Comparative analysis of the radionuclide composition in fallout after the Chernobyl and the Fukushima accidents

Konstantin Kotenko[1], Sergey Shinkarev[1], Yury Abramov[1], Evgeniya Granovskaya[1], Vladimir Yatsenko[1], Yury Gavrilin[1], Tetsuji Imanaka[2], Masaharu Hoshi[3]

[1]Burnasyan Federal Medical Biophysical Center, 123182, Zhivopisnaya Str, 46 Moscow, Russia
[2]Research Reactor Institute, Kyoto University, Kumatori-cho, Osaka 590-0494, Japan
[3]Research Institute for Radiation Biology and Medicine, Hiroshima University, 1-2-3 Kasumi, Minami-ku, Hiroshima 734-8553, Japan

Abstract. The nuclear accident occurred at Fukushima Dai-ichi Nuclear Power Plant (NPP) (March 11, 2011) similar to the accident at the Chernobyl NPP (April 26, 1986) is related to the level 7 of the INES. It is of interest to make a comparative analysis of radionuclide composition of fallout following the both accidents. Results of spectrometric measurements were used in that preliminary analysis. Two areas following the Chernobyl accident were considered: (1) the near zone of fallout – the Belarusian part of the central spot extended up to 60 km around the Chernobyl NPS and (2) the far zone of fallout – the "Gomel-Mogilev" spot centered 200 km to the north-northeast of the damaged reactor. In the case of Fukushima accident the near zone up to about 60 km considered. Comparative analysis has been done with respect to refractory radionuclides (^{95}Zr, ^{95}Nb, ^{141}Ce, ^{144}Ce), as well as of the following radionuclides ^{131}I, ^{103}Ru, ^{106}Ru, ^{134}Cs, ^{137}Cs, ^{140}La, ^{140}Ba and the results of such comparison have been discussed. With respect to exposure to the public the most important radionuclides are ^{131}I and ^{137}Cs. For the both accidents the ratios of ^{131}I/^{137}Cs in the considered soil samples are in the similar ranges: (3-50) for the Chernobyl samples and (5-70) for the Fukushima samples. Similar to the Chernobyl accident a clear tendency that the ratio of ^{131}I/^{137}Cs in fallout decreases with the increase of the ground deposition density of ^{137}Cs within the trace related to a radioactive cloud has been identified for the Fukushima accident. It looks like this is a universal tendency for the ratio of ^{131}I/^{137}Cs in fallout along the trace of a radioactive cloud. This tendency is important for objective reconstruction of ^{131}I fallout based on the results of ^{137}Cs measurements of soil samples carried out at late dates after the Fukushima accident.

Key Words: Fukushima, Chernobyl, fallout, ^{131}I, ^{137}Cs

1. Introduction

The nuclear accident occurred at Fukushima Dai-ichi Nuclear Power Plant (NPP) on March 11, 2011 resulted in release of huge amount of radionuclides into environment and large-scale radioactive contamination of territories. Similar to the accident at the Chernobyl NPP (April 26, 1986), which was the most severe accident ever to have occurred in nuclear industry so far [1], the Fukushima accident was also related to the level 7 of the INES. However, there are differences in these two accidents, including initial reasons of them, amount and dynamics of radioactive releases, as well as radionuclide composition in fallout.

After the Chernobyl accident more than a thousand soil samples taken mainly in the most contaminated territories were measured by gamma-ray spectrometry with a Ge detector within three months after the accident. Analysis of the results of spectrometric measurements allowed for revealing a clear tendency that the ratio of ^{131}I/^{137}Cs in fallout decreases with the increase of the ^{137}Cs ground deposition density. Those two radionuclides are of specific interest because they provide the main contribution to the public exposure (external and internal). The purposes of this paper are: (1) to make a comparative analysis of the radionuclide composition in fallout after the Chernobyl and the Fukushima accidents and (2) to check whether the dependency of the ratio of ^{131}I/^{137}Cs in fallout versus ground deposition density of ^{137}Cs revealed following the Chernobyl accident is confirmed in case of Fukushima accident.

2. Fallout after the Chernobyl accident

At least two different sets of radionuclide composition in fallout were identified for the Chernobyl accident: (1) the near zone of fallout – the Belarusian part of the central spot extended up to 60 km around the Chernobyl NPS formed due to dry deposition on April 26-27, 1986 and (2) the far zone of fallout – the "Gomel-Mogilev" spot centered 200 km to the north-northeast of the damaged reactor formed due to mainly wet deposition on April 28-29, 1986 [1, 2]. Those two zones are clearly seen in Figure 1 presenting the ground deposition density of ^{137}Cs released in the Chernobyl accident [1, 2].

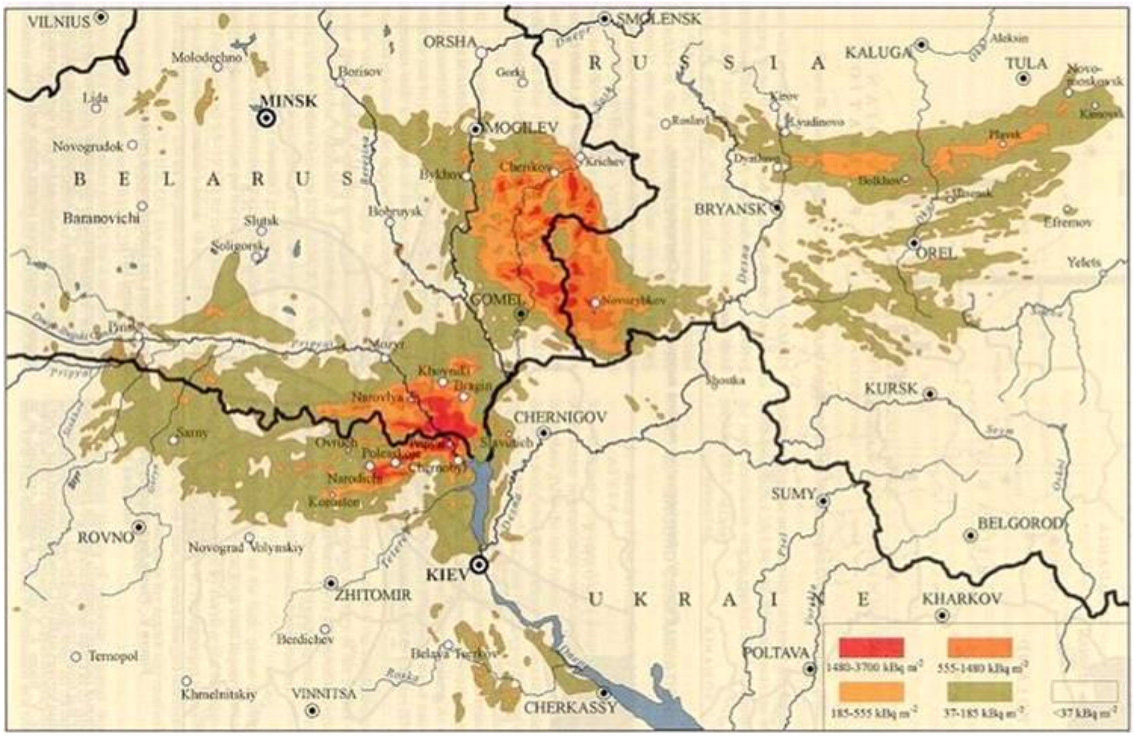

Fig.1. Ground deposition density of ^{137}Cs released in the Chernobyl accident [1, 2].

Analysis of the ratio of various radionuclides to ^{137}Cs showed that the deposition in the near zone reflected the radionuclide composition of the fuel. For that zone the average ratios of such refractory radionuclides as ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{141}Ce, ^{144}Ce to ^{137}Cs in ground deposition were in the range of 2-5; the ratios of ^{103}Ru and ^{106}Ru to ^{137}Cs is about 3 and 1, respectively; the ratios of ^{140}Ba and ^{140}La to ^{137}Cs is about 3, while the average ratios of ^{131}I and ^{132}Te to ^{137}Cs is about 20 [3, 4]. Here and below all the ratios are decay-corrected to the corresponding dates of the main fallout following the accident.

The "Gomel-Mogilev" spot in the far zone of fallout is characterized with a high contamination from ^{137}Cs occurred primarily on rainfall at the time the plume passed over. For that zone the average ratios of refractory radionuclides to ^{137}Cs in ground deposition are much less than that in the near zone and were determined in the range of (0.06-0.11); the average ratios of ^{103}Ru and ^{106}Ru to ^{137}Cs is about 2 and 0.7, respectively; the average ratios of ^{140}Ba and ^{140}La to ^{137}Cs is about 0.7, while the average ratios of ^{131}I and ^{132}Te to ^{137}Cs is about 10 [3, 4].

In order to analyze the dependency of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of ^{137}Cs in fallout only results of spectrometric measurements expressed in [Bq m^{-2}] measured within two months (earlier than June 24, 1986) following the accident were taken into account for each of two zones. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs ground deposition density in the near zone and far zone is shown in Figures 2 and 3, respectively. A general tendency that this ratio decreases with the increase of the level of ^{137}Cs in fallout is clearly seen.

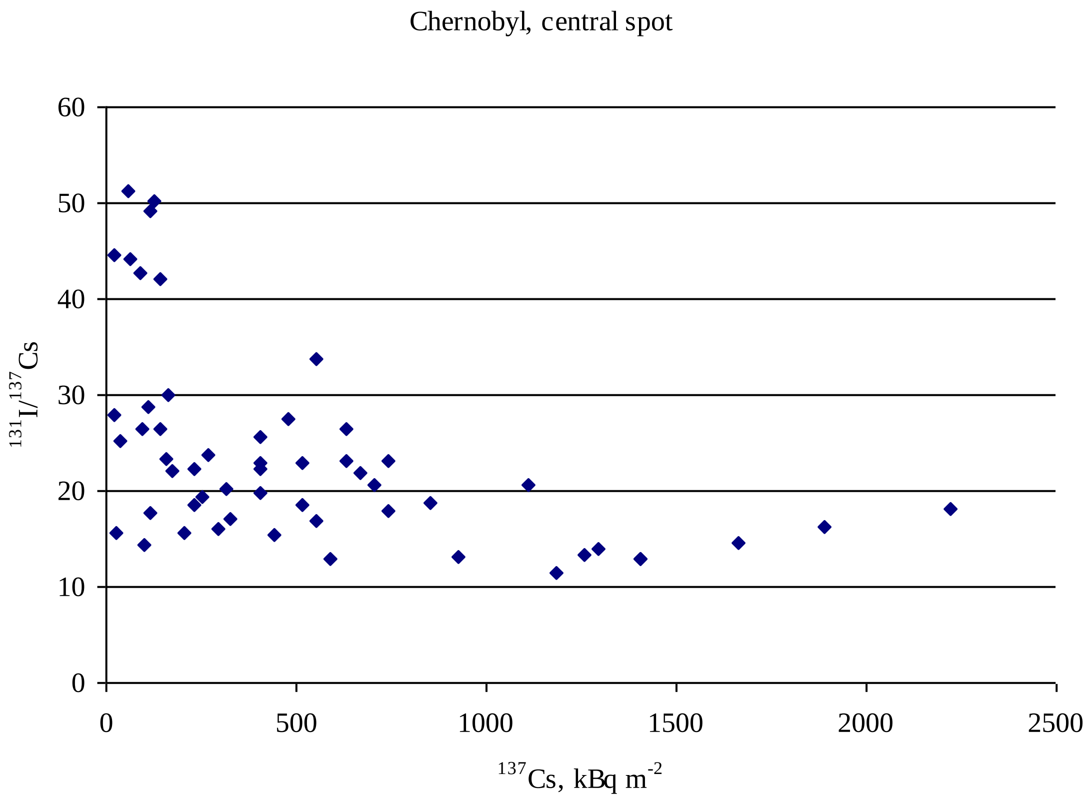

Fig.2. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs ground deposition density in the near zone following the Chernobyl accident.

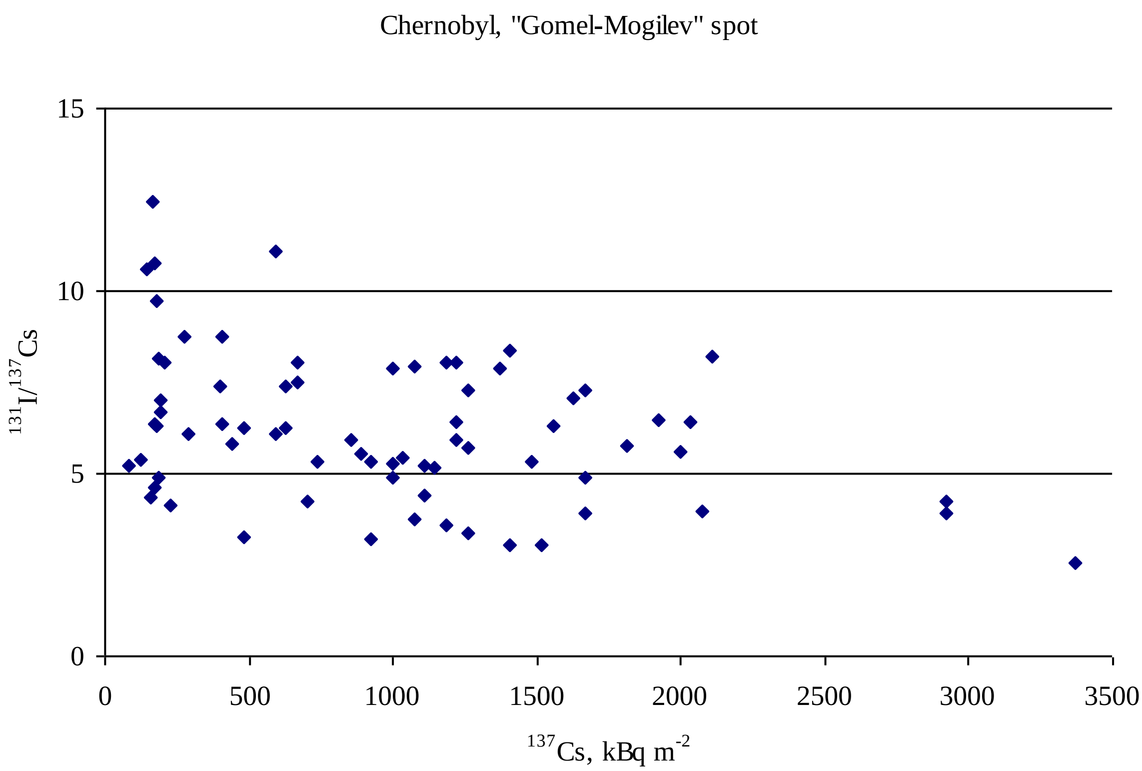

Fig.3. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs ground deposition density in the far zone following the Chernobyl accident.

3. Fallout after the Fukushima accident

In the case of Fukushima accident the results of spectrometric measurements published by Japanese specialists [5] who collected soil samples at the end of March 2011 in the near zone at distances (30-45) km from the Fukushima NPP at the north part on the northwestern radioactive trace mainly formed on March 15, 2011 due to wet deposition [6] were considered.

In the considered soil samples taken in the near zone fallout of the Fukushima accident such refractory radionuclides as ^{95}Zr, ^{95}Nb, ^{141}Ce, ^{144}Ce have not been reliably identified though peaks related to ^{99}Mo were recognized. It is very different from the Chernobyl fallout where ratio of such refractory radionuclides as ^{95}Nb and ^{144}Ce to ^{137}Cs in soil samples collected especially in the near zone was observed to have been greater that 10, while for the far zone those ratios were lower by one-two orders of magnitude. Radioactive isotopes of ruthenium were not revealed in the Fukushima fallout while in the Chernobyl fallout in both near and far zones the ratio of ^{103}Ru/^{137}Cs and ^{106}Ru/^{137}Cs were in the range (0.5-10) for the majority of soil samples. Ratio of ^{134}Cs/^{137}Cs is about (0.8-0.9) for the Fukushima fallout and on average about 0.5 for the Chernobyl fallout. Low content of ^{140}La and ^{140}Ba was identified in the Fukushima soil samples but in the major part of the Chernobyl samples the ratios of these radionuclides to ^{137}Cs exceed unity.

Investigation of the ratio of ^{131}I/^{137}Cs in fallout versus the level of the ^{137}Cs ground deposition density is of specific interest. Analysis of this dependency applied to a limited number of soil samples taken at the end of March 2011 [5] shows that in the range of ^{137}Cs fallout from 0.6 to 2.2 MBq m^{-2} the considered ratio decreases from 13 to 8. In order to involve more data into the analysis the results of spectrometric measurements of concentration of ^{131}I and ^{137}Cs in soil samples taken in the near zone in various directions from the Fukushima Dai-ichi NPP and measured from the end of March to the end of May 2011 were considered [7]. A map of the ^{137}Cs ground deposition density around the Fukushima Dai-ichi NPP is presented in Figure 4 [8].

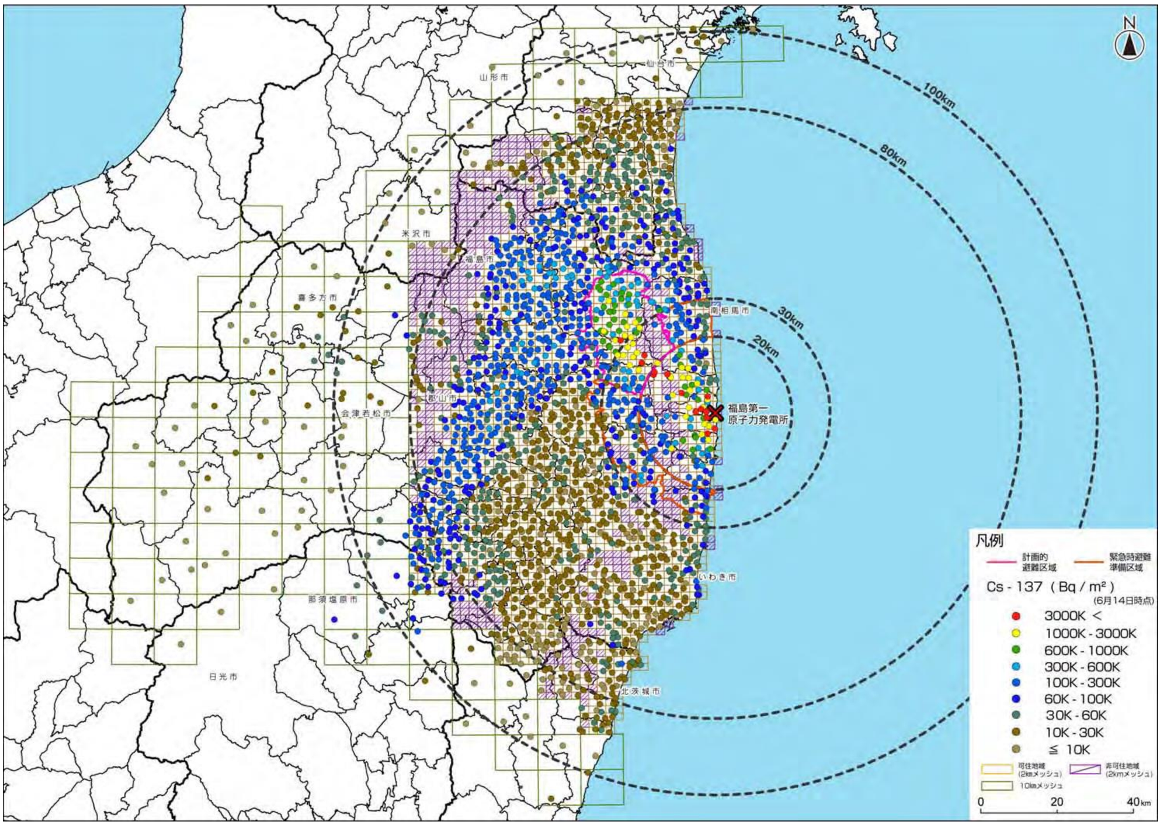

Fig.4. A map of the ^{137}Cs ground deposition density around the Fukushima Dai-ichi NPP [8].

According to available information all soil samples were (0-5) cm depth [7]. It implies a strong correlation between the results of actual measurements expressed in units [Bq kg^{-1}] and ground deposition density of each radionuclide in [Bq m^{-2}]. Analysis of a large bulk of spectrometric measurements of ^{131}I and ^{137}Cs concentration in soil samples available in [7] showed a similar tendency that was revealed for the Chernobyl accident according to which the ratio of $^{131}I/^{137}Cs$ in fallout decreases with the increase of the ground deposition density of ^{137}Cs. Examples of this dependency shown for soil samples taken in various directions from the Fukushima Dai-ichi NPP are given in Figures 5 through 8. The activity of ^{131}I was decay-corrected from the date of measurement to the date of the main fallout which was taken to be March 15, 2011 [6].

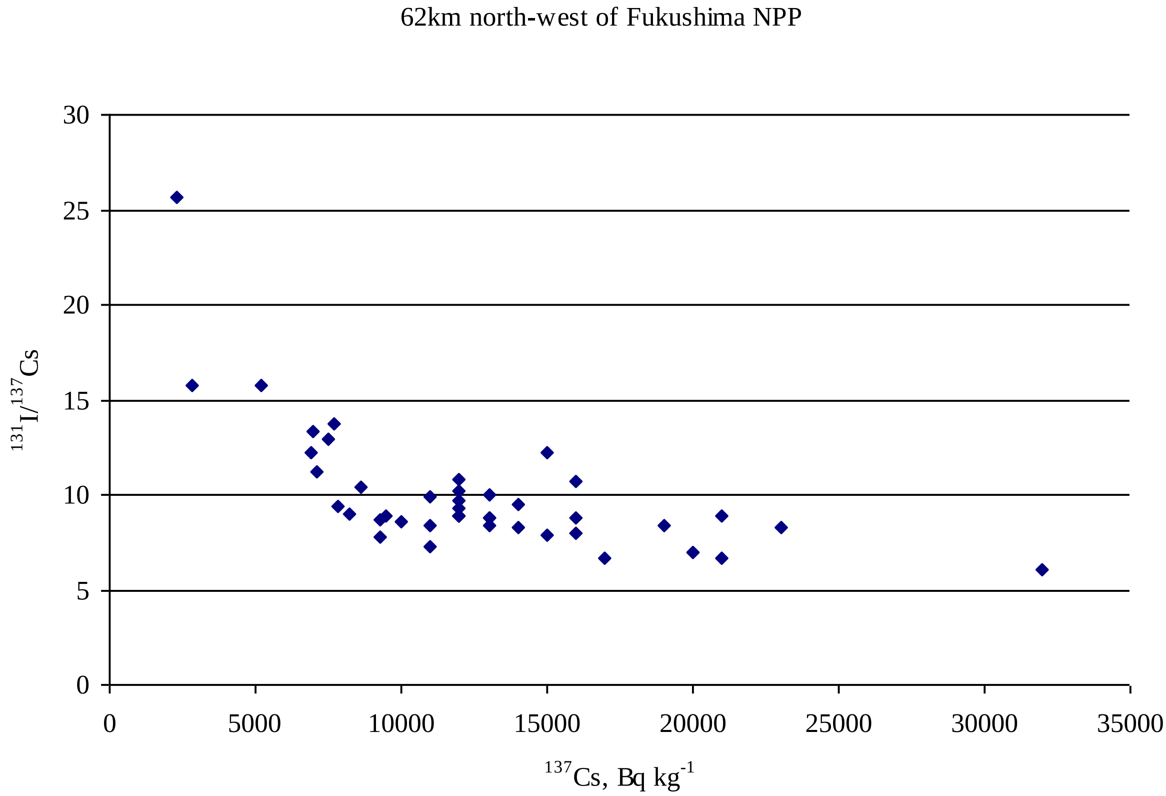

Fig.5. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs concentration in soil samples at 62 km north-west of the Fukushima Dai-ichi NPP.

4. Discussion

The most important distinction in the radionuclide composition of fallout after the Fukushima accident compared to that after the Chernobyl accident is a trace concentration of the refractory radionuclides in the Fukushima fallout, while for the Chernobyl fallout in the near zone the radionuclide composition is close to that in fuel. It can be explained by the fact that the reactors at the Fukushima Dai-ichi had containments which despite of their damage preserved substantial release of refractory elements into environment.

Similar to the Chernobyl accident a clear tendency that the ratio of $^{131}I/^{137}Cs$ in fallout decreases with an increase of the ground deposition density of ^{137}Cs within the trace related to a radioactive cloud has been identified for the Fukushima accident. It looks like this is a universal tendency for the ratio of $^{131}I/^{137}Cs$ in fallout along the trace of a radioactive cloud. The main reason for that can be explained by different efficiency of the scavenging processes from the atmosphere to the ground for ^{131}I and for ^{137}Cs, both for dry and for wet deposition.

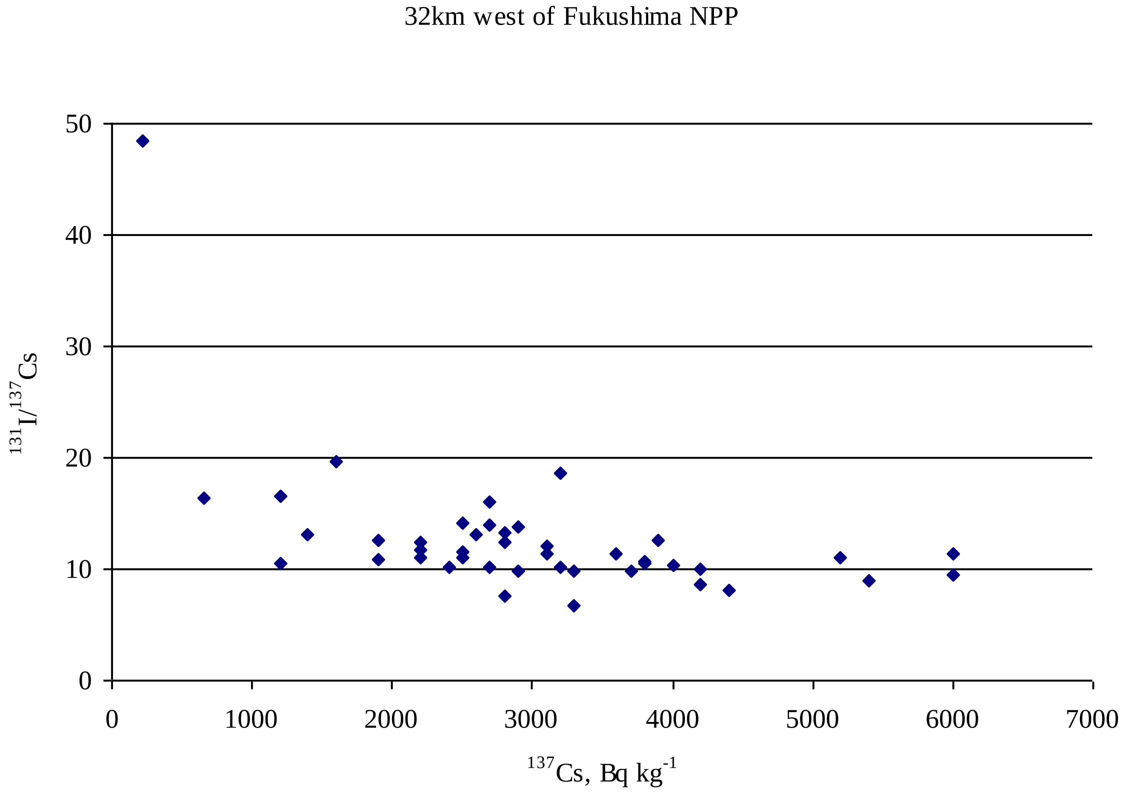

Fig.6. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs concentration in soil samples at 32 km west of the Fukushima Dai-ichi NPP.

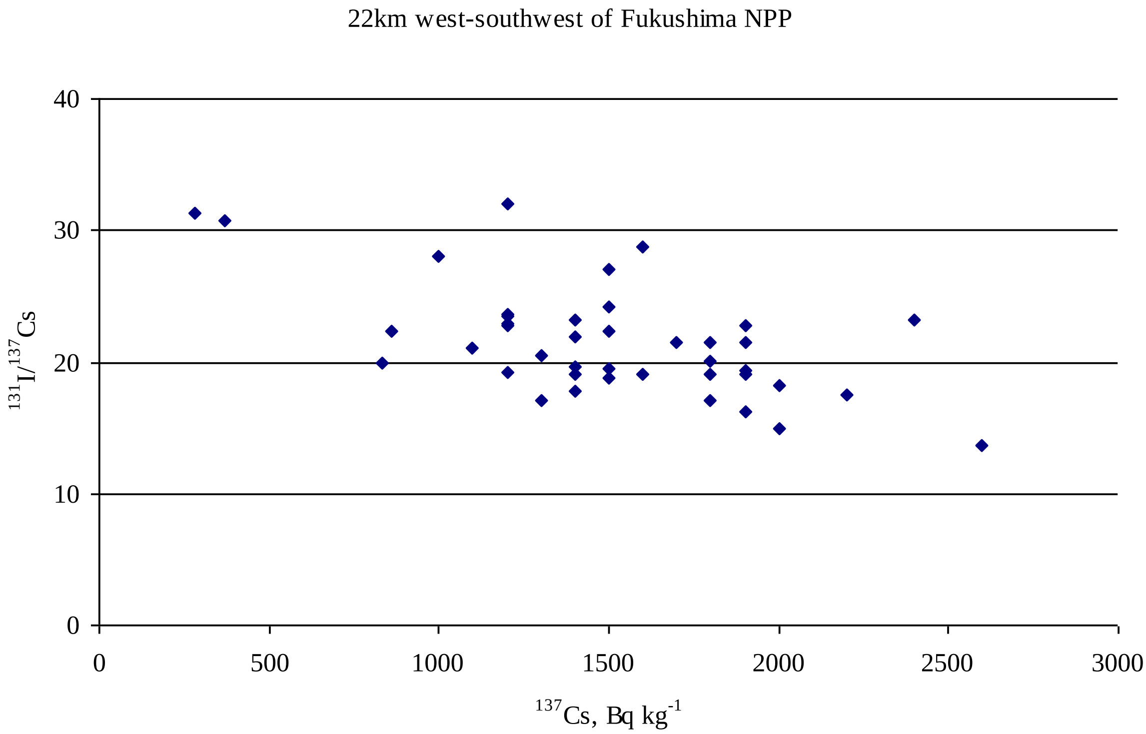

Fig.7. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs concentration in soil samples at 22 km west-southwest of the Fukushima Dai-ichi NPP.

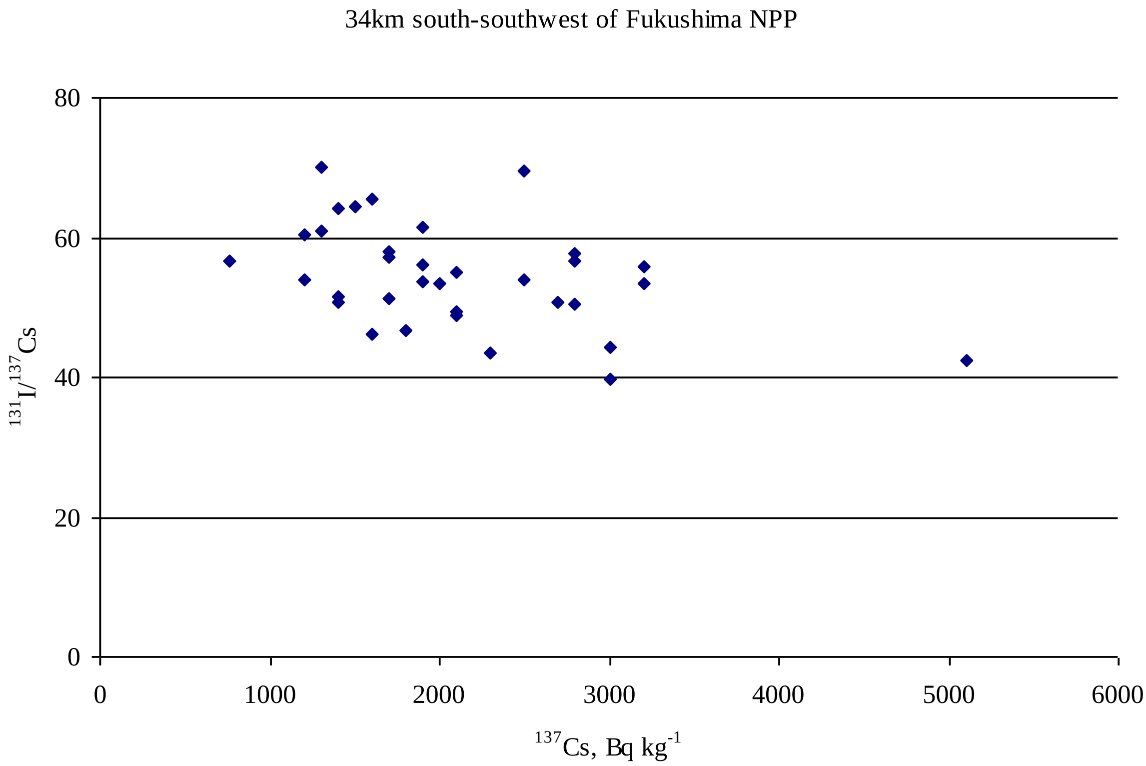

Fig.8. Variation of the ratio of $^{131}I/^{137}Cs$ in fallout versus the level of the ^{137}Cs concentration in soil samples at 34 km south-southwest of the Fukushima Dai-ichi NPP.

Cesium was likely to be predominantly attached to aerosols, while iodine was present in various forms: (1) elemental, which would be deposited more readily than aerosols in the case of dry deposition, (2) organic, which would deposit much less than aerosols, and (3) aerosols. The dry deposition velocity of elemental iodine is estimated to be greater than that of aerosols by a factor of 6 to 10, while the deposition velocity of organic iodine is by a factor of 10 less than that of aerosols [9]. A substantial part of ^{131}I might have been in a gaseous form. The measurements conducted at the distance of about 210 km from Fukushima NPP at the Japan Chemical Analysis Center in the Chiba Metropolitan Area showed that the ratio of gaseous iodine to total iodine in the air was in the range of (0.52-0.71) [10]. It is worth noting that under wet deposition the efficiency of scavenging processes from the air to the ground for aerosols is much higher than that for gaseous. So, in case of wet deposition, which results in a higher level of ^{137}Cs, the ratio of $^{131}I/^{137}Cs$ decreases compared to that under dry deposition.

5. Conclusion

In the near zone of the Fukushima accident such refractory radionuclides as ^{95}Zr, ^{95}Nb, ^{141}Ce, ^{144}Ce have not been reliably identified. For the Chernobyl fallout the activity of refractory radionuclides in soil samples is substantial, especially for samples collected in the near zone, for example, ratios of $^{95}Nb/^{137}Cs$ and of $^{144}Ce/^{137}Cs$ (decay-corrected to the date of the main fallout) reached 10 and even more, while for the far zone those ratios were lower by one-two orders of magnitude. Radioactive isotopes of ruthenium were not revealed in the Fukushima fallout while in the Chernobyl fallout in both near and far zones the ratios of $^{103}Ru/^{137}Cs$ and $^{106}Ru/^{137}Cs$ were in the range (0.5-10) for the majority of soil samples. Ratio of $^{134}Cs/^{137}Cs$ is about (0.8-0.9) for the Fukushima fallout and on average about 0.5 for the Chernobyl fallout. Low content of ^{140}La and ^{140}Ba was identified in the Fukushima soil samples but in the major part of the Chernobyl samples the ratios of these radionuclides to ^{137}Cs exceed unity.

With respect to exposure to the public the most important radionuclides are ^{131}I and ^{137}Cs. For the both accidents the ratios of $^{131}I/^{137}Cs$ in the considered soil samples are in the similar ranges: (3-50) for the Chernobyl samples and (5-70) for the Fukushima samples. Similar to the Chernobyl accident a clear tendency that the ratio of $^{131}I/^{137}Cs$ in fallout decreases with the increase of the ground deposition density of ^{137}Cs within the trace related to a radioactive cloud has been identified for the Fukushima accident. It looks like this is a universal tendency for the ratio of $^{131}I/^{137}Cs$ in fallout along the trace of a radioactive cloud. This tendency is important for objective reconstruction of ^{131}I fallout based on the results of ^{137}Cs measurements of soil samples carried out at late dates after the Fukushima accident.

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