32	22h00 – 04/30/04	n.d.	31 ± 17	/	22 ± 1.4
33	10h00 – 05/01/04	n.d.	43 ± 15	/	25 ± 1.0
34	22h00 – 05/01/04	n.d.	20 ± 14	/	29 ± 1.3
35	10h00 – 05/02/04	n.d.	35 ± 14	/	33 ± 1.7
36	22h00 – 05/02/04	n.d.	8 ± 14	/	38 ± 2.8
37	10h00 – 05/03/04	404 ± 69	55 ± 17	7.4	49 ± 1.7
38	22h00 – 05/03/04	n.d.	16 ± 15	/	38 ± 3.6
39	10h00 – 05/04/04	n.d.	17 ± 18	/	33 ± 0.4
40	22h00 – 05/04/04	78 ± 71	44 ± 21	1.8	37 ± 2.2
41	10h00 – 05/05/04	n.d.	33 ± 18	/	40 ± 1.8
42	22h00 – 05/05/04	110 ± 42	74 ± 13	1.5	37 ± 0.6
43	10h00 – 05/06/04	n.d.	80 ± 17	/	36 ± 1.1
44	22h00 – 05/06/04	89 ± 28	66 ± 14	1.4	32 ± 0.9
45	10h00 – 05/07/04	99 ± 43	55 ± 21	1.8	32 ± 0.6

*The errors reported are propagated errors arising from one sigma counting uncertainty due to detector calibration and background correction. n.d., not detected

£ Total ozone data are averaged every 12 hours. The errors reported are standard deviations from one sigma.

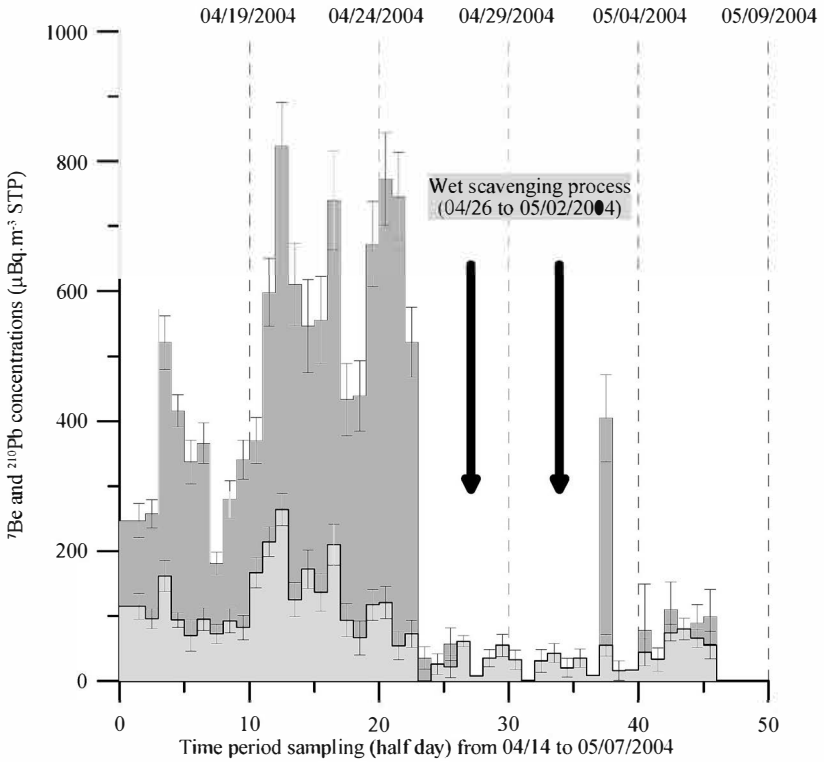

Fig. 3. Daily variation of atmospheric concentrations of ^{7}Be (dark gray) and ^{210}Pb (light gray) at Ny-Ålesund, Svalbard from April 14th to May 7th 2004. Vertical bars in each plot indicate counting error (1σ).

allowing the accumulation of radionuclides in the surface air (Paatero *et al.*, 2003). That is what Suzuki *et al.* (1996) observed during an atmospheric ^{210}Pb measurement program realized by a Japanese Arctic Glaciological Expedition in 1995, in Ny-Ålesund. From February 24th to March 14th 1995, they showed that ^{210}Pb atmospheric concentrations varied from 83 to 1204 μBq m^{-3} with an average of 325 μBq m^{-3} ($n=18$) (Suzuki *et al.*, 1996). In springtime, the polar front moves northward, leaving the major sources areas, and it isolates, with increasing efficiency, the Arctic from the mid latitudinal influences. As a consequence, the Arctic troposphere is relatively depleted in ^{210}Pb during the summer (Larsen and Sanderson, 1991; Paatero *et al.*, 2003). Our studied period corresponds to April–May, when the concentrations are usually still quite high (Paatero *et al.*, 2003). However, we can observe that our values are close to the ones observed during the summer period, *i.e.*, when the concentrations are usually low (Paatero *et al.*, 2003). If we consider other Arctic sites, we can see that our data are slightly lower than the values observed in summer time at Thule and Point Barrow—$\approx$100 μBq m^{-3}—(Larsen and Sanderson, 1991), and close to mean atmospheric concentrations—75 μBq m^{-3}—estimated by Samuelsson *et al.* (1986) around Svalbard archipelago. In fact, the meteorological conditions observed in April–May 2004, in particu-

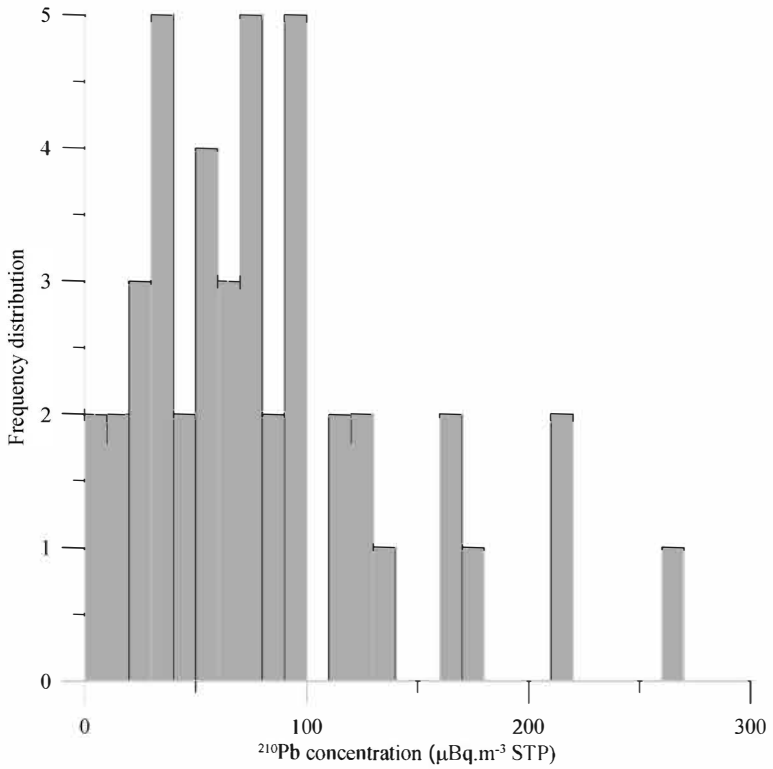

Fig. 4. Frequency distribution of daily ^{210}Pb activity concentrations in the air, in Ny-Ålesund, Svalbard from April 14th to May 7th 2004.

lar regarding previous years (warmer temperatures, early snow melting..) (Fig. 5), may induce that "spring" conditions could have been shifted by one to two months; this could explain the low observed values. In this particular case, the meteorological conditions normally occurred in summertime would have been established from April, with no return to spring conditions. However, another hypothesis, that we cannot exclude, could explain our data. The low concentrations could result from temporary exceptional meteorological conditions, only viable for the short period of our measurements. In this case, there would be a return to expected higher concentrations after this period. These particular conditions could temporary hide conditions that are more representative of atmospheric ^{210}Pb and ^{7}Be concentrations and composition in this area, at this period. We have to keep in mind that we are discussing a single period of observation at Ny-Ålesund, with no a priori assurances that this is a representative seasonal pattern of atmospheric ^{210}Pb and ^{7}Be concentrations and composition.

Moreover, as we can observed in Paatero *et al.* (2003) and Suzuki *et al.* (1996), there are considerable daily variations in the ^{210}Pb activity concentrations from one sample to another one. These important variations depict the episodic nature of ^{210}Pb activity concentrations in the ambient air due to the different associated air masses

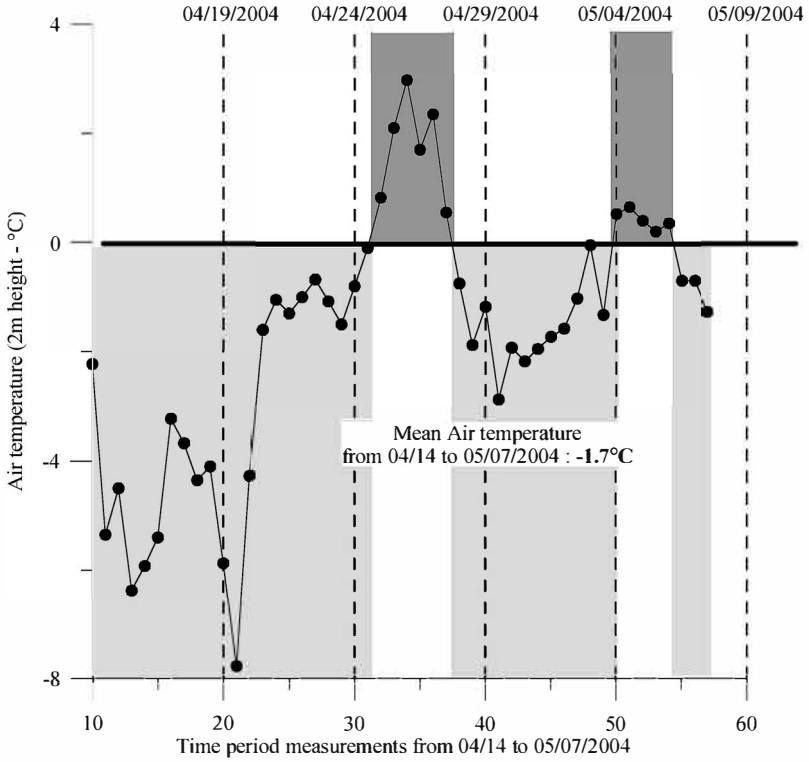

Fig. 5. Scatter plot showing the evolution of air temperature (2 m height, Koldewey Station AWI) in Ny-Ålesund, Svalbard from April 14th to May 7th 2004.

originating from different source regions. The relationship between atmospheric ^{210}Pb concentration and air pressure attests this hypothesis. According to correlation analysis, the ^{210}Pb atmospheric concentration and atmospheric pressure are positively correlated, with regression coefficient of 0.75 ([^{210}Pb Conc.$=4.642^{*}$ (Atmo. Pres.)$-$ 4647.643]; $n=43$) (Fig. 6). This may indicate that the ^{210}Pb atmospheric concentration increase as a function of air mass exchange over the Svalbard region, as described by Suzuki *et al.* (1996). In the same study, Suzuki *et al.* (1996) assert that rapid atmospheric transport, induced by high atmospheric pressure system, brings a large amount of "young" terrigenous aerosols from European continental areas to the atmosphere of Svalbard. However, we can observe that the highest ^{210}Pb concentration values measured during our studied period are below the values observed in air masses originating from free of ice, mid latitudinal European continental areas (Preiss *et al.*, 1996; Liu *et al.*, 2001; Paatero *et al.*, 2003). On the contrary, our values are quite close to the oceanic values (Samuelsson *et al.*, 1986; Preiss *et al.*, 1996). The meteorological parameters that are available, the wind direction in particular (Table 2), do not give information precise enough to determine the origin, oceanic or terrestrial, of the air masses fluxes over the Svalbard. According to three-days backward trajectories analysis, provided by NOAA ARL (2005), we observed that air masses ending to Ny-Ålesund

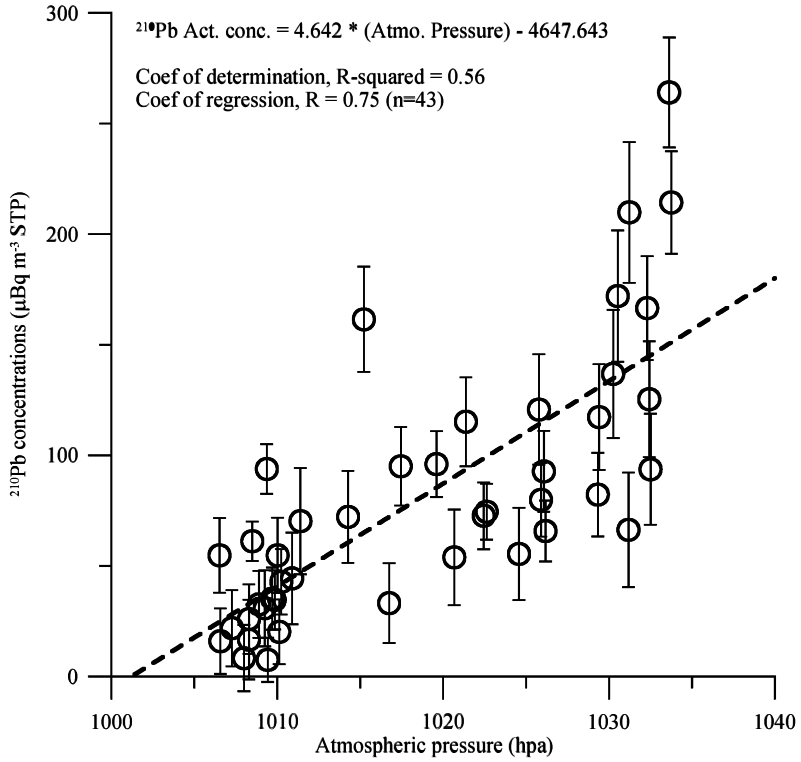

Fig. 6. Scatter plot showing the relationship between ^{210}Pb activity concentrations in the aerosol filter samples and atmospheric pressure in Ny-Ålesund, Svalbard from April 14th to May 7th 2004. Vertical bars in each plot indicate counting error (1σ). The regression coefficient is 0.75 (n=43).

have an oceanic origin, during our studied period. Figure 7 shows three backward trajectories ending in Ny-Ålesund on April 15th, 20th and 28th at 00:00 UTC, corresponding to the three main air masses arriving to Svalbard during the campaign. The second backward trajectory (Fig. 7b) shows the major marine air mass arriving to Ny-Ålesund, after remaining three days in Arctic ocean or Barents sea. We have to notice that this oceanic origin have represented more than 65% of the air masses over the Svalbard during our campaign. Although a non negligeable area of the glacial Arctic ocean is still frozen at this time of the considered year (April–May), air masses taking their origin above the ice shelf become enriched in marine aerosols along their journey; that is why they are called "marine air masses". The third backward trajectory (Fig. 7c) represents the minor marine air masses arriving to Ny-Ålesund. These air masses (15%) were coming from the Atlantic ocean, between Iceland and Feroe islands. There is no noticeable difference between the concentration values of ^{210}Pb measured in air masses coming from Atlantic or Arctic ocean. The only lack of ^{210}Pb source by marine areas can explain the low concentrations measured in air masses from the Atlantic ocean. Concerning air masses coming from Arctic ocean and Barents sea,

Table 2. Meteorological data from Ny-Ålesund Arctic site, Svalbard, from 14th of April to 7th of May 2004. Data are extracted from meteorological observatory program database, carried out by the Alfred Wegener Institute (AWI), at the Koldewey Station.

Measurements period (local time)	Wind direction (10m height) (degrees)	Wind speed (10m height) (m s^{-1})	Air pressure (to sea level) (hpa)	Air temperature (2m height) (° Celsius)	Present weather
22h00 – 04/14/04	158	5	1021.4	-5.4	
22h00 – 04/15/04	120	6	1019.6	-6.4	
10h00 – 04/16/04	108	14	1015.2	-5.9	Drifting snow
22h00 – 04/16/04	110	28	1009.4	-5.4	Drifting snow
10h00 – 04/17/04	108	23	1011.4	-3.2	Drifting snow
22h00 – 04/17/04	108	20	1017.5	-3.7	Drifting snow
10h00 – 04/18/04	100	13	1022.5	-4.4	
22h00 – 04/18/04	180	3	1026.1	-4.1	
10h00 – 04/19/04	268	4	1029.3	-5.9	
22h00 – 04/19/04	173	2	1032.3	-7.8	
08h30 – 04/20/04	123	3	1033.8	-4.3	Continuous snowflakes
22h00 – 04/20/04	205	4	1033.6	-1.6	Intermittent snowflakes
10h00 – 04/21/04	140	2	1032.4	-1.1	
22h00 – 04/21/04	133	3	1030.5	-1.3	
10h00 – 04/22/04	150	2	1030.3	-1.0	
22h00 – 04/22/04	188	5	1031.2	-0.7	Intermittent snowflakes
10h00 – 04/23/04	240	6	1032.5	-1.1	Intermittent snowflakes
22h00 – 04/23/04	165	4	1031.2	-1.5	Intermittent snowflakes
10h00 – 04/24/04	85	3	1029.4	-0.8	Intermittent snowflakes
22h00 – 04/24/04	173	4	1025.8	-0.1	
10h00 – 04/25/04	93	3	1020.7	+0.8	
22h00 – 04/25/04	113	8	1014.3	+2.1	Rain, not freezing
10h00 – 04/26/04	225	7	1010.7	+3.0	Rain, not freezing
22h00 – 04/26/04	190	2	1008.3	+1.7	Rain, shower
10h00 – 04/27/04	268	6	1007.3	+2.4	Drizzle
22h00 – 04/27/04	190	4	1008.5	+0.6	Intermittent snowflakes
10h00 – 04/28/04	143	3	1009.4	-0.8	Snow shower, heavy
22h00 – 04/28/04	118	4	1009.9	-1.9	Snow shower, heavy
10h00 – 04/29/04	193	2	1010.0	-1.2	Snow crystals
22h00 – 04/29/04	110	4	1008.9	-2.9	Continuous snowflakes
10h00 – 04/30/04	298	3	1008.5	-1.9	
22h00 – 04/30/04	258	2	1009.3	-2.2	Snow crystals
10h00 – 05/01/04	218	1	1010.3	-2.0	Continuous snowflakes
22h00 – 05/01/04	138	4	1010.1	-1.7	
10h00 – 05/02/04	155	4	1009.7	-1.6	
22h00 – 05/02/04	153	6	1008.0	-1.0	
10h00 – 05/03/04	105	11	1006.5	-0.1	
22h00 – 05/03/04	145	11	1006.6	-1.3	
10h00 – 05/04/04	133	10	1008.3	+0.5	
22h00 – 05/04/04	173	11	1010.9	+0.7	Continuous snowflakes
10h00 – 05/05/04	308	1	1016.8	+0.4	Intermittent snowflakes
22h00 – 05/05/04	248	2	1022.6	+0.2	
10h00 – 05/06/04	175	3	1025.9	+0.4	
22h00 – 05/06/04	325	2	1026.2	-0.7	Continuous snowflakes
10h00 – 05/07/04	198	1	1024.6	-0.7	

another phenomenon occurs. In springtime, the longer daylight duration induces an increase of vertical mixing in the atmosphere column. Particle aerosols carrying ^{210}Pb atoms, which were accumulated in the surface air during wintertime, are then transported into upper air and thus, lowering surface air concentrations. This process is discussed by Paatero and Hatakka (2000) in Northern Finland. The first backward trajectory (Fig. 7a) shows air masses which had a terrestrial origin. These air masses

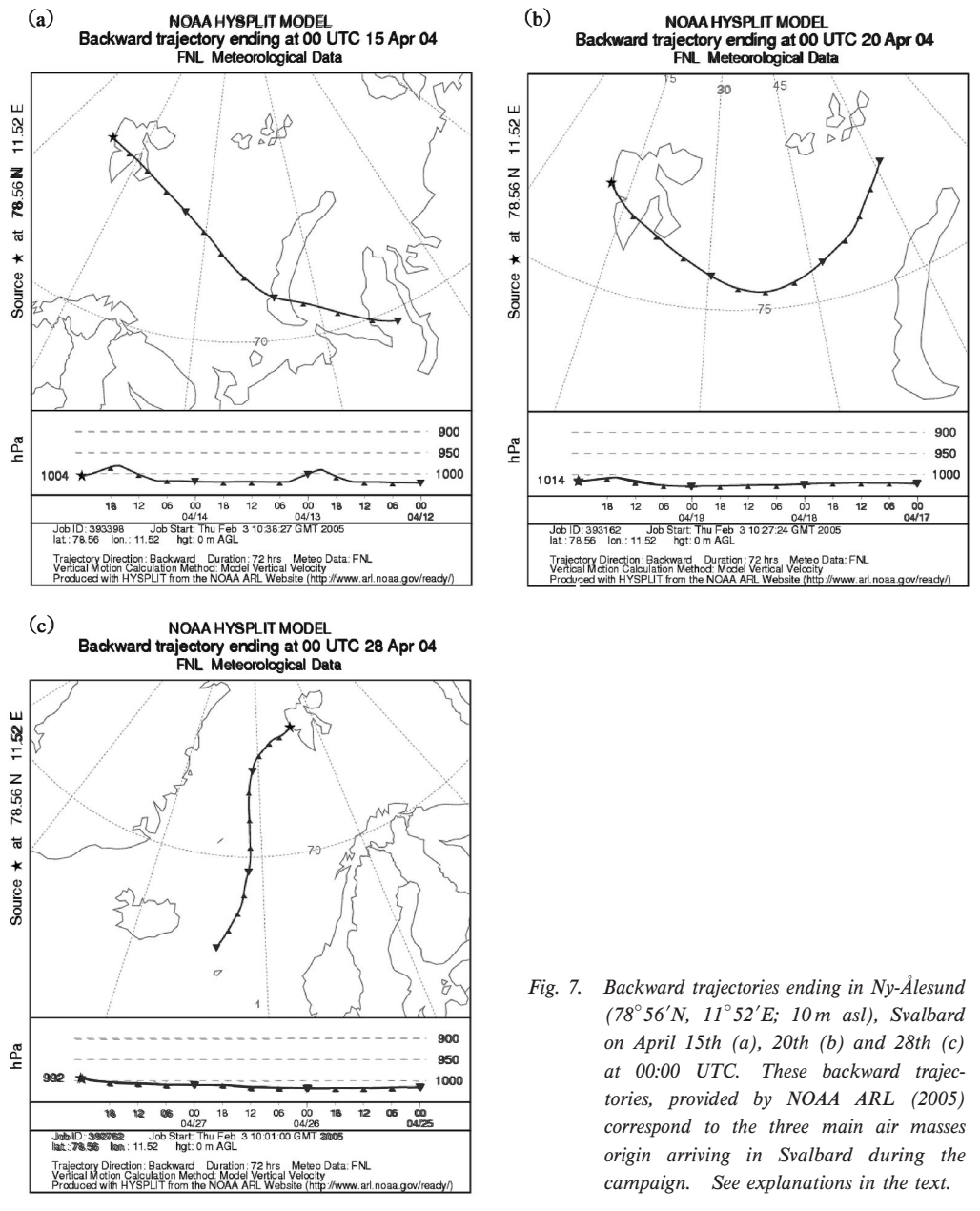

Fig. 7. Backward trajectories ending in Ny-Ålesund (78°56′N, 11°52′E; 10 m asl), Svalbard on April 15th (a), 20th (b) and 28th (c) at 00:00 UTC. These backward trajectories, provided by NOAA ARL (2005) correspond to the three main air masses origin arriving in Svalbard during the campaign. See explanations in the text.

passed above the Barents sea and obviously remained about two days at sea level. These last air masses come from Russia and Finland, *i.e.*, continental zones which are still frozen at this time of the year, thus being depleted in ^{222}Rn. This explains the low ^{210}Pb concentration values observed in these air masses. We have to keep in mind that variations could also be observed even if the air masses originate from same ^{210}Pb source regions in function of scavenging processes occuring during the transport.

Beryllium-7—As shown in Fig. 3, there are considerable variations in the daily ^{7}Be activity concentrations, during our measurement time period. Results give an atmospheric ^{7}Be mean concentration and a standard deviation ($\pm 1\sigma$) of $402 \pm 234\,\mu$Bq m^{-3} ($n=29$) ranging from 35 to $824\,\mu$Bq m^{-3}. The median value of the whole dataset is about $404\,\mu$Bq m^{-3} ($n=29$), *i.e.* close to the mean values, which implies a Gaussian distribution of data. We have also to keep in mind that mean and median concentrations values previously cited are only calculated in samples in which ^{7}Be has been detected. In the frame of this study, this radioelement is not detected in more than one third of the atmospheric samples, *i.e.*, from April 27th and May 2nd included (*i.e.*, 6 successive days), as well as four other filters sampled at the end of the field campaign. Number of samples below the detection limit are 16 (Table 1). We have to keep in mind that the non detection of ^{7}Be in these samples does not necessarily mean a complete depletion of the element in the sampled atmospheres. A sharp temporary decrease of ^{7}Be atmospheric concentrations due to variations of environmental and meteorological parameters, coupled to analytical detection limits can explain this phenomenon. It has been shown that the surface air concentration of ^{7}Be and its variability depend on several main factors and/or processes which are, latitude, season of sampling, altitude, wet scavenging, atmospheric exchanges and solar activity (Feely *et al*., 1981, 1989; Brost *et al*., 1991; Zanis *et al*., 1999; Baskaran and Shaw, 2001; Salisbury and Cartwright, 2005). The seasonal variations observed are then a complicated function of all these processes, since each of them can have its own seasonal or annual cycle (Brost *et al*., 1991). However, among the previously described parameters, the removal by the precipitation and the exchange of air masses represent the two most probable hypothesis that could explain the potential depletion of ^{7}Be (Zanis *et al*., 1999; Liu *et al*., 2001). Several authors (Brost *et al*., 1991; Baskaran *et al*., 1993; Koch *et al*., 1996; Zanis *et al*., 1999) show measurements and model results suggesting that wet deposition (*i.e.*, wet scavenging) accounts for more than 90% of the total deposition on the ground (Brost *et al*., 1991). Indeed, high relative humidity is associated with a higher condensation potential and consequently, a high wet scavenging rate of aerosols carrying ^{7}Be atoms. Therefore, it results low ^{7}Be activity concentrations in the air. The more the height of the tropospheric column of the wet deposition is, the more the decrease of the concentration of ^{7}Be—and ^{210}Pb—associated to aerosols, in surface air, will be great. Dibb and Jaffrezo (1993) show the influence of atmospheric exchanges and in particular of troposphere/boundary layer exchanges, on the variation of radionuclide concentration in surface air. In the case of this study, the abundant snow and rain precipitations, registered between April 26th and May 2nd—alternation between continuous heavy and moderate snowflakes and rain—can thus have been sufficient enough to lead to an efficient scavenging of aerosols in the lower troposphere column, and as a consequence,

a sharp drop of the radionuclide concentrations (Fig. 3). As a consequence, the filtered air volumes for each sample (~500 m^3/filter) were too low to detect subsisting ^{7}Be atoms in the lower tropospheric layer, through the employed device system of gamma counting. The significant decrease of atmospheric ^{210}Pb contents, during the same period of time, seems to confirm the hypothesis of the wet scavenging action on the evolution of natural radioactivity concentration (Fig. 3 and Table 1). The lower moisture content of the air masses previously centered above the study area the days before, so the lack of precipitation may prevent the removal of ^{7}Be and ^{210}Pb by wet deposition.

There is currently no database concerning surface atmospheric ^{7}Be concentrations over the Svalbard area. Nevertheless, few similar studies have been made in other areas of the Arctic zone (Dibb and Jaffrezo, 1993; Baskaran and Shaw, 2001). The mean ^{7}Be value, observed during our campaign, appears to be systematically below (5 to 10 times lower) the monthly mean concentrations measured over Greenland (Nord, Thule, Dye 3, Summit) and Alaska (Point Barrow) for the same period of time (Dibb and Jaffrezo, 1993). One has to be very cautious on the validity of the sites comparisons. Indeed, there are complex interactions between the origin of the radionuclide source areas over the considered zones and the small and large scale atmospheric circulation pattern,

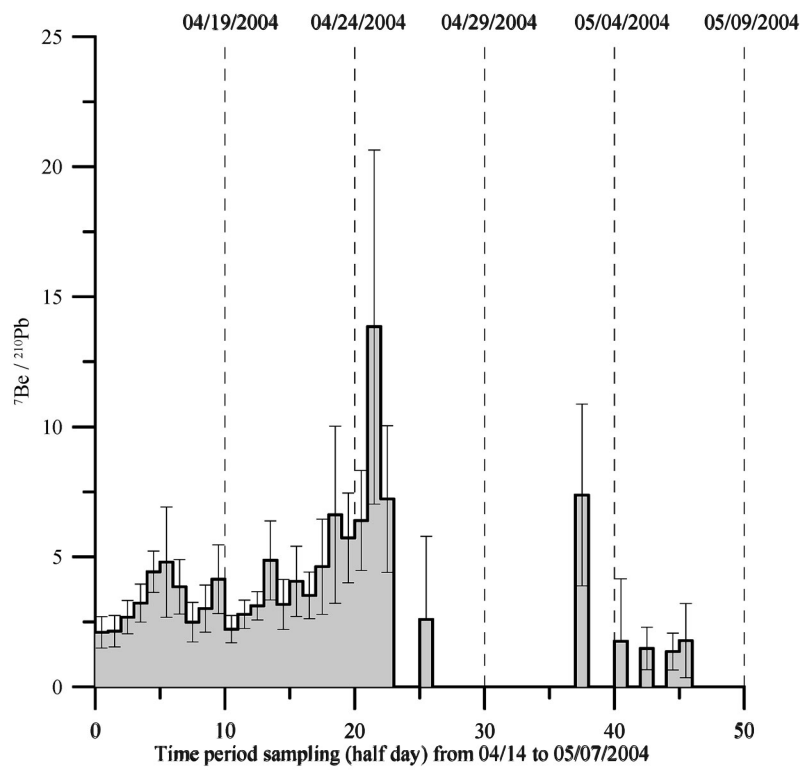

Fig. 8. Daily variation of atmospheric ^{7}Be/^{210}Pb ratio, in Ny-Ålesund, Svalbard from April 14th to May 7th 2004. Vertical bars in each plot indicate standard deviation (1σ).

during the period of study. An annual monitoring of ^{7}Be concentrations during future field campaigns, should help to better understand the parameters and processes that are responsible of the strong observed variabilities. More extensive database in Svalbard will enable us to address the issues on the sources and transport of Arctic aerosols.

^{7}Be/^{210}Pb ratio and ozone data—The study of the ^{7}Be/^{210}Pb ratio evolution coupled to the fluctuations of ^{7}Be concentration in the surface atmosphere, can allow to detect variations in the rate of transfer and exchange through the tropopause, and, as a consequence, the potential injection of stratospheric air masses into the troposphere will be found. The ^{7}Be/^{210}Pb activity ratio varied between a factor of 1.4 to 13.8 (mean 4.12±2.6) (Fig. 8). The order of magnitude of the mean ratio is in good agreement with those observed in Arctic region such as Greenland (Kap Tobin station) or Alaska (Poker flat and Eagle stations) and with those calculated, by simulation, using a chemical tracer model (Koch *et al*., 1996; Baskaran and Shaw, 2001). The sharp increase of the ratio (up to 13.8) from April 23rd to 25th 2004 could imply a vertical transport with injection of stratospheric air masses enriched in ^{7}Be, into the tropospheric

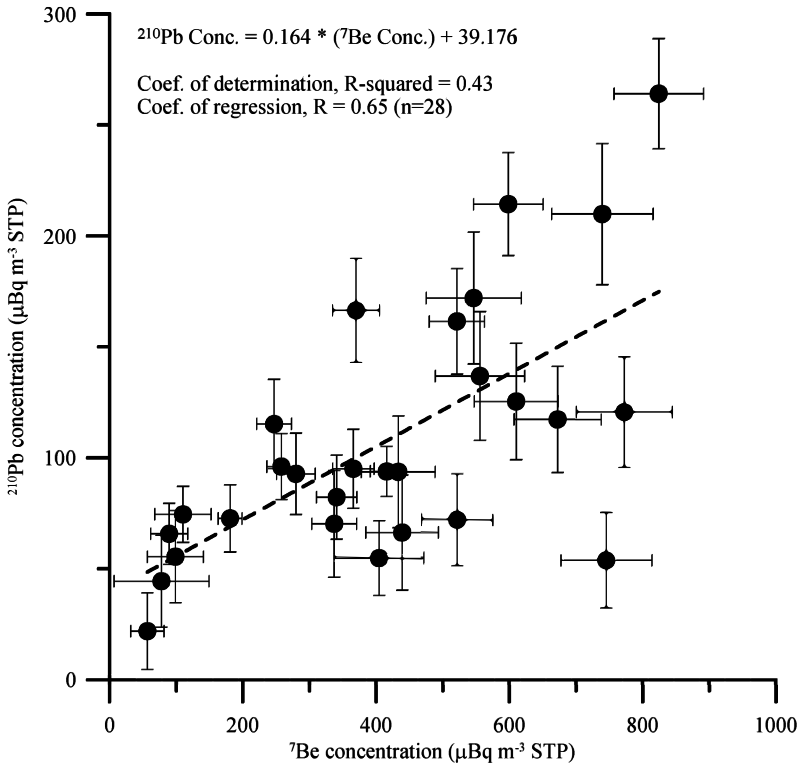

Fig. 9. Scatter plot showing the relationship between ^{210}Pb and ^{7}Be in the aerosol filter samples collected in Ny-Ålesund, Svalbard from April 14th to May 7th 2004. Vertical and horizontal bars in each plot indicate counting error (1σ). The regression coefficient is 0.65 (n=28).

air surface. However, the relative ozone stability in the surface atmosphere, through this period of time, seems to exclude this last hypothesis. Lead-210 concentrations alone evolve, with an observation of lower values (Fig. 3 and Table 1). The decrease, even small, of ^{210}Pb concentration is sufficient enough to considerably increase the ratio. So, the sharp increase of the ratio may be an artefact linked to the variability of studied radionuclides concentration values. The three-days air masses backward trajectories shows that the whole air masses, independently of their origin (marine or terrestrial), have circulated at sea level, without any apparent mixing with upper atmospheric layers. It must be reiterated that we are discussing about the potential influence of a stratospheric intrusion on lower atmospheric layers of the troposphere. We cannot draw any conclusions on stratospheric intrusions that may have happened in the upper atmospheric layers.

We have to notice that the surface concentrations of ^{7}Be plotted against ^{210}Pb in all the aerosols filter samples (Fig. 9) show a positive correlation between these two radionuclides ($r=0.65$, $n=28$). This fact implies that these two radionuclides have a relatively similar atmospheric removal behavior, despite different source origins. From similar data, many authors have shown that in most of the continental and coastal stations, as in our case, these two radionuclides cannot be simply used as two independent air mass tracers (Todd *et al*., 1989; Baskaran *et al*., 1993). This example shows how difficult it is to use ^{7}Be and ^{210}Pb radionuclides, as well as their ratio, as atmospheric tracer tools in the case of this study.

4. Conclusion

This paper summarized preliminary data series of half daily observations of airborne ^{210}Pb and ^{7}Be, in Ny-Ålesund, from April 24th to May 7th 2004. Although one has to be cautious regarding the comparisons of the data, ^{7}Be and ^{210}Pb concentrations values measured during our studied period are relatively low compared to those measured in other Arctic sites for the same period time (April–May) during previous studies. Lead-210 activities are close to the one measured above the ocean and Arctic continental zones during summer season (Samuelsson *et al*., 1986; Dibb and Jaffrezo, 1993; Paatero *et al*., 2003). Beryllium-7 activity concentrations, measured during our campaign, appears to be systematically 5 to 10 times lower than those observed in other Arctic sites (Greenland and Alaska) for the same period of time (Dibb and Jaffrezo, 1993). We have also to keep in mind that, as Paatero *et al*. (1998, 2003) observed for ^{210}Pb, there were considerable daily variations in natural radionuclide activity concentrations during measurements period. This remark is particularly valuable for ^{7}Be activities. These day-to-day variations are probably due to local synoptic-scale weather situations, as the effect of wet scavenging induced by snow and rain precipitation events. This last hypothesis suggests a slight tendency for higher ^{7}Be and ^{210}Pb concentrations to occur at the surface of snow mantle in the same time of wet scavenging in the air. A more detailed study of the flux deposits of these two radionuclide at the surface of the snow mantle, during these precipitation events, should allow to check this hypothesis. Part of the results, —^{7}Be and ozone concentrations—, show also the absence of direct ^{7}Be stratospheric injections during our sampling campaign, in the low troposheric air

masses. It is also confirmed by three-day backward trajectories of air masses reaching Svalbard during the field campaign. We can also notice that the three referenced air masses do not show considerable differences in ^{210}Pb and ^{7}Be concentrations values, despite their different origins.

It must be reiterated that we are discussing a short record period of ^{7}Be and ^{210}Pb measurements at Ny-Ålesund, with no a priori assurances that this is a representative seasonal pattern of atmospheric ^{7}Be and ^{210}Pb concentrations and composition. Some authors considered (Zanis *et al*., 1999) that three years of measurements at least are necessary to provide a robust seasonal natural radionuclide cycle. Nevertheless, we can wonder: are these preliminary results sufficient to hypothesize that year 2004 was an exceptional year with the establishment of summer meteorological conditions with a shift of one or two months? What are the real impact of both the particular local meteorological conditions that have occurred during the campaign and the seasonal variations occurring on longer temporal and spatial scales? Further studies with longer monitoring of atmospheric radionuclide composition, and also aerosol chemical composition along the year are planned to answer this question. It should help to better understand the parameters and processes that are responsible of the strong radionuclide concentration variabilities, and also better determine the sources and transport processes of Arctic aerosols in Svalbard.

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