Measurement Limits to $^{134}$Cs Concentration in Soil

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Abstract

We investigate the caesium concentrations in soils in mountain areas near Gori nuclear power plant in Korea, focusing on the measurement limits to the $^{134}$Cs. In order to lower the minimum detectable amount (MDA) of activity for the $^{134}$Cs, we have used the ammonium molybdophosphate (AMP) precipitation method to get rid of the $^{40}$K existing in natural radioactivity, which reduces the MDA of activity about ten times smaller than those without the AMP precipitation method. The MDA results for the $^{134}$Cs were found to be in the range between 0.015 and 0.044 Bq/kg-dry weight. In order to diminish the background, we also have measured a part of the soil samples in Yangyang, a small town in the east coast of Korea. However, it turns out that in order to detect the $^{134}$Cs in the samples the MDA should be reduced to the level of mBq/kg-dry weight.

Key words: $^{134}$Cs, $^{137}$Cs, ammonium molybdophosphate (AMP) precipitation method, minimum detectable amount (MDA)

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1 Introduction

It is of great importance to study the anthropogenic caesium radioisotopes $^{137}\text{Cs}$ and $^{134}\text{Cs}$, since their production and emission rates are much higher than other radioisotopes from nuclear fissions at nuclear power plants (NPP) and they have rather low movility in soils. Thus, $^{137}\text{Cs}$ are widely used as tracer radionuclides for monitoring the NPP-related environmental radioactivity. The $^{137}\text{Cs}$ nucleus ($t_{1/2} = 30.07\text{y}$) is a long-lived beta emitter decaying a 95% branching ratio to the metastable state ($t_{1/2} = 2.55\text{m}$) of $^{137}\text{Ba}^*$ at 661.7keV. While the $^{134}\text{Cs}$ nucleus is also a beta emitter, its decay time ($t_{1/2} = 2.065\text{y}$) is much shorter and emits many gamma-rays correlated with the beta electron. Due to these different life-times, it is possible to date the anthropogenic radioactivity with the ratio of $^{134}\text{Cs}$ to $^{137}\text{Cs}$ concentrations in soil measured. However, since the $^{134}\text{Cs}$ has much shorter half life-time compared to the $^{137}\text{Cs}$, the dating range depends solely on a minimum detectable amount (MDA) of activity for the short-lived $^{134}\text{Cs}$ radioisotope.

Moreover, it is well known that the $^{137}\text{Cs}$ contamination arises mainly from three different sources: Atmospheric nuclear weapon tests (NWT) in the period of the late 1950s and early 1960s, in particular, in the northern hemisphere (UNSCEAR, 2000), the Chernobyl accident taken place in May 1986, and discharges from nuclear power plants (NPPs). On the contrary, the $^{134}\text{Cs}$ does not come from the NWT, since it is induced only by the neutrons impinged on the fission radionuclide $^{133}\text{Cs}$, which rarely occurs in the course of a nuclear explosion due to a lack of the reaction time. However, the $^{134}\text{Cs}$ can be produced in reactors at NPPs. Thus, knowing the ratio of $^{134}\text{Cs}$ to $^{137}\text{Cs}$ concentrations in soil may determine even their origin. Note that the fallout ratio of the Chernobyl explosion is known to be about $^{134}\text{Cs}/^{137}\text{Cs} \simeq 50\%$ in 1986 (Thomas and Martin, 1986).

In order to determine the ultra-low level caesium concentrations quantitatively in soil, it is essential to understand the natural background originated mainly from uranium-, thorium-, actinium-series nuclides, and the potassium $^{40}\text{K}$. The $\gamma$-ray peaks of the $^{134}\text{Cs}$, in particular, those at 604.7keV and 795.6keV are overlapped with a great deal of $\gamma$-ray peaks from natural radionuclides. Moreover, strong peaks such as a 1460keV line of the $^{40}\text{K}$ at high energies produce the Compton-continuum background that covers the wide range of the gamma energies for caesium decays. Thus, we need to get rid of the natural background as much as possible. One of the best ways known to eliminate the $^{40}\text{K}$ is to use the ammonium molybdophosphase (AMP) precipitation method. AMP is an inorganic compound that selects the caesium exclusively and has high adsorption capacity (Suss and Pfrepper, 1981). This method has been widely applied to sea-water samples in order to enrich the Cs concentration. However, this radiochemical extraction for a soil sample should be emphasized by its
effective background suppression, thereby significantly lowering the minimum detection limit.

In the present work, we want to investigate the minimum detection limit to the measurement of the $^{134}\text{Cs}$ in surface soils in mountain areas near the Gori NPP in Korea, aiming at determining the ratio of the $^{134}\text{Cs}$ to $^{137}\text{Cs}$. In Korea, eighteen nuclear reactors are being operated at four different NPP sites. One of the NPP sites is located in Gori, a small town in the south-east coast of Korea, where four pressurized-water reactors are in operation. The first Gori reactor has been operated first time in Korea since 1978. The environmental radioactivity in the vicinity of the Gori NPP has been monitored last years. The samples were taken from soils, rain water, sea water, surface water, milk, seaweed, egg, etc. Recently, high concentrations of the $^{137}\text{Cs}$ were first reported in the soil samples from the mountain areas near the Gori NPP in the range of 50 − 460 Bq/kg-dry, while the $^{137}\text{Cs}$ concentrations in surface soils of Korea are known to be ranged in 7.86 − 70.1 Bq/kg-dry with a mean value of $33.2 \pm 16.1$ Bq/kg-dry (Kim et al., 1998).

Taking into account the fact that the Chernobyl accident took place more than thirty years ago, the $^{134}\text{Cs}$ concentration was reduced by three orders of magnitude, while a half of the original $^{137}\text{Cs}$ concentration is survived (Pourcelot et al., 2003). If the ratio of $^{134}\text{Cs}$ to $^{137}\text{Cs}$ is higher than the average ratio to be mapped out for the distant area from the Gori power plant, the origin should be investigated thoroughly. In this regard, it is of great significance to conduct the methodological study for the measurement of the ultra-low level concentration of $^{134}\text{Cs}$ in its own right.

The present work is organized as follows: In Section 2, we briefly explain the process of the experiment. In Section 3, we discuss the detection limits to the $^{134}\text{Cs}$ concentration. In the final Section, we summarize and draw conclusions of this work.

2 Experiment

2.1 Sampling and Preparation

Soil samples were collected from 96 sites at top areas of four different mountains near the Gori NPP: Mt Daleum (35°18′ N, 129°12′ E), Mt Ilgwang (35°16′ N, 129°12′ E), Mt Samgak (35°21′ N, 129°13′ E), and Mt Daewoon (35°24′ N, 129°13′ E). All these mountains are located within 7-12 km distance from the Gori NPP. Uncultivated soil was sampled from the 5 − 10 cm layer. Each sample was taken in close proximity to give approximately 0.5 to 1.5 kg. Since we
are mostly interested in measuring both $^{134}$Cs and $^{137}$Cs, we have selected the samples with the highest $^{137}$Cs concentration: $461 \pm 9 \text{ Bq/kg-dry}$. From that sample, we took two samples for the measurement and prepared them with different methods: Sample A was air-dried at 65°C for 10 hours and sieved with 2mm mesh size. It was then filled in a 450ml Marinelli beaker. On the other hand, sample B was heated at 450°C for 10 hours. One liter of 37% HCl solvent was added to sample B, and the solution was stirred with heat for 4 hours. Soil contents in the sample solution were sieved through two layers of the normal filter(5C, 125mmφ) and the glass-fiber filter(GF/C, 110mmφ) in the Buhner filtering system. Unfiltered sediment was washed out with hot water.

For precipitation with ammonium-molybdophosphate(AMP), the sample was stirred with the 80g AMP solution at PH2 for 10 hours. The sample was then sieved with a membrane filter of 0.45µm pore size, and after cleansing the sample bottle with 1% HCl solution, the cleansing solution was put to the filter to sieve remaining contents. After the AMP precipitation, the sample was dried with infrared light, and put into a standard 55ml Marinelli beaker for $\gamma$-ray measurement.

2.2 Gamma-ray Detection

We have measured the $^{137}$Cs concentration in the soil samples by directly measuring the 661.7keV, 604.7keV, and 795.8keV gamma rays with an HPGe detector. The HPGe $\gamma$-ray detector has a 30% relative efficiency at 1330keV of $^{60}$Co, and the schematic setup of the detector and the shielding system is shown in Fig. 1.

![Fig. 1. Schematic view of the measurement setup](image)

The HPGe detector was coupled to standard NIM electronics and a PC with
Gamma-Vision™ spectroscopy software. The system was shielded with a uniform lead castle of 10cm in thickness. Certified reference materials (CRM) of mixed gamma-ray sources from KRISS (Korea Research Institute of Standards and Science) were employed for the efficiency calibration of the system. The CRM provides with 13 gamma-ray peaks from 10 radioactive elements: $^{241}\text{Am}$ (59.5keV), $^{109}\text{Cd}$ (88.0keV), $^{57}\text{Co}$ (122.1keV, 136.5keV), $^{139}\text{Ce}$ (165.86keV), $^{51}\text{Cr}$ (320.08keV), $^{113}\text{Sn}$ (391.70keV), $^{85}\text{Sr}$ (514.00keV), $^{137}\text{Cs}$ (661.66keV), $^{88}\text{Y}$ (898.04keV, 1836.05keV), and $^{60}\text{Co}$ (1173.23keV, 1332.49keV).

Fig. 2. Efficiency curves of the HPGe detector for the 450ml dry-soil sample (square) and the 45ml AMP sample (circle), respectively.

Figure 2 represents efficiency curves of the HPGe detector, fitted with the following empirical logarithmic polynomials:

$$
\ln \varepsilon = \sum_{n=0}^{3} a_n (\ln E_\gamma)^n,
$$

where $E_\gamma$ denotes the gamma-ray energy. The $a_n$ are the fitting parameters of the polynomial. The efficiency for the 450ml sample is lower than that for the 45ml AMP sample in the low-energy region, because of a larger self-absorption of gamma-rays in the sample volume.

Figure 5 depicts the gamma-ray spectrum of the AMP sample overlaid with that of the dry-soil sample. The chemical separation shows a significant improvement in a signal-to-background ratio. The background is reduced dramatically by almost a order of magnitude due to the AMP method. The upward arrows in Fig. 5 designate the positions of gamma-ray energies from $^{134}\text{Cs}$ and $^{137}\text{Cs}$.
3 Detection Limits

We are now in a position to discuss the MDA of activity of samples. The criterion for limits of detectability is usually given by the MDA of activity. The most widely used definition of the MDA was first defined by Curie (Curie, 1968) as follows:

\[
MDA = \frac{2.71 + 4.65\sqrt{N_b}}{\varepsilon \cdot P_\gamma \cdot t},
\]

where \(N_b\) denotes the number of background events measured during the time \(t\). The \(\varepsilon\) represents the detection efficiency as defined in Eq. (1), and \(P_\gamma\) stands for a branching fraction for \(^{134}\text{Cs}\) emitting the respective gamma-ray. The minimum detectable activity depends on the square root of the number of background events which mainly come from three different reasons: The sample itself, Compton continuum, and natural radioactivity. Note that it actually depends on the inverse square root of the time, since the background events also increase as a function of time.

In Fig. 4 we draw the gamma-ray spectrum of the dry-soil sample measured at a normal laboratory without the AMP precipitation taken into account. The peak at 609 keV corresponds to that of the gamma-ray from \(^{214}\text{Bi}\). Because of the background, it is very difficult to identify the spectrum of the sample, in particular, in the higher region of the gamma-ray energy.
Fig. 4. Gamma-ray energy spectrum of the sample measured at a normal laboratory.

In Fig. 5, we present the full energy-range gamma-ray energy spectrum of the AMP-precipitated sample measured at the underground laboratory. As compared with Fig. 4, it is shown that the background is drastically reduced.

In order to reduce the background of the sample, we have brought the sample to an underground laboratory located in Yangyang, a small town in the east coast of Korea. The underground laboratory is specialized to search for dark matter for which it is essential to develop low-background measurement. It utilizes the space in a tunnel of Yangyang Pumped Storage Power Plant located deep under a mountain. Figure 5 shows the gamma-ray spectrum of the AMP-precipitated sample measured at the underground laboratory in Yangyang. As compared with Fig. 4 it is shown that the background is drastically reduced.

In Fig. 6, we present the full energy-range gamma-ray energy spectrum of the AMP-precipitated sample measured at the underground laboratory. As mentioned before, the AMP precipitation method has greatly reduced the MDA by almost one order of magnitude. It filters selectively out Cs isotopes from the sample. Moreover, the Compton continuum background from $^{40}$K is also shown to be noticeably reduced. However, the 795 keV peak from $^{228}$Ac
Fig. 6. Full energy-range gamma-ray energy spectrum of the AMP precipitation sample measured at a underground laboratory. It turns out that our natural background level is almost four orders of magnitude smaller than the level of Cs radioisotope concentrations.

is interfered with the 795.6 keV from $^{134}$Cs. Therefore, we found it difficult to resolve the peak from $^{134}$Cs even with very low background. The 604.7 keV peak from $^{134}$Cs can be well resolved from underlying background. The 609.3 keV peak from $^{214}$Bi appears in the vicinity of the 604.7 keV peak, but a good energy resolution of the HPGe detector helps the two peaks resolved clearly. It therefore should be noted that the 604.7 peak should be searched for in the $^{134}$Cs survey. It is then necessary to suppress background contributions underneath the 604.7 keV peak. The region near the peak is between the 661.7 keV $^{137}$Cs peak and its Compton edge. The yield in the region is thus mainly due to multiple Compton scattering of the 661.7 keV $^{137}$Cs gamma ray in a HPGe crystal. A possible experimental approach is a gamma-ray detection with a Compton suppressed system so that it may be possible to suppress such multiple Compton scattering events and to lower the MDA significantly. Figure 7 summarizes how to resolve peaks of $^{134}$Cs and $^{137}$Cs from the background.

Table I lists the values of the MDA from the present work, with and without the AMP precipitation method considered.

| Location | without AMP | with AMP |
|----------|-------------|----------|
| $^{134}$Cs | < 0.2 | < 0.015 |
4 Summary and Conclusion

In the present work, we investigated the caesium concentrations in soil samples taken from mountain areas in the vicinity of the Gori nuclear power plant, emphasizing the minimum detection limit to the measurement of $^{134}$Cs concentration. We carried out the sampling by taking $5-10$ cm layer of surface soil at the top areas of four different mountains within the $7-12$ km distance of the Gori nuclear power plant. We have used the ammonium molybdate-phosphate (AMP) precipitation method to get rid of the $^{40}$K existing in natural radioactivity, which reduces the minimum detectable activity (MDA) of activity about ten times smaller than those without the AMP precipitation method. Even though we were able to reduce the MDA for the $^{134}$Cs from 0.2 to 0.015 by one order of magnitude smaller, using the ammonium-molybdate-phosphate precipitation method in order to lower the background mainly arising from the natural radionuclide $^{40}$K, it is not possible to determine the $^{134}$Cs concentration quantitatively.

While it is of great difficulty to track down the origin of the $^{137}$Cs that are found in higher concentrations in soil samples near the Gori nuclear power plant, the results imply that the $^{137}$Cs found in the samples are at least 20 years old. Taking into account the fact that the Chernobyl accident took place 22 years ago from now, one can assume that the present $^{137}$Cs concentrations may have come from the Chernobyl explosion. Since it is known that the fallout ratio of the Chernobyl accident is about $^{134}$Cs/$^{137}$Cs $\simeq$ 50% in 1986 (Thomas and Martin, 1986), the present ratio should be dropped to be about 0.1%, considering that the half life-times of the $^{134}$Cs and $^{137}$Cs are, respectively, 2.065y and 30.07y. In fact, the fallout due to the Chernobyl accident did not affect Korea much, since the distribution of $^{137}$Cs is similar to those of
other countries (C.S. Kim et al., 1998). Thus, we can infer from it that the present concentration of the $^{134}$Cs should be less than 0.01 Bq/kg-dry which is definitely lower than the minimum detection limit reached in the present work.

In order to reduce the minimum detectable amount of $^{134}$Cs activity further, we need to get rid of the Compton continuum, using the Compton suppression method, which is now under investigation.

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