DOI: 10.31276/VJSTE.63(3).92-96

# Developing a database for Cs-137 radioactivity concentration and assessing residual levels in surface soils of Quang Ninh, Hai Phong, and Lang Son provinces

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Received 20 March 2021; accepted 10 June 2021

***Abstract:***

**The radioactivity concentration of Cs-137 was determined in 826 soil samples collected from Hai Phong, Quang Ninh, and Lang Son. The residual levels of Cs-137 have also been assessed and compared with previous publications to obtain a trend in Cs-137 in the surface soil. The actual measured average of the residual level of Cs-137 in the soil samples collected in this study was 236±146 Bq/m², while the value calculated using the published experimental formula was 407±77 Bq/m². The impact of Cs-137 deposition from the Fukushima nuclear accident (2011) in this area is confirmed to be negligible.**

***Keywords:*** **Hai Phong, Lang Son, Quang Ninh, radioactivity of Cs-137, residual level of Cs-137.**

***Classification number:*** **5.3**

## Introduction

Cs-137 is an artificial fission product with a relatively long half-life (30 years). As a result, Cs-137 can be used as an indicator of the environment impact of release from a nuclear power plant during normal operation or emergencies. Although the residual level of Cs-137 in soil in Vietnam was studied many years ago, recent developments of nuclear power plants near the North border of Vietnam, especially the operation of Fangchengang Nuclear Power Plant (NPP) at about 54 km from the border, has brought out the need to establish a database of radioactivity concentration (Bq/kg) and a reassessment of residual levels (Bq/m²) of Cs-137 near border areas. In this study, the radioactivity concentration and the residual level of Cs-137 in the soils of the Hai Phong, Quang Ninh, and Lang Son provinces are determined to fulfil this purpose.

## Materials and methods

A total of 826 soil samples were collected over a period of 3 years from 2017-2019 following standard procedures by VARANS TSC.

To mitigate time constraints, sample processing and analyses were performed simultaneously by three laboratories: VARANS TSC (526 samples; with 100 samples from Quang Ninh and 426 samples from Lang Son), the Institute for Nuclear Science and Techniques (220 samples; including 80 samples from Hai Phong and 140 samples from Quang Ninh), and the Center for Environmental Technology & Treatment - Ministry of Defense (80 soil samples from Quang Ninh).

### *Soil sampling and processing method*

Soil samples were taken from a flat-surfaced area based on a 4.5 km x 4.5 km grid in all accessible areas to ensure relatively uniform sample density over all three provinces. All the soils, dug from a 30 cm x 30 cm x 30 cm cubic hole, were mixed thoroughly. A depth of 30 cm was chosen to take into account the disturbance processes of soils. Then, the soils were reduced to about 1-1.5 kg by quartering and compartmentalization. Relevant information from the soil samples were also recorded.

The sample processing procedures of all involved organizations were standardized and included the

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following steps: crushing the sample to reduce soil particle size to 1 cm or less; removing foreign materials such as plant roots, pebble, and garbage; spreading the soil sample on tray then heating to about 100-105°C until the samples were completely dried out (weight variation less than 1%); grinding and sieving the dry soil to obtain particle sizes less than 1 mm; and finally weighing and compressing the samples into a confined box for measurements.

***Cs-137 analysis methods and quality assurance measures***

Technical specifications of each low-background gamma spectrometer used by each laboratory for sample measurement and analysis are shown in Table 1.

The energy and efficiency calibration of the HPGE detectors were developed using a multipurpose RGU-1 reference source and RGU_IAEA.Lib library provided by IAEA.

The geometry calibration file and correction curve for self-absorption due to sample density and composition were also generated using RGU-1 standard sources. The chemical composition of RGU-1 is similar to that of surface soil samples [1], which indicates that the contribution ratio of chemical elements in the two samples are similar. In addition, the self-absorption of the samples with different chemical compositions mainly influences the energy region of less than 186 keV. For the higher energy region, self-absorption correction is not necessary and thus was neglected [2]. Therefore, generation of the self-absorption correction curve using RGU-1 is a suitable method.

For VARANS TSC and INST, the quality assurance (QA) for sample analyses were performed by analysing the IAEA reference sample (Sample No.4, provided in the 2018 inter-laboratory comparison conducted by IAEA) with known activity of nuclides including K-40, Cs-137, Tl-208, Pb-210, Pb-212, Pb-214, Bi-214, Ra-226, and Ac-228, which followed IAEA's assessment method in which the trueness, precision of the analysis results is assessed separately [3]. For CTET, the QA was performed by inter-comparison of the analysis results from three laboratories for a set of three samples. The QA results show that all analysis results were evaluated as "acceptable" (the highest grade among three levels of evaluation: A "Acceptable", N "Not acceptable", and W "Warning").

## Results

***Development of database on Cs-137 radioactivity concentration in surface soil***

Figure 1 shows the spatial distribution of Cs-137 radioactivity concentration in soil layers up to 30 cm in depth on the GIS maps of the Hai Phong, Quang Ninh, and Lang Son provinces.

**Table 1. Technical specifications of low-background gamma spectrometers used by involved laboratories.**

| N⁰ | Parameter | VARANS TSC | Institute for Nuclear Science and Techniques (INST) | Center for Technology Environmental Treatment (CTET) |
|---|---|---|---|---|
| 1 | Lead chamber | ORTEC, Cylindrical, inner diameter: 28.0 cm; outer diameter: 50.4 cm | CANBERRA, Cylindrical, inner diameter: 27.9 cm; outer diameter: 50.8 cm | Cubical lead chamber with outer dimensions: 100 cm x 80 cm and inner dimensions: 50 cm x 50 cm |
| 2 | Detector manufacturer | 01 HPGe detector made by CANBERRA | 02 HPGe detectors made by CANBERRA | 01 HPGe detector made by CANBERRA |
| 3 | Type of detector and relative efficiency | GX4020, ≥40% | - GC5019, ≥50%<br>- BE6530, ≥65% | GC1520, 15% |
| 4 | Resolution | ≤2 keV at 1.33 MeV and ≤1.2 keV at 122 keV | - GC5019: ≤1.9 keV at 1.33 MeV and ≤1.0 keV at 122 keV<br>- BE6530: 1.9 keV at 1325 MeV and 0.684 keV at 122 keV | 1.66 keV at 1.33 MeV and 0.77 keV at 122 keV |
| 5 | Minimum detectable activity (MDA) to Cs-137 | 0.18 Bq/kg | 0.3 Bq/kg | 0.6 Bq/kg |

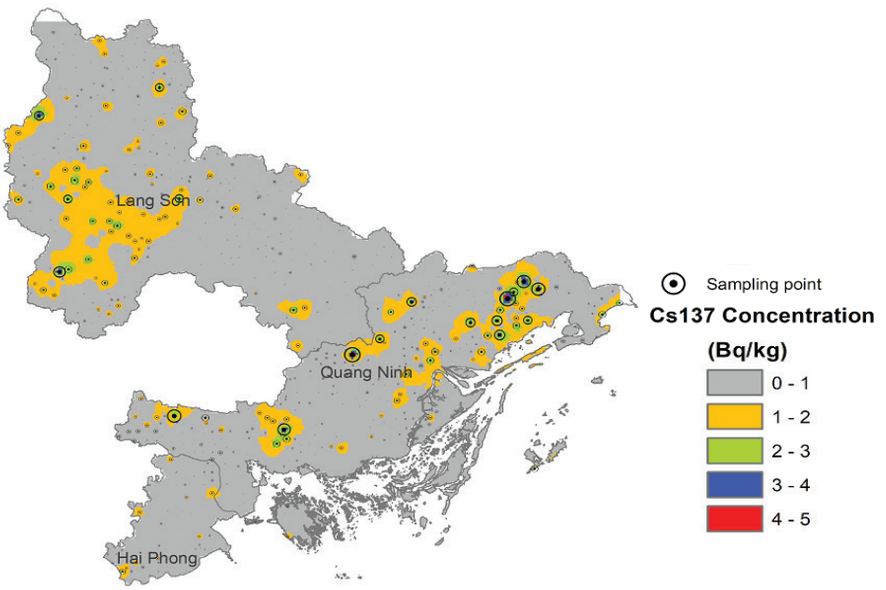


**Fig. 1. Spatial distribution of Cs-137 radioactivity concentration in soil samples collected in 03 surveyed provinces.**

The measured radioactivity concentrations of Cs-137 in Quang Ninh are higher than that of Hai Phong and Lang Son, albeit not much. The observation of high concentrations of Cs-137 is consistent with the high average annual rainfall in Mong Cai - Quang Ninh in comparison with that of the other surveyed areas [4].

Statistical distributions of Cs-137 radioactivity concentration in the 826 soil samples collected in Hai Phong, Quang Ninh, and Lang Son provinces are shown in Fig. 2.

The statistical parameters of the radioactivity concentration of Cs-137 in the 826 soil samples collected from all 3 provinces are shown in

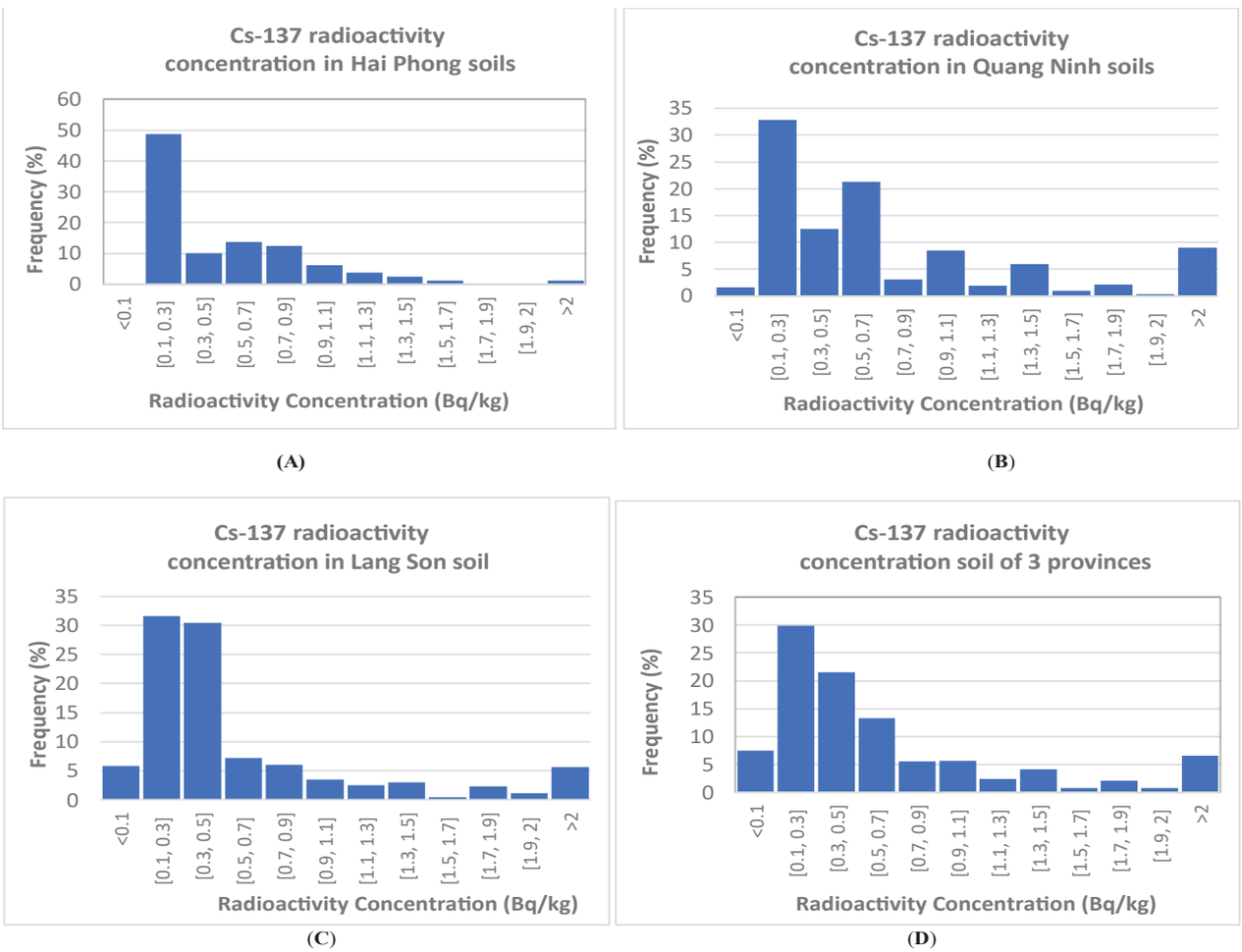


**Fig. 2. Statistical distributions of Cs-137 radioactivity concentration in 826 soil samples collected from (A) Hai Phong, (B) Quang Ninh, (C) Lang Son, and (D) the 3 provinces as a whole.**

**Table 2. Statistical parameters on radioactivity concentration of Cs-137 in 826 soil samples collected in Hai Phong, Quang Ninh, and Lang Son.**

| Statistical parameters | All 3 provinces | Quang Ninh | Lang Son | Hai Phong |
|---|---|---|---|---|
| Number of samples | 826 | 320 | 426 | 80 |
| Mean (Bq/kg) | 0.68 | 0.84 | 0.60 | 0.46 |
| Standard error (Bq/kg) | 0.03 | 0.05 | 0.03 | 0.05 |
| Median (Bq/kg) | 0.41 | 0.60 | 0.37 | 0.34 |
| Mode (Bq/kg) | 0.30 | 0.30 | 0.15 | 0.05 |
| Standard deviation (Bq/kg) | 0.75 | 0.90 | 0.65 | 0.46 |
| Kurtosis | 9.86 | 8.64 | 5.03 | 2.26 |
| Skewness | 2.73 | 2.71 | 2.18 | 1.33 |
| Minimum (Bq/kg) | 0.03 | 0.06 | 0.03 | 0.05 |
| Maximum (Bq/kg) | 5.70 | 5.70 | 4.14 | 2.32 |
| Confidence level (95%) | 0.05 | 0.10 | 0.06 | 0.10 |

Table 2. For all samples in which Cs-137 radioactivity concentration was below LOD, the LOD values were chosen instead.

***Comparison with results from previous publications***

In 2001, Prof. Pham Duy Hien and colleagues published on the residual density of Cs-137 in 292 undisturbed soil samples collected from 292 positions across Vietnam within 9.2-23.0$^0$N latitude [5]. The principle purpose of this study is to assess the current status of Cs-137 in the surveyed area in order to establish a baseline for radioactivity concentration of this nuclide as an initial reference value in case of future emergency. Therefore, the priority for selecting samples is that the sampling points are evenly distributed over all accessible areas of the 3 provinces of interest (latitude from 20.61 to 22.18$^0$N), which will take into account the land use in all localities. Thus, soil disturbance was not considered in our study.

To make an appropriate comparison, only 264 soil samples in our study were taken into account as samples with Cs-137 radioactivity concentration below LOD were neglected.

*Radioactivity concentration of Cs-137:* according to Ref. [5], the radioactivity concentration of Cs-137 in 292 soil samples varied from 0.5 to 18.0 Bq/kg with an average of 2.03 Bq/kg. In our study, these values ranged between 0.14 to 5.70 Bq/kg with an average of 1.14 Bq/kg, which is 56% of the average value from the previous research. According to the exponential decay principle, the expected residual levels of Cs-137 should be approximately 60-63% of the published values in 2001 (the addition of Cs-137 due to events such as the Fukushima accident could be ignored*).

The observed difference could be explained by processes that cause Cs-137 to redistribute to deeper layer of soil, i.e., natural erosion or man-made disturbance of the soil from cultivation or road renovation activities.

*Residual level of Cs-137:* the research by Prof. Pham Duy Hien's group [5] also proposed an experimental formula for determining the residual level of Cs-137 according to the latitude of the sampling location and rainfall:

$$Ln(I) - \varepsilon = (3.53 \pm 0.09) + (0.092 \pm 0.004)L + (0.62 \pm 0.03)AR$$

where I is the residual level of Cs-137 in soil sample (Bq/m$^2$); L is the latitude of the sampling location (degree); AR is the annual rainfall at the sampling location (m).

The variation of Ln(I) in the surveyed area is shown in Fig. 3. The residual level of Cs-137 in soil samples, calculated from the experimental formula with rainfall data taken from the nearest metrological stations, shows a smaller variation than the data collected under our study, although the upper limit of both data sets are quite similar.

*According to the annual measurement data under the Annual Environmental Monitoring Task done by the Atomic Energy Institute of Vietnam, Cs-137 in fallout samples were detected at their Lang Sơn city and Ha Long city stations over the period from 4/2011 to 9/2011. The residual level at this period was 0.14 Bq/m$^2$. After that, the residual levels of Cs-137 at the stations were below LOD.

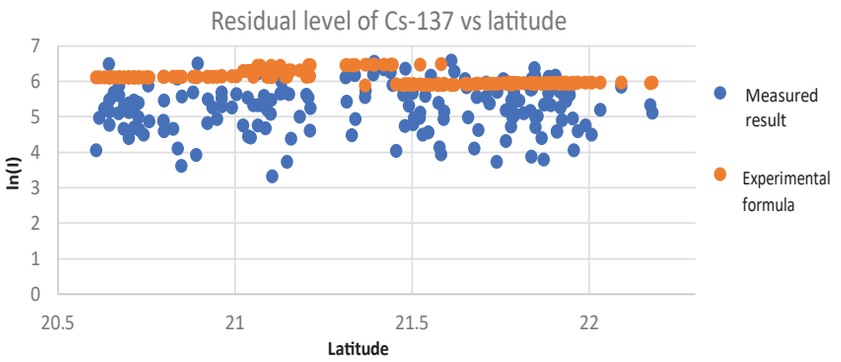


**Fig. 3. Comparison of the measured residual level of Cs-137 with calculation results from the experimental formula** [5].

The actual average measured residual level of Cs-137 in soil samples collected in this study was 236±146 Bq/m$^2$, while the one calculated from the experimental formula was 407±77 Bq/m$^2$. Although the measured values were generally more dispersed and lower than the calculated values due to the erosion and disturbance processes as mentioned above, they are still in agreement when one takes into account the standard deviation of both datasets.

## Discussion and conclusions

This study has provided comprehensive and detailed data on the current background level of Cs-137 radioactivity concentration in soil from Hai Phong, Quang Ninh, and Lang Son provinces. The data could be used as a reference value in case of future significant emergency from nuclear power plants near the north border of Vietnam.

The residual level of Cs-137 in soil samples collected from the surveyed provinces demonstrate a decreasing trend when compared to previous monitoring results, which indicates that the contribution of nuclear accidents that occurred in the past 20 years (including the Fukushima accident) are negligible.

The processes of natural erosion and anthropogenic disturbance of the topsoil are quite strong and widespread in the survey area. The data should be further updated regularly to quantitatively estimate the relationship between these processes and Cs-173 levels to provide a more accurate assessment of Cs-173 supplementary after a significant emergency.

The experimental formula, provided by Prof. Pham Duy Hien and colleagues in his publication in 2001, is still considered valid for the recent data taking into account the disturbed characteristics1 of the collected soils.

## ACKNOWLEDGEMENTS

The study was conducted as a part of the Ministerial Project titled Survey and Establishment of Environmental Radiation Database for localities potentially impacted in the early period of northern transboundary nuclear emergency for the purpose of emergency response as assigned by Ministry of Science and Technology to VARANS TSC from 2017 to 2019, with contribution by technical staff from VARANS TSC, INST and CTET in sample collection, processing, and analysis.

We would like to express a special thanks of gratitude to Dr. Nguyen Hao Quang for the valuable comments to elaborate this paper.

## COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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