Solid-liquid distribution coefficients (Kd-s) of geological deposits at the Chernobyl Nuclear Power Plant Site with respect to Sr, Cs and Pu radionuclides: a short review

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Abstract

A review is presented of data on solid-liquid distribution coefficients (Kd-s) of the main radiologically important radionuclides of the Chernobyl release within geological deposits at the Chernobyl Nuclear Power Plant (ChNPP) Site. The Kd values for Sr, Cs and Pu for Quaternary sandy deposits that form sedimentary cover at Chernobyl fall within the range of parameters reported in international sorption databases. In agreement with general knowledge on radionuclide geochemical behavior and affinity to soils, Kd-s increase in the sequence: Sr < Cs < Pu. Alluvial and fluvioglacial sandy deposits are characterized by larger Kd values then deposits of eolian genesis due to higher content of clay minerals in fine fractions. For Sr, laboratory batch tests have given Kd values that are in a reasonable agreement with in situ measurements. At the same time, the ^{90}Sr Kd-s obtained from groundwater transport model calibrations were noticeably lower than experimentally determined values, thus showing potential limitations of the Kd-approach. Monitoring data on mobility of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in groundwater in the Chernobyl zone on a whole are consistent with the radionuclide Kd-s summarized in this article. The highest concentrations in groundwater (based on data for 2012-2014) were observed for ^{90}Sr, while orders of magnitude lower concentrations were observed for ^{137}Cs and ^{239,240}Pu. At the same time, detection of ^{137}Cs and ^{239,240}Pu in groundwater at sites with a relatively deep groundwater table suggests the possibility of facilitated transport of small amounts of these radionuclides in the form of non-retarded colloids or complexes.

Keywords: sorption distribution coefficient, Chernobyl accident, strontium-90, cesium-137, plutonium-239/240, groundwater contamination

1. Introduction

Background

The Chernobyl accident on 26 April 1986 resulted in serious contamination of the adjacent area by fallout radionuclides. Radioactive fallout, including an important component of nuclear fuel particles, were deposited on land areas and on water bodies, such as the cooling pond of the Chernobyl Nuclear Power Plant (ChNPP) (Vakulovsky et al., 1994; Kashparov et al., 2003). During the course of the emergency post-accident clean-up of the Chernobyl NPP site, several engineered radioactive waste storage facilities and hundreds of waste dumps (e.g., shallow trenches with waste dug into the local sandy soil) were created at the ChNPP site in the proximity of the damaged ChNPP Unit 4 (Antropov et al., 2001; Molitor et al., 2017).

During the last decade, the 'Vector' complex for disposal of radioactive waste originating from decommissioning of ChNPP and remedial works in Chernobyl Exclusion Zone was created in the elevated area dividing the watersheds of the Uzh and Pripyat Rivers at about 14 km distance from the ChNPP (Bugai et al., 2017) (Fig.1). Recently, a decision was made to develop, on the basis of the Vector complex, a national centralized radioactive waste disposal and storage facility for wastes originating both in the nuclear sector, as well as radioactive wastes of industrial and medical origin in the Ukraine (Lisichenko et al., 2017).

During the more than 30-year period that has passed since the Chernobyl accident a number of studies were carried out in order to predict radionuclide transport in the geosphere from the radioactivity sources of accidental origin situated at ChNPP site (waste dump sites, cooling pond, Sarcophagus etc.) (e.g., Bugai et al., 1996, 2005; Kivva et al., 1996; Onishi et al., 2007; Molitor et al., 2017), as well as to carry out performance assessment analyses for the planned radioactive waste disposal and storage facilities of the Vector Complex (Shehtman et al., 1996; Bugai et al., 2017). In some instances, the studies mentioned above included experimental determination of site-specific parameters for radionuclide transport in the subsurface environment. In particular, the key parameter governing radionuclide transport in the unsaturated zone and groundwater is the solid-liquid distribution

coefficient (K_d), describing radionuclide partitioning between the pore water and soil matrix due to various chemical and physical process (e.g., ion-exchange, surface complexation, volume diffusion, etc.), which are commonly termed 'sorption'. The sorption studies encompassed the main radiologically important long-lived radionuclides of Chernobyl origin: strontium-90, cesium-137 and plutonium-238, -239 and -240 isotopes. The K_d determinations were carried out for different types (lithological, genetic) of geological deposits, and employed various experimental techniques.

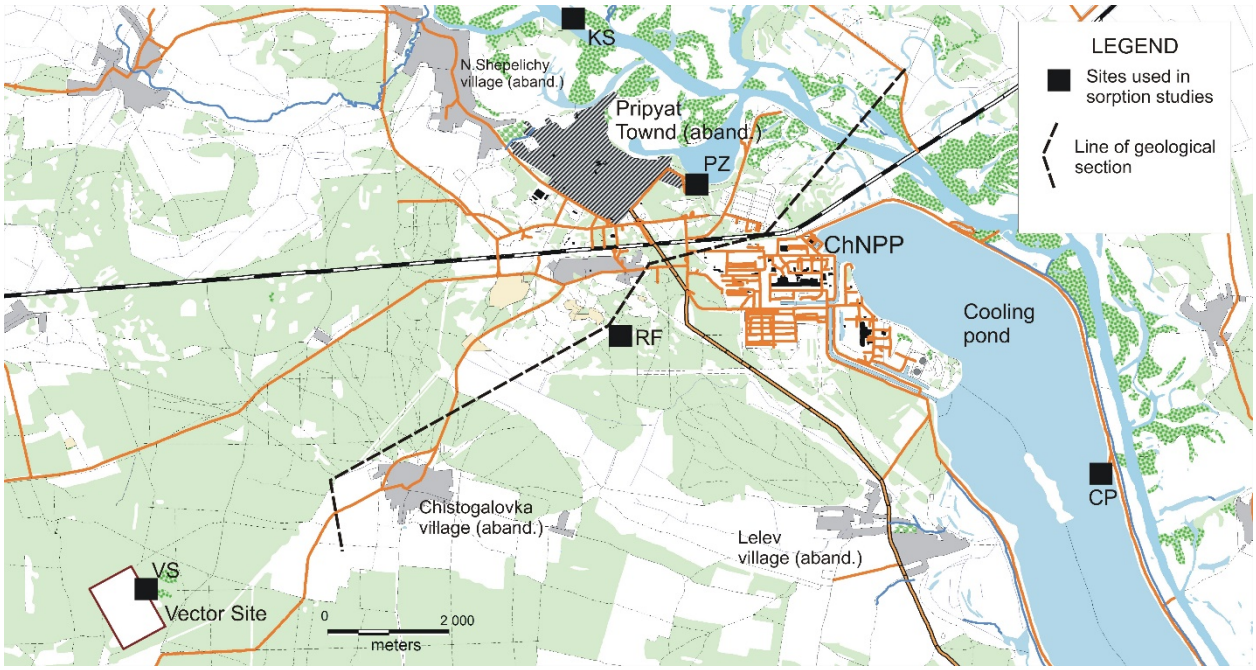

Fig.1. Map of the central part of the Chernobyl Exclusion zone showing sites that have been sampled in the reviewed sorption studies.

Objectives

The objective of this article is to carry out review of data on sorption distribution (partition) coefficients of the radiologically important (in the long term) radionuclides of the Chernobyl release for geological deposits at the ChNPP site. Particular attention is paid to the lithological characteristics of geological deposits (soils), and to methods of K_d determination. Where available, data are documented on physical and geochemical properties of deposits and of test solutions used in sorption tests. This allows a cross-comparison of sorption properties of various deposits, as well as of experimental methods for K_d determination.

Scope

In this review we focus on sorption properties of geological deposits that form the upper part of geological strata below the organic topsoil layer, and would usually host the first (from the surface) phreatic aquifer at the Chernobyl NPP site. This is the geological environment where migration of radionuclides typically occurred from the key sources of radioactivity such as near-surface radioactive waste disposal sites (or waste dumps) and/or contaminated water bodies (e.g., Chernobyl cooling pond).

The data for the more organic topsoil layer a (e.g., upper 20 - 50 cm depth range of soil) are not included in this review, as these data can be found elsewhere (e.g., (IAEA, 2010)). In addition, radionuclide migration in topsoil is influenced by a larger spectrum of factors (including bio-geochemical processes and radionuclide cycling in the 'soil – plant' system), and therefore represents a special subject, that was deemed to be outside of the scope of this article.

We also briefly summarize key geochemical process and factors that control the adsorption-desorption behavior of reviewed radionuclides on the matrix of geological deposits, and related specific experimental data (where available) for geological deposits in Chernobyl Exclusion Zone (CEZ).

This article makes available to the international scientific community data from a number of Russian- and Ukrainian- language publications and reports that, to the knowledge of the authors, have not been reviewed before in the English – language literature. The Chernobyl experience provides interesting insights into different methods of K_d determination, as well as validation of the experimentally determined K_d parameter values by calibration of subsurface radionuclide transport models. It also shows some limitations and pitfalls of the K_d approach for describing field behavior of specific radionuclides in the subsurface.

2. Chernobyl site geology and hydrogeology

Samples of geological deposits used for sorption studies reviewed in this publication were usually taken from near-surface excavations (pits with depth of up to approximately 1 m) or from boreholes drilled at the ChNPP site (with depth ranging from several meters to several tens of meters). We will first give a

95 brief overview of the geological structure and lithology composition of geological deposits forming the
96 upper part of sedimentary cover at the ChNPP site.

97 ***Geological structure***

98 The upper part of geological strata at ChNPP site that hosts the unconfined aquifer is composed of
99 Quaternary Pliocene and Pleistocene-Holocene sandy deposits (Matoshko et al. 2002, 2004) (Fig. 2). The
100 total thickness of these deposits is about 30 m. The above sediments are underlain by the regional low-
101 permeability aquitard layer of carbonate clays (marls) of the Kiev Suite of the Eocene (the thickness of
102 this aquitard layer is 10-20 m).

103 ***Geomorphology and lithology***

104 With regard to geomorphology, the following elements can be distinguished at the ChNPP site (see
105 Fig.2): (1) first terrace of the Pripyat River (where the ChNPP is located) which is composed mainly of
106 Lower - Middle Pleistocen alluvial sandy deposits; (2) Quaternary plateau which is composed of Pliocene
107 and Middle Pleistocene fluvial-glacial sandy and silty-clayey deposits (to the SW of ChNPP), and (3)
108 floodplain of Pripyat River composed of Holocene alluvial sandy deposits (to NE of ChNPP).

109 The Quaternary alluvial deposits are most often represented by strata of relatively homogeneous fine-
110 medium sands, with inter-bedded layers of loam and silt materials. Thickness of lenses of finer deposits
111 vary from several centimeters to 1–2 m and more (Matoshko et al., 2004).

112 The fluvial-glacial deposits that form upper part of geological section within the Quaternary Plateau (see
113 Fig.2) are composed of predominantly sands with inter-layers of silt and clayey moraine deposits. The
114 thickness of these layers vary from tenths of centimeters to 3-4 m or more (Shehtman et al., 1996; Bugai
115 et al., 2017).

116 Throughout the site, the alluvial and fluvioglacial deposits are covered from the surface by a layer of
117 eolian sandy deposits (with a typical thickness from less than one to several meters). This layer of eolian
118 sands has formed during the Late Pleistocene due to of wind erosion of fine fractions of alluvial (or
119 fluvioglacial) sandy deposits, with wind transportation of deposited particles to short distances. It is
120 composed of well-sorted fine sands (Matoshko et al., 2004).

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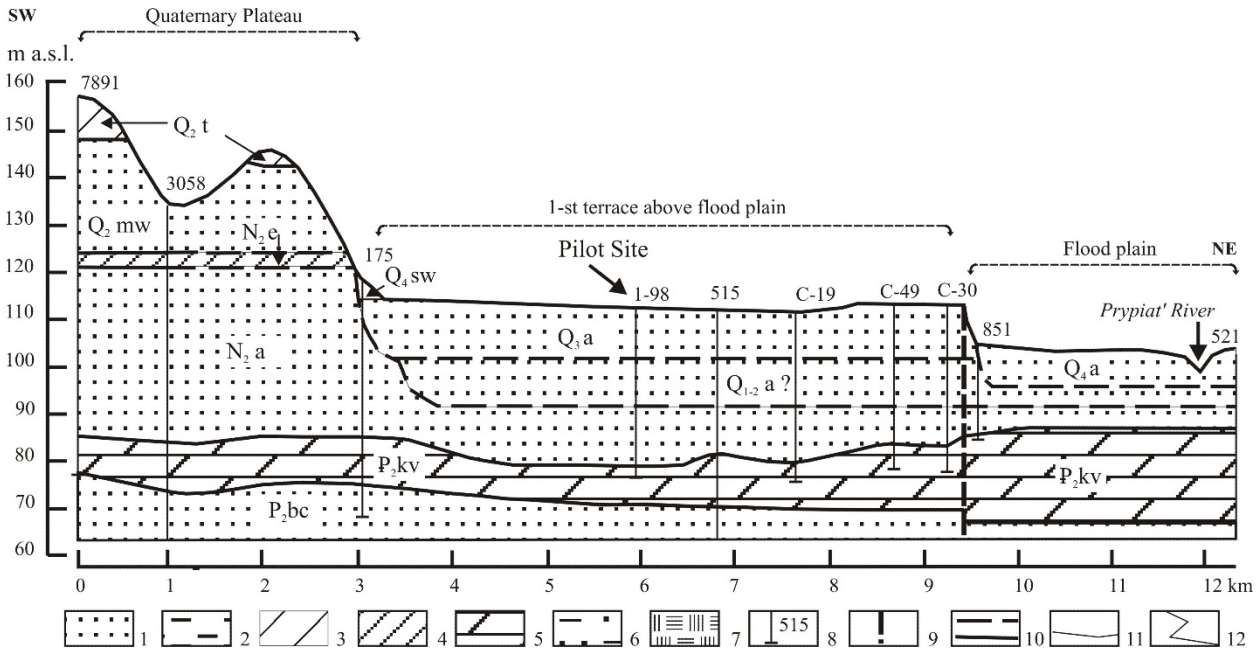

Fig.2. Geologic cross-section of the ChNPP site (Position of the cross-section is shown in Fig. 1) (Matoshko et al., 2004). Legend: 1 – sands, 2- silts, 3 – basal till, 4 – clay, 5 – marl, 6 – inter-bedding of sands and silts, 7 – peat and peaty sand, 8 – boreholes (numbered), 9 – inferred fault, 10 – boundaries between suites: supposed (upper) and established (lower), 11 - boundaries between depositional facies, 12 – facial replacement, 13 – groundwater level (generalized). Indices: Q_4 - Holocene, Q_{3-4} - Upper Pleistocene - Holocene unstratified, Q_3 - Upper Pleistocene, Q_{1-2} - Lower Pleistocene - Middle Pleistocene unstratified, N_2 - Pliocene, P_2 – Eocene; kv - Kyiv, bc – Buchack. Genetic types of deposits: a - alluvial, mw – melt-water, eol - aeolian, e - presumably waste mantle, sw – slopewash. Facies: ob – overbank, ch – channel, a-ch – abandoned channel.

Bulk density of sandy deposits usually ranges from 1.6 to 1.8 g/cm^3, while porosity usually ranges from 33 to 42% (Matoshko et al., 2004; Bugai et al., 2017). The eolian deposits have low cation-exchange capacity (CEC) values, which are usually less than 1 meq/100 g, while the alluvial and fluvio-glacial deposits have CEC values in the range from 2 to 10 meq/100 g (depending on content and mineralogy of the clay fraction) (Matoshko et al., 2004; Bugai et al., 2017). Based on data of (Kuznetvov, 1973; Olkhovik et al., 1992; Matoshko et al., 2004) Quaternary alluvial sandy deposits of the study area are predominantly composed of quartz with admixtures of the feldspar (up to ~10 %). The clay fraction of alluvial deposits is composed of kaolinite, montmorillonite and hydromica (illite), as well as fine-dispersed calcite, quartz, and amorphous ferrous oxides. In eolian sands, the content of quartz reaches 98–99%, while the clay-size (finest) fraction is almost totally composed of finely-dispersed quartz.

Hydrogeology

The sandy Quaternary deposits host the first unconfined aquifer from the surface, which is recharged by meteoric waters. The depth to groundwater ranges from 0 – 2 m in floodplain areas to 5-6 m within the first terrace of Pripyat River, and up to 12-14 m and more within the Pripyat River and Uzh River water divide area within the Quaternary Plateau. The thickness of the unconfined aquifer varies from 15 m to 30-40 m; the transmissivity is estimated at 100-400 m^2/day. The groundwater is usually calcium/magnesium - bicarbonate type; the total dissolved solids typically vary from 100 to 500 ml/g, the pH value is in the range 5.6 - 6.2 (Dzhepo and Skalskyy, 2002).

More details on the Chernobyl NPP site geology and hydrogeology can be found in (Dzhepo and Skalskyy, 2002; Matoshko et al. 2002, 2004)

Specific locations at ChNPP site sampled in the course of sorption studies

The sorption studies reviewed in this article were carried out on geological deposit samples taken from several distinct locations in the Chernobyl zone listed in Table 1. These sites represent important locations from the perspective of radionuclide migration from radioactive sources of accidental origin to the surrounding near-surface geological environment (including the "Red Forest" waste dump site and Chernobyl cooling pond). These sites encompass all the main geomorphological areas of the ChNPP site (see Fig.1 and 2) as discussed in previous paragraphs.

Table 1. Lists of specific sites that have provided samples for sorption studies reviewed in this article.

Site*	**Abbreviation**	**Geomorphological area**	**Genetic type of deposits**
Cooling pond	CP	Pripyat River flood plain	alluvial
Krasnensky Starik	KS	Pripyat River flood plain	alluvial
Red Forest	RF	First terrace of Pripyat River	Eolian, alluvial
Pripyat Zaton	PZ	First terrace of Pripyat River	Eolian, alluvial
Vector Site	VS	Quaternary Plateau	Fluvial-glacial, alluvial

Remark: Several sites ('Colling pond', 'Pripyat Zaton', 'Krasnensky Starik') in Table 1 are named after adjacent water bodies; however, it should be stressed that samples of geological deposits at these sites were taken from surrounding geological formations, not from bottom sediments of these water bodies

3. Data sources

The sorption studies reviewed in this article were usually carried out in order to provide parameters for radionuclide transport modelling and risk assessment analyses as a part of environmental impact assessment studies for major radioactivity sources of the accidental origin such as the radioactively contaminated cooling pond (e.g., Bugai et al., 1997; Bugai et al., 2005) and/or "Red Forest" waste dumps (e.g., Radium Institute, 1992; Olkhovik et al., 1992), and also to support safety assessments for the planned radioactive waste repositories at Vector Site (e.g., Shehtman et al., 1996).

The most comprehensive sorption studies in the Chernobyl zone during last two decades were carried out within the international radioecological research projects Chernobyl Pilot Site Project (CPS Project, 1999–2004) and Experimental Platform in Chernobyl (EPIC, 2004–2012) (Dewière et al., 2005; Van Meir et al., 2009; Kashparov et al., 2012; Bugai et al., 2012). These projects studied radionuclide migration from near-subsurface radioactive waste burial (trench no.22) at "Red Forest" radioactive waste dump site containing nuclear fuel particles. Apart from sorption studies on deposits collected immediately from the trench no.22 site, sorption experiments were also carried on samples collected from the geological analog of the CPS Site, the ''Pripyat Zaton'' exposure (e.g., Devol-Brown et al., 2002; Szenknectet al., 2005), which represents the natural cliff of the river terrace extending along the Pripyat Zaton Inlet of the Pripyat River at 2 km distance from the CPS/EPIC site (see Fig. 1). Here the upper part of the geological cross-section is situated in unsaturated conditions, which simplified access to deeper soil layers and sample collection.

4. Distribution coefficient (K_d) as a sorption parameter

Definition of Kd

The sorption distribution coefficient (Kd) is an empirical parameter defined as the ratio of the quantity of the contaminant of interest adsorbed per unit mass of solid (geological deposit) to the quantity of the contaminant remaining in aqueous phase (groundwater) per unit volume at chemical equilibrium (or at "quasi-equilibrium" state reached during the experiment). Inherent in the K_d- model (or "linear

isotherm" adsorption model) is the assumption that adsorption of the contaminant of interest on solid phase is linearly dependent of its concentration in the aqueous phase. This assumption typically holds only for low contaminant concentrations in solution.

K_d approach for quantifying sorption: advantages and limitations

Process that are termed 'sorption' (i.e., binding of solute species to matrix of geological deposits) in a general case can include various physical and chemical mechanism, including: ion exchange, surface complexation, surface (co-) precipitation of mineral phases, incorporation to mineral structure either by volume diffusion or crystal growth (inclusion) (Neumann, 2012). With the Kd approach, all of the interactions listed above of solute with solid phase are bulked into one empirical parameter (i.e., the distribution coefficient).

An important aspect of the K_d is that values are sensitive not only to the nature and properties of the mineral matrix, but also to geochemistry of the aqueous phase e.g., presence of ions competing for adsorption sites, solution ionic strength, pH, redox potential, presence of organic ligands, mineral (or organic) colloids. One other underlying assumption for application of the Kd sorption model in radionuclide transport calculations is that radionuclide sorption process is instantaneous and fully reversible. The "non-ideal" behavior of radionuclides in field conditions (e.g., relatively slow "kinetically controlled" solute exchange between aqueous phase and solid phase; or partly irreversible sorption - so called "sorption hysteresis") can lead to deviations between Kd- based modeling predictions and field observations of radionuclide migration in the subsurface.

To address the listed difficulties, the last decades have seen an increasing amount of research on developing mechanistic geochemical contaminant transport and sorption models (e.g., see recent review by (Steefel et al., 2015)). However mechanistic models are much more demanding in terms of geochemical expertise of the modeler, input data on solution geochemistry and solid matrix surface properties, and on databases of thermodynamic constants for relevant geochemical and bio-chemical reactions. The above requirements often limit the practical application of mechanistic sorption models.

Therefore, despite the limitations discussed above, the Kd model still represents probably the most common approach for quantifying interactions of radionuclides with the solid matrix of geological materials in risk assessment and performance assessment analyses. In some instances, the Kd model is an interim measure until sufficient data is generated for a more complex model (M.I. Sheppard and S.C. Sheppard, 2002). The advantage of the Kd approach includes relatively simple and well established methods of experimental determination (e.g., batch laboratory tests; see below), as well as availability of extensive published databases of Kd values for various soils and geological materials (e.g., (Sheppard and Thibault, 1990; Serne, 2007; IAEA, 2010)). The modeler, however, needs to be aware of the above limitations and potential pitfalls of the Kd model.

5. Methods of Kd determination

Several methods have been used to measure (or estimate) radionuclide Kd values for geological deposits in the Chernobyl exclusion zone. These methods are briefly described below.

Laboratory batch tests

The method that has been used most often for sorption studies is the laboratory batch method. In this method, the known mass of soil is added to laboratory vessel containing the known concentration (or activity) of radionuclide of interest in soluble form. The vessel is sealed and stirred until equilibrium is supposedly reached between the solid and aqueous phase (typically, from several hours to several days, though this does not necessarily ensure full equilibrium). Upon the completion of experiment, the solutions and solid phase area separated (e.g., by centrifuging), and the radionuclide concentration in the residual solution is measured. The Kd value is then calculated from radionuclide concentration loss from solution (due to soprption on solid phase) using simple mass balance equations (Olkhovik et al., 1992; Devol-Brown et al., 2002). The pitfall of laboratory batch tests is potential dependence of resulting Kd values on 'liquid – solid' phase ratio (due to changes in specific surface area of solid phase per unit volume of liquid, changes in chemical composition of liquid phase, etc.), as this ratio can be in batch tests much higher than in natural (field) conditions (Konoplev et al., 1988). To address this difficulty,

batch tests can be carried out at relatively small 'liquid – solid' ratio (e.g., 1:1), and using the pre-equilibration of solid phase with the solution of known chemical composition.

Field in-situ partition tests

In this method, the water saturated geological core sample is collected using auger drill directly from the contaminated aquifer. Upon extraction from the aquifer, the aqueous phase is separated from the solid phase directly in the field (e.g., by means of application of a vacuum to the water saturated core sample). The soil sample containing residual pore water is sealed. The solute sample is stabilized, and directed along with soil sample to laboratory. In the laboratory, the solid samples are analyzed for residual water content, and both solute and soil samples are analyzed for radionuclide content. These data allow an in situ Kd value to be calculated. This method is relatively experimentally simple, and was used quite often in the Chernobyl zone which offered important opportunities for sampling contaminant plumes in groundwater (e.g., Gudzenko et al., 1994; Bugai et al., 2001; Levchuk et al., 2009).

Column tests

The column test method uses an inert (i.e., not interacting with soil) tracer (e.g., tritium) as well as a solution containing radionuclide in soluble form, both of which are filtered through a column packed with soil. Observation of breakthrough curves at the exit from column allows estimation of the radionuclide retardation factor as a ratio of velocities of inert tracer and radionuclide of interest. Next, radionuclide Kd value is calculated from the retardation factor and known soil parameters (bulk density, porosity). Column tests were used in the reviewed Chernobyl studies only in a few instances (e.g., Szenknect, 2003), as this method requires a more complex experimental setup, and can take long time (in case radionuclide has a large Kd and respective retardation factor value)

Groundwater transport model calibration

In case groundwater monitoring data are available for radionuclide migration in the subsurface from the radioactivity source with the well-known (or reasonably accurately estimated) release history (e.g., release occurs from a contaminated surface water body in which activity concentration was monitored),

264 the Kd can be estimated by means of calibrating a groundwater transport model. The Kd value obtained
265 from model calibration can be considered as an 'effective' transport parameter that represents space-
266 averaged (and/or time-averaged) value, and it may be dependent on the scale, time interval and model
267 conceptualization of the monitored radionuclide transport process in the subsurface. In the case of
268 Chernobyl exclusion zone, such model calibration studies were carried out for ^{90}Sr transport from the
269 Chernobyl NPP cooling pond (Bugai et al., 2005), and for ^{90}Sr transport from trench no.22 at Red Forest
270 site (Bugai et al., 2012).

271 The detailed discussion of experimental procedures, advantages and limitations of different methods of
272 Kd determination can be found in (US EPA, 1999a).

273 **6. Sorption parameters of radionuclides**

274 In this section, Kd values for individual radionuclides are presented as compiled from the reviewed
275 literature. Compilation of Kd values is presented in the form of set of tables (for specific radionuclides).
276 Each entry of the table provides information on the geological sample collection site in Chernobyl zone,
277 lithology of studied samples of deposits, experimental method of Kd determination, number of samples
278 analyzed, mean and range (minimum, maximum) of measured/estimated Kd values, and reference to
279 the source of data (publication, report). In the case that an article provides information on Kd values for
280 several types of deposits, the Kd values are further sub-divided in accordance with the lithological
281 features of deposits (e.g., 'sand', 'sandy loam', etc.). Lithological characteristics of geological deposits
282 (e.g., as 'sand', 'fine sand', 'sandy loam') are based on descriptions which are provided in the initial
283 reference.

284 In addition, each line in table includes a unique ID corresponding to the site, lithological type of deposits
285 and specific reference for the reported Kd values. Based on this ID, additional information related to
286 experimental conditions of Kd determination including physical and hydraulic characteristics of deposits
287 (e.g., grain size distribution, sediment density, porosity, hydraulic conductivity etc.) and on chemistry of
288 test solution (e.g., ion composition, pH etc.) can be found in the supplementary Annex to this article
289 (when such information is available in the original reference). The list of provided supplementary data

varies from on reference to other reference (depending on information available in respective reference).

In the presented compilation of Kd data, we use the commonly utilized assumption that all isotopes of the same radionuclide (e.g., ^{238}Pu, ^{239}Pu and ^{240}Pu; or ^{85}Sr and ^{90}Sr) have the same Kd values (e.g., (EPA, 1999a; IAEA, 2010)).

Strontium

Based on literature data, the strontium sorption on soils is usually governed by ion exchange mechanism on regular exchange sites (RES) in clays and organic matter (Gil-García et al., 2009, 2011a). In accordance with the ion-exchange sorption model, the radiostrontium Kd is essentially dependent on the CEC of deposits (in particular fine fraction, clay minerals with large surface area) and it is also sensitive to the presence of competing cations (Ca, Mg, stable Sr) in solution (Patterson and Spoel 1981; Lefevre et al. 1996; Szenknect, 2003). A fraction of the radiostrontium (usually relatively small) may become fixed with time by the solid phase (an ageing effect). Such fixation effects were reported for ^{90}Sr sorption on iron hydroxide coatings on mineral sand grains (Jackson and Inch, 1989) and for organic substrates of soils (Boyer et al., 2018).

Strontium-90 was the most mobile radionuclide in soils, groundwater and surface water of the Chernobyl Zone during the post-accident period compared to cesium and plutonium radioisotopes (Bugai et al., 1996). Therefore, sorption behavior of radio-strontium was studied in a larger number of studies, that resulted in bigger statistics of Kd values for this radionuclide. Radiostrontium Kd estimates for sand deposits of various genesis vary from less than 1 ml/g to about ~20 ml/g; the values for clay - loam deposits reach up to ~80 ml/g (Table 2). It should be noted that in a number of reviewed studies, laboratory sorption tests have used ^{85}Sr rather than ^{90}Sr.

313 Table 2. Compilation of Kd values for ^{90}Sr / ^{85}Sr based on studies conducted on geological deposits from
314 the Chernobyl Exclusion Zone.

ID	Site	Type of deposits	Method	Number of samples	Kd, ml/g Mean (range)	Reference
RF1A	Red Forest	sand	batch	3	2.8 (1.9-4.0)	Radium Institute, 1992
RF2A	Red Forest	Fine sand	batch	7	2.7 (1.1-4.4)	Olkhovik et al., 1992
RF2B	Red Forest	Sandy loam	batch	3	7.0 (4.5-10.5)	Olkhovik et al., 1992
RF3A	Red Forest	Fine sand	In situ	6	2 (0.2-5)	Bugai et ai., 2002
RF4A	Red Forest	Fine sand	Model calibration	1	0.5 - 1	Dewiere et al., 2005
VS1A	Vector	Coarse sand	batch	2	4.7 (4 - 5.5)	Shehtman et al., 1996
VS1B	Vector	Medium sand	batch	10	3 (1 – 6.5)	Shehtman et al., 1996
VS1C	Vector	Fine sand	batch	32	3.7 (1 – 10)	Shehtman et al., 1996
VS1D	Vector	Silty sand	batch	15	5.8 (2.2 – 12)	Shehtman et al., 1996
VS1E	Vector	Sandy loam	batch	10	19.5 (5 – 45)	Shehtman et al., 1996
VS1F	Vector	Clay loam	batch	8	34.6 (9.7 – 77)	Shehtman et al., 1996
PZ1A	Pripyat Zaton	Fine sand	batch	1	9	Devol-Brown et al., 2002
PZ2A	Pripyat Zaton	Fine sand	batch	2	2.8 – 2.9	Bugai et ai., 2001
PZ2B	Pripyat Zaton	Medium sand	batch	1	6.4	Bugai et ai., 2001
PZ2C	Pripyat Zaton	Silty sand	batch	3	35 (19 – 50)	Bugai et ai., 2001
PZ3A	Pripyat Zaton	Fine sand	batch	1	12.5	Szenknect et al., 2005
CP1A	Cooling pond	Medium sand	In-situ	6	4.3 (1.9 - 7.4)	Gudzenko et al., 1994
CP2A	Colling pond	Medium sand	Model calibration	6 (locations)	0.8 (0.5 – 1.3)	Bugai et al., 2005
KS1A	Krasnensky Starik	Fine sand (alluvial)	In situ	2	17.2 (12.3 – 22.1)	Gudzenko et al., 1994

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316 Laboratory stirred flow-through reactor experiments have shown that radiostrontium sorption on eolian
317 sand deposits from Chernobyl Pilot Site in the “Red Forest” is a ‘‘fast’’ and reversible process
318 (characteristic sorption time <1 min; desorption 100%) (Szenknect et al, 2005). In field conditions, about

10% of the ^{90}Sr in sand cores collected from alluvial aquifer at cooling pond that was exposed to contamination for a period of about 10 years (i.e., the time elapsed since the Chernobyl Accident in April 1986) was found to be in a fixed from (not extractable by a 0.1 M $CaCl_2$ solution) likely reflecting sorption ageing effect (Bugai et al., 1997).

Cesium

Radiocesium isotopes (e.g., cesium-134, cesium-137) are generally known to have relatively low mobility in the subsurface environment due to high sorption coefficients on soils and geological deposits. Retention of Cs radionuclides on soils is known to be essentially governed by adsorption reactions by ion exchange mechanism, where certain types of micaceous clay minerals play an important role. In particular, sorption of Cs on illite is known to be highly selective, and can be partly irreversible (this last process is called "fixation"), or desorption process can be very slow; competitive effects from K^+ and NH_4^+ ions are also important factors (Cornell, 1993; Gil-García et al., 2009, 2011b). In Fukushima soils, similar selective and partly irreversible adsorption of Cs on weathered biotite clay mineral has been observed in laboratory tests (Mukai et al., 2016). Apart from migration in dissolved ionic form, upon certain conditions (oxic conditions, low ionic strength, high flow rates) Cs can be transported in groundwater in the form of colloids (both of inorganic and organic nature), however the mass fraction of radionuclide transported in colloidal form usually is quite low (Kersting, 2012).

The ^{137}Cs (along with ^{90}Sr) is the most radiologically important radionuclide of the Chernobyl release, therefore its sorption behavior has been studied in a relatively large number of studies in the CEZ. Cesium Kd estimates for sand vary from less than 7 ml/g to 670 ml/g; the Kd values for clay - loam deposits fall to the range of 300 - 8300 ml/g (Table3). Being less mobile, ^{137}Cs was usually found in groundwater of the CEZ during post-accident period at several orders of magnitude lower concentrations compared to ^{90}Sr (Bugai et al., 1996; Dzhepo and Skalskyy, 2002). Presumably due to fixation on clay minerals, the amount of mobile (and bio-available) forms of ^{137}Cs has shown a tendency to decrease in soils of Chernobyl zone following the accident (Ivanov and Kashparov, 2003).

345 Table 3. Compilation of Kd values for ^{137}Cs based on studies conducted on geological deposits from the
346 Chernobyl Exclusion Zone.

ID	Site	Type of deposit	Method	Number of samples	Kd, ml/g Mean (range)	Reference
RF1B	Red Forest	Sand	batch	2	32.5 (27 – 38)	Radium Institute, 1992
RF1C	Red Forest	Sandy loam	batch	2	200 (160 – 240)	Radium Institute, 1992
RF1D	Red Forest	Clay loam	batch	1	311	Radium Institute, 1992
VS1A	Vector	Coarse sand	batch	2	155 (150 – 160)	Shehtman et al., 1996
VS1B	Vector	Medium sand	batch	10	270(120 - 640)	Shehtman et al., 1996
VS1C	Vector	Fine sand	batch	32	300 (90 – 800)	Shehtman et al., 1996
VS1D	Vector	Silty sand	batch	15	480 (170 – 1080)	Shehtman et al., 1996
VS1E	Vector	Sandy loam	batch	10	1260 (210 – 4500)	Shehtman et al., 1996
VS1F	Vector	Clay loam	batch	8	2070 (300 - 8300)	Shehtman et al., 1996
PZ1A	Pripyat Zaton	Fine sand	batch	1	43	Devol-Brown et al., 2002
PZ4A	Pripyat Zaton	Fine sand	batch	4	500 (410 – 580)	Szenknect, 2003
PZ4B	Pripyat Zaton	Fine sand	column	1	1220 (R=6600)	Szenknect, 2003

347

348 Plutonium isotopes

349 Plutonium has the most complex geochemistry among the reviewed radionuclides. The adsorption of
350 Pu by soils depends upon its oxidation state, groundwater pH, complexation with dissolved organic
351 ligands, and other factors. Pu can exist in the aquatic environment in four different valence states (+3,
352 +4, +5, +6). The reduced forms of Pu (+3, +4) are generally 2- 3 orders of magnitude less mobile than the
353 oxidized forms (+5, +6) due to adsorption and precipitation reactions. Relatively high Kd values
354 determine generally low mobility in groundwater in the far-field (EPA, 1999b; Cantrell and Felmy, 2012).
355 The mobility of Pu in the subsurface can be significantly promoted by colloid formation and

356 complexation reactions, leading to facilitated transport of low-retarded anionic forms of Pu associated
357 with low-molecular organic compounds; at the same time, the fraction of fast migrating forms of Pu is
358 usually relatively low (Kersting, 2012; Santschi et al., 2017).

359 Adsorption parameters of Pu isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu) on local geological deposits from the
360 ChNPP site have been analyzed in relatively fewer studies compared to Sr and Cs. Less interest in Pu can
361 be due to less abundance of Pu in the radioactivity source-term in CEZ (^{239,240}Pu:^{137}Cs activity ratio in
362 dispersed nuclear fuel particles was 0.012 in 1995 (Antropov et al., 2001)) and due to generally known
363 lower mobility in soils and the aquatic environment, so that Pu did not pose an immediate hazard for
364 contaminating water resources in the CEZ. Plutonium Kd values for sand estimated by different methods
365 vary from 7 ml/g to 670 ml/g; the Kd values for clay - loam deposits vary from 200 - 6100 ml/g (Table4).

366 Table 4. Compilation of Kd values for Pu isotopes based on studies conducted on geological deposits
367 from Chernobyl Exclusion Zone.

ID	Site	Type of deposits	Method	Number of samples	Kd, ml/g Mean (range)	Reference
RF1E	Red Forest	Sand	batch	4	360 (150 – 670)	Radium Institute, 1992
RF5A	Red Forest	Fine sand	In-situ	3	25 (7-47)	Levchuk et al., 2009
VS1A	Vector	Coarse sand	batch	1	180	Shehtman et al., 1996
VS1B	Vector	Medium sand	batch	6	180 (70 – 380)	Shehtman et al., 1996
VS1C	Vector	Fine sand	batch	16	187 (50 – 490)	Shehtman et al., 1996
VS1D	Vector	Silty sand	batch	6	255 (100 – 410)	Shehtman et al., 1996
VS1E	Vector	Sandy loam	batch	6	1415 (200 – 2900)	Shehtman et al., 1996
VS1F	Vector	Clay loam	batch	4	2850 (910 – 6100)	Shehtman et al., 1996

368

369

370 **7. Discussion**

371 Typical ranges of values of Kd values of Sr, Cs and Pu radionuclides for geological deposits from the CEZ
372 are summarized in Table 5. The same table also lists the mean Kd values based on a recent
373 comprehensive statistics of sorption parameters of radionuclides summarized for radioecological
374 modeling purposes in the IAEA TRS-472 report (IAEA, 2010).

375 The following general observations can be made. In general, the site-specific radionuclide Kd values for
376 Quaternary sandy geological deposits from the CEZ (listed in Table 5) fall within the range of values for
377 the respective types of deposits reported in the literature (Sheppard and Thibault, 1990; Serne, 2007;
378 IAEA, 2010). In agreement with general knowledge on radionuclide geochemical behavior and affinity to
379 soils, the radionuclide sorption distribution coefficients increase in the sequence: Sr < Cs < Pu.

380 It can be noted also, that typical radionuclide Kd values for sandy deposits in the CEZ are on the whole
381 lower than mean values for same type of soils from the IAEA TRS-472 report. This can be explained by
382 the specific origin and respective mineralogical composition of sandy deposits in CEZ, where a large
383 proportion of the Kd determinations included in Tables 2 - 4 were carried out on eolian sand deposits
384 from upper soil layers, where fine fractions are represented by dispersed quartz (rather than clay
385 minerals) and are characterized by relatively low CEC values and sorption capacity (see Section 2).

386 Radionuclide Kd data for different deposit lithologies in Chernobyl Exclusion zone (based on combined
387 Kd data set analyzed in this article; see the electronic supplement) are shown in graphical format in Fig.
388 3. An interesting observation is significantly higher variability of Kd values for deposit containing larger
389 percentage of loam and clay fractions compared to deposits with sandy material. The possible
390 explanation of higher variability of Kd values for clay loam and sandy loam deposits is variability of
391 content in fine fractions of these deposits of clay minerals controlling radionuclide sorption process
392 (e.g., hydromica as opposed to fine dispersed calcite or quartz).

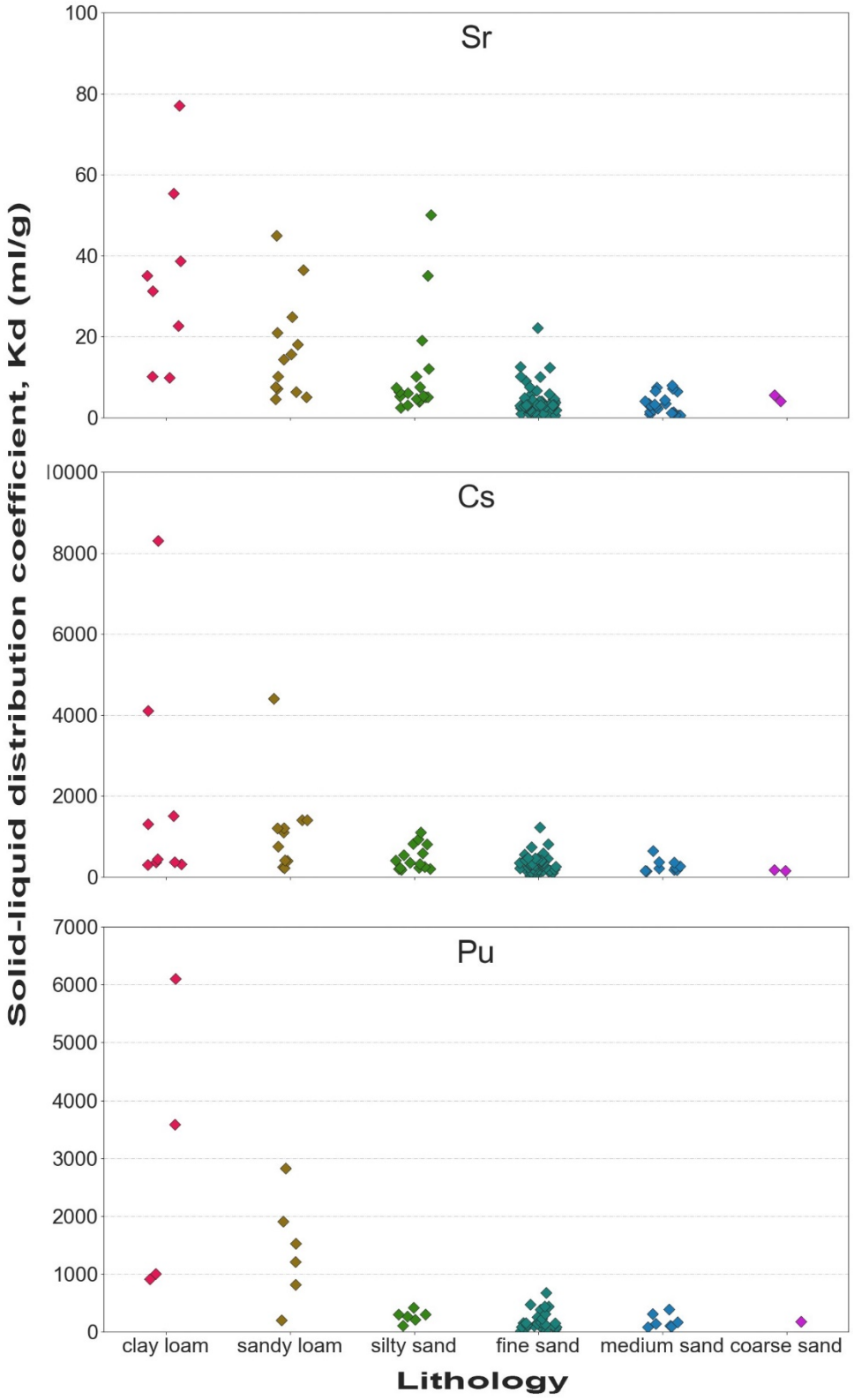
393

394 Table 5. Summary table of typical Kd values of Sr, Cs and Pu isotopes based on studies conducted on
395 geological deposits from Chernobyl Exclusion Zone.

Radionuclide	**Sediment type**	**Method**	**Typical Kd values *, ml/g**	**Min Kd, ml/g**	**Max Kd, ml/g**	**TRS-472 Kd (mean), ml/g**
Sr						
	Sand	Batch	2 - 12,5	1.0	12	22
	Sand	In-situ	2 - 17	0.2	22.1	
	Sand	Model calibr.	0.5 - 1	0.5	1.3	
	Loam / Clay	Batch	7 - 35	4.5	77	69
Cs						
	Sand	Batch	32.5 - 480	27	1080	530
	Loam/Clay	Batch	311 - 2070	210	8300	370
Pu						
	Sand	Batch	180 - 360	50	670	400
	Sand	In-situ	25	7	47	
	Loam/Clay	Batch	1415 - 2850	200	6100	1100

396 Note: * - Range of reported 'mean' Kd values from different studies

397



398

399 Fig.3. Radionuclide Kd data for different lithologies of geological deposit in the Chernobyl Exclusion

400 zone.

401 The Chernobyl case study allows us to carry out inter-comparison of results of Kd determination using
402 different methods. For Sr, Kd determinations using the batch methods and in situ partition tests have
403 produced Kd values that are in a reasonable agreement (see Table 2 and Table 5).

404 At the same time, ^{90}Sr Kd values obtained from groundwater transport model calibrations for trench
405 no.22 at "Red Forest" and for the Chernobyl cooling pond were noticeably lower than experimentally
406 determined sorption parameters (see Table 2 and Table 5).

407 In the case of trench 22, low 'effective' Kd value obtained from groundwater transport model calibration
408 (that described ^{90}Sr transport from the trench to the eolian sand aquifer on a time scale of ~20 y) was
409 explained by the evolution of geochemical conditions in groundwater during the simulated period (Bugai
410 et al., 2012): elevated concentrations of competing cations (Ca, Mg, stable Sr) in the leachate plume
411 emerging from the trench in the early aftermath of the Chernobyl accident (caused by biogeochemical
412 degradation of organic matter inside the burial) presumably has lowered sorption of ^{90}Sr on aquifer
413 deposits, compared to the time of experimental determinations of Kd in the in-situ and batch tests .

414 The ^{90}Sr Kd values of an order of ~ 1 ml/g obtained from calibration of the groundwater transport model
415 describing radionuclide transport from the contaminated cooling pond to the Pripyat River in the alluvial
416 sand aquifer (Bugai et al., 1997, 2005) are noticeably lower than Kd values of ~ 2 -7 mL/g determined
417 from in situ partition tests using cores from the contaminated aquifer near the cooling pond (Gudzenko
418 et al., 1994) (see Table 1). This was attributed to partly non-exchangeable (or kinetically-limited)
419 sorption - desorption reactions of ^{90}Sr on aquifer sediments: some part of the ^{90}Sr adsorbed from an
420 initially high-concentration solution emerging from the pond is retained on sediments resulting in a
421 higher apparent in situ Kd (Bugai et al., 1997). Similar ^{90}Sr non-equilibrium sorption process affecting
422 groundwater transport was reported by Lyon and Patterson (1984), who studied ^{90}Sr transport in
423 groundwater in sand aquifer at Chalk River Nuclear Laboratories site in Canada.

424 The above examples illustrate some potential pitfalls that can be encountered when applying
425 experimentally determined Kd values in groundwater transport models.

426 It is also worth noting relatively low plutonium Kd values obtained in in-situ partition tests compared to
427 values reported from batch tests (see Tables 4, 5). Low Pu Kd values from in-situ tests for sandy deposits
428 from "Red Forest" site likely reflect the influence on sorption of Pu association with colloids and low-
429 molecular organic compounds, which were present in groundwater due to degradation of organic debris
430 inside the trench no.22 (Levchuk et al., 2009).

431 ***Cross-checking Kd-s with groundwater monitoring data in CEZ***

432 Monitoring data on comparative mobility of radionuclides in groundwater in the 10-km zone of the
433 ChNPP reported by State Special Enterprise "Ecocenter", which is responsible for environmental
434 radiological monitoring in CEZ and is carrying groundwater observations at about 140 monitoring wells
435 (Table 6), are broadly consistent with the radionuclide Kd values listed in Table 5. The highest
436 concentrations in groundwater (based on published observation data for 2012-2014) were observed for
437 ^{90}Sr (which have the lowest Kd values), while orders of magnitude lower groundwater concentrations
438 are observed for ^{137}Cs and Pu isotopes. More details on groundwater contamination in Chernobyl zone
439 during the post-accident period can be found in (Bugai et al., 1996, 1997, 2012; Dzhepo and Skalskyy,
440 2002; Kireev et al., 2013).

441 Table 6. Radionuclide concentrations in groundwater in the 10-km zone of ChNPP in 2012 – 2014
442 (measured in monitoring wells of State Special Enterprise "Ecocenter").

Radionuclide	**Max concentrations, Bq/L**	**Typical concentrations, Bq/L**	**Reference**
Sr-90	60 - 170	0.5 - 5	Kireev et al., 2013
Cs-137	0.1 – 0.5	0.01 – 0.05	Kireev et al., 2013
Pu-239+240	0.001 – 0.004	0.00005 – 0.0005	S.Obrizan, Ecocenter, pers.comm

443

444 Though Cs and Pu activity concentrations in groundwater are generally low, these radionuclides are
445 detected in groundwater at sites in CEZ with a relatively deep groundwater table (e.g., several meters) in
446 concentrations (see Table 6) that are still much higher than simple retardation calculations using Kd

447 model would suggest. In some instances, “artificial” contamination of deep aquifer strata during well
448 construction, or facilitated transport from contaminated soil surface along an imperfect contact in the
449 borehole between the well column and surrounding geological deposits cannot be excluded. However,
450 similar data were reported for the experimental site near the trench no.22 in Chernobyl “Red Forest”
451 where special precautions were taken to remove contaminated topsoil layer prior to well installation,
452 and to isolate monitoring well screens using bentonite seals (Bugai et al., 2012; Levchuk et al., 2009), as
453 well as for one-point-in-time groundwater sampling results at radioactive waste dump sites in Chernobyl
454 zone using direct push-drill techniques, where collected groundwater samples suffered minimal
455 disturbance (Antropov et al., 2001, Appendix H). Therefore, the discussed above occurrence of small
456 amounts of Cs and Pu radionuclides in groundwater in our opinion can be most likely explained by
457 facilitated transport of small amounts of these radionuclides from the contaminated (by radioactive
458 fallout) soil surface in the form of non-retarded colloids or complexes, as observed at some other
459 radioactively contaminated sites worldwide (Kersting, 2012; Santschi et al., 2017).

460 It should be noted that a relatively high mobility of ^{90}Sr in groundwater compared to ^{137}Cs and Pu
461 radionuclides has been observed at many other sites worldwide including, for example, Chalk River
462 Nuclear Laboratories in Canada (Jackson and Inch, 1980; Melnyk et al., 1984), Hanford site in the USA
463 (Gilmore et al., 2007; Hartman et al., 2007) and Sellafield in the UK (Sellafield Ltd., 2017).

464 **8. Conclusions**

465 Studies of radionuclide sorption on geological deposits that were carried during last three decades in
466 Chernobyl Exclusion Zone provide various experimental evidence (from laboratory, and from the field)
467 on Sr, Cs, and Pu radionuclide interactions with geological media; these are briefly summarized in this
468 article.

469 The presented compilation of Kd values can be used to complement existing Kd databases, and it can be
470 used as a source of sorption parameters for other sites with analogous geology and hydrogeology
471 worldwide. It can be also used for performance assessment of existing ‘legacy’ radioactive waste burial
472 sites and planned radioactive waste disposal facilities at Vector Complex in Chernobyl zone.

The Chernobyl experience also can be useful to provide a better understanding of potential limitations and pitfalls of the Kd based modeling approach (as discussed in this article). The CEZ as an 'open sky laboratory' offers opportunities and can provide valuable monitoring and experimental data on in-situ interactions of radionuclides with geological materials in challenging scientific areas (e.g., facilitated transport of radionuclides in association with organic ligands and colloids, radionuclide fixation and 'ageing' process by deposits, non-equilibrium adsorption-desorption process, etc.). Unfortunately, until now sorption studies in CEZ have been rather narrowly focused and limited in scope. It is our hope that the scientific community will benefit in future from research opportunities offered by Chernobyl Zone.

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