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# Distribution and Source of $^{129}\text{I}$ , $^{239,240}\text{Pu}$ , $^{137}\text{Cs}$ in the environment of Lithuania

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## Abstract

Fifty five soil samples collected overall Lithuania in 2011 and 2012 were analyzed for  $^{129}\text{I}$ ,  $^{137}\text{Cs}$  and Pu isotopes in order to investigate the level and distribution of artificial radioactivity in Lithuania. The activity and atomic ratio of  $^{238}\text{Pu}/^{239,240}\text{Pu}$ ,  $^{129}\text{I}/^{127}\text{I}$  and  $^{131}\text{I}/^{137}\text{Cs}$  were used for identifying the origin of these radionuclides. The  $^{238}\text{Pu}/^{239+240}\text{Pu}$  and  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in the soil samples analyzed varied in range of 0.02 - 0.18 and 0.18 - 0.24, respectively, suggesting the global fallout as the major source of Pu in Lithuania. The values of  $10^{-9}$  to  $10^{-6}$  for  $^{129}\text{I}/^{127}\text{I}$  atomic ratio revealed that the source of  $^{129}\text{I}$  in Lithuania is global fallout in most cases though several sampling sites shows a possible impact of reprocessing releases. Estimated  $^{129}\text{I}/^{131}\text{I}$  ratio in soil samples from the southern part of Lithuania shows negligible input of the Chernobyl fallout. No correlation of the  $^{137}\text{Cs}$  and Pu isotopes with  $^{129}\text{I}$  was observed, indicating their different sources terms. Results demonstrate uneven distribution of these radionuclides in the Lithuanian territory and several sources of contamination i.e. Chernobyl accident, reprocessing releases and global fallout.

## 1. Introduction

The activity and atomic ratios of long-lived radionuclides are useful parameters for identifying their origin because their values depend on the sources and generation processes [1–5]. Among them  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{238,239,240}\text{Pu}$  are the key radionuclides for prediction of the sources of radioactive contamination. The activity ratio of Pu isotopes enables distinguishing the global fallout from nuclear weapons testing, the Chernobyl fallout or other sources[6], [7]. Currently the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  activity ratio is not a precise tool to differentiate from one another the global fallout or unexploded weapons-grade Pu. Because the SNAP-9A satellite accident in 1964 and the Chernobyl accident in 1986 released, remarkable amounts of  $^{238}\text{Pu}$  and emerged in the environment[4]. Differentiation of radioactive fallout from various sources has been investigated in ice cores [8], sea water [9], [10] atmosphere and soil [2, 4,11] by means of the Pu isotope ratios. In general, it is theoretically estimated and confirmed by experimental measurement that  $^{240}\text{Pu}/^{239}\text{Pu}$  atomic ratio of  $\sim 0.40$  is due to the Chernobyl fallout [12]–[14], the values from 0.03 to 0.18 represent the nuclear test global fallout[2], [15] and the  $^{240}\text{Pu}/^{239}\text{Pu}$  mass ratio of about 0.05-0.10 is in weapons-grade Pu [2].

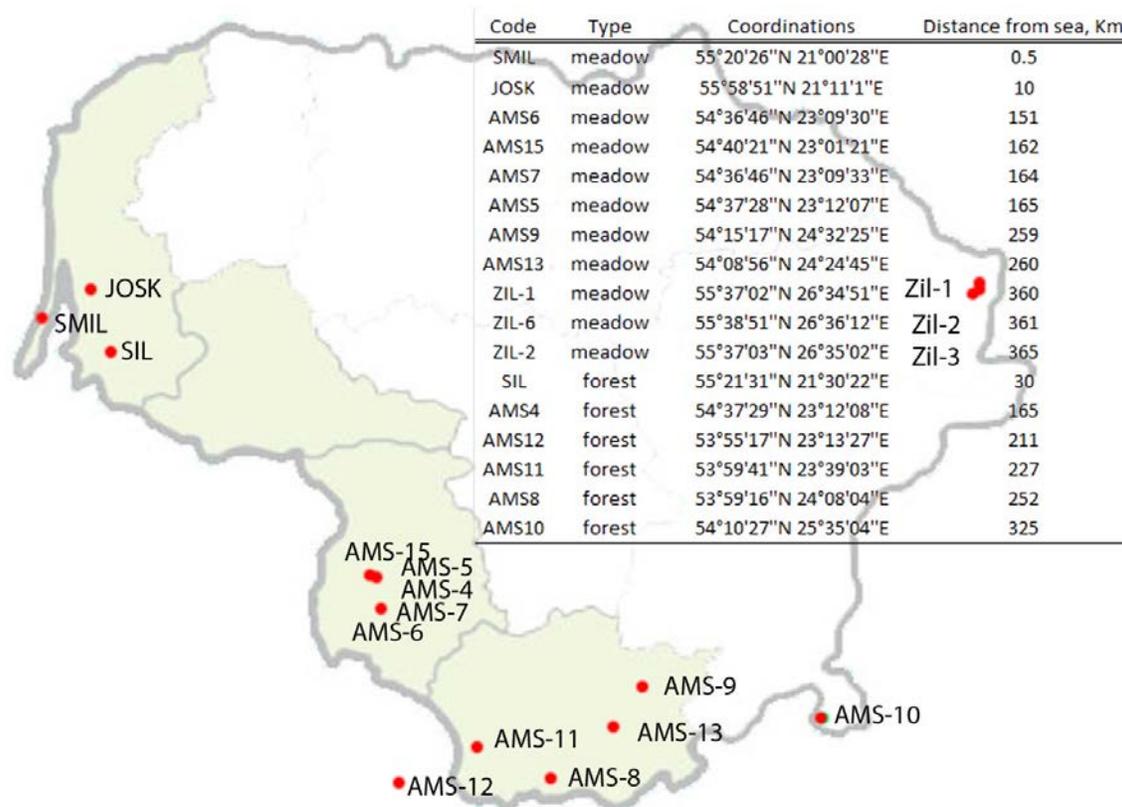
The ratio of fission and activation products  $^{134, 135, 137}\text{Cs}$  varies significantly depending on the reactor and fuel type making it an important indicator of the source identification [16]. The half-life of cesium-137 is relatively short (30.1y), nevertheless it is still possible to use this radionuclide for identification of source of the contamination. The main source of this isotope in the environment were the Chernobyl accident and nuclear weapon testing[17]–[20]. The  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio of 0.018 and  $^{238}\text{Pu}/^{137}\text{Cs}$  activity ratio of 0.00068 the representative value of global fallout in soil were determined by Bunzl and Kracke in 1988 [21]. The  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio of 0.12 to 0.28 was found in the marine and lake sediments as the source of global fallout [17]. The  $^{239,240}\text{Pu}/^{137}\text{Cs}$  activity ratio for the Chernobyl accident releases was observed to be 6.8 in the mushroom sampled in the 30km zone of the Chernobyl NPP[18]. Subsequent events such as the wild fire in the Chernobyl forest [22] are possibly secondary contamination source.

Iodine-129 is a naturally occurring long-lived radioisotope of iodine (15.7 My) formed by the cosmic ray reaction with Xe and fission of  $^{238}\text{U}$ , [23] it is supposed that the stable  $^{127}\text{I}$  in the atmosphere and biosphere are mainly originated from the releases of iodine from the sea [24]–[26]. The concentration of iodine-129 in the environment is low and the natural  $^{129}\text{I}/^{127}\text{I}$  atomic ratios was theoretically estimated to be  $(0.04\text{--}3.0)\times 10^{-12}$ , the measured value in the pre-nuclear marine sediment is  $1.5\times 10^{-12}$  [23], [27], [28]. While  $^{129}\text{I}/^{127}\text{I}$  ratios in pre-nuclear soil show a big variation, this might attributed to the high different concentration of  $^{127}\text{I}$  in different types of soil. Since 1945, large amount of  $^{129}\text{I}$  was released to the environment through human nuclear activities, such as nuclear weapons testing, nuclear fuel reprocessing and nuclear accident, the  $^{129}\text{I}/^{127}\text{I}$  atomic ratio can be therefore used for identifying the origin of iodine from the pre- nuclear period to the period of human activities. Atmospheric  $^{129}\text{I}/^{127}\text{I}$  ratio exceeded  $10^{-9}$  in the Northern Hemisphere and due to the releases of nuclear weapons testing, while this value achieved to  $10^{-6}$  in European seawater and the North Atlantic seawater due to the discharges of two European reprocessing plants in Sellafield (UK) and La Hague (France) [29]–[31]. Fan et al. (2013)[32] reported that the major sources of  $^{129}\text{I}$ , contributing more than 90% to the total inventory of  $^{129}\text{I}$  in the current environment, were European nuclear fuel reprocessing plants. Zhang et al. (2013) [33] present  $^{129}\text{I}/^{127}\text{I}$  ratios which vary from  $10^{-12}$  to  $10^{-4}$  in pre-nuclear samples and highly contaminated areas, respectively. Lehto et al. (2012) [34] show that  $^{129}\text{I}$  concentrations in the Baltic Sea water have reached  $4\times 10^9$  atoms  $\text{L}^{-1}$ , and the elevated  $^{129}\text{I}$  level was interpreted as the liquid discharges from Sellafield and La Hague reprocessing plants, which was transported to the Baltic Sea through water exchange between the North Sea and the Baltic Sea. Hou et al. (2009) reported that the concentration of  $^{129}\text{I}$  in the precipitation in Denmark varied from  $0.28\times 10^9$  to  $5.63\times 10^9$  atoms  $\text{L}^{-1}$  and the  $^{129}\text{I}/^{127}\text{I}$  ratios varied from  $5.04\times 10^{-8}$  to  $76.5\times 10^{-8}$  with an average value of  $30.1\times 10^{-8}$ . The high  $^{129}\text{I}$  level was interpreted as re-emission of  $^{129}\text{I}$  from the North Sea, where the  $^{129}\text{I}/^{127}\text{I}$  ratio in the seawater is up to  $10^{-6}$  [35].

This work aims to distinguish the possible sources of radioactive contamination in Lithuania by measuring the activity concentrations of various radionuclides in the soil samples collected across the country. The additional data of  $^{129}\text{I}$  are useful tool to obtain the new knowledge on the sources of contamination. For the first time  $^{129}\text{I}$  was measured in the soil of Lithuania, which provide essential data for further application of these radionuclides for investigation of environmental process.

## 2. Material and method

### 2.1 Sample description, preparation and measurement technique



**Fig. 1.** Map of Lithuania divided into two zones (zone-I white color, and zone-II gray color). According to Nedveckaite et al. (1986) zone-II was highly contaminated by the radioactive cloud from Chernobyl. Most of the samples were collected in zone-II for iodine-129 measurement.

Fifty-five top soil samples (0-5 cm) were collected from different locations across Lithuania in 2011 and 2012 representing western, south-western and southern parts of Lithuania (Fig.1, Table S-1 in supplementary material), and analyzed for determination of Pu isotopes and  $^{137}\text{Cs}$ . Among them, 17 samples were analyzed for  $^{129}\text{I}$  (Fig.1). Sampling areas were selected with the most probable chance of contamination: the southern and western parts of Lithuania area close to the Baltic Sea are regions which are under the influence of the global fallout and the Chernobyl accident. According to U. N. S. C.[14] about 76% of the total activity from the atmospheric nuclear weapons tests was deposited in the northern hemisphere with the major part (~98%) in the region from  $0^\circ\text{N}$  to  $70^\circ\text{N}$ . Moreover, the Baltic seashore is a specific area where not only the fallout from the Chernobyl accident contributed to the artificial radioactivity levels to a minor degree but also it may be under the influence of the contaminated North Sea water [36]. Hou et al. (2002) showed that liquid discharges from nuclear fuel reprocessing plants in La Hague (France) and Sellafield (United Kingdom) in the North Atlantic Ocean coming to the Baltic Sea through the strait of Denmark were enriched by  $^{129}\text{I}$  [37]. Another sampling

area was selected in radius of 10 km around the Ignalina Nuclear Power Plant (INPP) which may be the potential source of radioactive contamination.

Four measurements techniques were used: accelerated mass spectrometry (AMS) for  $^{129}\text{I}$ , inductively coupled plasma mass spectrometry (ICP-MS) for  $^{127}\text{I}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , alpha spectrometry for  $^{238}\text{Pu}$ , and  $^{239,240}\text{Pu}$  and gamma spectrometry for  $^{137}\text{Cs}$ . For each measurement technique samples were prepared in a different way which are described below.

### *2.2 Determination Iodine isotopes*

Determination of  $^{129}\text{I}$  in soil samples were carried out in Xi'an AMS Center, China. About 5 g samples were precisely weighed to a quartz boat,  $^{125}\text{I}$  (100 Bq) was spiked. The sample in the boat was combusted in a tube furnace at  $800\text{ }^{\circ}\text{C}$ , iodine released from the sample was trapped in 30 ml of  $0.4\text{ mol L}^{-1}\text{ NaOH}-0.05\text{ mol L}^{-1}\text{ NaHSO}_3$  solution. The recovery of iodine was monitored using gamma spectrometry by measuring  $^{125}\text{I}$  in the trap solution, and compared with  $^{125}\text{I}$  spiked to the sample.  $^{127}\text{I}$  in the trap solution was measured applying ICP-MS by using 1.0 ml of solution and diluted 10 times using deionized water. 1.0 mg of  $^{127}\text{I}$  carrier (prepared by dissolution of iodine crystal in  $0.4\text{ mol L}^{-1}\text{ NaOH}$ , the iodine crystal was purchased from Woodward company, UAS, which has very low level of  $^{129}\text{I}$ ) was added to the remained trap solution, and iodine was separated by solvent extraction using  $\text{CHCl}_3$  after converting all iodine species to iodide using  $\text{KHSO}_3$  at pH1-2 and oxidizing iodide to  $\text{I}_2$  using  $\text{NaNO}_2$ . Extraction was repeated and organic phases were combined. Iodine in  $\text{CHCl}_3$  phase was back extracted using  $0.01\text{ mol L}^{-1}\text{ KHSO}_3$ , the separated iodine in iodide form was precipitated as  $\text{AgI}$  by addition of 1 ml of  $1.0\text{ mol L}^{-1}\text{ AgNO}_3$  to the back extracted solution.  $\text{AgI}$  precipitate was separated by centrifuge and dried at  $70\text{ }^{\circ}\text{C}$ , then mixed with Nb powder and pressed into the copper holder.  $^{129}\text{I}/^{127}\text{I}$  ratios in the prepared targets were measured by AMS using 3MV Tandem AMS system (HVEE) in Xi'an AMS center.  $\text{I}^{5+}$  ions sputtered from the ion source were chosen for the measurement, where  $^{127}\text{I}^{5+}$  was measured as charges (current) using a Faraday cup and  $^{129}\text{I}$  was measured using a gas ionization detector. All samples were measured for 6 cycles and 5 min per sample in each cycle. Procedure blank sample was prepared using the same procedure as the samples, and  $^{129}\text{I}/^{127}\text{I}$  in the blanks were measured to be  $(2.2\pm 0.3)\times 10^{-13}$ , which is 2 orders of magnitude lower than the samples. A detailed information about the procedure could be found in [38], [39].

### *2.3 Determination of Plutonium Isotopes*

Plutonium was extracted by acid leaching from the soil matrix and purified using the anion-exchange (Bio-Rad AG-1 $\times$ 8 100-200 mesh, Bio Rad Company) method followed by the extraction chromatography technique. The analytical procedure has been reported elsewhere [40]. The purified plutonium sample was used to electrodeposit plutonium on stainless steel disks. The chemical yield of plutonium in separation procedures was monitored by measuring  $^{242}\text{Pu}$  as a yield tracer. The ORTEC Octate-Plus  $\alpha$ -spectrometer with eight  $600\text{mm}^2$  detectors (resolution in the range of 25-27 keV; efficiency  $\sim 20\%$ ) was used to measure activity of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  on the disk. The detection limit of  $^{239+240}\text{Pu}$  for counting time of 90000 s was estimated to be about 10 mBq. After the measurements the

disks were rinsed with ultrapure grade nitric conc. acid (Merck, Germany) the solution was diluted with 18.2 MΩ de-ionized water to 2 % w/w nitric acid.

$^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in 2%  $\text{HNO}_3$  solution were measured with the sector field inductively coupled plasma mass spectrometer (ELEMENT2, Thermo Fisher Scientific). For a better sensitivity and background performance the APEX (Elemental Scientific, USA) desolvation nebulizer was used. The detection limit for Pu Pu isotopes ( $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ) is estimated based on 3 times standard deviation of the blank to be  $5 \times 10^{-14} \text{ g g}^{-1}$ .

#### *2.4 Determination of Cesium-137*

Soil samples were prepared in plastic containers of the standard geometry for gamma spectrometry measurement. The reference standards with radionuclides ( $^{57}\text{Co}$ ,  $^{139}\text{Ce}$ ,  $^{113}\text{Sn}$ ,  $^{85}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$  and  $^{65}\text{Zn}$ ) free of the coincidence-summing effects were used for the efficiency calibration. The gamma spectrometer was equipped with the HPGe coaxial gamma-X-ray detector (GMX-series with a 0.5 mm thickness Be window) made by Ortec (USA). The relative efficiency of the detector was 33 %, the energy resolution at 1332.5 keV was 1.8 keV. All samples were counted for the fixed time of 50,000 s. The detection limit for  $^{137}\text{Cs}$  is 0.185 Bq. Additionally, activity concentrations of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  were measured in the IAEA-TEL-2014-03 reference soil sample for analytical quality control.

### **3. Results and discussion**

#### *3.1 Distribution and sources of Iodine isotopes*

The concentrations of  $^{127}\text{I}$  and  $^{129}\text{I}$  in soil collected across Lithuania in 2012 are given in Table 1. The  $^{127}\text{I}$  concentrations vary from  $0.4 \pm 0.06$  to  $6.66 \pm 0.19 \mu\text{g g}^{-1}$ . The concentration of  $^{129}\text{I}$  in meadow soil samples was  $(2.01-10.21) \times 10^8 \text{ atoms g}^{-1}$ , whereas the concentration of  $^{129}\text{I}$  in the forest soil varied  $(4.72 \pm 0.07-34.16 \pm 0.56) \times 10^8 \text{ atoms g}^{-1}$ . An extremely high concentration of  $34.16 \pm 0.56 \times 10^8 \text{ atoms g}^{-1}$  for  $^{129}\text{I}$  was obtained in sample SIL taken at a location 30 km from the Baltic Sea in deciduous forest. One of  $^{129}\text{I}$  sources in this area might be the Baltic Sea, where marine discharges of  $^{129}\text{I}$  from two European reprocessing plants was transported along the European coast in the North Sea and then the Kattegat and Danish Strait, a high  $^{129}\text{I}$  level in the Baltic Sea water has been reported.  $^{129}\text{I}$  in the Baltic Sea water, as well as in the North Sea where even higher  $^{129}\text{I}$  concentration was reported might be emitted to the atmosphere, and afterward deposited to the land.[5], [34], [41], [42]. Predominant wind in Lithuania is mostly westwards, meaning that gaseous  $^{129}\text{I}$  re-emitted from the Baltic Sea and North Sea can be transported from the Baltic Sea and deposit to the terrestrial system in Lithuania [36], [43].

A rather different distribution of  $^{129}\text{I}$  in the meadow and forest soil was observed in present study. This might be related to the composition of the upper layer soil as well as geographic circumstance. The relative higher  $^{129}\text{I}$  concentration in meadow and forest soil might attributed to their better vegetation coverage of soil surface, atmospheric deposited iodine (dry deposited and washout of rainfall) can be better retained in the soil without significant loss by blowing out by wind or wash out by water flood in rain season. In forest soil, besides atmosphere deposition, degraded tree leaves absorbed  $^{129}\text{I}$  from the atmosphere fall onto the soil surface might be extra source of  $^{129}\text{I}$  in the soil, which make the  $^{129}\text{I}$  concentration in forest soil higher than other soil type. Retention of the radionuclide in the soil layer

depends on various factors, one of which is the organic matter content in soil [23], [36], [44]. The organic matter content in forest samples ranged from 9 to 89%, while in the meadow samples it ranged from 4 to 14%. Different concentrations of iodine in the forest and meadow soil could also be influenced by soil pH. The pH values of forest soil vary from 3.54 to 5.63. However, no correlation between the concentration of  $^{129}\text{I}$  and geochemical parameters (organic matter and pH) of the soil samples was observed. For forest soil (sample AMS8) with a very high organic matter content of 89%, the concentration of  $^{129}\text{I}$  is only  $11.90 \pm 0.18 \times 10^8$  atoms/g compared to a  $^{129}\text{I}$  concentration of  $34.16 \pm 0.56 \times 10^8$  atoms  $\text{g}^{-1}$  in soil SIL-with the organic matter amount of only 9%. The concentrations of  $^{129}\text{I}$  in the soil samples of the study area are therefore the consequences of a different source and retention feature of the soil.

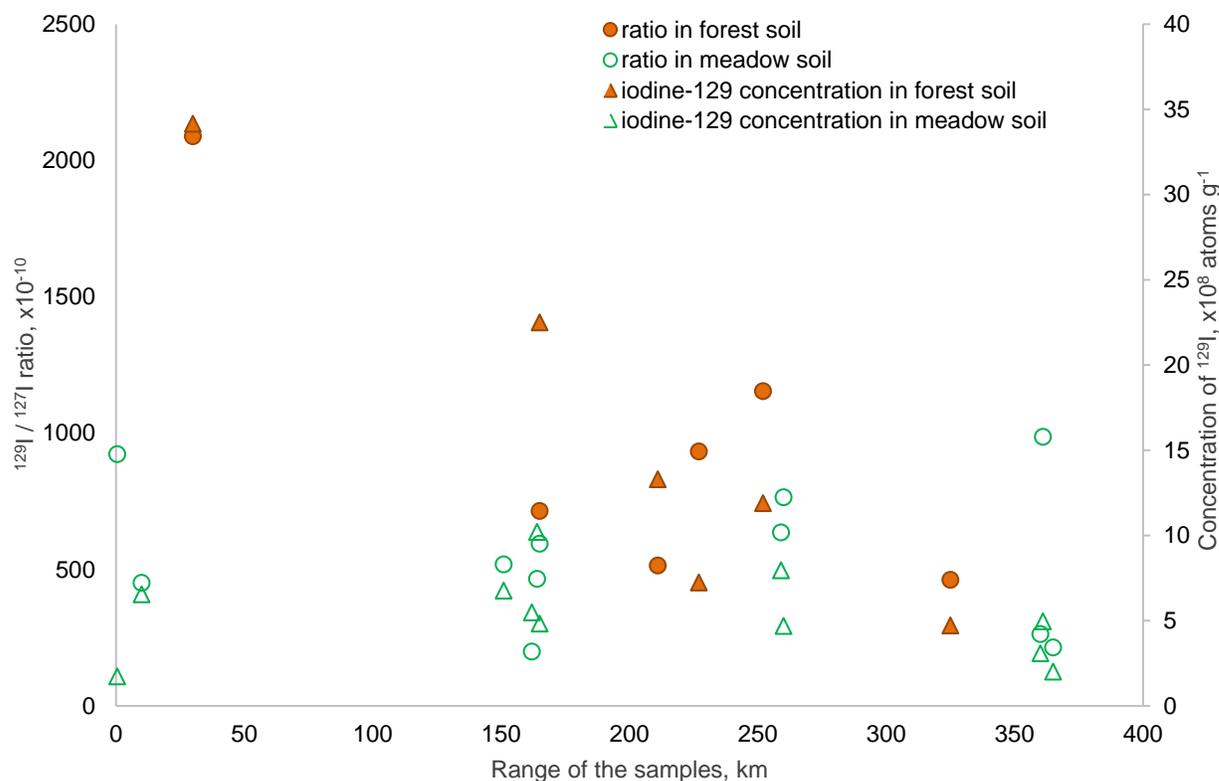
$^{131}\text{I}$  and  $^{134,137}\text{Cs}$  activity concentrations in some environmental samples collected in Lithuania in 1986 straight after the Chernobyl NPP accident were measured [45]. The higher  $^{131}\text{I}$  concentration in milk was found in the southern and western parts of Lithuania [46]. High levels of  $^{131}\text{I}$ ,  $^{103}\text{Ru}$  and  $^{134}\text{Cs}$  in the ground level air ( $45.2 \text{ Bq m}^{-3}$ ,  $20.3 \text{ Bq m}^{-3}$  and  $27.9 \text{ Bq m}^{-3}$ , respectively) were measured in Preila (the Curonian Spit), which is higher than those in Vilnius by a factor of 2–3 [47].

**Table 1.** Concentrations and atomic ratios of iodine isotopes in the soil samples

Code	Soil type	Distance to the Baltic Sea, km	$^{127}\text{I}$ Conc, $\mu\text{g g}^{-1}$	$^{129}\text{I}$ Conc, $\times 10^8$ atoms $\text{g}^{-1}$	$^{129}\text{I}/^{127}\text{I}$ ratio, $\times 10^{-8}$ at at $^{-1}$
SIL	forest	30	$3.45 \pm 0.03$	$34.16 \pm 0.56$	$20.89 \pm 0.37$
AMS4	forest	165	$6.66 \pm 0.19$	$22.57 \pm 0.34$	$7.15 \pm 0.23$
AMS12	forest	211	$5.46 \pm 0.13$	$13.35 \pm 0.21$	$5.15 \pm 0.15$
AMS11	forest	227	$1.64 \pm 0.08$	$7.24 \pm 0.12$	$9.33 \pm 0.48$
AMS8	forest	252	$2.17 \pm 0.06$	$11.90 \pm 0.18$	$11.54 \pm 0.35$
AMS10	forest	325	$2.15 \pm 0.05$	$4.72 \pm 0.07$	$4.63 \pm 0.07$
SMIL	meadow	0.5	$0.40 \pm 0.06$	$1.73 \pm 0.06$	$9.23 \pm 1.41$
JOSK	meadow	10	$3.06 \pm 0.16$	$6.55 \pm 0.12$	$4.52 \pm 0.25$
AMS6	meadow	151	$2.75 \pm 0.07$	$6.76 \pm 0.11$	$5.20 \pm 0.15$
AMS15	meadow	162	$4.80 \pm 0.06$	$5.48 \pm 0.09$	$2.00 \pm 0.04$
AMS7	meadow	164	$4.61 \pm 0.10$	$10.21 \pm 0.16$	$4.67 \pm 0.13$
AMS5	meadow	165	$1.71 \pm 0.08$	$4.83 \pm 0.07$	$5.96 \pm 0.29$
AMS9	meadow	259	$2.64 \pm 0.06$	$7.96 \pm 0.12$	$6.36 \pm 0.18$
AMS13	meadow	260	$1.29 \pm 0.03$	$4.69 \pm 0.08$	$7.65 \pm 0.21$
ZIL-1	meadow	360	$2.47 \pm 0.01$	$3.08 \pm 0.09$	$2.64 \pm 0.07$
ZIL-6	meadow	361	$1.06 \pm 0.06$	$4.96 \pm 0.08$	$9.87 \pm 0.21$
ZIL-2	meadow	365	$1.98 \pm 0.02$	$2.01 \pm 0.05$	$2.14 \pm 0.06$

Fig. 2 shows the dependence of the concentration of  $^{129}\text{I}$  in soil on the distance from the sea to the sampling site, a significantly decreased  $^{129}\text{I}$  concentration with the distance to the Baltic Sea can be observed. The soil samples collected in the area around the INPP (samples ZIL 1, 2, 6, Table 1) shows relative lower concentrations of  $^{129}\text{I}$  ( $(1.7-3.1) \times 10^8$  atoms  $\text{g}^{-1}$ ) compared to other samples, especially those collected close to the Baltic Sea. While  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in these three samples shows a big variation ( $(263.64-987.25) \times 10^{-10}$ ), this is mainly attributed to the very low concentration of  $^{127}\text{I}$  in the

sample SIL-6, cause an increased  $^{129}\text{I}/^{127}\text{I}$  atomic ratio in this sampling site. The highest  $^{129}\text{I}/^{127}\text{I}$  atom ratio of  $2 \times 10^{-7}$  was observed in the sample SIL which was collected at a site nearby the Baltic Sea with a distance of only 30 km to the Baltic Sea, the highest  $^{129}\text{I}$  concentration of  $34 \times 10^8$  atoms  $\text{g}^{-1}$  was also determined in this sample. A similar high  $^{129}\text{I}$  concentrations and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios were also determined in other two sampling sites (SMIL and JOSK) with distance to the Baltic of less than 30 km. For the samples collected in the other sites with distance to the Baltic Sea of more than 200 km, and remote from the INPP, a relative lower  $^{129}\text{I}$  concentration ( $(4.7-22.6) \times 10^8$  atoms  $\text{g}^{-1}$ ) and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios ( $(2.0-11.5) \times 10^{-8}$ ) were observed, which is similar to the  $^{129}\text{I}/^{127}\text{I}$  ratio of ( $10^{-9} - 10^{-8}$ ) in the environment considered as a representative ratio for nuclear weapons testing contribution in terrestrial areas[23], [31], [34]. These results might indicate that a very limited contribution of INPP to the  $^{129}\text{I}$  in the environment compared to the other sources, even for the surrounding area of INPP. A significant contribution of the Baltic Sea, with a clear contribution of the reprocessing releases to the  $^{129}\text{I}$  level, especially to the sampling sites close to the Baltic Sea, i.e. re-emission of  $^{129}\text{I}$  to the atmosphere from the seawater in the highly contaminated area by the reprocessing plants, such as the Baltic Sea, and North Sea is responding to the increased  $^{129}\text{I}$  level in these area, although the global fallout of  $^{129}\text{I}$  from the nuclear weapons testing also has a significant contribution to the  $^{129}\text{I}$  inventory in this area[34], [48].

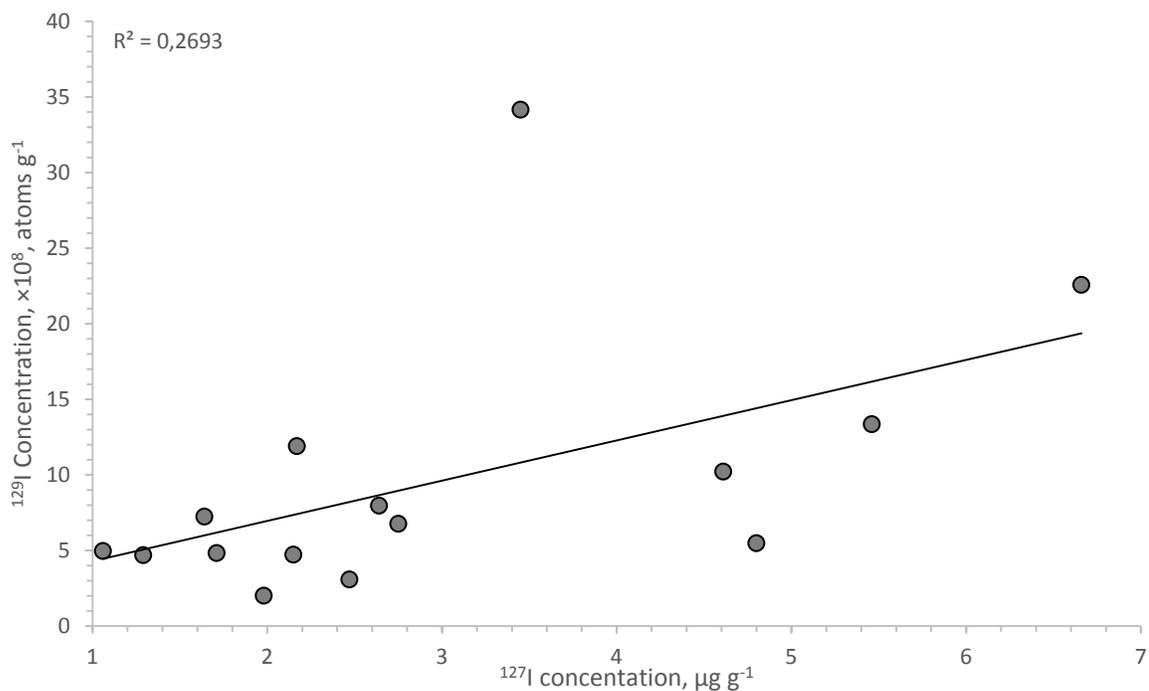


**Fig.2.** A plot of  $^{129}\text{I}/^{127}\text{I}$  atomic ratio and concentration of  $^{129}\text{I}$  in soil samples against the distance from sampling site to the Baltic Sea.

The concentration of  $^{127}\text{I}$  in the investigated soil sample shows small variation with an average of  $2.98 \pm 0.41 \mu\text{g g}^{-1}$ . It has been reported that the  $^{129}\text{I}$  transfer factors from soil to grass are quite different compared to natural  $^{127}\text{I}$  [49], this might be attributed to the different species of  $^{127}\text{I}$  and  $^{129}\text{I}$  in soil due

to their different sources. Our results do not show a significant correlation (Fig. 3) between  $^{127}\text{I}$  and  $^{129}\text{I}$  in all soil samples, this should result from the different sources of these two isotopes in the soils samples investigated in this work. Daraoui et al. (2012) reported that  $^{127}\text{I}$  and  $^{129}\text{I}$  undergo different sorption processes in the soil because of different histories of the two iodine isotopes (“old” and “modern”) in the soil [23]. “Old” iodine is originated in the soils over a long time and had a  $^{129}\text{I}/^{127}\text{I}$  ratio of  $\sim 10^{-12}$ , while “modern” iodine after 1940 has increased the  $^{129}\text{I}/^{127}\text{I}$  ratio by several orders of magnitude.

It is interesting to mention that we observe a negative correlation between  $^{129}\text{I}$  and  $^{137}\text{Cs}$  ( $R=-0.95$ ) in forest soil, but no correlation if we consider all data (meadow and forest soil). This result suggest that the sources of these two isotopes is completely different.  $^{129}\text{I}$  emission from reprocessing plants and re-emission from North Sea and the Baltic Sea is a major source of  $^{129}\text{I}$  in Lithuania, the two European reprocessing plants at La Hague (France) and Sellafield (UK), as well as the North Sea and the Baltic Sea located in east of Lithuania, which cause a decreased deposition and concentration of  $^{129}\text{I}$  in soil with the increased distance from the sampling sites to the Baltic Sea. While Chernobyl accident is a major source of  $^{137}\text{Cs}$  in Lithuania, cause an inhomogeneous deposition of  $^{137}\text{Cs}$  in Lithuania, with a trend of higher in the south part (see discussion below). In addition, no correlation between  $^{129}\text{I}$  and plutonium isotopes was observed, this can be attributed to that plutonium isotopes in Lithuania is mainly attributed to the global fallout from the nuclear weapons testing, with small contribution from the Chernobyl accident, causing a relative homogeneous distribution in Lithuania (see discussion below).



**Fig.3.** Correlation between  $^{129}\text{I}$  and  $^{127}\text{I}$  in soil analyzed in this work.

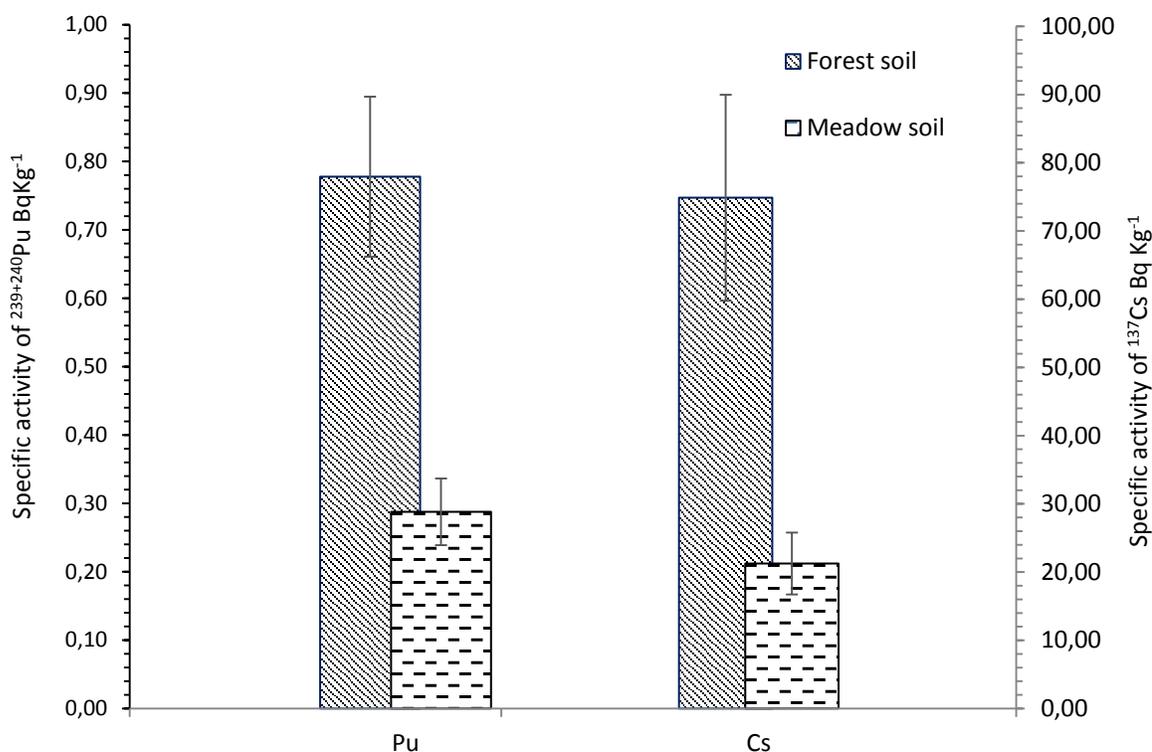
### 3.2 Distribution and sources of Pu isotopes and $^{137}\text{Cs}$ in Lithuania

The activity concentrations of plutonium isotopes ranged from 0.01 Bq kg<sup>-1</sup> to 0.25 Bq kg<sup>-1</sup> for <sup>238</sup>Pu and from 0.05 Bq kg<sup>-1</sup> to 1.80 Bq kg<sup>-1</sup> for <sup>239,240</sup>Pu in 39 meadow and forest soil samples collected in autumn of 2011. <sup>238</sup>Pu/<sup>239,240</sup>Pu activity ratios and <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios were calculated to be 0.02-0.18 and 0.18-0.24, respectively. In meadow soil, a relative lower <sup>238</sup>Pu activity concentration of 0.01–0.05 Bq kg<sup>-1</sup>, and <sup>239+240</sup>Pu concentration of - 0.07–0.53 Bq kg<sup>-1</sup> were measured, whereas a high activity concentrations were determined in forest soils, varying in 0.05-0.09 Bq kg<sup>-1</sup> of <sup>238</sup>Pu and 0.74–1.80 Bq kg<sup>-1</sup> of <sup>239,240</sup>Pu.

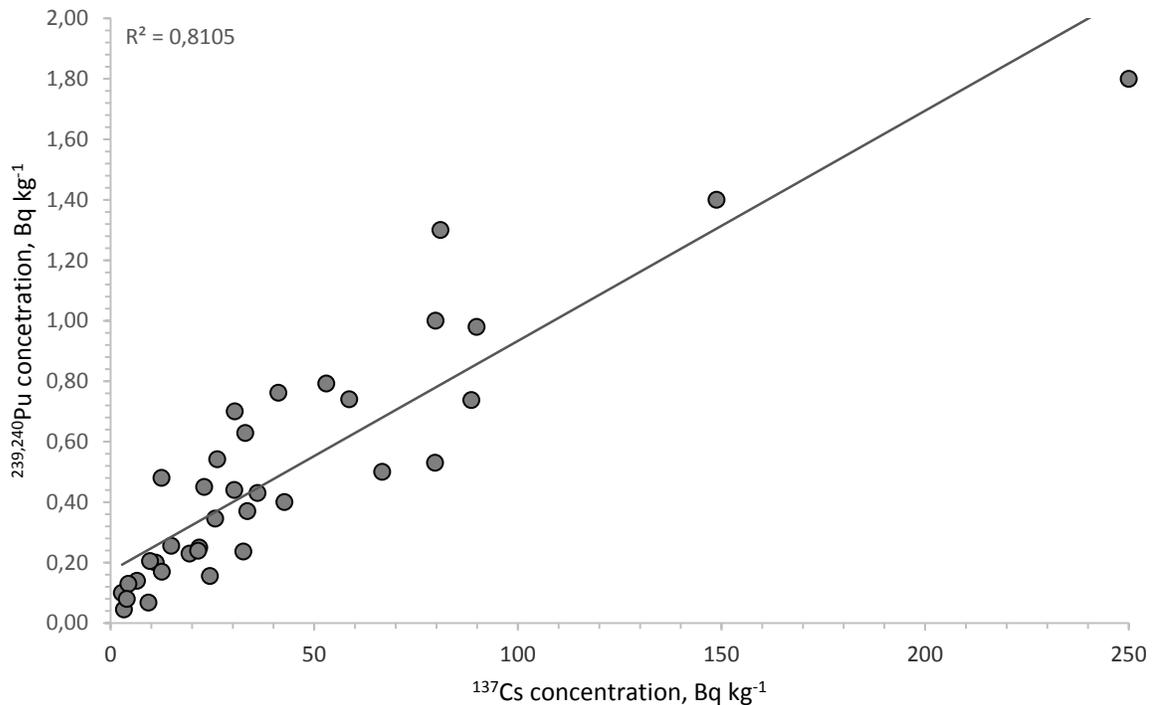
Higher <sup>239,240</sup>Pu activity concentrations have been reported to be 0.05–1.30 Bq kg<sup>-1</sup> in the top layer soils in Lithuania in 1995 and the activity ratio of <sup>238</sup>Pu/<sup>239,240</sup>Pu were in 0.3–0.45 in some samples, showing a significant impact of the Chernobyl NPP accident [50]. Following study of lakes sediments collected in 1999 revealed that up to 31% of plutonium was originated from the Chernobyl accident [51][52].

Another study of top layer meadow soil samples collected all over Lithuania in 2008 showed less <sup>239,240</sup>Pu activity concentration prevailing, namely within 0.10–0.40 Bq kg<sup>-1</sup> [53]. All these studies indicates that the highest Chernobyl fallout was detected in the southern, south-eastern regions of Lithuania where the primary radioactive plume travelled [54].

<sup>238</sup>Pu/<sup>239,240</sup>Pu activity ratios and <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios did not show any significant difference in undisturbed meadow and forest soil samples while the concentrations of <sup>239,240</sup>Pu isotopes and <sup>137</sup>Cs shows a strong correlation (Fig. 5), this might indicate their similar deposition pattern and similar behavior in environment. <sup>137</sup>Cs/<sup>239,240</sup>Pu ratio was similar in both type of soil samples although <sup>239,240</sup>Pu and <sup>137</sup>Cs concentrations were almost three times higher in forest than in the meadow soil (Fig. 4).



**Fig. 4.** Average activity concentration of <sup>239+240</sup>Pu and <sup>137</sup>Cs in meadow and forest soil.



**Fig.5.** Correlation between the  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  activity concentrations.

The activity concentrations of  $^{137}\text{Cs}$  range in 4.3-53.0  $\text{Bq kg}^{-1}$  in the meadow soil and 6.65-250.0  $\text{Bq kg}^{-1}$  in forest soil samples (Fig. 4). High  $^{137}\text{Cs}$  specific activities of  $148.8 \pm 11.8$ ,  $210.6 \pm 16.8$  and  $250.0 \pm 20.0$   $\text{Bq kg}^{-1}$  were found in forest soil samples, especially in AMS4, AMS8 (southern part of Lithuania), and in AMS10 (southeastern part), respectively. Elevated  $^{137}\text{Cs}$  concentration in these area might attribute to the deposition of the Chernobyl accident [55]. On the other hand,  $^{137}\text{Cs}$  was deposited world-wide from atmospheric fallout as a result of the above-ground nuclear weapons tests in the 1950s and 1960s, the major source of  $^{137}\text{Cs}$  on the Lithuanian territory before the Chernobyl NPP accident was the global fallout, especially from nuclear weapon tests in Novaya Zemlya, in the northern part of Russian Federation [52].

Ylipieti et al (2008) did a comprehensive investigation on  $^{137}\text{Cs}$  activity concentration in the top-layer forest soils in the Baltic States, Finland [56] as well as northwestern part of Russian Federation. The results showed that the global fallout of  $^{137}\text{Cs}$  was still clearly detectable in the humus layer (0-3 cm) in the Baltic States (Eastern coast of the Baltic Sea), especially in northeast Estonia and southern Finland. An average  $^{137}\text{Cs}$  activity concentration of  $6.8 \pm 1.8$   $\text{Bq kg}^{-1}$  was measured before Chernobyl accident [55]. The highest deposition area in the Baltic States was northeastern part of Estonia with a  $^{137}\text{Cs}$  deposition density of 550-720  $\text{Bq m}^{-2}$ . After the Chernobyl accident  $^{137}\text{Cs}$  activity concentration in the 0-20 cm arable soil increased to 6.7-28.5  $\text{Bq kg}^{-1}$  [57].  $^{137}\text{Cs}$  measurement in the Baltic coast [58] pre- and post-Chernobyl showed a highly heterogeneous distribution of  $^{137}\text{Cs}$  with the highest values of 380-440  $\text{Bq kg}^{-1}$ , while pre-Chernobyl values were up to 4-8  $\text{Bq kg}^{-1}$  only.

### 3.3 Temporal variation of radionuclides level in Lithuania.

Nedveckaitė et al. (1989) have measured  $^{131}\text{I}$  and other radionuclides in meadow/grass in Lithuania on 12 May, 1986[45] and observed that the most contaminated area was the south/southwestern part of Lithuania. According to the Chernobyl contamination level, Lithuanian territory was divided into two zones (zone I and zone II, Fig.1). In zone I (the northern and central region of Lithuania) only  $^{103}\text{Ru}$  and  $^{131}\text{I}$  were detected with the average activity concentration of  $^{131}\text{I}$  of  $1 \text{ kBq kg}^{-1}$ . Zone II was the most contaminated one, where the lowest activity concentration of  $^{131}\text{I}$  was  $1.2 \text{ kBq kg}^{-1}$  and the highest activity concentration of  $^{131}\text{I}$  was  $35 \text{ kBq/kg}$ . Besides  $^{131}\text{I}$  and  $^{103}\text{Ru}$ ,  $^{141}\text{Ce}$ ,  $^{144}\text{Ce}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{140}\text{La}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$  were also detected, demonstrated a significant impact of the Chernobyl accident in this area.

The samples measured in this work were mainly collected from zone II. Chernobyl derived  $^{129}\text{I}/^{131}\text{I}$  atomic ratio of 12-19 has been estimated theoretically and by measurements of  $^{129}\text{I}$  and  $^{131}\text{I}$  in samples collected in the highly contaminated area of Chernobyl accident [12]. The estimated  $^{129}\text{I}/^{131}\text{I}$  atomic ratio in this work is much higher than the Chernobyl derived  $^{129}\text{I}/^{131}\text{I}$  atomic ratio (Fig.6) by a factor up to 10 and in certain areas even by a factor 60 (fig 6. Range 30 and 360 km) indicating that Chernobyl accident has a limited contribution to the  $^{129}\text{I}$  inventory in Lithuania.

To quantitatively estimate the contribution of Chernobyl to  $^{129}\text{I}$ , we assume that  $^{129}\text{I}$  in Lithuania originated from both Chernobyl and global fallout and  $^{131}\text{I}$  is only originated from Chernobyl accident, our estimated ratio can be presented as:

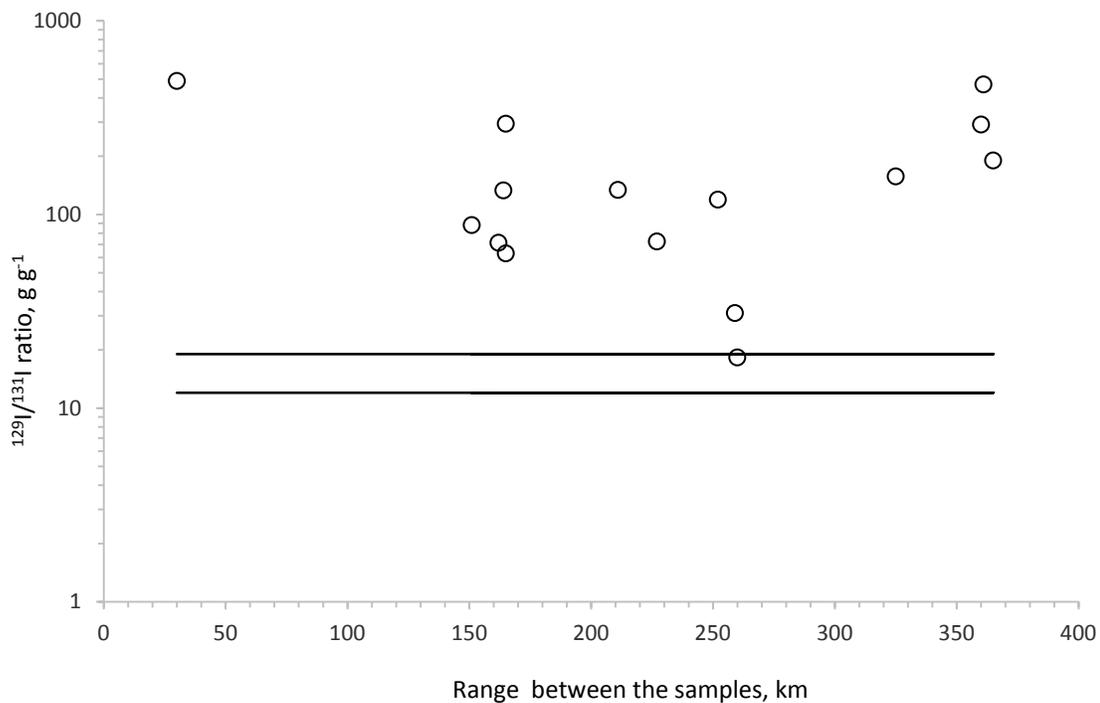
$$R_{est} = \frac{(I_{Ch}^{129} + I_{Gl}^{129})}{I_{Ch}^{131}};$$

$$\frac{I_{Ch}^{129}}{I_{Ch}^{131}} = R_{theo} \text{ is Chernobyl derived ratio, using the value reported by Mironov et al. (2002),}$$

Then global fraction of  $^{129}\text{I}$  is:

$$I_{Gl}^{129} = (R_{est} - R_{theo}) \cdot I_{Ch}^{131}.$$

The measured  $^{129}\text{I}/^{131}\text{I}$  ratio in most of the samples shows that more than 95% of  $^{129}\text{I}$  originated from the global fallout which includes nuclear weapon testing, reprocessing facilities, etc. Nevertheless, there are a few exception, e.g. the sampling point at 265 km, where 10% of  $^{129}\text{I}$  originated from the global fallout and the rest — of the Chernobyl fallout. The isotopic ratio of  $^{238}\text{Pu}/^{239,240}\text{Pu}$  at this point is 0.28 suggesting ~30% plutonium originated from the Chernobyl accident.

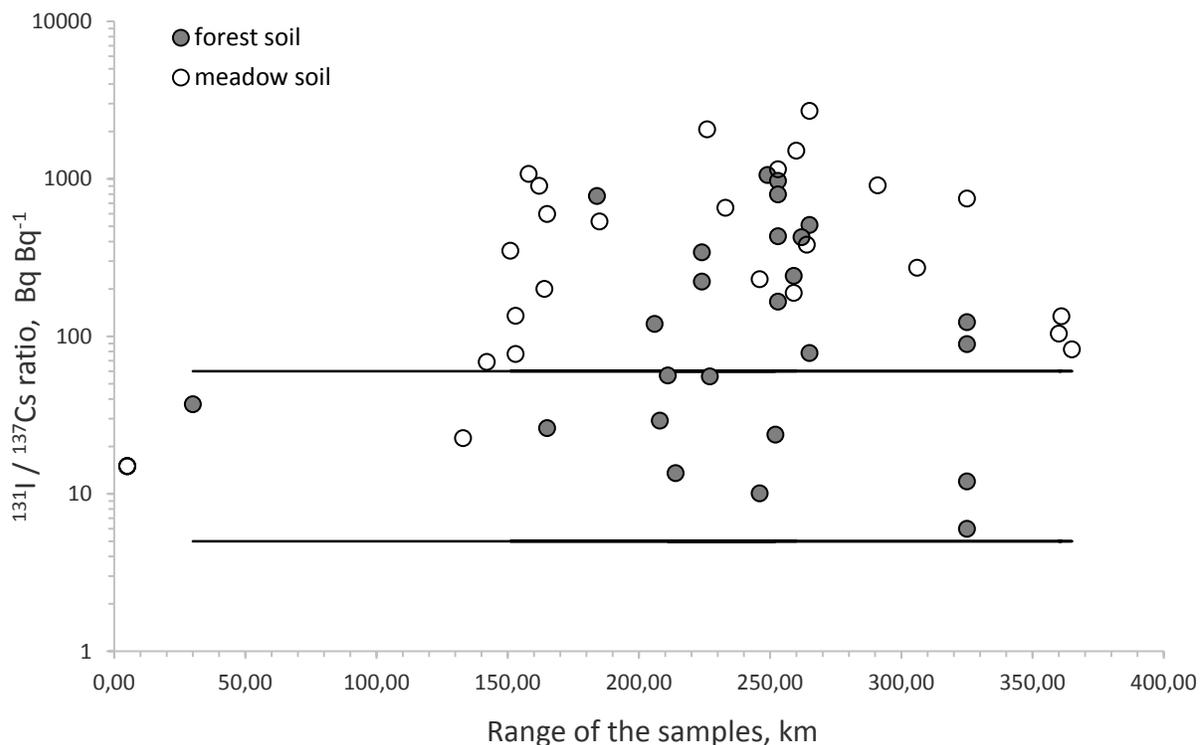


**Fig.6.** Estimated  $^{129}\text{I}/^{131}\text{I}$  atomic ratios in the investigated soil samples. Solid lines indicates the range of the Chernobyl derived ratio reported by Mironov et al. 2002 [12].

Chernobyl derived  $^{131}\text{I}/^{137}\text{Cs}$  activity ratio of 10.4 and  $^{129}\text{I}/^{137}\text{Cs}$  atomic ratio of 0.135 have been reported by Mironov et al. 2002. However higher  $^{131}\text{I}/^{137}\text{Cs}$  activity ratios of 40-92 have been observed in the samples taken in the Baltic States and Poland [59], [60]. This was interpreted as a significant fractionation between  $^{131}\text{I}$  and  $^{137}\text{Cs}$  during long distance transport and deposition of Chernobyl radioactive plume [12] such a fractionation was also observed in Fukushima derived contamination observed in Japan and Europe [61], [62].

$^{131}\text{I}/^{137}\text{Cs}$  ratios (decay corrected to 1986) in Lithuania are reconstructed using the measured  $^{137}\text{Cs}$  in this work and the reported  $^{131}\text{I}$  data [45] (Fig.7, open and full circles represent forest and meadow soil respectively). Large variation of  $^{131}\text{I}/^{137}\text{Cs}$  activity ratios of 6 - 2700 was observed.  $^{131}\text{I}/^{137}\text{Cs}$  activity ratios of 5-92 in soil have been reported [59], [60], which are shown in Fig. 7 (solid lines). The estimated  $^{131}\text{I}/^{137}\text{Cs}$  activity ratio in this work do not fall into the range of reported values (between two lines), most of them higher than the reported values. This indicates reduced  $^{137}\text{Cs}$  concentration in the measured samples comparing to the reported results. It might be attributed to the gradual removal of  $^{137}\text{Cs}$  from the soil by either erosion of soil or downwards migration of  $^{137}\text{Cs}$  in the soil column, disturbs of the soil due to human activity might be another reason causing a reduced  $^{137}\text{Cs}$  concentration in the top surface soil. Filled circles in Fig. 7 represent samples collected in the areas where minimal human impact on the soil (agriculture activity) occurred in the past 30 years. The  $^{131}\text{I}/^{137}\text{Cs}$  activity ratios in these samples are close to the range of two solid lines in Fig.7, confirming that human activity might be the major influence to the enhanced  $^{131}\text{I}/^{137}\text{Cs}$  ratios in the soil samples. Considering the high  $^{137}\text{Cs}$  levels in some Lithuanian soil samples are due to the Chernobyl fallout, we can estimate an average activity concentration of  $^{137}\text{Cs}$  of about  $500 \text{ Bq Kg}^{-1}$  (corrected to 1986) in these soil samples in Lithuania. With the reported  $^{129}\text{I}/^{137}\text{Cs}$  atomic ratio of 0.135 and  $^{131}\text{I}/^{137}\text{Cs}$  atomic ratio of 10.4 [12], the concentrations of

Chernobyl derived  $^{131}\text{I}$  and  $^{129}\text{I}$  in 1986 can be reconstructed to be  $\sim 4.6 \times 10^7$  atoms  $\text{g}^{-1}$  for  $^{129}\text{I}$  and  $\sim 2.5 \times 10^6$  atoms  $\text{g}^{-1}$  for  $^{131}\text{I}$ . The concentration of  $^{129}\text{I}$  in soil AMS10 (which contains the highest concentration of  $^{137}\text{Cs}$  250 Bq kg) was determined to be  $4.72 \pm 0.07 \times 10^8$  atoms  $\text{g}^{-1}$ , which is one order of magnitude higher than Chernobyl derived  $^{129}\text{I}$  indicating a negligible contribution of Chernobyl accident to the  $^{129}\text{I}$  inventory in Lithuania, meanwhile it also confirm the domination source of  $^{129}\text{I}$  is either from the Baltic Sea and North Sea where received high marine discharges of  $^{129}\text{I}$  from reprocessing plants, as well as the direct atmospheric releases from the two European reprocessing plants.



**Fig.7.** The estimated activity ratios of  $^{131}\text{I}/^{137}\text{Cs}$ . Solid lines indicates the range of the Chernobyl derived ratio reported by [59], [60].

## 5. Conclusions

The level of key long-lived radionuclides  $^{137}\text{Cs}$ ,  $^{129}\text{I}$  and Pu isotopes were analyzed in soil samples collected across Lithuania in 2011-2012. The ratios of these radionuclides in soil samples show that there are two major sources in Lithuania, i.e. the Chernobyl accident and reprocessing releases. Though the main contamination source for Pu is global fallout and Chernobyl fraction in most contaminated areas are 30%.

The  $^{129}\text{I}/^{127}\text{I}$  atomic ratio in the soil samples range from  $10^{-9}$  to  $10^{-6}$ , indicating that the main contamination source of  $^{129}\text{I}$  in Lithuania is not the Chernobyl accident but other sources – most probably the release from European nuclear reprocessing facilities.

The reconstructed  $^{129}\text{I}/^{131}\text{I}$  atomic ratios in most of the soil samples shows that more than 95% of  $^{129}\text{I}$  originated from the fallout, while in the southern part of Lithuania there are some locations indicating only 10% of  $^{129}\text{I}$  originated from the global fallout.

No correlation between  $^{137}\text{Cs}$  and  $^{129}\text{I}$  isotopes in all soil samples suggest that the contamination source and the origin of these two isotopes are different. The data show that the concentration of  $^{137}\text{Cs}$

contrary to the concentration of  $^{129}\text{I}$  did not increase over the past 30 years. It even decreases mostly due to a human (agriculture activity) activity.

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## Supplementary data

Table S-1. Additional sampling information.

Nr.	Code	Samling site Nr.	Coordinates	Soil type	Distance to the Sea, km
1	1.AN	111026.DIR.KAPČIAM_M	53°59'05"N 23°39'10"E	Forest	265
2	2.AN	111026.DIR.KAPČIAM_M'	53°59'41"N 23°39'03"E	Forest	206
3	3.AN	111026.DIR.KAPČIAM_P"	54°00'11"N 23°41'47"E	Meadow	185
4	4.AN	111026.DIR.KAPČIAM_P'	54°00'19"N 23°39'19"E	Meadow	246
5	5.AN	111026.DIR.KAPČIAM_P	54°00'54"N 23°39'20"E	Meadow	246
6	6.AN	111025.DIR.SUVALKAI_M	54°01'49"N 22°56'28"E	Forest	153
7	7.AN	111025.DIR.GLEBOKI_BROD_M	53°58'27"N 23°16'03"E	Forest	153
8	8.AN	111025.DIR.PLASKA_M	53°55'17"N 23°13'27"E	Forest	142
9	9.AN	111026.DIR.DIEVENIŠKĖS_P	54°09'59"N 25°36'39"E	Meadow	158
10	10.AN	111026.DIR.MARCINKONYS_M	54°03'06"N 24°24'01"E	Forest	184
11	11.AN	111026.DIR.DIEVENIŠKĖS_M	54°09'40"N 25°36'46"E	Forest	214
12	12.AN	111026.DIR.SEN.VARĖNA_P	54°15'17"N 24°32'25"E	Meadow	208
13	13.AN	111026.DIR.SEN.VARĖNA_P	54°15'17"N 24°32'25"E	Meadow	203
14	14.AN	111025.DIR.LAZDIJAI_P	54°11'18"N 23°28'58"E	Meadow	222
15	15.AN	111025.DIR.IGLIŠKĖLIS_P	54°33'48"N 23°31'54"E	Meadow	30
16	16.AN	111025.DIR.VIRBALIS_P	54°37'31"N 22°50'32"E	Meadow	222
17	17.AN	111025.DIR.VILKAVIŠKIA_P'_6'	54°36'46"N 23°09'33"E	Meadow	224
18	18.AN	111025.DIR.VILKAVIŠKIA_P_6	54°36'46"N 23°09'30"E	Meadow	224
19	19.AN	111026.DIR.DIEVENIŠKĖS_40m.P	54°10'36"N 25°35'20"E	Meadow	226
20	20.AN	111026.DIR.DIEVENIŠKĖS_100m.M	54°10'27"N 25°35'04"E	Forest	233
21	21.AN	111026.DIR.VAISIEJAI_P	54°06'21"N 23°40'08"E	Meadow	249
22	22.AN	111026.DIR.MARCINKONYS_P	54°03'06"N 24°24'04"E	Meadow	253
23	23.AN	111025.DIR.LIEPONYS_M	54°34'43"N 24°21'37"E	Forest	253
24	24.AN	111026.DIR.LEIPALINGIS_P	54°04'47"N 23°54'19"E	Meadow	253
25	25.AN	111026.DIR.DRUSKININKAI_M	54°01'52"N 23°58'55"E	Forest	253
26	26.AN	111025.DIR.SKRIAUDUPIS_P	54°37'28"N 23°12'07"E	Meadow	253
27	27.AN	111026.DIR.ŠALČININKAI-P	54°18'16"N 25°21'32"E	Meadow	265
28	28.AN	111025.DIR.GRAŽIŠKIAI_P	54°26'31"N 23°00'00"E	Meadow	265
29	29.AN	111026.DIR.STAKIAI_P	54°17'45"N 25°32'10"E	Meadow	262
30	30.AN	111025.DIR.PRIENAI_M	54°37'05"N 23°58'43"E	Forest	264
31	31.AN	111026.DIR.LATEŽERIS_M_SMĖL	53°59'16"N 24°08'04"E	Forest	259
32	32.AN	111025.DIR.SKRIAUDUPIS_M	54°37'29"N 23°12'08"E	Forest	291
33	33.AN	111026.DIR.LATEŽERIS_M	53°59'16"N 24°08'04"E	Forest	306
34	34.AN	111026.DIR.EIŠIŠKĖS_P	54°11'45"N 24°50'52"E	Meadow	325
35	35.AN	111026.DIR.LATEŽERIS_ŠK_P	53°59'02"N 24°14'08"E	Meadow	325
36	36.AN	111026.DIR.MARCINK-VARĖNA_M	54°05'02"N 24°24'16"E	Forest	325
37	37.AN	111026.DIR.LATEŽERIS_M'	53°59'23"N 24°08'04"E	Forest	325
38	38.AN	111026.DIR.LATEŽERIS_M_PAKAL	53°59'16"N 24°08'04"E	Forest	133