*Radioprotection*, Suppl. 1, vol. **40** (2005) S329-S334

DOI: 10.1051/radiopro:2005s1-049

# Fate of long-lived radioactive halogens, ($^{36}$Cl, $^{129}$I), in agricultural ecosystems: Field investigations

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**Abstract.** Field experiments in the Chernobyl exclusion zone have been carried out to determine the behaviour of the radioactive halogens $^{36}$Cl and $^{125}$I (as a surrogate for $^{129}$I) in the soil-plant system and along human food chains. The investigations on the migration in four types of soil of the two radionuclides showed that the vertical transport of chlorine was the most rapid: 9 months after the contamination the residual fractions of $^{36}$Cl in the 20 cm arable layer varied from 1% to 3% according to soil type. For iodine this percentage was less than 3% in any case.

The soil to plant transfers were quantified for various agricultural crops (cereals, grass, root vegetables, leaves vegetables and fruit vegetables) and for several kinds of soils. The root uptake of $^{36}$Cl by plants was particularly important. As a result of this very high biological assimilation about 60% of the contamination was extracted from the soils by the plants after one vegetation period. For iodine the concentration ratios values are 2 or 3 order of magnitude lower than for chlorine.

## 1. INTRODUCTION

Chlorine-36 ($T_{1/2} = 3\ 10^5$ years) and iodine-129 ($T_{1/2} = 15.7\ 10^6$ years) are two long-lived radionuclides which can be discharged into the environment during the nuclear fuel cycle, in particular during processing irradiated fuel, and from nuclear waste disposal. They are biologically important elements. Radioecological parameters are needed for assessment models to estimate their impact on the natural environment and the human health. This paper describes studies of radiochlorine and radioiodine behaviour in soils and root uptake by plants that were conducted in the natural conditions of the Chernobyl Exclusion zone and in laboratory over a 3 years period using four different soil types (podzoluvisol, greyzem and typical and meadow chernozem).

## 2. MATERIALS AND METHODS

An experimental site has been established in spring 2000 along the western radioactive fallout area of the Chernobyl accident in a restricted territory that would not be used in the future due to its high contamination by long-lived radionuclides [1]. It measures 20x20 m and is surrounded by a 1.5 m high fence to prevent wild animals intrusion. Within this area four pits (4x4 m with a depth of 0.5 m) were dug, the indigenous soil removed and replaced by the 20-cm arable layer of four soils originating from different agricultural land of Ukraine (Table 1). Each pit was then divided into four sub-parcels (2x2 m) to permit the simultaneous cultivations of four plant species on each type of soil.

Water solutions of radiochlorine and radioiodine were sprayed onto the sub-parcel surface and carefully mixed into the 20-cm soil layer. $^{125}$I was used as a surrogate for $^{129}$I because it is more conveniently measured. Due to the high mobility in soils of $^{36}$Cl and the short $^{125}$I half-life ($T_{1/2}$ = 60 days), analogous process of soils contamination was performed at the beginning of the vegetation periods in 2000, 2001 and 2002.

From 2000 to 2003 radishes, lettuce, beans and wheat were cultivated. During each year there were 3-4 radish harvests, 1-2 lettuce harvests, 1 bean harvest and 1 winter wheat (2001) and spring wheat (2002) harvest. In spring 2003 potatoes, onions, peas, wheat, oat, clover and ryegrass were planted.

Soil samples were taken from each parcel to measure radionuclides concentrations at the planting and harvesting stages.

During the experiment, data concerning air and soil temperature, rainfall, atmospheric pressure, air and soil humidity, intensity of solar radiation, and wind direction and velocity were recorded by the meteorological stations located at a distance of 2 km.

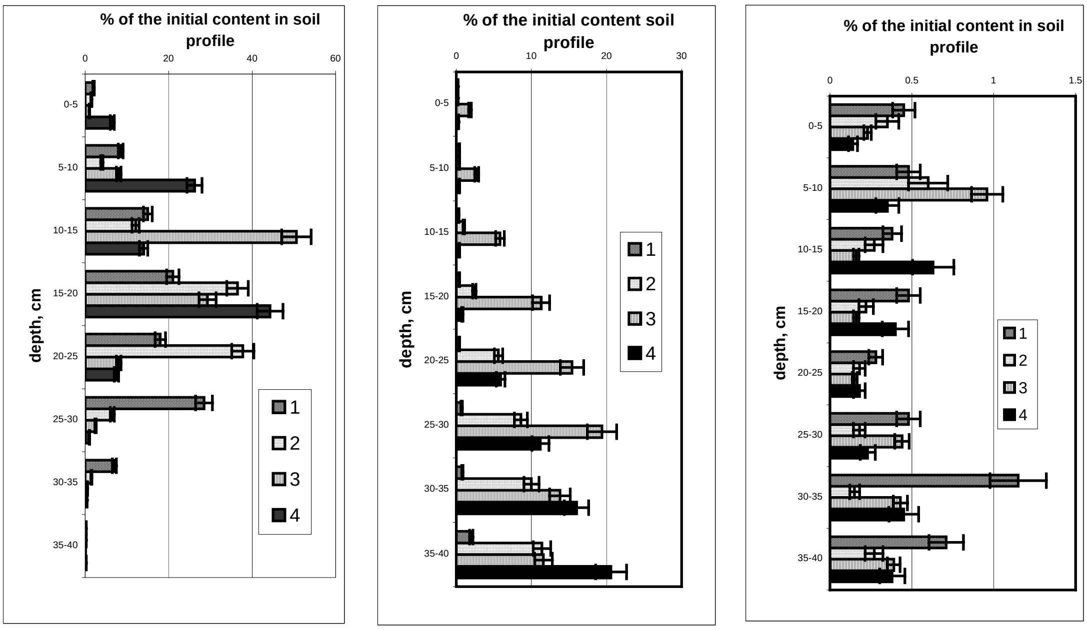
**Table 1.** Main agrochemical characteristics of soils.

| | **Soil type** | | | |
|---|---|---|---|---|
| | **Podzoluvisol** | **Greyzem** | **Meadow Chernozem** | **Typical Chernozem** |
| Stable iodine content, mg/kg | 0.4±0.3 | 1.0±0.4 | 2.3±0.5 | 5.3±0.8 |
| Exchangeable calcium, meq/100 g | 1.6±0.1 | 5.9±0.3 | 12.0±0.4 | 22±1 |
| Exchangeable magnesium, meq/100 g | 0.16±0.01 | 0.62±0.02 | 1.03±0.05 | 1.7±0.1 |
| Sum of exchangeable bases, meq/100 g | 3.0±0.5 | 9.2±0.2 | 25.4±0.5 | not determined |
| Cation exchange capacity (meq/100 g) | 5.8±0.6 | 14±1 | 26±1 | 35±2 |
| $pH_{H2O}$ | 6.3±0.3 | 7.9±0.1 | 8.2±0.2 | 8.6±0.1 |
| $pH_{KCl}$ | 5.2±0.6 | 6.9±0.3 | 7.0±0.3 | 7.3±0.1 |
| Organic matter, % | 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 |

## 3. RESULTS AND DISCUSSION

### 3.1 $^{36}$Cl behaviour in soils

Results of measurements taken in 2000-2001 concerning $^{36}$Cl vertical distribution in soil profiles (Fig. 1) illustrated that the radionuclide migrates very intensively from the arable layer in deeper layers in all the soils studied (values are normalized to the specific activity of soil after injection). Vertical distributions of $^{36}$Cl in the 40-cm soil layer also feature a decrease of the vertical migration intensity in a soil sequence: podzoluvisol >greyzem ≈ typical chernozem >meadow chernozem.




28 days after injection 96 days after injection 269 days after injection

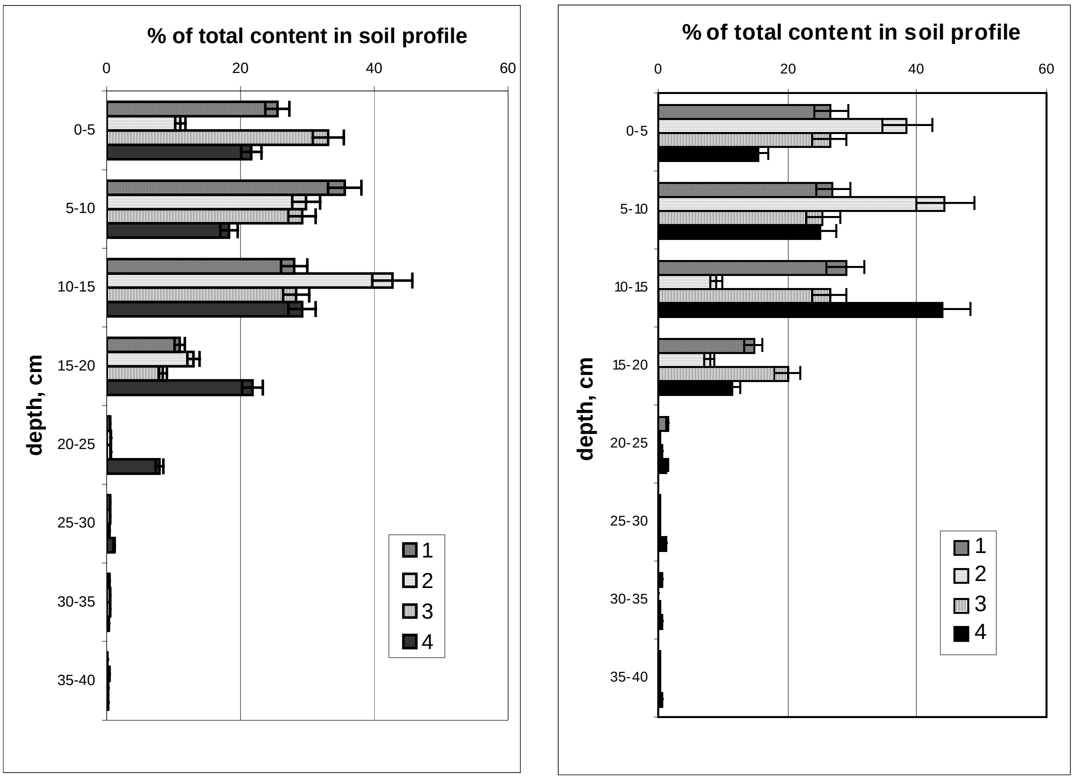
**Figure 1.** Vertical distribution of $^{36}$Cl in soil profiles 28, 96 and 269 days after radiochlorine injection in the 20-cm upper layer of soils: 1 - podzoluvisol; 2 - greyzem; 3 - meadow chernozem; 4 - typical chernozem.

On day-96 after the injection, the fractions of radionuclide remaining in the 20-cm arable layer were only 1%, 5%, 2% and 30%, respectively. On day 269 after the injection, these fractions were 1.5-1.8% of the initial amount. The half-decrease period for $^{36}$Cl specific activity in the arable layer of all soils varied in the range 13-50 days.

It was found that the relative content of radiochlorine in the 20-cm arable layer of all soils in 2000-2001 decreased as a function of the cumulative rainfall ($R^2$=0.7-0.85) and could be expressed by the exponential function $\exp(-(0.015\pm0.001)\cdot PR)$, where rainfall PR is given in mm. Intensive rains observed during the first month following the initial radionuclide injection entailed a sharp decrease of the radionuclide content in the upper layer of soil, while the further decrease noted over the next two months was not so significant.

## 3.2 $^{125}$I behaviour in soils

The vertical $^{125}$I distributions in the studied soil profiles indicated that there had been an insignificant degree of vertical radionuclide migration in the arable layer. Specific $^{125}$I activity values measured in the 20 cm arable soil layer are presented in figure 2 (results are normalized relative to the soil's specific activity following initial radionuclide spraying, taking radioactive decay into account).




28 days after injection | 269 days after injection

**Figure 2.** Vertical distribution of $^{125}$I in soil profiles 28 and 269 days after radioiodine injection in the 20-cm upper layer of soils: 1 - podzoluvisol; 2 - greyzem; 3 - meadow chernozem; 4 - typical chernozem.

Less than 3% of the total radioiodine content had migrated from the arable layer (to a depth of more than 20 cm) over a 269 days period. It corresponds to an effective diffusion coefficient of less than $2{\cdot}10^{-10}$ cm$^2$ s$^{-1}$. The results obtained are in good agreement with the radioiodine migration estimates for the arable soil layer (0.5-5%), based on the literature [2],[3].

### 3.3 Root uptake of $^{36}$Cl and $^{125}$I by vegetation

Soil-to-plant transfers were studied from 2000 to 2003. The concentration ratio values (CR= (specific activity of radionuclide in vegetation, Bq kg$^{-1}$)/(specific activity of radionuclide in the arable soil layer, Bq kg$^{-1}$)) for the various soil types and plants are calculated with regard to the fresh weight of the vegetation at the moment of mature plant harvest.

In order to compare the CR values obtained with those reported in the literature, the vegetation concentration coefficients were also measured following drying to the air-dry state (t=80°C until a constant mass is reached) and ashing (t=500° C during 16 hours). The CR for air-dry or ash-residue states can be determined by multiplying the value calculated on the fresh weight basis with the corresponding coefficient. These coefficients for radish (roots), lettuce (leaves) and beans (pods) were 20±3, 19±2, 5±1 following drying to an air-dry state, and 127±9, 106±11, 92±13 following ashing, respectively.

Crop productivity (for crops at fresh weight) at the moment of harvesting was of 1-2 kg m$^{-2}$ for lettuce leaves and radish roots, 0.1-0.4 kg m$^{-2}$ for bean pods and 0.3-0.4 kg m$^{-2}$ for wheat grains (1-2 kg m$^{-2}$ for straw).

### 3.3.1 *Soil-to-plant transfer of $^{36}Cl$*

$^{36}Cl$ and natural stable chlorine have been measured in soils and vegetation samples.

No reliable evidence of dependence of radiochlorine CR on the soil type was observed. CR values for $^{36}Cl$ reach 15±10 in radish roots, 30±15 in lettuce leaves, 15±11 in bean pods, 23±11 in wheat seeds and 210±110 in wheat straw, 2.6±0.4 (11±2 for dry plants) in green peas, 1.5±0.5 (10±4) in onions, 8±1 (33±4) in potatoes, 90±26 in clover hay, 158±88 in ryegrass hay, 17±6 (18±6) and 174±67 in seeds and straw of oats, respectively. These data correlate well with the results obtained earlier under laboratory conditions [4] and with the literature data for stable chlorine [5] and for $^{36}Cl$ [6].

Therefore, assuming that the productivity of radish, lettuce and aboveground parts of wheat is about 1-2 kg $m^{-2}$, it may be deduced that more than half of $^{36}Cl$ activity in arable soil layer soil will pass into these plants during one period of vegetation in the absence of the intensive vertical migration of chlorine with the convective water flux. The fraction of radiochlorine transported into radish roots and into wheat seeds can reach 5-10% and 2-5%, respectively.

The specific content of stable Cl did not vary significantly in plants from one harvest to another and was not dependent on the soil type: radish roots - 440±130 mg $kg^{-1}$, lettuce leaves - 620±95 mg $kg^{-1}$, bean pods - 310±85 mg $kg^{-1}$, wheat seeds - 580±125 mg $kg^{-1}$. The specific content of stable chlorine in wheat straw was approximately one order of magnitude higher than in seeds (4900±1400 mg $kg^{-1}$). In addition, values of stable chlorine specific activity in all soils were close 20-40 mg $kg^{-1}$).

Furthermore, chlorine ions were mainly retained in the soil through absorption by vegetation. $^{36}Cl$ content in soil under the wheat was practically unaltered during the entire vegetation period.

In order to assess chlorine release from the dead vegetation into soil solution and its further involvement in the migration processes, a batch experiment was carried out. In this experiment, we studied the kinetics of $^{36}Cl$ transfer into distilled water from the straw of cereals that had been grown in 2003. The straw was cut into 2-3 cm-long pieces. Samples of wheat straw were added with 300 ml of distilled water. They were mixed for 20 min to wet the straw, then for 1 min mixings just before the solution sampling. 5 ml samples of the solution were periodically taken to measure $^{36}Cl$'s activity and determine its full activity in the solution. Almost all $^{36}Cl$ had passed into the solution for 98 hours.

### 3.3.2 *Soil-to-plant transfer of $^{125}I$*

The results obtained indicated that the bioavailability of radioiodine quickly decreased during the first 30-50 days following injection due to absorption by the soil, and changed slightly during the following period. They indicated that the higher radioiodine CR values were observed for all of the plants in podzoluvisol (2-15 times higher than in other soils). No consistent difference in the plant radioiodine accumulation was observed relative to greyzem, meadow chernozem and typical chernozem soils. The organic matter and stable iodine contents and the acidity of soil did not appear to have a significant influence on the bioavailability of radioiodine.

CR values in radish roots and salad leaves varied slightly over time and ranged from 0.01-0.03 for podzoluvisol and 0.001-0.005 for other soils. CR values for green bean pods (0.003-0.004 for podzoluvisol and 0.0002-0.0007 for other soils) are one order of magnitude lower than in radish and salad (this difference decreased by a factor of 2-3 as compared to the CR values in the air-dry state). The lowest CR values are obtained in wheat seeds: 0.001 for podzoluvisol and 0.0001-0.0003 for other soils. Specific activity of radioiodine in wheat straw is two orders of magnitude higher than in seeds. Relative to the CR values obtained and the productivity of the studied species, the relative uptake of radioiodine by plants during the vegetation period can be estimated. 0.06-0.01% of radioiodine is transfered from podzoluvisol to the radish (including leaves) and salad. For other soils, this value varies from 0.06-0.002%, depending upon the time period between injection and harvesting. For bean pods, the values obtained for podzoluvisol and other soils are of 0.0002-0.0003% and 0.00006-0.0001%, respectively. Relative uptake of radioiodine by the aboveground biomass of wheat is 0.01-0.1%.

Obtained CR values even for the first year of radioiodine incubation in soil are much lower than a value cited by IAEA (0.02) for edible parts of common crops [7].

## 4. CONCLUSIONS

$^{36}$Cl has a high migration ability and its transfer from soil to plants is extremely high.

Variations in the dependence of chlorine content in the arable layer on the rainfall shows that other parameters influence $^{36}$Cl migration with the convective flux, such as those determining the moisture movement: soil humidity, temperature, evapotranspiration etc. All these factors must be taken into account when modeling the dynamics of the chlorine concentration in the arable layer of soil.

Chlorine ions are mainly temporarily retained in the soil through sorption by vegetation. Following organic matter decomposition, chlorine passes into the soil solution and is thus, once again, available for plants.

In contrast with chlorine, the iodine behaviour in the soil-plant system is soil type dependent. Its mobility in soils and bioavailability are weak in situations where oxic conditions prevail. Therefore to forecast the long-term behaviour of iodine in anoxic media and especially when assessing the impact of nuclear waste disposals it would be necessary to take into account that, in other biogeochemical conditions, iodine solubility and mobility could be higher.

## References

[1] Kashparov, V., Colle, C., Zvarich, S., Yoschenko, V., Levchuk, S., Lundin, S., (2004). *Journal of environmental radioactivity*, (In press).

[2] Tikhomirov, .A. (1983). Radioecology of iodine. Moscow, Book Company Energoatomizdat, 88 p.

[3] Whitehead, D.C. (1984). *Environmental International* **10**, 321-339.

[4] Colle, C., Mauger, S., Massiani, C., Kashparov, V., Grasset G., (2002). Proceedings of the International Congress ECORAD 2001, Aix-en-Provence (France), 3-7 September 2001. Special issue, *Radioprotection* **37** C1, 491-496.

[5] Sheppard, S.C., Evenden, W.G., Macdonald, C.R., (1999). *Journal of environmental radioactivity*, **43**, 65-76.

[6] Bostock, A., Ashworth, D., Shaw, G., 2002. Proceedings of the International Congress ECORAD 2001, Aix-en-Provence (France), 3-7 September 2001. Special issue, *Radioprotection* **37** C1, 341-346.

[7] IAEA (1994) Handbook of parameters values for the prediction of radionuclides transfer in temperate environments. Technical Reports Series n°364, 74 pp., ISBN 92-0-101094-X.