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Modelling Environmental Impacts of Cesium-137 under a Hypothetical Release of Radioactive Waste

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Abstract

Waste tanks at the nuclear facility located at Sellafield, UK, represent a nuclear source which could release radionuclides to the atmosphere. A model chain which combines atmospheric transport, deposition as well as riverine transport to sea has been developed to predict the riverine activity of ^{137}Cs. The source term was estimated to be 9×10^4 TBq of ^{137}Cs, or 3 % of the assumed total ^{137}Cs inventory of the HAL (Highly Active Liquid) storage tanks. Air dispersion modelling predicted ^{137}Cs deposition reaching 127 kBq m^{-2} at the Vikedal catchment in Western Norway. Thus, the riverine transport model predicted that the activity concentration of ^{137}Cs in at the river outlet could reach 9,000 Bq m^{-3} in the aqueous phase and 1,000 Bq kg^{-1} in solid phase at peak level. The lake and river reaches showed different transport patterns due to the buffering effects caused by dilution and slowing down of water velocity.

Keywords

Risk assessment, catchment modelling, atmospheric deposition, radionuclide transport, SNAP, INCA

1. Introduction

Nuclear accidents like the Chernobyl accident in 1986 and the Fukushima accident in 2011 cause acute release of hazardous radionuclides, such as ^{131}I, ^{134}Cs, ^{137}Cs, ^{239,240}Pu, ^{238}Pu and ^{241}Am into terrestrial, aquatic and marine environments (Reponen et al. 1993; Shozugawa et al. 2012; Zheng et al. 2012). ^{137}Cs, a radioactive isotope of cesium mostly formed as the fission products of ^{235}U in nuclear reactors and weapons, is of particular environmental concern (Miro et al. 2012). Cs is relatively soluble compared to other radionuclides (Ciffroy et al. 2009) and its 30 year decay half-life causes Cs to persist in the aquatic environment. The Chernobyl accident prompted a large effort to quantify the radionuclides transport, including ^{137}Cs, from catchment to freshwater (Monte 1997; Monte 1998; Zheleznyak et al. 1997; Zheleznyak et al. 1992).

The investigation of the impacts to the environment under hypothetical accident is a common practice for the Norwegian Environmental Agency and Norwegian Radiation Protection Authority (NRPA) in order to develop accident preparedness plan (Thørring et al. 2010). Those agencies identified that the Sellafield nuclear waste storage site posed a potential risk to terrestrial environment in Norway, an accident might occur at the facility. Given the mobility and environmental relevance of ^{137}Cs, we focus our risk assessment exercise on this element, and assess its residence-time and distribution in a river basin after fallout of ^{137}Cs under such a scenario. In order to evaluate of the impacts following accidental contamination in a timely manner, models for predicting the behavior of radionuclide transport in rivers basins arose as essential tools for prioritizing and implementing effective risk management strategies, many progresses have been made to develop tools for understanding and predicting the behavior of radionuclides in environment (Monte et al. 2004). There are usually two types of modelling approaches, namely ‘holistic’ and ‘reductionistic’. The holistic approaches tend to describe the processes based on empirical equations by first order compartment systems, for instance, the MARTE model (Monte 2001) and the ECOPRAQ model (Hakanson et al. 2002). While the reductionistic approaches are instead aimed at describing the processes in great details according to primary laws from fundamental disciplines such as physics and chemistry, for instance BIOMOVS II (Konoplev et al. 1996). Although the reductionistic models are good tools for the understanding of the overall migration process but it can be difficult for practical purposes due its high requirements for site specific data. The combination of both approaches usually stands as a good compromise between fast computation time and good process-oriented understanding. We thus introduce the semi-distributed processes-oriented model based on the already established catchment model platform – INtegrated CAtchment model (INCA) (Whitehead et al. 1998a; Whitehead et al. 1998b), the purpose of the study is to provide a tool for fast evaluation of the potential risk for a freshwater ecosystem under a hypothetical nuclear accident.

2. Materials and methods

2.1. Study site receiving ^{137}Cs fallout after an accident at Sellafield Nuclear Facility

It is not straightforward to formulate a set of objective criteria for the definition of hypothetical accidents. Considering the problem from the environmental perspective, maximum deposition over Norwegian territory has been used as the main criterion, to represent a worst case scenario. Western Norway is identified as the most seriously affected region under the hypothetical accident (Thørring et al. 2010; Ytre-Eide et al. 2009). The Vikedal River Catchment in Western Norway discharging into the North Sea (Fig. S2) was chosen for calibrating and testing the model. Vikedal is an important salmon river in Norway (Hesthagen et al. 1999), the coastal area which receives runoff from Vikedal River is also an important aquaculture area. This site is relevant because a potential nuclear accident and subsequent fallout could seriously damage the local environment and harm the local economy. The Vikedal Catchment has a total surface area of 118 km^2. 30% is covered by forests, 7% is cropland and 63 % is mountain shrub. The model describes a simplified river network structure which is then divided into 5 reaches (Fig. S2). The five reaches represent the five divisions of the river which drains the five corresponding sub-catchments (VK1, VK2, VK3, VKT and VKM).

2.2. Modelling methods

We built the INCA-RAD model based on previously developed INCA platform (Supplementary Material). Briefly, after its deposition on the land, it will be distributed among soil water and soil particles. The surface and subsurface flow will carry the dissolved partitions of ^{137}Cs in the stream and ^{137}Cs bonded to soil particles will be transported by flow following erosion. ^{137}Cs in-stream processes contain mainly sorption and desorption between suspended particulate matters (SPM) and aqueous phase, SPM sedimentation and resuspension (Fig. S1), more detailed description of the model is provided in Supplementary Material. To consider ^{137}Cs fate and transport, the mineral fraction of the soil is considered relevant for ^{137}Cs sorption, where the partitioning between water and particles is given by the partition coefficient K_D (Eq. 1):

$$K_D = \frac{A_i}{C_i} \text{ m}^3 \text{ kg}^{-1} \quad (1)$$

where K_D = partition coefficient, m^3 kg^{-1}

C_i = total dissolved adsorbate remaining in water at equilibrium, Bq m^{-3}

A_i = amount of adsorbate on the solid at equilibrium, Bq kg^{-1}

A decay constant is obtained by using the half-life of ^{137}Cs of 30.17 years (Eq. 2):

$$k_{decay} = \frac{Ln(2)}{\tau} \text{ d}^{-1} = 6.2 \times 10^{-5} \text{ d}^{-1} \quad (2)$$

2.3. Source term development

Sellafield is a nuclear fuel reprocessing and nuclear decommissioning site located in Northwest UK (Fig. S2). There have been 21 major incidents in the past half a century which resulted in off-site radiological releases with a rating on the International Nuclear Event Scale (INES) above level 3 (Webb et al. 2006). Total activity of ^{137}Cs is assumed to be between $1.9*10^6 – 3.0*10^6$ TBq (Supplementary Material).

3. Results and discussion

3.1. Atmospheric deposition of ^{137}Cs

^{137}Cs is assumed to be in aqueous state during the transport as the source tank contains liquid wastes. Fig. 1 shows the total predicted deposition during transport of ^{137}Cs from Sellafield to Norway. It is shown that the highest deposition (>500 kBq m^{-2}) happens within UK. The Norwegian west coast receives deposition between 50 and 500 kBq m^{-2} (yellow areas in Fig. 1). Specifically, the study sites Vikedal catchment received a total of 127 kBq m^{-2} during the 48-hour period, which is of similar magnitude to what was received in Norway after the Chernobyl accident (50-200 kBq m^{-2} deposition over Norway). After the Chernobyl accident, central Norway and especially mountainous regions were affected by relatively high levels of ^{137}Cs deposition. The maximum, above 50 kBq m^{-2} ^{137}Cs, was observed in the Valdres and Jotunheimen areas (Baranwal et al. 2011). The helicopter measurements made in 2011 over Jotunheimen have revealed that the deposition in 1986 was above 200 kBq m^{-2} in the most contaminated areas (Skuterud et al. 2014).

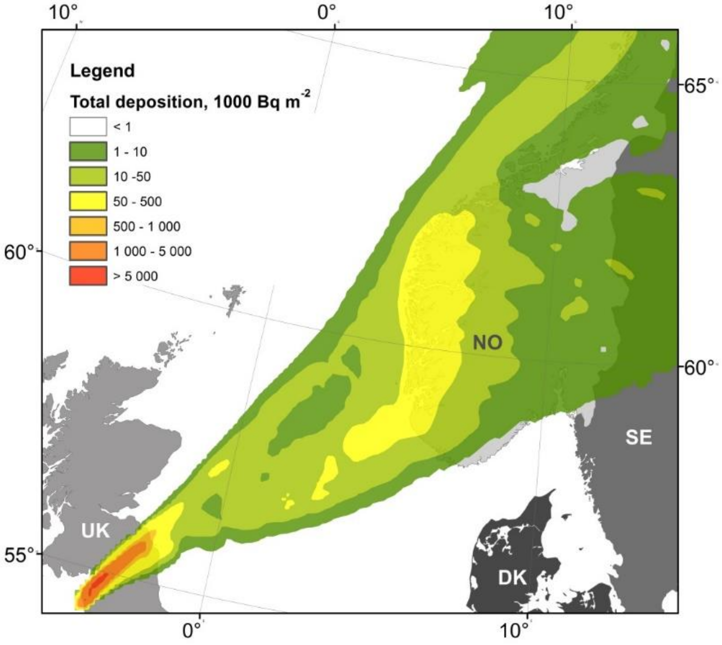

Fig. 1. Total deposition of ^{137}Cs under worst case scenario in Bq m^{-2} simulated by the SNAP model

3.2. ^{137}Cs transport in the aquatic compartments

The activity of ^{137}Cs is simulated for both river water column and river bed sediment for all the five reaches (Fig. S2). The results show distinct differences of the transport patterns between the river reaches (VK1, VK2, VKT and VKM) and the lake reach (VK2) (Fig. 2). Fig. 2a shows the

^{137}Cs activity of both water and sediment of VK2. The specific activity in water of VK2 reached ~ 3800 Bq m^{-3} three months after the accident, and then decreased gradually to < 500 Bq m^{-3} after five years. Meanwhile, the specific activity in the sediment of VK2 reached 1100 Bq kg^{-1} after four years then progressively decreased. Fig. 2b shows the ^{137}Cs activity in both water and sediment of VKM. Compared with the lake reach, activity in both water and sediment of the river reach shows a fast response to ^{137}Cs deposition. The specific activity in water of VKM increased to its top about 9000 Bq m^{-3} only 10 days after the accident, and the activity decreased by half only one year after the accident. The specific activity in sediment of VKM also shows a similar pattern as that in river water of VKM. In general, the activity in river reaches shows more dynamic fluctuations than that of the lake reach, consistent with the lake residence (~ 200 days) time buffering inflowing ^{137}Cs. At the same time, activity in river reaches is heavily affected by the precipitation, as heavy rainfall generates high surface flow which carries much of particle-bound ^{137}Cs into the river within a few hours.

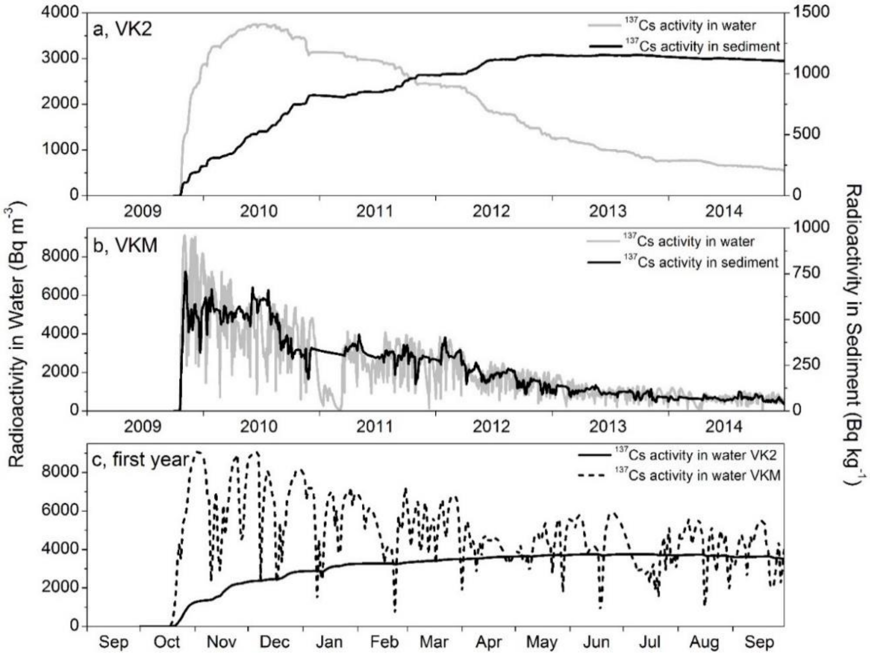

Fig. 2. Simulated specific activity of ^{137}Cs in sediment (black lines) and water (gray lines) in Reach VK2 (panel a) and VKM (panel b), as well as comparison between ^{137}Cs in water in reach VK2 (solid line) and VKM (dashed line) during the first year after the accident (panel c)

3.3. Sensitivity analysis

From the modelling results, the main key processes controlling the ^{137}Cs transport in a watershed includes the partitioning of ^{137}Cs between aqueous and solid phases, soil particles erosion, sediment transport in river and hydrologic processes. A sensitivity analysis exercise was therefore carried out to quantify the relative importance of the above-mentioned processes which translate into nine parameters (Table S1) in INCA model. A range of upper and lower boundaries of the parameters were given based on literature or previous INCA model experiences. For example, the most sensitive parameter is the partition coefficient between water and suspended particles (K_D). Cs forms few stable complexes and is likely to exist in water as the free Cs^{+} ion, which adsorbs rather strongly to most minerals, especially clays (USEPA 1999). In the simulation, the K_D value is determined based on geometric mean of 219 field experiments (IAEA 2010), the maximum and minimum of the reported values were used to define the upper and lower boundary for the K_D in sensitivity analysis to examine the variance of the results given the range of a selected parameter.. The model results are also very sensitive to easily accessible fraction. This parameter describes how much percentage of particle complexation sites is easily accessible which is usually on the surface of particles, and the rest of sites are in the inner part of the particles which are less accessible. Bigger easily accessible fraction could result in the fast equilibrium of partitioning. Hydrologic residence time, which determines how fast the rainfall becomes surface and sub-surface flow, also influence model results. The current model is a generic tool to quickly evaluate the potential risks related to a hypothetical accident, it has limited information on site specific data, however better knowledge of mineralogy of the sediment grains, the organic matter content, Fe content in sediment and solute composition in water at the study site could further improve modelling results to be more accurate.

3.4. Comparison with past accidents

Table 1 summarizes some of the reported ^{137}Cs activity in lakes that were affected by the Chernobyl or Fukushima accident. The simulated results of this study show similar scale of water ^{137}Cs activity to that of the Finnish lakes, at a comparable spatial scale. Four months after the Chernobyl accident, ^{137}Cs in Lake Päijänne water reached 1650 Bq m^{-3} (Vetikko and Saxen 2010). Here we predict 3800 Bq m^{-3} in Lake Vikedal, which is of comparable magnitude. However, ^{137}Cs activity in the sediments is predicted to be one order of magnitude lower than that observed in the Finnish lakes (Table 1). Our results are close to that from Japanese studies (Table 1), specifically those at Lake Akimoto (Matsuda et al. 2015), Lake Hibara, Lake Agari-Onuma, Lake Teganuma and Lake Inbanuma (Fukushima and Arai 2014). The reason for the different activity levels of ^{137}Cs between our study and those on Finish lakes is likely caused by two main reasons. First of all, the Vikedal river is a very clear river where the SPM concentrations at most of the time are around 1 mg/L (https://vannmiljo.miljodirektoratet.no), the lack of SPM greatly limits the transport of ^{137}Cs into sediment. Secondly, Lake Vikedal is located at relatively upper reach of the Vikedal River, which means that the lake doesn't have a big catchment area. Therefore, the particle output from the catchment to Lake Vikedal is also relatively small compared with Finish Lake Päijänne.

Environmental media concentration limits (EMCLs) represent, for a selected media (water or sediment) the activity that would result in a dose-rate to the most exposed organism equal to that of the selected screening dose-rate (10 µGy h^{-1} for ERICA). Recently, such values of EMCLs were updated for ^{137}Cs, using the ERICA Integrated Approach (Andersson et al. 2009), to 51 Bq m^{-3} for water, and 1.75×10^4 Bq kg^{-1} for sediment (Brown et al. 2016). Under the hypothetical accident considered here, water ^{137}Cs activities are in general over the EMCL, however sediment ^{137}Cs activities are below the EMCL. This implies that the aquatic organisms such as insect larvae, which is the reference organism in ERICA for freshwater EMCLs, may be at eco-toxicological risk after exposure to aqueous ^{137}Cs, while benthic organism may not.

Table 1 Comparison of the activity predicted by INCA-RAD ad Vikedal with that observed following in Finland, Ukraine and Japan following actual accidents.

Lake	Time elapsed (yr)	Water (Bq m^{-3})	Sediment (Bq kg^{-1} d.w.)	References
Simulated activity in Norway following hypothetical accident in Sellafield				
Vikedal	0.25	3800	250	This study
Vikedal	4	500	1100	This study
Measured activity in Finland following the Chernobyl accident				
Päijänne	0.33	1650	-	(Vetikko and Saxen 2010)

Päijänne	10	-	18530	(Vetikko and Saxen 2010)
Vehkajärvi	16	260 – 290	17000 – 20000	(Saxen 2007)
Siikajärvi	16	290 – 320	13000 – 18000	(Saxen 2007)
Measured activity in Ukraine following the Chernobyl accident				
Glyboke	30	4	126000	(Ganzha et al. 2014)
Measured activity in Japan following the Fukishima accident				
Hayama	1 – 2	66.2	17340	(Matsuda et al. 2015)
Akimoto	1 – 2	24.5	2357	(Matsuda et al. 2015)
Tagokura	1 – 2	1.6	301	(Matsuda et al. 2015)
15 lakes	0 – 2	-	23 – 26000	(Fukushima and Arai 2014)

4. Conclusions

Environmental modelling is a powerful tool for authorities concerned with the environmental consequences of a low-frequency, high risk nuclear accident. Here, the hypothetical accident at Sellafield has shown to lead to elevated activity of ^{137}Cs in both water and sediment in Western Norway. The levels of ^{137}Cs specific activity are comparable to those measured in Norway and Finland after past accidents, and may pose a risk to aquatic organisms. ^{137}Cs in sediment decreases more slowly than that in water phase due to strong adsorption of Cs 137 on particulate matters in the sediment.

The combination of atmospheric dispersion modelling using SNAP and of catchment hydrochemical modelling using the augmented INCA model INCA-RAD proves a useful tool for supporting scientific research and management decision making on the interactions between climate events, land use, biogeochemistry and radionuclide deposition. These results further highlight the usefulness of parsimonious hydrochemical modeling to assess the risk posed by deposition of ^{137}Cs.

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