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Anthropogenic radioactivity assessment of the Adamello Glacier ice core

Introduction

Glaciers are a key point in understanding the changes that the climate is undergoing, as they have a major influence on the hydrological cycle and on the net surface energy budget. Drilling ice cores provides the possibility to access information on temperature, precipitations and atmospheric composition from the time snow fell and ice formed. Alpine glaciers are characterized by a high accumulation rate and therefore have a high temporal resolution. Because of the proximity to urbanized areas, these glaciers are more sensitive to anthropogenic disturbances. Ice cores can show radioactive contamination, both natural and anthropogenic, due mainly to wet deposition processes, and to a lesser extent to dry fall-out (Pinglot et al., 2001). The analysis of radionuclides in alpine samples gives the possibility to trace circulation in the Northern Hemisphere, giving information on potential sources and residency times. Some radioactive nuclides can be linked to defined events, thus providing a temporal marker horizon that can be used to reconstruct accumulation rates in the glacier and date the ice core. This work of thesis focused on the detection of ^{137}Cs and ^{3}H. ^{137}Cs is a very effective tracer of anthropogenic influence on environments as it is a man made isotope which does not occur naturally. The release of ^{137}Cs in the atmosphere is linked to nuclear tests, which took place before the Partial Test Ban Treaty (PTBT) entered into force in 1963, and to nuclear accident such as Chernobyl in 1986 or Fukushima in 2011. Tritium can be used to study the melting effects in glaciers if the samples have a high depth resolution (Kang et al., 2015). This happens to be the case in alpine glaciers, where the accumulation rate is high enough to have a good signal concerning the last century. It has been proven that there is a well defined peak corresponding to the year 1963 linked to the tritium that was sent in the stratosphere during thermonuclear bomb testing(Pinglot et al., 2003). Subsequently, it has often been observed a seasonal variation of tritium due to the remaining part in the stratosphere and linked to the fact that in spring the exchange between troposphere and stratosphere is enhanced (Van Der Wel et al., 2011). The Chernobyl accident (1986) also altered the natural activity of tritium, but the detection of this horizon has yet proven to be more difficult due to lower levels of tritium release, a more local contamination, and melting processes in the glacier.

Analysis were performed on the chips of the ice core extracted from the Adamello glacier during the summer of 2016. This ice core was drilled in the framework of the POLLiCE project, with the target of studying the climate of the last century. It is the first ice core that has been drilled from Italy's biggest glacier.

Material and Methods

During this work of thesis two techniques have been used: gamma ray spectroscopy, for the detection of ^{137}Cs, and liquid scintillation counting for total beta activity analysis and tritium identification. This mainly depended on the fact that the two target isotopes behave differently during radioactive decay; the former decays emitting gamma rays, while the latter decays emitting beta particles.

Gamma ray spectroscopy was performed using a High-Purity Germanium (HPGe) detector. Water samples from the ice core were analyzed with HPGe detector with counting times of 160 hours on average. A peak at 661.6 keV, corresponding to gamma ray emission of ^{137}Cs, was recorded in some samples. Total beta activity analysis was performed with spectrometer Quantulus 1220 (TM) on aqueous samples extracted from chips of the ice core. Performing total beta protocol, a clear excess was found in some samples.

Chips corresponding to the central part of the ice core were, after the first round of analysis, filtrated using a vacuum-driven Millipore Filtration System. Polycarbonate 0.45 µm filters were used. Filtrated water was used to perform additional analysis with liquid scintillation counting. 40 ml were taken and passed through a Tritium Eichrom column in order to separate tritium from all the other radionuclides potentially present in the samples. Eluate was then used to prepare samples. Filters were also analyzed through gamma ray spectroscopy, using a Germanium Well detector. This kind of detector allows samples to be inserted directly inside the germanium. With this set-up, small samples can be measured with very high efficiencies. Filters were analyzed with counting times of 24 hours on average. This analysis provides the possibility to assess the contribution of particulate matter to the total radioactivity measured in the ice core and to better understand the differences showed by beta and gamma spectroscopy.

Absolute efficiencies for the HPGe detector containing the samples were calculated with MonteCarlo method using the Geant4 code, which is a toolkit for the simulation of the passage of elementary particles through matter (Agostinelli et al., 2003). For samples that showed an ^{137}Cs excess, the specific activity was calculated, whilst for samples without any excess a detection limit was calculated.

The use of detectors for both gamma ray spectroscopy and liquid scintillation counting requires a certain number of preliminary analysis, to be done before the environmental samples are measured. Standard samples have been prepared and analyzed for both gamma ray spectroscopy and liquid scintillation counting. Two different standard sources have been measured with different Germanium detectors: a multigamma standard source and a ^{134}Cs/^{137}Cs standard source. In particular, the multigamma standard source was used to validate the Monte Carlo method. Two sets of standard samples have been prepared for Liquid Scintillation Counting, both containing ^{3}H. The second set was passed through Eichrom columns prior to measuring. These standards were used to construct a caibration curve for tritium detection. In all cases samples were prepared in a way that they were in the most similar condition as the environmental samples.

Results and Conclusions

Results from gamma ray spectroscopy conducted on the Adamello ice core show a clear contamination of cesium, which can be attributed to thermonuclear bomb testing. The highest peak, found at a depth of 32.0 m ± 0.3 m (see Figure 1) corresponds with no doubt to the year 1963. The other 5 peaks, found between 20.72 m ± 0.34 m and 30.72 m ±0.31 m, are of more difficult attribution. The hypothesis here formulated, is their attribution to resuspension events followed by subsequent deposition on the glacier. The main assumption is that the peaks are to be attributed to the same events which generated the 1963 peak. The countries which continued testing after PTBT are France and China. France tested at Mururoa and Fangataufa sites, in French Polynesia, from 1966 to 1974, for a total yield of 10.1 Mt (Warner and Kirchmann, 2000). China tested at Lob Nor, a desert site in the north-western part of the country, from 1964 to 1980 for a total yield of 20.7 Mt (Warner and Kirchmann, 2000). Considering that the total yield for year 1962 was 163.8 Mt, although it is possible for radioactive fallout from chinese and french experiments to have reached the Alps, the activity values would be far lower than those detected. The geographical position and relatively low elevation of Adamello glacier make the glacier susceptible to local events of resuspension from surrounding mountain ranges and Po plain.

Moreover, results from filtration show that ^{137}Cs is mostly bound to particulate matter, as we were not able to detect an activity in samples after filtration. This poses a problem in the determination of the specific activity. The Monte Carlo simulations used for evaluation of the efficiency, assumed the cesium to be distributed uniformly in the sample. This assumption is now proven to be wrong. For what concerns total beta analysis, analyzing the shape of the spectra that had shown excess in respect to the background and comparing it with spectra of standard samples containing tritium, we assessed that the main contribution to the total beta activity was the presence of tritium. The analysis of samples passed through an Eichrom column, has proven this to be true. LSC analysis has shown a double peak, between 28.8 $\pm$ 0.3 m and 30.7 $\pm$ 0.3 m, at a slightly shallower depth than the cesium peak, as shown in Figure 1. If the double peak is to be attributed to 1963, this delay could be due to the different atmospheric path followed by the two elements. In fact, as mentioned above, cesium travels bound to particulate matter, while tritium is incorporated in water molecules.

To conclude, gamma ray spectroscopy has proven to be a powerful tool analysis, especially due to its straightforward identification of radioisotopes. Its main limitation, especially when you have to deal with a great number of samples, such as in ice cores, is the longer measuring time required. This work of thesis has underlined the possibility to limit gamma ray spectroscopy analysis on the particulate matter contained in the ice. This, while requiring an additional step in the preparation of samples (filtration), gives the opportunity to use a well-type detector, which has an higher efficiency and requires lower measuring times. In fact, approximately one day measurement for this kind of samples has proven to be an adequate measuring time, in contrast to 4 to 5 days necessary with the classical HPGe detector. This would also eliminate the error in the estimate of efficiency due to the non-uniformity of cesium in the water. Moreover, the highest sensibility was achieved with the Well detector, reaching the value of 0.0012 Bq, while the HpGe detector used for the analysis of the aqueous samples reached a detection limit of 0.020 Bq/kg.

Liquid scintillation counting has also given good results. Its main advantage, in respect to gamma ray spectroscopy in general, is the lower measuring time required, which gives the possibility to obtain relatively quickly a complete profile of the ice core . Total beta activity has proven itself to be non essential. The impossibility to determine the exact contribution of different isotopes to the spectrum and the consequent difficulty in constructing a calibration curve are the main problems of this kind of analysis. Knowing that the main contribution to the beta signal is tritium, it seems more appropriate from now on to perform a tritium analysis straight away. Once again, this would require additional preparation of the samples, which have to be filtrated to eliminate solid impurities and passed through an Eichrom column before measuring.

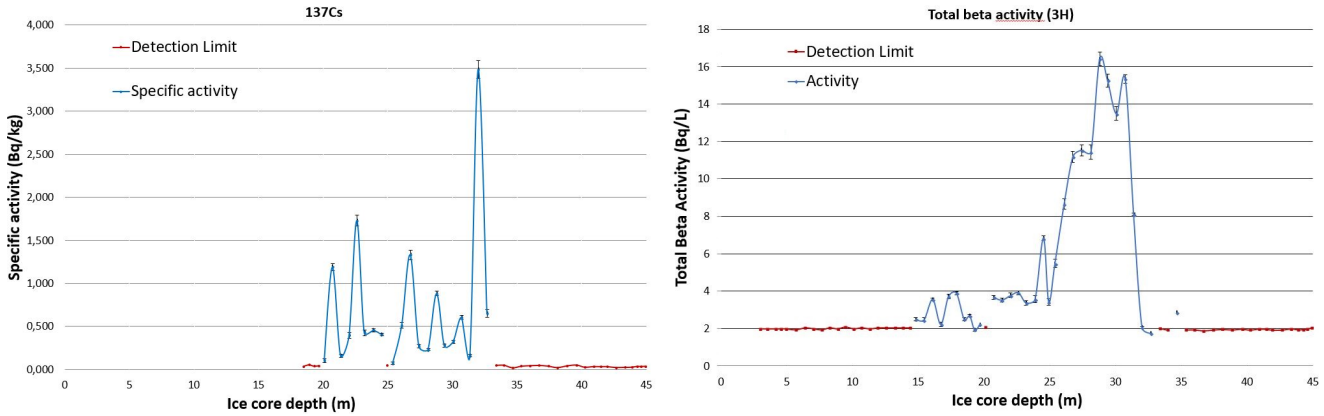

Figure 1: Cesium profile (right) and Total beta Activity (left) in the Adamello ice core.

Bibliography

Agostinelli, S., J. Allison, K. a. Amako, J. Apostolakis, H. Araujo, P. Arce, M. Asai, D. Axen, S. Banerjee, G. Barrand, et al.

2003. Geant4—a simulation toolkit. *Nuclear instruments and methods in physics research section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 506(3):250–303.

Kang, S., F. Wang, U. Morgenstern, Y. Zhang, B. Grigholm, S. Kaspari, M. Schwikowski, J. Ren, T. Yao, D. Qin, et al.

2015. Dramatic loss of glacier accumulation area on the tibetan plateau revealed by ice core tritium and mercury records. *The Cryosphere*, 9(3):1213–1222.

Pinglot, J. F., J. O. Hagen, K. Melvold, T. Eiken, and C. Vincent

2001. A mean net accumulation pattern derived from radioactive layers and radar soundings on austfonna, nordaustlandet, svalbard. *Journal of Glaciology*, 47(159):555–566.

Pinglot, J. F., R. A. Vaikmae, K. Kamiyama, M. Igarashi, D. Fritzsche, F. Wilhelms, R. Koerner, L. Henderson, E. Isaksson, J.-G. Winther, et al.

2003. Ice cores from arctic sub-polar glaciers: chronology and post-depositional processes deduced from radioactivity measurements. *Journal of Glaciology*, 49(164):149–158.

Van Der Wel, L., H. Streurman, E. Isaksson, M. Helsen, R. Van De Wal, T. Martma, V. A. Pohjola, J. C. Moore, and H. Meijer

2011. Using high-resolution tritium profiles to quantify the effects of melt on two spitsbergen ice cores. *Journal of glaciology*, 57(206):1087–1097.

Warner, F. and R. Kirchmann

2000. *Nuclear Test Explosions, Scope 59: Environmental and Human Impacts*, volume 59. John Wiley & Sons Incorporated.