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1	Dependency of radioiodine root uptake by crops on soil characteristics
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The aim of this study was to quantify the parameters of root uptake of radioiodine 28 by agricultural crops under steady state conditions depending on the main soil 29 characteristics. For this purpose, a long-term (483-days) pot experiment was conducted 30 under natural conditions in the Chornobyl Exclusion Zone to grow radish in soils of four 31 different types with added isotope ¹²⁵I. The experiment demonstrated an increase in root 32 uptake of radioiodine by radish roots in the following sequence of soil types: clay soil < 33 loam soil << sandy soil (Chernozem ~ Phaeozem < Greyzem << Podzoluvisol). The 34 obtained results have been analyzed in conjunction with the results of our previous 35 36 studies to identify the factors determining the parameters of root uptake of radioiodine by the studied crop species. The ¹²⁵I concentration ratios (*CRs*) in edible parts of crop 37 species (radish roots: from 0.003 for Chernozem to 0.02 for Podzoluvisol; lettuce 38 leaves: 0.004-0.04; bean pods: 0.0003-0.004; wheat straw: 0.01-0.1; and wheat seeds: 39 0.0001-0.001) anticorrelated with the characteristics of the soils studied: the distribution 40 coefficients Kd of ¹²⁵I (from 112 L kg⁻¹ for clay soils to 19 L kg⁻¹ for sandy soil, $R^2=0.56$ -41 0.97) and Kd' of stable iodine (93-19 L kg⁻¹, R^2 =0.43-0.74), stable iodine concentration 42 in soil (6.2-0.5 mg kg⁻¹, R²=0.71-0.88), and humus content (4.0-0.8%, R²=0.44-0.78). 43 The obtained steady-state CR values and their dependence on the soil characteristics 44 can be used to model the root uptake of ¹²⁹I, a long-lived radiological contaminant, and 45 to predict its accumulation in human food and animal feed. 46

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Keywords: radioiodine; concentration ratio; transfer factor; root uptake; activity
 concentration; pot experiment.

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51 Highlights

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⁵³ Radioiodine root uptake into plants has been studied in a long-term pot experiment

- ⁵⁴ Radioiodine concentration ratios CRs were determined for four different soil types
- ⁵⁵ Radioiodine CRs inversely correlate with iodine distribution coefficients in soil
- 56 Radioiodine CRs inversely correlate with stable iodine and humus contents in soil

59 **1. Introduction**

Long-term radioactive waste storage facilities in various geological formations 60 can be a source of radioactive contamination of the environment with long-lived 61 radionuclides, such as ¹²⁹I ($T_{1/2}$ =15.7.10⁶ y). To date, the major sources of ¹²⁹I releases 62 into the environment are nuclear weapon tests with an estimated input of 10¹¹-10¹² Bq 63 (Tikhomirov, 1983; Deitermann et al., 1989), and nuclear fuel cycle plants, especially 64 those that reprocess irradiated fuel (Daly et al., 1974; Robens et al., 1988; Wershofen et 65 al., 1991; Satoh et al., 2019). The Chornobyl (1986) and Fukushima (2011) accidents 66 resulted in releases of (4-5)·10¹⁰ Bg and (0.7-6)·10¹⁰ Bg of ¹²⁹I, respectively 67 (Steinhauser et al., 2014) and caused a sharp increase in the ¹²⁹//¹²⁷ I ratio in the 68 contaminated areas (Szidat et al., 2000; Hoshi et al., 1994; Sahoo et al., 2009; Miyake 69 et al., 2012). 70

lodine is an important element involved in the metabolism in animal and human 71 organisms. The United Nations Scientific Committee on the Effects of Atomic Radiation 72 (UNSCEAR) considers several pathways of iodine intake to humans (UNSCEAR, 2016): 73 inhalation (average daily intake of 0.29 μ g d⁻¹); ingestion of surface water (5.3 μ g d⁻¹); 74 ingestion of marine fish and shellfish (11 μ g d⁻¹); deposition onto crops that are directly 75 consumed by humans or ingested by dairy and beef cattle and then subsequently 76 ingested by humans (6.6 µg d⁻¹); and root uptake from soil and surface waters followed 77 by ingestion of crops and animal products (200 µg d⁻¹, i.e., major contribution). Due to 78 its long half-life period, ¹²⁹I can significantly influence the radioecological situation on the 79 Earth and contribute to the collective human exposure doses to in the long term. 80

UNSCEAR estimates the average daily intake of ¹²⁹I into human based on the concentration ratio or the transfer factor of radionuclides from soil and water to food without considering the intake rate of stable iodine (¹²⁷I). According to these estimates, for equal rates of atmospheric releases of the long-lived volatile radionuclides (³H, ¹⁴C,

⁸⁵Kr and ¹²⁹I). ¹²⁹I provides the largest contribution to annual collective doses to 85 population (UNSCEAR, 2016). The soil-to-plant transfer of radioactive isotopes of iodine 86 and, consequently, their intake into the human body may depend on the stable iodine 87 content in soil (Korobova et al., 2010a). Therefore, assessment of the concentration 88 ratio (CR, the ratio of the activity concentration of radionuclide in a plant. Bq kq⁻¹, to its 89 concentration in the soil, Bg kg⁻¹; referred also as the transfer factor, TF (IAEA, 2010)) 90 of radioiodine for agricultural plants in various soil conditions is an important scientific 91 and practical task. 92

After the contamination of pasture vegetation and cattle fodder with radioactive 93 iodine isotopes released by the Chornobyl accident, the short-lived ¹³¹I isotope 94 contributed more than 90% of the dose absorbed by the human thyroid gland due to 95 intake of radionuclides with milk (Bazyka et al., 2016). Since the physical-chemical 96 properties of ¹³¹I and ¹²⁹I are identical, analysis of the long-lived ¹²⁹I contents in soil and 97 components of animal and human diets is the most efficient method for retrospective 98 assessment of the doses from ¹³¹I in the early stages after the Chornobyl and 99 100 Fukushima accidents (Michel et al., 2015; Muramatsu et al., 2015; Balonov et al., 2018).

lodine metabolism in humans and animals is well studied (Howard et al., 1996; 101 Voigt and Kiefer, 2007; Vandecasteele et al., 2000). Also, the experimental data on 102 foliar contamination of vegetation and resulting translocation of the short-live iodine 103 radioisotope (¹²⁵I) to edible parts of various crops are available (Henner et al., 2013; 104 Hurtevent et al., 2013). These data were acquired to assess the consequences of an 105 irrigation with contaminated water. Long-term prediction of human intake of ¹²⁹I that 106 radioactive waste repositories could release requires knowledge of relevant factors 107 driving root uptake of radioiodine by edible parts of crops under different soil-climate 108 conditions. However, until recently, relatively little attention has been paid to the 109 radioiodine root uptake studies (Prister et al., 1979; Tikhomirov et al., 1980; Tikhomirov, 110 1983; Kashin, 1987; Haushild and Aumann, 1989), as well as to the behavior of iodine 111

in soil (Prister et al., 1977; Prokhorov et al., 1978; Katagiri et al., 1997; Sheppard et al.,
1992, 1995; Muramatsu and Yoshida, 1996, Yoshida et al., 1998, Sheppard, 2003).
Literature data are rather limited and contradictory, and reported root uptake
parameters of radioiodine in agricultural ecosystems vary within two orders of
magnitude (IAEA, 2010).

Quasi-equilibrium distribution in soil of ionic forms of radioiodine (iodide and 117 iodate) is achieved by relatively rapid processes (Whitehead, 1973; Bors et al., 1991; 118 Yoshida et al, 1992; Sheppard et al, 1995). The results of numerous studies of sorption 119 dynamics in soil are in agreement to indicate that iodine is absorbed mainly by organic 120 matter of soil and, to a lesser extent, by sesquioxides of iron and aluminum (Whitehead, 121 1974a, 1974b, 1978; Bors et al., 1988; Christiansen and Carlsen, 1989; Muramatsu et 122 al., 1990, 2002; Muramatsu and Yoshida, 1996; Sheppard and Thibault, 1992; 123 Sheppard et al., 1995; Yoshida et al., 1998; Bostok et al., 2002, 2003). 124

Studies of iodine radioisotopes transfer in different soils have not allowed to 125 establish a reliable correlation between radioiodine bioavailability and main soil 126 properties (Tikhomirov et al., 1980; Muramatsu et al., 1990; Evans and Hammad, 1995; 127 Marumatsu, 1996). Radioiodine concentration ratios obtained in field experiments 128 (Prister et al., 1977; Deitermann et al., 1989; Hauschild, 1989; Muramatsu et al., 1993, 129 2002) for major crop species ranged from 0.1 to 5 in vegetative plant organs and from 130 0.0005 to 0.07 in reproductive organs i.e. fruits. ¹²⁹I environmental concentrations are 131 very low and their measurement is a challenge. In this regard, another iodine 132 radioisotope, 125 I (T_{1/2}=59.16 days), is more convenient for studying radioiodine 133 behavior in the soil-plant system experiments. Most of the available studies of the 134 radioiodine isotopes transfer into plants (Deitermann et al., 1989; Hauschild, 1989; 135 Muramatsu et al., 1993, 2002; Kashparov et al., 2005a) were conducted during the 136 initial dynamic phase of remobilization between the soil components, i.e. without 137 reaching an apparent steady state. The long-term dynamics of radioiodine soil-to-plant 138

transfer and concentration ratios depending on soil characteristics remain poorlystudied.

In a previous micro-field experiment carried out under natural conditions in the Chornobyl Exclusion Zone, we studied the dynamics of root uptake of ¹²⁵I by plants from four types of soils, namely Podzoluvisol, Greyzem, Phaeozem and Chernozem (Kashparov et al., 2005a). In that study, injection of 500 Bq of ¹²⁵I per kilogram of soil enabled reliable measurements of its concentrations in the soil, soil solution, and plants samples only within half a year after the experiment began because of its rapid decay and fixation by soil solid phases.

This paper reports the results of a long term (483 days, 18.04.2002-14.08.2003) 148 pot experiment on ¹²⁵I uptake by radish root in natural conditions achieved through a 149 thousand times higher soil contamination with ¹²⁵I than in the earlier study by Kashparov 150 et al. (2005a). The main objectives of this study were to obtain parameters of 151 152 radioiodine transfer from soil to radish roots and to identify the most reliable soil parameters governing the long-term bioavailability of radioiodine in soil. We also 153 compared data obtained from this pot experiment (radish) and original non-published 154 data from the above-mentioned micro-field experiment (lettuce leaves, bean pods, 155 wheat grain and straw) to analyze variability of radioiodine uptake parameters. 156

157

158 **2. Material and methods**

159 **2.1 Soil characteristics and experimental design**

In this experimento we used the same soils (Podzoluvisol, Greyzem, Phaeozem and Chernozem according to FAO-WRB classification (IUSS, 2006)) as in our previous micro-field studies in the Chornobyl Exclusion Zone (Kashparov et al., 2005a, 2005b, 2007a, 2007b). These soils with contrasting agrochemical characteristics (texture, pH, organic matter content (OMC), cation exchange capacity (CEC), stable iodine content) are most typical for continental Ukraine and were taken from a 20-cm arable layer of

agricultural lands in different regions of Ukraine (Table 1). Soil characteristics in Table 1 166 are from Kashparov et al. (2005a); they were determined according to methods 167 published by Arynushkina (1970) and Lisoval et al. (1984). Stable iodine content in each 168 soil was determined using the method of Proskuryakova (1966) for soils, and the size 169 distributions of the soil particles were obtained using a laser granulometer (HR 850-B). 170 The values presented are average values obtained from 5-12 sample measurements 171 per soil type. According to the IAEA (2010) soil classification, the studied soils can be 172 classified as Sand soil (Podzoluvisol), Loam soil (Greyzem) and Clay soil (Phaeozem 173 and Chernozem) (Table 1). 174

After homogenization, the soils were placed into cylindrical pots 19.5 cm in 175 diameter and 20 cm high. Each pot contained approximately 5 kg of soil at natural field 176 humidity (soil moisture contents were 11-23%, Table 1). On April 18th, 2002, ¹²⁵I in KI 177 form (Amersham Pharmacia Biotech UK Limited, England) at 7.4 MBg per pot (N=2, in 178 179 duplicate for each soil type) was simultaneously injected into all soil pots, which were then kept in field at the experimental site in the Chornobyl Exclusion Zone (Kashparov 180 et al., 2005a, 2018). After injection, the soil in each pot was thoroughly mixed and 181 sampled for radioactive and stable iodine isotope measurements. The ¹²⁵I activity 182 concentrations and stable iodine concentrations in the soil pots at the initial moment 183 after injection are presented in Table 2. 184

185

186 **2.2 Plants cultivation**

After the injection of ¹²⁵I, we again homogenized the soils in the pots and sowed radish (*Raphanus sativus L.*, French Breakfast variety) according to standard agronomic recommendations. The distance between the seeds was 4-5 cm. The vegetation pots were placed on the ground and kept in natural weather conditions throughout the experiment. When necessary, watering was applied to allow maintaining biomass growth during the vegetation periods.

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2.3. Sampling and measurements

Soil from each vegetation pot was sampled after each harvest. For this purpose, 196 we used a cylindrical soil corer (N=5, 1 cm in diameter, 20 cm long) allowing the soil 197 core to be sampled to the full depth of the pot (about 100 g from each vegetation pot). 198 Soil solution was extracted by centrifugation from 20 g aliguots taken from soil core 199 samples, and ¹²⁵I activities and stable iodine concentrations in the extracts were 200 measured. The aliquot weights were measured before and after centrifugation. The soil 201 solution extracts were then adjusted to a fixed volume with distilled water to measure 202 125 I activity by scintillation γ -spectrometry using a Gamma Track 1185 Series (USA) low-203 background modular automatic γ -counting system with passive shielding and 204 scintillation Nal detector in the well. Stable iodine content in soil was measured 2 times, 205 before the ¹²⁵I injection and after the end of the experiment (483 days) by means of 206 kinetic rhodanide-nitrite method for soils (Proskuryakova, 1966). After sampling the 207 aliquots for extraction of soil solution, the remaining portions of the soil cores were 208 thoroughly homogenized and divided into 5 subsamples of 18 g each. In each 209 subsample, ¹²⁵I activity was determined as the average of 10 measurements by means 210 of scintillation γ -spectrometry. Once measured, the soil samples were returned to the 211 same place from where they were taken from the pots, then the soils in the pots were 212 carefully mixed, and radish was sown again. 213

In the earlier micro-field experiment (Kashparov et al., 2005a), we observed a sharp decrease in the bioavailability of ¹²⁵I within 80 days after its injection due to its rapid fixation to organic compounds of soils. Accordingly, the radionuclide uptake into radish root decreased during the first and second vegetation periods. For this reason, in the present study, the first harvesting was carried out 83 days after injection of ¹²⁵I, i.e., when the equilibrium in the soil–soil solution system was expected to be reached (Table

3). Once harvested, the radish roots were washed with distilled water to remove soil particles, homogenized and placed into 20 mL test-tubes. For each vegetation pot, ¹²⁵I activities were measured in 3-5 samples by means of scintillation γ -spectrometry. To determine the content of stable iodine in the radish root samples, the method reported by Titova (1981) for plants was used. All measurements were performed in fresh samples to avoid iodine volatilization. Iodine concentrations in the soil were converted to dry weight taking into account the soil moisture contents in the samples.

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229

228 2.4 Data processing

All measured activities of ¹²⁵I were decay corrected on the date of injection.

Distribution coefficients (Kds, L kg⁻¹) of ¹²⁵I and stable iodine were determined as 230 ratios of ¹²⁵I activity concentrations or stable iodine concentrations in dry soil (Bg kg⁻¹ or 231 mg kg⁻¹, respectively, dry weight (DW)) to their concentrations in soil solutions (Bq L^{-1} or 232 mg L⁻¹). Concentration ratios (CRs, dimensionless) of ¹²⁵I and stable iodine were 233 calculated as the ratios of ¹²⁵I activity concentrations or stable iodine concentrations in 234 radish roots (Bg kg⁻¹ or mg kg⁻¹, respectively, fresh weight (FW)) to their concentrations 235 in soil (Bq kg⁻¹ or mg kg⁻¹ DW). CR corresponds to the transfer factor (F_{ν}) under 236 equilibrium conditions (IAEA, 2010). 237

We calculated *CR* values based on the measured soil parameters. Under the equilibrium hypothesis, the radioiodine concentration in the plant is assumed to be linearly correlated with its concentration in the soil solution (IAEA, 2010), i.e.

$$A_{radish} = k \cdot A_{sol} \tag{1}$$

where A_{radish} is the average ¹²⁵I activity concentration in radish (kBq kg⁻¹), A_{sol} is the average ¹²⁵I activity concentration in soil solution (kBq L⁻¹), and *k* is the bioaccumulation factor (i.e., the plant to soil solution concentrations ratio, L kg⁻¹).

CR of ¹²⁵I can be defined as follows:

246
$$CR = A_{radish}/A_{soil} = k \cdot A_{soil}/A_{soil} = k/Kd$$
 (2)

where A_{soil} is the ¹²⁵I activity concentration in soil (kBq kg⁻¹).

The stable iodine concentration ratio (*CR*) is defined as follows:

CR' = k'/Kd'

where k' is the bioaccumulation factor (L kg⁻¹) and Kd' is the distribution coefficient (L kg⁻¹) for stable iodine.

(3)

The conventional statistical methods (*t*-test, non-parametric test, regression analysis) were used to establish the relationships between the studied parameters.

254

255 3. Results and discussion

3.1 lodine isotope concentrations in the soil and plant samples

By the end of the experiment, approximately 20% of the injected ¹²⁵I activities 257 was lost (Table 2). Taking into account the estimate uncertainties, there was no 258 difference in losses between the 4 soil types. We found no reliable change of the stable 259 iodine contents of the soils over the entire 483-day period of the experiment (Table 2). 260 Prister et al. (1977) reported high rates of radioiodine volatilization (up to 80%) after its 261 application onto the ground surface without further soil mixing. In contrast, in our 262 previous micro-field experiments under natural conditions in the Chornobyl Exclusion 263 Zone (Kashparov et al., 2005a), release of radioiodine from the arable layer of all soils 264 was not reliably detected, because the experimental design suppressed evaporation of 265 the injected radionuclide (soils were moistened and thoroughly mixed to a depth of 10 266 cm following radioiodine injection and to a depth of 20 cm after 1 hour). 267

For all soil types, average stable iodine concentrations in soil solutions were quite close (0.03-0.09 mg L⁻¹), and average ¹²⁵I activity concentrations in soil solutions and radish roots in Podzoluvisol were 5-8 and 3-5 times higher, respectively, compared with other soil types (Tables 2 and 3). Average FW concentrations of stable iodine in radish roots were similar in all soils (0.02-0.03 mg kg⁻¹) and did not depend on its concentrations in soil.

In winter, when air temperature dropped below 0°C, the ¹²⁵I activity 274 concentrations in soil solutions decreased in all soil types (Table 2). As a result, the 275 values of the distribution coefficient Kd of 125 I in soils in winter were higher than the 276 average for each soil (Table 4), and seasonal changes in Kds were inversely 277 proportional to changes in the ¹²⁵I concentrations in soil solutions. The concentration 278 decreased by an order of magnitude in Podzoluvisol, , whereas this effect was much 279 less pronounced in other soil types. The "freezing" effect was also noted in a study 280 conducted by Bostock et al. (2003). These authors concluded that the decrease in ¹²⁵I 281 extractability due to freezing was indicative of a microbially mediated sorption process. 282 Such an assumption, however, does not explain the absence of a similar decrease in 283 the concentrations of stable iodine in the soil solution sampled in winter (Table 2). On 284 the other hand, the values of ¹²⁵I activity concentration in the soil solution varied 285 significantly during the observation period, even if winter samples were excluded (by an 286 order of magnitude in Podzoluvisol, and by a factor of 3 in other soils; Table 2). Several 287 studies have shown such variations even within a single vegetation period because ¹²⁵I 288 activity concentrations in soil solution are sensitive to variations in environmental factors 289 such as temperature, soil moisture content, and microbial activity factors (Muramatsu et 290 al., 1991, 1993, Yoshida et al., 1998, Bostock et al., 2003). The concentrations of stable 291 iodine in the soil solution in the studied soils varied 3-5-fold during the observation 292 period (Table 2). 293

In the plant samples, the concentrations of both isotopes in each soil differed greatly between harvests (Table 3). Average concentrations of stable iodine in the radish root samples did not differ between the soils, while for ¹²⁵I the concentrations in roots of radish planted in Podzoluvisol were significantly higher (t-test p<0.05) than in other three soils. The factors influencing iodine uptake by plants are analyzed below.

299

300 3.2 Bioaccumulation factors and concentration ratios of iodine in radish roots

301 and *Kd* in soils

Non-parametric analysis revealed a rather weak correlation ($r_s=0.48$, p=0.03) 302 between the ¹²⁵I activity concentrations in radish roots (Table 3) and in soil solutions 303 (Table 2) at harvesting times. The weak correlation can be explained by the fact that the 304 measurement results indicate the concentrations in the soil solution A_{sol} at the moments 305 of sampling, while the concentrations in radish roots A_{radish} were formed by intake of the 306 radionuclide during the whole vegetation period, when the values of A_{sol} could differ 307 from those measured at harvesting. However, a strong linear correlation (R²=0.98) 308 occurs between the averaged values of these two variables for all soil types (with the 309 winter values excluded), i.e., when the influence of temporal variation in the 310 concentrations is eliminated. The average ratio of the concentrations, i.e., the ¹²⁵I 311 bioaccumulation factor for all soils (eq. 1), can be estimated as $k=0.35\pm0.07$ L kg⁻¹ 312 (R=0.99). A close average value of the bioaccumulation factor, with greater variation 313 between the soil types, $k = 0.40 \pm 0.24$ L kg⁻¹, was found for stable iodine. 314

The ¹²⁵I concentration ratios (CRs) for radish roots in all soils (Table 4) agree well 315 with the data obtained in the micro-field experiments (Kashparov et al., 2005a). In 316 Podzoluvisol and Greyzem, the mean values of ^{125}I CR were about half as high as I_{st} 317 CR', whereas in Phaeozem these values did not differ significantly, and in Chernozem 318 they were similar. CR variations over the observation period were greater for ¹²⁵I (up to 319 a factor of 4) than for Ist. For both isotopes, the concentration ratios decreased in the 320 order Podzoluvisol>Grevzem>Phaeozem>Chernozem. Except for Podzoluvisol, the 321 average *Kds* of ¹²⁵I were higher than the *Kd*'s of the stable isotope. Since concentration 322 ratios are inversely correlated with distribution coefficients (eq. 2 and 3), the average 323 Kds and Kd's increased in the same order by soil type. Overall, the values of both 324 analyzed parameters, CR and Kd, in each soil were close for the two isotopes, 325 suggesting that the quasi-equilibrium state of ¹²⁵I in the soils was approaching. The 326

average concentration ratios of both isotopes can be approximated by the same inverse 327 dependence on the distribution coefficients with a coefficient equal to the average of the 328 k and k' values in all soils, 0.39 \pm 0.18 L kg⁻¹ (Fig. 1a). The only outlier is the point for l_{st} 329 in Podzoluvisol; without this data, R^2 of the correlation increases to 0.97. For steady 330 state conditions, CR of ¹²⁵I can be predicted based on the apparent Kd' of stable iodine. 331 In our observations, CR was negatively correlated ($R^2=0.74$, Fig. 1b) to Kd' with the 332 coefficient of 0.31 \pm 0.08 L kg⁻¹, which is a mean of the k' values in three soils (without 333 Podzoluvisol). 334

The average background (natural) concentrations of stable iodine in soil solutions of the studied soils (A'_{sol}) remained in the range 0.03-0.09 mg L⁻¹ (Table 2), i.e., were weakly dispersed and almost constant, leading to consider the proportionality of ¹²⁵I *CR* to the stable iodine concentration (A'_{soil} , Table 2):

$$CR = k'/Kd' = k' A'_{sol}/A'_{soil} = C_l/A'_{soil}$$
(4)

Therefore, ¹²⁵I *CR* in radish roots is negatively correlated to the stable iodine concentration in the soil (R^2 = 0.71, Table 5) as shown in Fig. 1c, and

342 with $C_1=0.014\pm0.004$ mg kg⁻¹.

343 Tikhomirov (1983) suggested a linear dependence of the stable iodine 344 background level in soil A'_{soil} on humus content *H*, %:

 $345 \qquad A'_{soil}=a\cdot H+b \tag{5}$

with *a* = 0.43 and *b*=0.25

In this pot experiment, the coefficients of this linear equation were different when plotting data from Tables 1 and 2: a=1.6 and b=-0.76, with a pairwise correlation coefficient r= 0.97 between A'_{soil} and H. ¹²⁵I *CR* in radish roots and H in soil were negatively correlated (R²= 0.78, Table 5, Fig. 1d).

In our experiment, the accuracy of the *CR* assessment did not differ significantly (RMSE=0.003-0.004) between the *CR(Kd'*), *CR(A'_{soil})*, and *CR(H)* variants (Fig. 1b-d).

354 **3.3 Comparison of** *CR* **values for different species**

¹²⁵I CRs were measured for lettuce (L. sativa), beans (P. vulgaris) and wheat (T. 355 aestivum) grown on the same four soil types (Podzoluvisol, Greyzem, Phaeozem and 356 Chernozem) from 2000 to 2002 in the micro-field experiment conducted under natural 357 conditions in the Chornobyl Exclusion Zone (Kashparov et al., 2005a). The averaged 358 ¹²⁵I CR values for the lettuce leaves, bean pods and wheat grain and straw obtained 40 359 days after iodine injection, i.e. on the completion of its rapid absorption stage by soil 360 components, were analyzed for correlation with the averaged values of the soil 361 characteristics studied in this pot experiment (i.e., Kd of ¹²⁵I and Kd' of stable iodine, 362 stable iodine concentrations in soil A'_{soil}, and humus contents H). The corresponding 363 correlation coefficients and parameters values for each crop are presented in Table 5. 364 For all crops, ¹²⁵I CRs are negatively correlated with the studied soil characteristics. 365 Figs. 2 and 3 show the dependencies of ^{125}ICR on ^{125}IKd and stable iodine content in 366 soil A'_{soil}, respectively. 367

In lettuce leaves, bean pods, and wheat straw, the highest correlations were observed (\mathbb{R}^2 >0.86) between ¹²⁵I *CRs* and *A*'_{soil}, whereas for radish roots and wheat grain, predictions based on *Kds* were more accurate (\mathbb{R}^2 >0.94) (Figs. 2 and 3, Table 5). To a lesser extent than for radish, ¹²⁵I *CRs* in all crops also correlated with the other variables studied, *Kd*' and *H* (\mathbb{R}^2 =0.43-0.72; Table 5).

Under the same soil conditions, the highest accumulation of ¹²⁵I occured in wheat straw (Table 5). The *CRs* of ¹²⁵I in lettuce leaves and radish roots (FW) are approximately twice as low, and in bean pods and wheat grain are 30 and 60 times lower than in wheat straw, respectively. The ¹²⁵I *CRs* for radish (roots), lettuce (leaves), bean (pods) and wheat (grain) in air-dry state can be recalculated from the values presented in Table 5 using the FW/DW ratios of 20±3, 19±2, 5±1, 1.1±0.1, respectively, which were obtained in our earlier studies (Kashparov et al., 2005a, 2007a).

381 **3.4** Use of the obtained parameters for radiological assessments

The ratios between the ¹²⁵I activity concentrations in wheat grain and straw are in good agreement with the results reported by Prister et al. (1979). The parameters and equations for obtaining the ¹²⁵I *CR* values in wheat straw can be used for estimating the transfer of the long-lived radiological contaminant ¹²⁹I into perennial cereal forage grass, cattle forage, and products for human consumption (milk and meat).

Thus, based on the data obtained in this long-term experimental study (root 387 uptake of ¹²⁵I under natural conditions in steady state), the correlations between ¹²⁵I 388 CRs in various edible crop species and the characteristics of the main soil types of 389 continental Ukraine and the corresponding model parameters were assessed. This 390 makes it possible to estimate the root uptake of radioactive iodine in the studied wide 391 range of soil conditions and to predict its accumulation in the edible parts of the studied 392 crops at harvesting. Root uptake of radioiodine by plants was anticorrelated with the 393 394 stable iodine contents in soil (Fig.1 and 3, Table 4 and 5). It means that at the same levels of ¹²⁹I deposition, its highest concentrations will be found in plants grown on 395 sandy soils, which are characterized by the lowest contents of stable iodine and humus. 396

As it was shown earlier, under steady state conditions CR=CR'=A'radish/A'soil'. 397 Therefore, a meaning of C_l in Table 5 is a constant concentration of stable iodine in a 398 certain plant, which does not depend on iodine abundance in soil and is determined by 399 the plant physiology. In our pot experiment, the measured ¹²⁷I concentrations in radish 400 roots in all soils during the observation period were $0.02\pm0.01 \ \mu g \ g^{-1}$ and were 401 independent of soil type and its concentration in the soil (Table 3). This value is close to 402 C_l of radish, 0.014±0.004 (Table 5), which indicates approaching the equilibrium 403 between the two iodine isotopes studied. Available literature data (Tikhomirov, 1983; 404 Kashin, 1987) show that stable iodine concentrations in various plant species on the 405 continental territory (i.e., located more than 200 km from the sea) of the former USSR 406 vary less than twofold and correspond to the concentrations obtained in our study. They 407

do not depend on soil type and the contents of stable iodine or humus in soil, which show variations within an order of magnitude. Higher concentrations of stable iodine in plants were observed in coastal regions as a result of interception and foliar uptake of iodine aerosols (Tikhomirov and Rusina, 1983). Therefore, in the case of irrigated agriculture, foliar pathway may provide additional uptake of stable iodine into plants, which depends on its concentration in irrigation water.

414

415 **4. Conclusions**

In this long-term study conducted under natural conditions, correlations between 416 ¹²⁵I CRs in various crop species and characteristics of the main soil types of continental 417 Ukraine (distribution coefficients of ¹²⁵I and stable natural iodine, stable iodine 418 concentrations in soil and humus contents) under stationary conditions were assessed. 419 The strongest correlations were found between ¹²⁵I CR and ¹²⁵I Kd and stable iodine 420 concentrations in the soil. This allows estimating the root uptake of ^{125,129}I by the studied 421 crop species and predicting the radioiodine isotopes accumulation in the edible parts in 422 the range of soil conditions under study. The transfer parameters of radioiodine root 423 uptake by radish roots from four different soils increased in the following order: clay soil 424 < loam soil << sandy soil (Chernozem ~ Phaeozem < Greyzem << Podzoluvisol). 425

Among the plant species studied in the same soil conditions after the completion 426 of the rapid sorption phase, the highest uptake of ¹²⁵I occurs in wheat straw, while its 427 uptake in lettuce leaves and radish roots (FW) is two times lower, and in bean pods and 428 wheat grain is 30 and 60 times lower than in wheat straw, respectively. The parameters 429 and equations for obtaining ¹²⁵I CR values in wheat straw and grain can be used to 430 assess the transfer of radioiodine (including ¹²⁹I) to cereals and forage fed to cattle, and 431 thus to estimate ^{125,129}I activity concentrations in human diet components (milk and 432 meat) in case of a release of radioiodine into the environment. 433

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644 **Table captions**

645

Table 1. Average agrochemical characteristics of soils (mean $\pm 2\sigma$) and size class distribution parameters (from Kashparov et al., 2005a).

648

Table 2. ¹²⁵I activity concentrations and stable iodine (I_{st}) concentrations in the soil samples, and soil moisture contents.

651

Table 3. ¹²⁵I activity concentrations and stable iodine (I_{st}) concentrations in the radish root samples.

654

Table 4. Distribution coefficients (*Kd*) and concentration ratios (*CR*) for ¹²⁵I and stable iodine (127 I) in radish roots.

657

Table 5. Parameters of relationships between ¹²⁵I *CR* for crops and main characteristics of the studied soils, and associated coefficients of determination (R^2).

660

661 **Figure captions**

662

Fig. 1. Correlations between concentration ratios of iodine isotopes in radish roots and soil properties: (a) *CR* and *CR*' vs distribution coefficients *Kd* (125 I) and *Kd*' (I_{st}), (b) *CR* vs *Kd*', (c) CR vs *A*'_{soil}, and (d) CR vs humus content *H*. Open and shaded symbols indicate the mean values for I_{st} and ^{125}I , respectively, and vertical and horizontal bars show standard deviations. R² and RMSE denote the coefficient of determination and root mean square error RMSE of approximation, respectively. In (a), the values in parentheses are R² and RMSE calculated without considering the data for stable iodine
 in Podzoluvisol. Solid and dotted lines represent the approximating trends and the
 associated 95% confidence intervals.

672

Fig. 2. ¹²⁵I *CR* in lettuce leaves (a), bean pods (b), wheat straw (c) and grain (d) vs ¹²⁵I
distribution coefficients *Kd* in the studied soils (based on the data from Kashparov et al.
(2005a). Solid and dotted lines represent the approximating trends and the associated
95% confidence intervals.

677

Fig. 3. ¹²⁵I *CR* in lettuce leaves (a), bean pods (b), wheat straw (c) and grain (d) vs stable iodine concentrations in the studied soils A'_{soil} (based on the data from Kashparov et al. (2005a)). Solid and dotted lines represent the approximating trends and the associated 95% confidence intervals.

Table 1. Average agrochemical characteristics of soils (mean $\pm 2\sigma$) and size class

Characteristics of soils		Sc	oil type	
FAO-WRB classification	Podzoluvisol	Greyzem	Phaeozem	Chernozem
(IUSS, 2006)				
CIS countries classification (Arynushkina,	Soddy-	Grey forest	Meadow	Typical
1970)	podzolic		Chernozem	Chernozem
IAEA (2010) classification	Sand	Loam	Clay	Clay
Coordinates of soil origin	51.0126° N	50.294°N	50.3038° N	47.9279° N
	30.0009° E	29.744° E	29.7407° E	30.5382° E
Stable iodine concentration before ¹²⁵ I	0.4±0.3	1.0±0.4	2.3±0.5	5.3±0.8
injection, mg kg ⁻¹				
Soil moisture at injection, %	11	14	18	23
Exchangeable calcium (Ca),	0.16±0.01	0.59±0.03	1.20±0.04	2.2±0.1
mg-eq kg ⁻¹				
Exchangeable magnesium, mg-eq kg ⁻¹	0.016±0.001	0.062±0.002	0.103±0.005	0.17±0.01
Sum of exchangeable bases, mg-eq kg ⁻¹	0.30±0.05	0.92±0.02	2.54±0.05	not determined
Cation exchange capacity, mg-eq kg ⁻¹	0.58±0.06	1.4±0.1	2.6±0.01	3.5±0.2
Hydrolytic acidity	1.5±0.5	0.46±0.04	0.5±0.1	0.37±0.04
The water extract, pH_{H2O}	6.3±0.3	7.9±0.1	8.2±0.2	8.6±0.1
The salt extract, pH_{KCI}	5.2±0.6	6.9±0.3	7.0±0.3	7.3±0.1
Humus (<i>H</i>), %	0.8±0.2	1.1±0.1	2.8±0.2	4.0±0.2
Total nitrogen (N _{total}), %	0.058±0.004	0.056±0.005	0.18±0.02	0.27±0.01
Alkaline-hydrolyzed nitrogen (N _{hydr}), mg	0.55±0.03	0.72±0.05	0.96±0.05	1.1±0.1
kg ⁻¹				

distribution parameters (from Kashparov et al., 2005a).

Size class distribution parameters

Median diameter, µm	141.2	48.9	23.2	11.6
10% mass, μm	<22.4	<3.0	<3.8	<1.7
90% mass, μm	<513	<462	<43.6	<31.6
>500 μm, % mass	8	6	0	0

		5	Sampling da	ate (days af	ter ¹²⁹ l inject	ion into the	soil)			
Parameter	June, 3	July, 10*	Aug, 15*	Oct, 2*	Jan, 30	May, 23*	June, 27*	Aug, 14	mean±2SD	mean±SD ¹
	(46)	(83)	(119)	(167)	(287)	(400)	(435)	(483)		
				Podzol	uvisol					
Mean I_{st} concentration (A'_{soil}), mg kg ⁻¹ DW (N=8) 0.5±0.1										
¹²⁵ I activity concentration at the injection, kBq kg ⁻¹ DW 1560±80										
Mean ¹²⁵ I activity concentration in soil in the period 46-483 days after the injection (A_{soil}), kBq kg ⁻¹ DW (N=8) 1190±60										
Mean ¹²⁵ I activity fraction remaining in soil in the period 46-483 days after the injection, % (N=8) 76±7										
¹²⁵ I in soil solution (A_{sol}), kBq L ⁻¹	163±8	178±18	167±8	78±4	11±3	19±4	101±6	51±9		108±53
I_{st} in soil solution (A'_{sol}), mg L ⁻¹	-	0.05±0.02	-	0.02±0.01	0.04±0.03	0.01±0.01	0.05±0.01	0.03±0.01		0.03±0.02
Soil moisture, %	11.6	11.9	7.8	14.6	8.3	16.3	13.4	16.3		13±3
				Grey	zem					
Mean I _{st} concentration (A' _{soil}), mg k	g⁻¹ DW (N	=8)							1.4±0.3	
¹²⁵ I activity concentration at the inje	ection, kBq	kg⁻¹ DW							1650±80	
Mean ¹²⁵ I activity concentration in s	soil in the p	eriod 46-483	days after	the injection	(A _{soil}), kBq kg	g⁻¹ DW (N=8)			1240±140	
Mean ¹²⁵ I activity fraction remaining	in soil in	the period 46	-483 days a	after the inject	tion, % (N=8))			75±10	
¹²⁵ I in soil solution (A_{sol}), kBq L ⁻¹	20±1	12±1	34±2	28±2	8±4	18±8	29±5	16±8		22±8
I_{st} in soil solution (A'_{sol}), mg L ⁻¹	-	0.08±0.05	-	0.07±0.03	0.05±0.04	0.04±0.01	0.04±0.01	0.2±0.1		0.09±0.07
Soil moisture, %	17.4	11.9	10.2	14.8	14.5	14.6	13.6	15.3		14±2

Table 2. ¹²⁵I activity concentrations and stable iodine (I_{st}) concentrations in the soil samples, and soil moisture contents.

¹ Without the Jan, 30 data (winter period with the temperature below 0° C)

Phaeozem											
Mean I _{st} concentration (A' _{soil}), mg kg ⁻¹ DW (N=8) 2.9±1.1											
¹²⁵ I activity concentration at the injection, kBq kg ⁻¹ DW 1740±90											
Mean ¹²⁵ I activity concentration in soil in the period 46-483 days after the injection (A_{soil}), kBq kg ⁻¹ DW (N=8) 1470±120											
Mean ¹²⁵ I activity fraction remaining in soil in the period 46-483 days after the injection, % (N=8) 84±9											
¹²⁵ I in soil solution (A_{sol}), kBq L ⁻¹	11.7±0. 6	31±3	17±2	11.5±0.6	8±4	10±4	15±5	10±4		15±8	
I_{st} in soil solution (A'_{sol}), mg L ⁻¹	-	0.10±0.07	-	0.03±0.02	0.05±0.07	0.08±0.01	0.03±0.01	0.03±0.01		0.05±0.03	
Soil moisture, %	19	16.2	12.5	19.3	18.9	19.5	18.5	19.9		18±3	
				Chern	ozem						
Mean I _{st} concentration (A' _{soil}), mg l	kg⁻¹ DW (N	=8)							6.2±1.3		
¹²⁵ I activity concentration at the inj	ection, kBo	kg⁻¹ DW							1850±90		
Mean ¹²⁵ I activity concentration in	soil in the	period 46-483	days after	the injection	(A _{soil}), kBq kç	g⁻¹ DW (N=8)			1510±120		
Mean ¹²⁵ I activity fraction remainin	g in soil in	the period 46-	483 days	after the inject	tion, % (N=8)			82±8		
¹²⁵ I in soil solution (A_{sol}), kBq L ⁻¹	20±1	7±1	21±6	11±2	10±8	14±13	14±10	22±7		15±6	
I_{st} in soil solution (A'_{sol}), mg L ⁻¹	-	0.18±0.11	-	0.06±0.04	0.14±0.10	0.09±0.02	0.06±0.02	0.04±0.02		0.09±0.06	
Soil moisture, %	25.1	22.2	15.2	22.3	22.4	24.3	21.8	28.5		23±4	

* Radish roots harvest

		maanteD									
Parameter	July, 10 (83)	July, 10 (83) Aug, 15 (119)		May, 23 (400)	<i>I</i> lay, 23 (400) June, 27 (435)						
Podzoluvisol											
¹²⁵ I in radish roots (<i>A_{radish}</i>), kBq kg ⁻¹ FW	25±6	44±23	12.3±0.6	37±22	12±6	26±14					
I _{st} in radish roots (<i>A</i> [,] _{radish}), mg kg ⁻¹ FW	0.03±0.02	-	-	0.03±0.02	0.02±0.01	0.03±0.02					
Greyzem											
¹²⁵ I in radish roots (<i>A_{radish}</i>), kBq kg ⁻¹ FW	19±2	10.1±0.5	5.4±0.4	7±4	5±0.5	9±6					
I _{st} in radish roots (<i>A</i> ['] _{radish}), mg kg ⁻¹ FW	0.02±0.01	-	-	0.03±0.03	0.02±0.01	0.02±0.01					
		Phaeo	zem								
¹²⁵ I in radish roots (<i>A_{radish}</i>), kBq kg ⁻¹ FW	7±5	9.6±0.5	2.7±0.4	13±4	3±2	7±4					
I _{st} in radish roots (<i>A</i> [,] _{radish}), mg kg ⁻¹ FW	0.03±0.02	-	-	0.01±0.01	0.02±0.01	0.02±0.01					
		Cherno	ozem								
¹²⁵ I in radish roots (<i>A_{radish}</i>), kBq kg ⁻¹ FW	6±3	5.8±0.3	2.4±0.3	4±3	5±3	5±2					
I _{st} in radish roots (<i>A</i> ['] _{radish}), mg kg ⁻¹ FW	0.02±0.01	-	-	0.01±0.01	0.03±0.01	0.03±0.01					

Table 3. ^{125}I activity concentrations and stable iodine (Ist) concentrations in the radish root samples.

	Sampling date (days after ¹²⁵ I injection into the soil)											
Parameter	June, 3 (46)	July, 10 (83)	Aug, 15 (119)	Oct, 2 (167)	Jan, 30 (287)	May, 23 (400)	June, 27 (435)	Aug, 14 (483)	mean±SD			
Podzoluvisol												
¹²⁵ I <i>Kd</i> , L_kg⁻¹	7.2±0.5	6.5±0.7	7.0±0.5	15±1	109±27	62±21	12±1	23±4	19±9			
l _{st} <i>Kd</i> ′, L kg⁻¹		9±5		20±12	13±13	36±22	9±4	19±13	19±11			
¹²⁵ I CR		0.021±0.005	0.04±0.02	0.010±0.005		0.03±0.02	0.010±0.005		0.022±0.012*			
I _{st} CR'		0.06±0.04				0.06±0.04	0.04±0.02		0.05±0.03			
Greyzem												
¹²⁵ I <i>Kd</i> , L kg⁻¹	63±8	103±14	37±5	45±6	162±87	70±31	42±8	77±39	62±23*			
l _{st} <i>Kd</i> ′, L kg⁻¹		18±12		19±10	32±30	36±14	41±16	7±4	24±14			
¹²⁵ I CR		0.015±0.002	0.008±0.001	0.004±0.001		0.006±0.003	0.004±0.001		0.008±0.005*			
I _{st} CR'		0.014±0.007				0.02±0.02	0.014±0.007		0.017±0.012			
				Pha	leozem							
¹²⁵ I <i>Kd</i> , L kg⁻¹	125±12	47±6	86±11	127±12	174±80	147±57	96±30	147±60	111±36*			
l _{st} <i>Kd</i> ′, L kg⁻¹		29±23		91±58	60±91	36±15	91±45	97±50	69±33			
¹²⁵ I CR		0.005±0.003	0.007±0.001	0.002±0.001		0.009±0.003	0.002±0.001		0.005±0.003			
I _{st} CR'		0.010±0.007				0.003±0.003	0.007±0.004		0.007±0.005			

Table 4. Distribution coefficients (*Kd*) and concentration ratios (*CR*) for 125 I and stable iodine (127 I) in radish roots.

¹ Without the Jan, 30 data (winter period with the temperature below 0°C)

Chernozem											
¹²⁵ I <i>Kd</i> , L kg⁻¹	76±7	203±32	73±21	142±24	147±113	111±105	108±78	68±22	112±48		
l _{st} <i>Kd</i> ′, L kg⁻¹		34±22		103±72	44±32	69±21	103±38	155±83	93±45		
¹²⁵ I CR		0.004±0.002	0.004±0.001	0.002±0.001		0.003±0.002	0.003±0.002		0.003±0.001		
I _{st} CR'		0.003±0.002				0.002±0.002	0.005±0.002		0.003±0.002		

* Significant difference between the values for 125 I and I_{st} (p<0.05).

Table 5. Parameters of relationships between ${}^{125}ICR$ for crops and main characteristics of the studied soils, and associated coefficients of determination (R^2).

1251 CD	CR=k/k	CR=k/Kd		CR=k'/Kd'		oil	CR=C _H /H		
ICR	k	R^2	k'	R^2	Cı	R^2	C _H	R ²	
Radish roots*	0.4±0.1	0.97	0.3±0.1	0.74	0.014±0.004	0.71	0.013±0.004	0.78	
Lettuce leaves**	0.5±0.2	0.76	0.5±0.3	0.46	0.02±0.01	0.86	0.02±0.01	0.51	
Bean pods**	0.034±0.005	0.61	0.03±0.02	0.43	0.0012±0.0004	0.87	0.0013±0.0009	0.47	
Wheat straw**	1.0±0.2	0.56	1.0±0.7	0.45	0.04±0.02	0.88	0.04±0.03	0.44	
Wheat grain**	0.016±0.005	0.94	0.013±0.006	0.66	0.0006±0.0004	0.78	0.0006±0.0002	0.72	

* this study; ** based on the data from Kashparov et al. (2005a).



Fig.1. Correlations between concentration ratios of iodine isotopes in radish roots and soil properties: (a) *CR* and *CR*' vs distribution coefficients *Kd* (¹²⁵I) and *Kd*' (I_{st}), (b) *CR* vs *Kd*', (c) CR vs A'_{soil} , and (d) CR vs humus content *H*. Open and shaded symbols indicate the mean values for I_{st} and ¹²⁵I, respectively, and vertical and horizontal bars show standard deviations. R² and RMSE denote the coefficient of determination and root mean square error RMSE of approximation, respectively. In (a), the values in parentheses are R² and RMSE calculated without considering the data for stable iodine in Podzoluvisol. Solid and dotted lines represent the approximating trends and the associated 95% confidence intervals.



Fig.2 ¹²⁵I *CR* in lettuce leaves (a), bean pods (b), wheat straw (c) and grain (d) vs ¹²⁵I distribution coefficients *Kd* in the studied soils (based on the data from Kashparov et al. (2005a)). Solid and dotted lines represent the approximating trends and the associated 95% confidence intervals.



Fig. 3. ¹²⁵I CR in lettuce leaves (a), bean pods (b), wheat straw (c) and grain (d) vs stable iodine concentrations in the studied soils A'soil (based on the data from Kashparov et al. (2005a)). Solid and dotted lines represent the approximating trends and the associated 95% confidence intervals.