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Determination of ^{129}I and ^{127}I Concentration in Soil Samples from the Chernobyl 30-km Zone by AMS and ICP-MS

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Chernobyl Soil/^{129}I/AMS/ICPMS/Iodine.

A large amount of radioiodine isotopes (mainly ^{131}I, $t_{1/2}$ = 8 days) was released from the accident at Chernobyl Nuclear Power Plant (CNPP) in April-May 1986. An increase in childhood-thyroid cancer in the contaminated areas in Belarus, Russia and the Ukraine was demonstrated to be caused by radioiodine released at the time of the accident. However, there is a lack of quantitative data on the ^{131}I levels in the local environment (e.g. air, plant, soil). At this point, a long-lived iodine isotope, ^{129}I ($t_{1/2}$ = 15.7 million years), also released with a certain ratio to ^{131}I from CNPP, could be used for estimating the ^{131}I levels in the environment. In this paper we present analytical results of the ^{129}I concentrations and ^{129}I/^{127}I atom ratios in soil samples collected from the CNPP exclusion zone (30-km zone), with the aim of assessing current contamination levels and distribution patterns. For the analysis of the iodine fraction in the investigated soil samples, the pyrohydrolysis method was utilized for separation of ^{127}I and ^{129}I nuclides, and subsequently their concentration was determined using inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS), respectively. The concentration of ^{129}I and the ^{129}I/^{127}I atom ratio in the surface soil samples in the 30 km-zone of CNPP ranged from 4.6 to 170 mBq/kg, and from 1.4×10^{-6} to 13×10^{-6}, respectively. These values are significantly higher than those from global ^{129}I fallout, indicating that most of the measured ^{129}I was due to the deposition of the accident. Stable iodine concentrations in this area were found to be very low (below 1 ppm) for most of the samples, suggesting the environmental iodine levels in this area to be potentially low. The ^{129}I/^{137}Cs activity ratio in surface and sub-surface soils was not so constant, i.e., in the range $(7.3–20.2) \times 10^{-7}$. This might be due to the different behavior of deposition and/or migration of these nuclides in soil. These results suggest the obtained data of ^{129}I to be useful for the reconstruction of the ^{131}I deposition in the contaminated areas.

INTRODUCTION

Iodine is an important nutrient element with one stable isotope, ^{127}I, one long-lived radioisotope, ^{129}I ($T_{1/2}$ 15.7 Ma) and several short-lived radioisotopes, e.g., ^{125}I ($T_{1/2}$ 60 d) and ^{131}I ($T_{1/2}$ 8.04 d).[1)] Iodine is concentrated in the thyroid gland of the human body. Following the accident of Chernobyl Nuclear Power Plant (CNPP) in April 1986, a large amount of radionuclides were released into the environment. Because most of the short-lived iodine isotopes (e.g. ^{132}I, ^{133}I) disintegrated to non-iodine progeny before they were transported through the atmosphere and incorporated into the thyroids of humans, the major hazard is expected from ^{131}I. This nuclide was detected widely near the vicinity of the accident site.

An increasing number of childhood-thyroid cancers was observed in Belarus and the Ukraine after some years of the accident.[2,3)] This is thought to be predominantly due to ^{131}I released from CNPP. However, there is a lack of data on ^{131}I as well as on its distribution pattern and deposition, especially in those regions where the incidence of thyroid cancer among children was found to be increasing. There are also not enough quantitative data to understand the relationship between the occurrence of thyroid cancer and the doses due to ^{131}I. Nowadays, it is not possible to measure ^{131}I derived from CNPP, due to its short half-life.

In order to assess the ^{131}I concentration levels following

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the accident, data on ^{137}Cs ($T_{1/2}$ = 30 y) was often used as an analogue in the contaminated areas. Since the chemical behavior of iodine and cesium in the environment is different, however, it is difficult to estimate the ^{131}I levels from those of ^{137}Cs. In the present paper we take advantage of a long-lived radioiodine, ^{129}I, which was also produced by fission in the reactor and released into the atmosphere together with ^{131}I during the accident. Ermilov *et al.* estimated the ^{129}I/^{131}I ratio in the reactor at Chernobyl accident to be 11–15 as atomic ratio.4) The ^{129}I is expected to remain after other short-lived isotopes of iodine have decayed to insignificant levels. Since iodine is accumulated in soil, the ^{129}I level in surface soil is expected to be elevated in those areas contaminated with Chernobyl fallout. Therefore, if one can measure ^{129}I concentrations in soil samples collected from a contaminated area, one may estimate the ^{131}I levels at the time of the accident and, in this way, contribute in reconstructing radiation doses through the ^{129}I data. ^{129}I is produced in nature by spontaneous fission processes in uranium and thorium within the Earth's crust and by cosmic-ray induced spallation of xenon in the atmosphere. Large amounts of ^{129}I and other fission products were released into the environment, due to atmospheric testing of nuclear weapons mainly in the late 1950s and early 1960s. The inventory of natural ^{129}I is approximately in the range of 100–260 kg.5) Atmospheric bomb testing added approximately 50 to 150 kg,6,7) while the Chernobyl accident contributed less than 2 kg.8,9) In addition, a vast amount of man-made ^{129}I has been released chronically into the environment by reprocessing plants.10,11)

Increase of the ^{129}I concentration in the environment can be quantified in terms of the ratio of ^{129}I/^{127}I. Prior to 1945, levels of the ^{129}I/^{127}I in the hydrosphere and biosphere showed in between 5×10^{-12} and 2.5×10^{-10}.12,13) The pre-anthropogenic ^{129}I/^{127}I ratio in recent sediments is reported as 1.5×10^{-12}.14) After man-made ^{129}I has entered the environmental iodine cycles, the global ^{129}I/^{127}I background ratio has been increased to the range 10^{-10}–10^{-5}. Recently it was reported by Rao and Fehn that most of the artificial ^{129}I was retained in the surface soil layer, and as a result 85 to 90% of ^{129}I is still present within the top 15 cm of soil, due to the high sorption characteristics of soil for iodine.15) Therefore, pre-Chernobyl ^{129}I must be taken into account for estimation of ^{131}I in the Chernobyl area.

Retrospective evaluation of ^{131}I deposition density and thyroid dose has been carried out in Poland through measurement of the ^{129}I concentration in the environmental samples.16) Therefore, it would be highly advantageous if a geographical distribution of ^{129}I (or the ^{129}I/^{127}I ratio) in regions contaminated by the Chernobyl accident be established, enabling the reconstruction of the Chernobyl ^{131}I dose from the ^{129}I data. There are some reports on ^{129}I in the literature that describe such efforts.17–20) However, the number of analytical results published so far is not sufficient to understand the levels and distribution of ^{129}I in the contaminated areas. As far as the detection of ^{129}I is concerned, the development of AMS is preferred over neutron activation analysis, due to its high sensitivity for detection of ^{129}I at low concentrations.21–23) Recently, we have developed analytical procedures for ^{129}I by using AMS at the Micro Analysis Laboratory, Tandem Accelerator, MALT facility of the University of Tokyo.24,25) This technique was used in the present study, to analyse ^{129}I/^{127}I ratios in Chernobyl soil samples. Since accumulation of radioiodine in the thyroid depends on stable iodine consumption, we have also measured concentrations of stable ^{127}I in the same soil samples using ICP-MS.

The main aim of the present study is to obtain information on the ^{129}I levels in soil samples collected in the 30-km zone around the Chernobyl reactor and explore the possibility to reconstruct the ^{131}I deposition at the time of the accident. Concentrations of stable iodine in soils of the Chernobyl area are also studied.

MATERIALS AND METHODS

All soil samples were collected within the 30-km CNPP zone using a cylindrical sampler with a diameter of 37 mm. The sampling depth was 20 cm, and the soil layers taken were from depths 0–2; 2–4; 4–6; 6–8; 8–10; 10–15; and 15–20 cm. Each sample consisted of a mixture of 5 sub-samples collected at a distance of 2–5 m between each other. The soil samples were dried, thoroughly homogenized followed by sieving prior to chemical analyses. Soil conditions around "Chernobyl 30-km zone" are characterized by dominating soddy podzolic soils (36.4% of total surface). Soil forming rocks are alluvial and fluvioglacial sedimentations of sandy and consolidated sandy-mechanic composition. These types are characterized by water penetration, and consequently precipitation removed nutrients from soil. Thus, soil fertility is low. Such territories are usually covered by forests. The S1 soil sample is slightly soddy-podzolic and of almost dust-forming sandy type. It was taken about 12 km from the CNNP. The S3 and S4 sampling places are located near the red forest. Sampling point S8 is located over a beak-water terrace of an ancient valley of a glacial water flow. There the soil is of break-water (creek) type, and peat soddy gleyic sandy loam. Sampling point S10 is characterized by soddy and slightly-podzolic consolidated sandy soil.

Three sets of soil samples were used for the analysis of ^{129}I. The first set of samples (S1, S3, S4, S8 and S10 in Fig. 1) collected in 2001 from forests near the Pripyat river and the Red Forest area. The sampling distances from the CNPP were between 4 and 12 km. We have selected S8 for a vertical profile study from surface to depth of 20 cm as a second set of samples, was located about 8 km north of the CNPP, and its uranium isotope ratios were already reported elsewhere.26) The soil is very sandy, almost without stones, and its clay and humus contents are low. The third set of

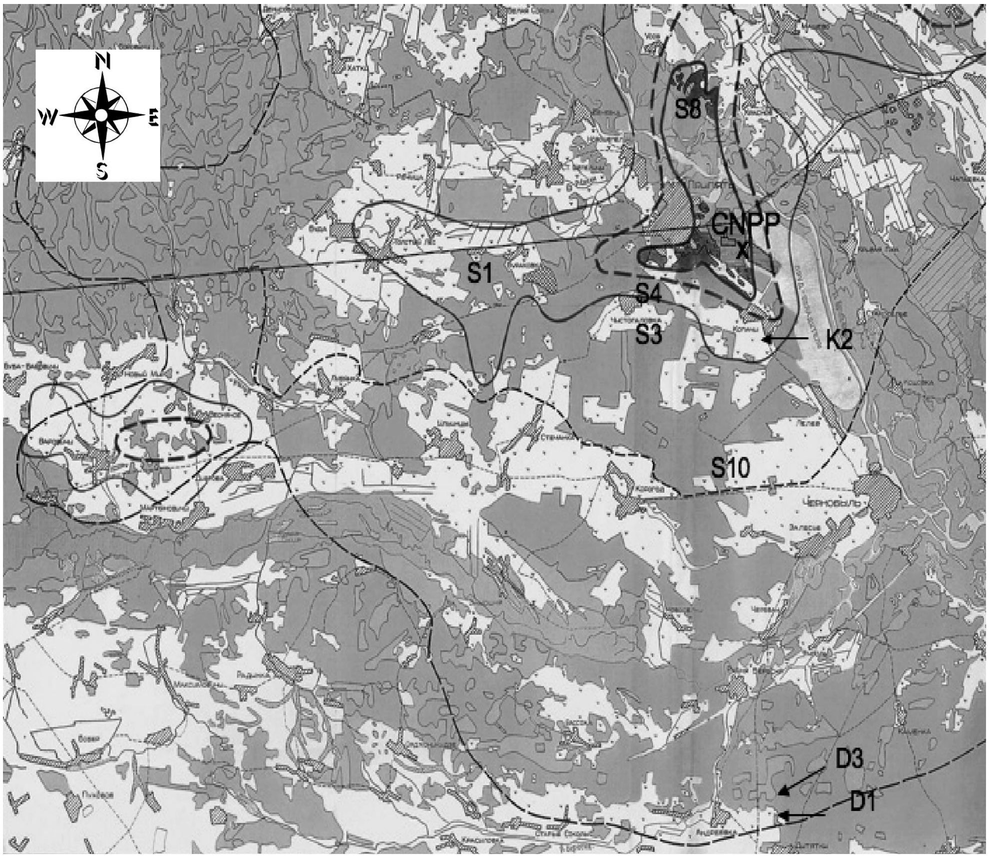

Fig. 1. Map showing Sampling stations.

samples was also forest soil collected in 1994/95 from Kopachy (6 km south east of the CNPP, K2 in Fig. 1), Dityatki-1 (28.5 km south of the CNPP, D1) and Dityatki-1 (26 km south of the CNPP, D3). These samples had originally been collected by a German group and used for ^{137}Cs analysis[27] and later for ^{239}Pu and ^{240}Pu analysis.[28] Composite soil samples, i.e., mixtures of L (litter), Of (fermentated organic layer) and Oh (humificated organic layer), were prepared in consideration of the depth of each layer.

The analytical methods used in this study include: (i) separation of iodine by pyrohydrolysis, (ii) measurement of stable iodine by ICP-MS, and (iii) a sequential solvent extraction and back extraction and a final preparation of AgI targets for the determination of ^{129}I/^{127}I ratios by AMS.

The separation procedure of iodine from soil used in this study is based on a method developed by Schnetger and Muramatsu,[29] Muramatsu and Wedepohl[30] and Chai and Muramatsu.[31] A mass of about 1 g of soil is mixed with V_2O_5 in a ceramic boat and placed in a quartz tube. The sample is then heated at 1000°C under a flow of oxygen gas. Finally, the evaporated iodine is collected with a trap containing TMAH (tetramethyl ammonium hydroxide: 1%) and Na_2SO_3 (0.1%) solutions.

For the analysis of stable iodine, part of the trap solution was taken and diluted to a concentration range between 5–100 ppb I using de-ionized water with TMAH (0.5%) and an internal standard (Cs: 20 ppb) being added. The diluted sample solution was determined for iodine by ICP-MS (Agilent 7500) using mass of 127. Detection limit of iodine by this method in soil sample (0.5 g) is about 5 ppb (ng/g).

For the analysis of ^{129}I, iodine carrier (I^-: 5–10 mg) and Na_2SO_3 solution (1 ml of 2% solution) was added to the trap solution. Then the iodine fraction was purified by solvent extraction and subsequent back extraction. For this, carbon tetrachloride was used as a solvent. By the addition of sodium nitrite solution (1%) under acidic condition, iodide was transformed to elemental iodine (I_2) and extracted into organic phase. Iodine was back extracted into aqueous solution (containing Na_2SO_3) as I^-. After NaCl (5 mg) was mixed, a silver nitrate solution was added to co-precipitate chloride and iodide ions. Finally, the precipitate was washed with ammonia solution to dissolve AgCl. The obtained AgI precipitation was dried under an infrared lamp (or in an oven at 70°C) and mixed with Ag powder and pressed into the cathode cone to prepare AMS target. Measurements of ^{129}I/^{127}I ratio were performed at MALT (Micro Analysis Laboratory, Tandem accelerator), the University of Tokyo. In this system, we have selected 4.0 MV for the accelerator terminal voltage and charge state 7+ and used a gas ionization detector for the final detection. We have also made a specific set up of electrostatic cylindrical analyzer (ECA) to reduce ^{127}I interference, which is the most serious interference for the detection of ^{129}I. The system background achieved was $^{129}\text{I}/^{127}\text{I} < 3 \times 10^{-14}$. Details of the separation procedures of iodine fraction from soil samples were described in Muramatsu *et al.*[25] and those for the AMS measurement conditions were described in Matsuzaki *et al.*[24]

RESULTS AND DISCUSSION

The analytical results obtained for the sample sets 1–3 collected from the Chernobyl 30-km zone are shown in Tables 1–3. The results are discussed in the following sequence: (1) stable iodine (^{127}I) concentrations, (2) ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios, and (3) relevance of the measured ^{129}I concentrations to those of ^{137}Cs and ^{131}I.

Table 1. Concentration of stable iodine and ^{129}I in Chernobyl soil (sample set 1) Sample locations are shown in Fig. 1. Surface soil samples (0–5 cm) were used.

Sample point	Distance from CNPP (km)	Iodine concentration (ppm)*	^{129}I concentration (mBq/kg)**	$^{129}\text{I}/^{127}\text{I}$ atom ratio
S1	12 km W	0.53	4.6	1.3E-06
S3	4 km SW	0.97	12.7	2.0E-06
S4	4 km SW	4.13	129	4.7E-06
S8	8 km N	0.21	46.6	3.3E-05
S10	12 km S	0.42	22.7	8.1E-06

(Note) * Mean of two determinations, error range was in most cases within 5%.
** Standard error by AMS measurements was approximately 5%.

Table 2. Vertical distribution of stable iodine and ^{129}I in Chernobyl soil (sample set 2) Sample location is shown in Fig. 1.

Sample Point	Depth (cm)	Iodine concentration (ppm)*	^{129}I concentration (mBq/kg)**	$^{129}\text{I}/^{127}\text{I}$ atom ratio
S8 0–2	0–2	0.209	49.2	3.5E-05
S8 2–4	2–4	0.212	46.6	3.3E-05
S8 4–6	4–6	0.177	39.3	3.3E-05
S8 6–8	6–8	0.162	20.5	1.9E-05
S8 8–10	8–10	0.127	9.7	1.2E-05
S8 10–15	10–15	0.119	3.9	4.9E-06
S8 15–20	15–20	0.103	1.9	2.8E-06

(Note) * Mean of two determinations, error range was in most cases within 5%.
** Standard error by AMS measurements was approximately 5%.

Concentrations of stable iodine in soil

The detection limit for stable iodine by the proposed method is about 0.1 ng/ml in solution, or about 2 ppb (0.002 ppm) for a solid sample of 1 g. In order to examine the analytical accuracy, we have also analyzed standard reference materials (e.g. JLK-1, JR-1) for which iodine values are reported, and a good agreement between the measured and the recommended (or certified) values was obtained.[31]

The stable iodine concentrations in the first sample set (Table 1) range from 0.24 to 4.13 ppm. The iodine concentrations in most of the samples are less than 1 ppm except one, and the mean concentration is 0.54 ppm. The second sample (Table 2) is mostly of podzol soil type, and the iodine concentrations were found to be very low and fairly uniform with depth, i.e., they range from 0.10 to 0.21 ppm with a mean of 0.16 ppm-from all seven layers. In the third sample set (Table 3), the iodine concentration range from 0.48 to 3.0 ppm (mean 1.7 ppm). We also analyzed stable iodine concentrations in IAEA-375 (Chernobyl soil) and obtained 1.47 ± 0.08 ppm (± standard deviation, one σ of 4 independent determinations). For the stable iodine concentration, there is no certified value reported by IAEA for this material, although there is a certified value for ^{129}I (see below). The above-mentioned results for stable iodine concentrations in the different Chernobyl soils indicate that the iodine levels in this region are markedly lower than those from other areas. For example, the world average value is ~5 ppm[32] and the average for Japanese upland soils (andosol soil type) is about 30 ppm.[33,34] Given the fact that there exists only few data on iodine in soils of the Chernobyl area, our results provide useful information to understand the levels of this element in this region: for example that the Chernobyl area is deficient in iodine in soil, and thus agricultural products may also be low in iodine. This in turn suggests that the iodine daily intake by the inhabitants of this region might be very low. Shiraishi *et al.*[35] reported - through analysis of total diets collected from 106 locations in the Northwest of the Ukraine that was contaminated by the Chernobyl accident - that daily intake of iodine is in the range of 0.0028–0.372 mg (median value: 0.028 mg) which is significantly smaller than the ICRP Reference Man value of 0.2 mg per day, and also below the level of the recommended dietary intake value (RDI) of 0.15 mg per day (ICRP 1975).[36] Note that it might be possible that deficiency of iodine could result in an increased radioiodine transfer rate into thyroid after the Chernobyl accident, which in turn could be one reason for the higher occurrence of childhood-thyroid cancer in this area.

^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios in soil

The detection limit of AMS for $^{129}\text{I}/^{127}\text{I}$ isotope ratio is in the order of 10^{-14},[23] which is much better than that by

Table 3. Concentrations of stable iodine, ^{129}I, ^{137}Cs and Pu in Chernobyl soil (sample set 3) and their inter-nuclide ratios.

(a)

Sample	Distance from CNPP	Iodine concentration (ppm)*	^{127}I (atom/kg)	^{129}I concentration (mBq/kg)**	^{129}I (mBq/m^2)	^{129}I (atom/kg)	^{129}I/^{127}I ratio
K2 L/Of/Oh 94	6 km S	2.12	1.01E+19	168	874	1.20E+14	1.20E-05
K2 L/Of/Oh 95	6 km S	1.75	8.33E+18	149	775	1.07E+14	1.28E-05
K2 Ah/B 94/95	6 km S	0.48	2.28E+18	12.7	1100	9.12E+12	4.00E-06
D3 L/Of/Oh 95	26 km S	3.00	1.42E+19	25.0	110	1.79E+13	1.26E-06
D3 B-94/95	26 km S	1.80	8.55E+18	2.4	89.0	1.72E+12	2.01E-07
D1 L/Of/Oh 95/94	29 km S	1.74	8.28E+18	16.9	74.5	1.21E+13	1.46E-06
D1 Ah/B 94/95	29 km S	1.09	5.18E+18	1.7	147	1.21E+12	2.34E-07

Note: Details of the sampling places and some other information are described elsewhere.[28)]

(b)

Sample	Density (kg m^{-2})	^{137}Cs (kBq/kg)	^{137}Cs (atom/kg)	$^{239+240}$Pu (Bq/kg)	^{129}I/^{137}Cs atom ratio	^{129}I/^{137}Cs activity ratio	^{129}I/ $^{239+240}$Pu activity ratio	^{137}Cs/ $^{239+240}$Pu activity ratio
K2 L/Of/Oh 94	5.2	230	3.15E+14	1430	0.38	7.31E-07	1.18E-04	161
K2 L/Of/Oh 95	5.2	179	2.45E+14	1130	0.43	8.32E-07	1.32E-04	158
K2 Ah/B 94/95	86.5	11.4	1.56E+13	76.7	0.58	1.12E-06	1.66E-04	149
D3 L/Of/Oh 95	4.4	20.0	2.74E+13	172	0.65	1.25E-06	1.45E-04	116
D3 B-94/95	37.1	1.8	2.53E+12	13.4	0.68	1.30E-06	1.79E-04	138
D1 L/Of/Oh 95/94	4.4	19.1	2.62E+13	137	0.46	8.86E-07	1.24E-04	139
D1 Ah/B 94/95	86.9	0.84	1.15E+12	6.3	1.06	2.02E-06	2.69E-04	161

(Note) * Mean of two determinations, error range was in most cases within 5%.

** Standard error by AMS measurements was approximately 5%.

neutron activation analysis (NAA). Since the method requires addition of 1 mg of stable iodide carrier to the sample, the practical detection limit in our measurements works out to be about 1×10^{-11} in terms of the ^{129}I/^{127}I ratio, or about 0.01 mBq/kg in terms of activity concentration, which is below the detection limit for common surface soil samples. We have determined ^{129}I in a standard reference materials supplied by IAEA (IAEA-375: Chernobyl soil) for validation of the analytical method, and obtained the concentration of ^{129}I as 1.64 ± 0.13 mBq/kg (mean of 4 determinations), which agrees well with the recommended value of 1.7 ± 0.4 mBq/kg. The ^{129}I/^{127}I ratio for this material was calculated to be 1.72×10^{-7}. Stable iodine concentration data obtained by ICP-MS were used for the calculation.

In nature, ^{129}I is produced from cosmic-ray induced reactions with atmospheric Xe, and from spontaneous fission of uranium. The ^{129}I/^{127}I ratio in the pre-atomic age was estimated to be 1.5×10^{-12}.[14,37)] Due to atomic weapons testing, ^{129}I is also produced artificially and added into the environment. As a result, the ^{129}I/^{127}I ratio in the environment increased. Because iodine accumulates readily in soil, surface soil samples show a high ^{129}I level, i.e., commonly in the order of 10^{-10}–10^{-8}, in terms of the ^{129}I/^{127}I ratio. Specifically, surface soil samples from surroundings of nuclear reprocessing plants show ^{129}I/^{127}I ratios up to 10^{-5}. In our previous studies,[38)] we measured ^{129}I in soil samples collected from Nagano Prefecture (Japan), which is far from nuclear facilities, and reported the ^{129}I concentration as 0.13 mBq/kg and the ^{129}I/^{127}I ratio as 6.5×10^{-9}. Mironov *et al.*[18)] have reported ^{129}I concentrations in pre-Chernobyl soils collected from Belarus as 0.094 mBq/kg (mean of 5 samples).

Our results on the ^{129}I levels for the surface soil samples collected from the Chernobyl area are shown in Tables 1–3. The ^{129}I concentrations and the ^{129}I/^{127}I atom ratios range from 1.7–168 mBq/kg and from (0.3–43) × 10^{-6}, respectively. The ^{129}I contents per unit area for the sample set 3 are in the range 74–1100 mBq/m^2 (Table 3). These values are significantly higher than those obtained for samples from background areas or from the pre-Chernobyl era (see discussion below).

The results of the vertical distribution of ^{129}I (Table 2) show that the ^{129}I concentration decreases systematically from the surface into the depth, i.e. from 49.2 mBq/kg (0–2 cm) to 1.9 mBq/kg (15–20 cm). However, there is no significant difference in the concentration of ^{129}I for the first 2 layers (0–2 and 2–4 cm). The ^{129}I/^{127}I ratio also decreases with depth from 3.6×10^{-5} to 2.8×10^{-6}. About 85% of the total inventory of ^{129}I (0–20 cm) is present in the first 6 cm from the surface. This is in a good agreement with earlier reports that most of the ^{129}I deposited after the Chernobyl accident retained in the surface soil layer.[16] The highest ^{129}I concentration (168 mBq/kg) found in this study was in the sample collected from a place 6 km from CNPP (Table 3). However, its ^{129}I/^{127}I ratio (1.2×10^{-5}) was lower than that of the 8 km sample (^{129}I/^{127}I ratio: 3.5×10^{-5} and ^{129}I concentration: 49.2 mBq/kg, as shown in Table 2). This is explained by the lower stable iodine concentration in the later sample.

The use of ^{129}I for the reconstruction of ^{131}I deposition requires the knowledge whether the ^{129}I from the Chernobyl fallout can be distinguished from the non-Chernobyl component of the ^{129}I deposition, which is due to global weapons testing fallout and aerial discharges from nuclear reprocessing plants and subsequent atmospheric transport. Based on the analysis of a pre-Chernobyl 20 cm deep soil (podzol) sample collected in 1985 from a pasture 400 km north of Chernobyl, Mironov *et al.*[18] reported that the average pre-Chernobyl concentrations of ^{129}I and ^{137}Cs were 26 mBq/m^2 and 2.4 kBq/m^2, respectively. It is interesting to note that these pre-Chernobyl ^{129}I and ^{137}Cs concentrations are in agreement with those reported elsewhere,[15] which were based on the analysis of a sample from Ostrovskie (Russia) in 1993, a site which was not contaminated due to CNPP accident. Michel *et al.*[20] estimated the pre-Chernobyl inventory for ^{129}I to be about 44 mBq/m^2 based on samples from the Moscow and Zhitomir districts in Russia. This value is much higher than that estimated for the global weapons fallout inventory of ^{129}I which varies in range from 1.4 to 2.9 mBq/m^2.[12,13,15] There is some discrepancy in pre-Chernobyl ^{129}I concentrations measured in samples from eastern and western Europe which can be attributed to the ^{129}I emissions of reprocessing plants.[39,40] Compared to the levels and variations of the pre-Chernobyl ^{129}I deposition, the fraction of Chernobyl ^{129}I in the samples investigated in the present study is much higher. Therefore it is concluded that it is possible to use the ^{129}I data from the present study for estimation of the ^{131}I deposition in the highly contaminated area, due to the Chernobyl accident.

Relevance of the measured ^{129}I concentrations in terms of ^{131}I and ^{137}Cs concentrations

Both ^{131}I and ^{129}I are produced by the fission of ^{235}U in a nuclear reactor. The amounts of their production are affected mainly by the type of nuclear fuel and its burn-up time. The ^{131}I/^{129}I activity ratio in the fallout at the time of the Chernobyl accident was estimated to be 3.8×10^7 (UNSCEAR2000).[41] Michel *et al.* (2005)[20] performed a detailed literature survey on measured and theoretical data of the ^{131}I/^{129}I ratio following the Chernobyl accident. They reported the best estimated value for the ^{131}I/^{129}I ratio to be $5.3 \pm 0.3 \times 10^7$ in terms of an activity ratio, or 13.6 ± 2.8 in terms of an atom ratio. Using the ^{131}I/^{129}I ratio estimated by Michel *et al.*[20] and the average pre-Chernobyl level (26 mBq/m^2) reported by Mironov *et al.*,[18] we have estimated the ^{131}I deposition based on our ^{129}I levels listed in Table 3 (74.5–1100 mBq/m^2). The range of the ^{131}I deposition at the accident time in this area was obtained to be 2.6–57 mBq/m^2.

Because of the high ^{137}Cs concentrations in those areas contaminated after the Chernobyl accident, the distribution of this nuclide was well studied. Therefore, ^{137}Cs is usually used as a proxy for retrospective ^{131}I dosimetry. The ^{129}I/^{137}Cs activity ratio calculated from the inventory data of UNSCEAR 2000 is 3.1×10^{-7} (or 0.16 as ^{129}I/^{137}Cs atom ratio). Concentrations of ^{137}Cs in the soil samples measured in our previous study are also shown in Table 3b after correction for radioactive decay to May 1986.[27,28] Using these data, we have compared the concentrations of ^{129}I (as a proxy of ^{131}I) in surface soils (L/Of/Oh layer) that were highly contaminated, with those of ^{137}Cs. The resulting ^{129}I/^{137}Cs activity ratio varies from $(0.73–2.0) \times 10^{-6}$ (or as atom ratio from 0.38 to 1.1) which corresponds to a variation factor of about 2.8, within the 30 km zone where the fractions of the pre-Chernobyl ^{137}Cs and ^{129}I to the total inventory are very small and can therefore be neglected. Mironov *et al.*[18] studied the use of ^{129}I and ^{137}Cs in soil for estimation of the ^{131}I deposition in Belarus and mentioned that ^{137}Cs can provide reasonably good estimates (+/–50%) for ^{131}I. In contrast, our results showed a larger variation than their values. Straume *et al.* studied ^{137}Cs and ^{129}I concentrations in soil samples collected from contaminated areas in Belarus. They obtained ^{129}I/^{137}Cs atom ratios of 0.35–1.1 in the central spot (close to Chernobyl; Pogonne, Tulgovichi, etc.) and 0.05–0.12 in the Gomel-Mogilov spot. Our results on the ^{129}I/^{137}Cs ratios are similar to their central spot values. Hou *et al.*[19] reported ^{129}I/^{137}Cs atom ratios in Belarus, Russia and Sweden. Their ^{129}I/^{137}Cs ratios were mostly in the range 0.1–0.3. These values are somewhat smaller than our values, possibly because their sampling places were significantly farer from the Chernobyl reactor compared to our sample locations, which were collected within the 30-km zone. In addition, since radioactive materials were released from the Chernobyl reactor at different stages, the composition of the nuclides at individual releases might have been changed during the course of the accident. Representative value of I/Cs ratio is difficult to obtain even after 20 years from the accident, since the rations differ among the soil layers because of the different deposition pattern as well as different chemical behavior between I and Cs in the environment system.

In Table 3b we have also listed Pu concentration data from our previous paper,[28] and compared it with the data of ^{129}I and ^{137}Cs. It can be seen that ^{129}I/Pu ratios varied by a factor of 2.3, while the ^{137}Cs/Pu ratios are consistent within a factor of 1.4. Because the chemical behavior of ^{131}I and ^{137}Cs at the time of their release into the environment were different, there should be restrictions to use ^{137}Cs as a proxy for ^{131}I. Use of ^{129}I should be better to estimate the ^{131}I deposition. Although the analysis of ^{129}I is more difficult and time consuming than that of ^{137}Cs, it is worthwhile.

The results of these analyses on ^{129}I can be useful for tracing the total radioiodine released into the environment during the accident in 1986. Accurate measurement of ^{129}I in soil samples from Ukraine could be used to determine the retrospective ^{131}I dosimetry for the exposure of thyroid. However, for a correct estimate of the ^{129}I release form the Chernobyl accident, the pre-accident ^{129}I deposits from nuclear weapons test and emissions from reprocessing plants must be quantified.

CONCLUSION

In this study, ^{129}I/^{127}I ratios in some Chernobyl soil samples were successfully analyzed using a combination of AMS and ICP-MS. It was demonstrated that less than 1 g of soil is enough for a quantitative estimation of the ^{129}I/^{127}I ratio using AMS, which is much less than required by the classical methods (e.g. NAA, β counting). AMS can accurately measure ^{129}I/^{127}I ratios down to 10^{-14}. The concentration of ^{129}I and the ^{129}I/^{127}I atom ratio in surface soil samples from the 30-km-zone of CNPP ranged from 4.6 to 170 mBq/kg and from 1.4×10^{-6} to 13×10^{-6}, respectively. These values are significantly higher than pre-Chernobyl values of ^{129}I, indicating that most of the measured ^{129}I was due to the fallout of the accident. The ^{129}I/^{137}Cs activity ratio in surface and sub-surface soils was not so constant, i.e., in the range $7.3–20.2 \times 10^{-7}$. This might be due to the different behavior of deposition and/or migration of these nuclides. These results suggest that the obtained data of ^{129}I could be useful in reconstructing the ^{131}I deposition in the contaminated areas. Information on the stable iodine concentrations in the Chernobyl area was also obtained in this study. The data obtained indicate that the concentration level of stable iodine in soil is very low (below 1 ppm) for most of the samples, suggesting a potentially low iodine intake of the population from agricultural products in this area.

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