Supplementary material to: "Cryoconite as an efficient monitor for the deposition of radioactive fallout in glacial environments"

Giovanni Baccolo[1,2], Edyta Łokas[3], Paweł Gaca[4], Dario Massabò[5,6], Roberto Ambrosini[7], Roberto S. Azzoni[7], Caroline Clason[8], Biagio Di Mauro[1], Andrea Franzetti[1], Massimiliano Nastasi[1,9], Michele Prata[10], Paolo Prati[5,6], Ezio Previtali[1,9], Barbara Delmonte[1], Valter Maggi[1,2]

[1] Environmental and Earth Sciences Department, University of Milano-Bicocca, Milano, 20126, Italy.
[2] INFN section of Milano-Bicocca, Milano, 20126, Italy.
[3] Department of Nuclear Physical Chemistry, Institute of Nuclear Physics Polish Academy of Sciences, Kraków, 31-342, Poland.
[4] Ocean and Earth Science, University of Southampton, National Oceanography Centre, Southampton, SO14 3ZH UK.
[5] Physics Department, University of Genoa, Genoa, 16146, Italy.
[6] INFN section of Genoa, Genoa, 16146, Italy.
[7] Department of Environmental Science and Policy, University of Milan, Milano, 20133, Italy.
[8] School of Geography, Earth and Environmental Sciences, University of Plymouth, Plymouth, PL48AA, UK.
[9] Physics Department, University of Milano-Bicocca, Milano, 20126, Italy.
[10] Laboratory of Applied Nuclear Energy, University of Pavia, Pavia, 27100, Italy.

Correspondence to: G. Baccolo (giovanni.baccolo@unimib.it)

In the following list the references whose data were used to create Fig. 4, are reported. Studies concerning sites where nuclear explosion tests and accidents occurred, were not taken into account. In addition to the listed studies, also the following references cited in the main text were used: Aarkrog and Dahlgaard, 1984; Kim et al., 1997, Kirchner et al., 2002.

- Aleksiayenak, Y. V., Frontasyeva, M. V., Florek, M., Sykora, I., Holy, K., Masarik, J., Brestakova, L., Jeskovsky, M., Steinnes, E., Faanhof, A., Ramatlhape, K. I.: Distributions of 137Cs and 210Pb in moss collected from Belarus and Slovakia. J. Environ. Radioactiv. 117, 19-24, 2013.
- Ali, A. A., Ghaleb, B., Garneau, M., Asnong, H., Loisel, J.: Recent peat accumulation rates in minerotrophic peatlands of the Bay James region, Eastern Canada, inferred by 210Pb and 137Cs radiometric techniques. Appl. Radiat. Isotopes 66, 1350-1358, 2008.
- Al Hamarneh, I., Wreikat, A., Toukan, K.: Radioactivity concentrations of 40K, 134Cs, 137Cs, 90Sr, 241Am, 238Pu and 239+ 240Pu radionuclides in Jordanian soil samples. J. Environ. Radioactiv. 67, 53-67, 2003.
- Baskaran, M., Asbill, S., Schwantes, J., Santschi, P., Champ, M. A., Brooks, J. M., Adkinson, D., Makeyev, V.: Concentrations of 137Cs, 239,240Pu and 210Pb in Sediment Samples from the Pechora Sea and Biological Samples from the Ob, Yenisey Rivers and Kara Sea. Mar. Pollut. Bull. 40, 830-838, 2000.

- Belivermiş, M. and Çotuk, Y.: Radioactivity measurements in moss (*Hypnum cupressiforme*) and lichen (*Cladonia rangiformis*) samples collected from Marmara region of Turkey. J. Environ. Radioactiv. 101, 945-951, 2010.
- Belivermiş, M., Kiliç, Ö., Çayir, A., Coşkun, M., Coşkun, M.: Assessment of 210Po and 210Pb in lichen, moss and soil around Çan coal-fired power plant, Turkey. J. Radional. Nucl. Chem. 307, 523-531, 2016.
- Breban, D. C., Moreno, J., Mocanu, N.: Activities of Pu radionuclides and 241Am in soil samples from an alpine pasture in Romania. J. Radional. Nucl. Chem. 258, 613-617, 2003.
- Caridi, F., Belvedere, A., D'Agostino, M., Marguccio, S.: 137Cs activity concentration in mosses in the Calabria region, south of Italy. J. Instrum. 12, DOI: 10.1088/1748-0221/12/05/P05001, 2017.
- Duffa, C., Renaud, P., Goutelard, F.: Activities and transfers of Pu and Am in rice samples from Camargue, France. J. Radioanal. Nucl. Chem. 252, 247-248, 2002.
- Ekdal, E., Karali, T., Saç, M. M.: 210Po and 210Pb in soils and vegetables in Kucuk Menderes basin of Turkey. Radiat. Meas. 41, 72-77, 2006.
- Figueira, R. C. L., Tessler, M. G., de Mahiques, M. M., Cunha, I. I. L.: Distribution of 137Cs, 238Pu and 239 + 240Pu in sediments of the southeastern Brazilian shelf–SW Atlantic margin. Sci. Total Environ. 357, 146-159, 2006.
- Gascò, C., Antón, M. P., Pozuelo, M., Meral, J., Gonzáles, A. M., Papucci, C., Delfanti, R.: Distributions of Pu, Am and Cs in margin sediments from the western Mediterranean (Spanish coast). J. Environ. Radioactiv. 59, 75-89, 2002.
- He, Q. and Dwelling, D. E.: The distribution of fallout 137Cs and 210Pb in undisturbed and cultivated soil. Appl. Radiat. Isotopes 48, 677-690, 1997.
- Heldal, H. E., Varskog, P., Føyn, L.: Distribution of selected anthropogenic radionuclides (137Cs, 238Pu, 239,240Pu and 241Am) in marine sediments with emphasis on the Spitsbergen–Bear Island area. Sci. Total Environ. 293, 233-245, 2002.
- Huh, C. A. and Su, C. C.: Distribution of fallout radionuclides (7Be, 137Cs, 210Pb and 239,240Pu) in soils of Taiwan. J. Environ. Radioactiv. 77, 78-100, 2004.
- Ishikawa, Y., Sato, N., Yoshihara, K.: 207Bi and other fallout nuclides in sea sediments in relation to ignition loss of samples. J. Radioanal. Nucl. Chem. 137, 67-74, 1989.
- Jenkins, C. E., Wogman, N. A., Rieck, H. G.: Radionuclide distribution in olympic national park, Washington. Water Air Soil Poll. 1, 181-204, 1972.

- Jia, G., Triulzi, C., Nonnis Marzano, F., Belli, M., Vaghi, M.: The fate of plutonium, 241Am, 90Sr and 137Cs in the Antarctic ecosystem. Antarctic Science 12, 141-148, 2000.
- Joshi, S. and McNeely, R.: Detection of fallout 155Eu and 207Bi in a 210Pb-dated lake sediment core. J. Radioanal. Nucl. Chem. 122, 183-191, 1988.
- Kahraman, A., Kaynak, G., Akkaya, G., Gürler, O., Yalçın, S.: Radioactivity measurements in epiphytic lichens of Uludağ Mountain in Western Anatolia. J. Radional. Nucl. Chem. 295, 1057-1066, 2013.
- Ketterer, M. E., Hafer, K. M., Mietelski, J. W.: Resolving Chernobyl vs. global fallout contributions in soils from Poland using Plutonium atom ratios measured by inductively coupled plasma mass spectrometry. J. Environ. Radioactiv. 73, 183-201, 2004.
- Komura, K.: Bismuth-207 in environmental samples. Radioisotopes 34, 555-558, 1985.
- Krmar, M., Wattanavatee, K., Radnović, D., Slivka, J., Bhongsuwan, T., Frontasyeva, M. V., Pavlov, S. S.: Airborne radionuclides in mosses collected at different latitudes. J. Environ. Radioactiv. 117, 45-48, 2013.
- LaBrecque, J. J. and Cordoves, P. R.: Determination and spatial distribution of 137Cs in soils, mosses and lichens near Kavanayen, Venezuela. J. Radional. Nucl. Chem. 273, 401-404, 2007.
- Lee, M. H. and Lee, C. W.: Association of fallout -derived 137Cs, 90Sr and 239,240Pu with natural organic substances in soils. J. Environ. Radioactiv. 47, 253-262, 2000.
- Lee, M. H., Lee, C. W., Boo, B. H.: Distribution and characteristics of 239,240Pu and 137Cs in the soil of Korea. J. Environ. Radioactiv. 37, 1-16, 1997.
- Lee, S. H., Povinec, P. P., Wyse, E., Pham, M. K., Hong, G. H., Chung, C. S., Kim, S. H., Lee, H. J.: Distribution and inventories of 90Sr, 137Cs, 241Am and Pu isotopes in sediments of the Northwest Pacific Ocean. Mar. Geol. 216, 249-263, 2005.
- Lee, S. H., Oh, J. S., Lee, J. M., Lee, K. B., Park, T. S., Lujaniene, G., Valiulis, D., Śakalis, J.: Distribution characteristics of 137Cs, Pu isotopes and 241Am in soil in Korea. Appl. Radiat. Isotopes 81, 315-320, 2013.
- Li, C. X., Le Roux, G., Sonke, J., van Beek, P., Souhaut, M., Van der Putten, N., De Vleeschouwer, F.: Recent 210Pb, 137Cs and 241Am accumulation in an ombrotrophic peatland from Amsterdam Island (Southern Indian Ocean). J. Environ. Radioactiv. 175-176, 164-169, 2017.
- Lindhal, P., Roos, P., Eriksson, M., Holm, E.: Distribution of Np and Pu in Swedish lichen samples (Cladonia stellaris) contaminated by atmospheric fallout. J. Environ. Radioactiv. 73, 73-85, 2004.
- Łokas, E., Mietelski, J. W., Ketterer, M. E., Klesczc, K., Wachniew, P., Michalska, S., Miecznik, M.: Sources and vertical distribution of 137Cs, 238Pu, 239+240Pu and 241Am in peat profiles from southwest Spitsbergen. Appl. Radiati. Isotopes 28, 100-108, 2013.

- Łokas, E., Bartmiński, P., Wachniew, P., Mietelski, J. W., Kawiak, T., Środoń, J.: Sources and pathways of artificial radionuclides to soils at a High Arctic site. Environ. Sci. Pollut. R. 21, 12479-12493, 2014.
- Machart, P., Hoffman, W., Türk, R., Steger,F.: Ecological half-life of 137Cs in lichens in an alpine region. J. Environ. Radioactiv. 97, 70-75, 2007.
- Mietelski, J. W., Olech, M. A., Sobiech-Matura, K., Howard, B. J., Gaca, P., Zwolak, M., Błażej, S., Tomankiewicz, E.: 137Cs, 40K, 238Pu, 239+240Pu and 90Sr in biological samples from King George Island (Southern Shetlands) in Antarctica. J. Environ. Radioactiv. 31, 1081-1089, 2008.
- Mietelski, J. W., Gaca, P., Olech, M.: Radioactive contamination of lichens and mosses collected in South Shetlands and Antarctic Peninsula. J. Radional. Nucl. Chem. 245, 527-537, 2000.
- Mietelski, J. W., Kubica, B., Gaca, P., Tomankievicz, E., Błaźej, S., Tuteja-Krysa, M., Stobiński, M.: 238Pu, 239+240Pu, 241Am, 90Sr and 137Cs in mountain soil samples from the Tatra National Park (Poland). J. Radioanal. Nucl. Chem. 275, 523-533, 2008.
- Mróz, T., Łokas, E., Kocurek, J., Gąsiorek, M.: Atmospheric fallout radionuclides in peatland from Southern Poland. J. Environ. Radioactiv. 175-176, 25-33, 2017.
- Navas, A., Soto, J., Machìn, J.: 238U, 226Ra, 210Pb, 232Th and 40K activities in soil profiles of the Flysch sector (central Spanish Pyrenees). Appl. Radiat. Isotopes 57, 579-589, 2002.
- Outula, I.: Effect of industrial pollution on the distribution of Pu and Am in soil and on soil-to-plant transfer of Pu and Am in a pine forest in SW Finland. J. Radioanal. Nucl. Chem. 257, 267-274, 2003.
- Popov, L., Mihailova, G., Naidenov, I.: Detemination of activity ratios of 238,239,240Pu, 241Am, 134,137Cs, and 90Sr in Bulgarian soils. J. Radioanal. Nucl. Chem. 285, 223-237, 2010.
- Quank, N. H., Long, N. Q., Lieu, D. B., Mai, T. T., Ha, N. T., Nhan, D. D., Hien, P. D.: 239+240Pu, 90Sr and 137Cs inventories in surface soils of Vietnam. J. Environ. Radioactiv. 75, 329-337, 2004.
- Rezzoug, S., Michel, H., Fernex, F., Barci-Funel, G., Barci, V.: Evaluation of 137Cs fallout from the Chernobyl accident in a forest soil and its impact on Alpine Lake sediments, Mercantour Massif, S.E. France. J. Environ. Radioactiv. 85, 369-379, 2006.
- Schertz, M., Michel, H., Barci-Funel, G., Barci, V.: Transuranic and fission product contamination in lake sediments from an alpine wetland, Boréon (France). J. Environ. Radioactiv. 85, 380-388, 2006.
- Schuller, P., Voigt, G., Handl, J., Ellies, A., Oliva, L.: Global weapons' fallout 137Cs in soils and transfer to vegetation in south–central Chile. J. Environ. Radioactiv. 62, 181-193, 2002.
- Shabana, E. I. and Al-Shammari, H. L.: Assessment of the global fallout of plutonium isotopes and americium-241 in the soil of the central region of Saudi Arabia. J. Environ. Radioactiv. 57, 67-74, 2001.

- Skuterud, L., Gwynn, J. P., Gaare, E., Steinnes, E., Hove, K.: 90Sr, 210Po and 210Pb in lichen and reindeer in Norway. J. Environ. Radioactiv. 84, 441-456, 2005.
- Testa, C., Jia, G., Degetto, S., Desideri, D., Guerra, F., Meli, M. A., Rosselli, C.: Vertical profiles of 239,240Pu and 241Am in two sphagnum mosses of Italian peat. Sci. Total Environ. 232, 27-31, 1999.
- Uğur, A., Özden, B., Saç, M. M., Yener, G.: Biomonitoring of 210Po and 210Pb using lichens and mosses around a uraniferous coal-fired power plant in western Turkey. Atmos. Environ. 37, 2237-2245, 2003.
- Wattanawatee, K., Krmar, M., Bhogsuwan, T.: A survey of natural terrestrial and airborne radionuclides in moss samples from the peninsular Thailand. J. Environ. Radioactiv. 177, 113-127, 2017.

	Morteratsch	Forni
^{137}Cs (Bq kg^{-1})	2,650 ± 3,800	2,000 ±2,800
^{207}Bi (Bq kg^{-1})	9.4 ± 6.6	5.7 ± 2.4
^{238}Pu (Bq kg^{-1})	2.6 ± 2.5	0.22 ± 0.08
^{239,240}Pu (Bq kg^{-1})	78 ± 77	4.9 ± 0.9
^{241}Am (Bq kg^{-1})	30 ± 36	4.4 ± 1.6
^{40}K (Bq kg^{-1})	810 ± 55	750 ± 200
^{238}U (Bq kg^{-1})	68 ± 14	61 ± 22
^{234}Th (Bq kg^{-1})	88 ± 15	65 ± 17
^{214}Pb (Bq kg^{-1})	57 ± 7	44 ± 11
^{214}Bi (Bq kg^{-1})	57 ± 8	45 ± 12
Supp. ^{210}Pb (Bq kg^{-1})	69 ± 6	55 ± 17
Unsupp. ^{210}Pb (Bq kg^{-1})	2,700 ± 750	6,100 ± 1,850
^{232}Th (Bq kg^{-1})	65 ± 9	72 ± 6
^{228}Ac (Bq kg^{-1})	47 ± 5	53 ± 12
^{224}Ra (Bq kg^{-1})	34 ± 12	38 ± 17
^{212}Pb (Bq kg^{-1})	50 ± 3	54 ± 12
^{212}Bi (Bq kg^{-1})	52 ± 7	60 ± 15
^{208}Tl (Bq kg^{-1})	49 ± 5	50 ± 12
Organic Carbon (m/m %)	4.7 ± 0.8	3.6 ± 0.5
Elemental Carbon (m/m %)	0.49 ± 0.25	0.21 ± 0.12

Tab. S1 Average composition of cryoconite from the Morteratsch and Forni glaciers. Average data (and standard deviations) concerning the activity concentration of radionuclides and of carbonaceous matter are reported with respect to the two glaciers considered in this study.

Decay Chain	Radionucl.	$T_{1/2}$	γ-line energy (keV)	Eff. · B.R. (%)	Minimum Detectable Activity (Bq kg^{-1})	Average Uncertainty (%)
^{238}U – natural	^{210}Pb	22.3 yr	46.5	2.8	17.3	5.9
none – artificial	^{241}Am	432.2 yr	59.5	28.2	1.7	13
^{238}U – natural	^{234}Th	24.1 d	92.3-92.8	2.8	17.7	13
^{232}Th – natural	^{212}Pb	10.64 hr	238.6	24.6	1.9	7.8
^{232}Th – natural	^{224}Ra	3.66	241.0*	25.8	21.1	35
^{238}U – natural	^{214}Pb	26.8 min	295.2	9.5	4.7	12
^{232}Th – natural	^{228}Ac	6.15 hr	338.3	4.2	11.5	17
^{238}U – natural	^{214}Pb	26.8 min	351.9	15.0	3.1	9.1
^{232}Th – natural	^{208}Tl	3.05 min	583.2	3.2	7.4	16
^{238}U – natural	^{214}Bi	19.9 min	609.3	5.2	4.6	13
none – artificial	^{137}Cs	30.07 yr	661.7	20.4	0.7	5.5
^{232}Th – natural	^{212}Bi	60.55 min	727.3	1.1	11.2	27
^{232}Th – natural	^{208}Tl	3.05 min	860.6	0.6	17.1	39
^{232}Th – natural	^{228}Ac	6.15 hr	911.2	3.7	2.9	13
none – artificial	^{207}Bi	31.55 yr	1063.7	4.5	2.6	32
none – natural	^{40}K	$1.3 \cdot 10^9$ yr	1460.8	1.3	6.6	7.4
^{238}U – natural	^{214}Bi	19.9 min	1764.5	1.6	4.4	15
^{232}Th – natural	^{208}Tl	3.05 min	2614.5	0.7	7.9	28

Tab. S2 Details about γ-spectrometry. For each of the analysed nuclides the relevant analytical information is reported. B.R. corresponds to branching ratio. For the emission at 241 keV from 224Ra (marked by an asterisk), a correction was needed to remove an interfering contribution from 214Pb.