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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 ¹³⁷Cs. The source term was estimated to be 9×10^4 TBq of ¹³⁷Cs, or 3 % of the assumed total ¹³⁷Cs inventory of the HAL (Highly Active Liquid) storage tanks. Air dispersion modelling predicted ¹³⁷Cs deposition reaching 127 kBq m⁻² at the Vikedal catchment in Western Norway. Thus, the riverine transport model predicted that the activity concentration of ¹³⁷Cs 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 ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ^{239,240}Pu, ²³⁸Pu and ²⁴¹Am into terrestrial, aquatic and marine environments (Reponen et al. 1993; Shozugawa et al. 2012; Zheng et al. 2012). ¹³⁷Cs, a radioactive isotope of cesium mostly formed as the fission products of ²³⁵U 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 ¹³⁷Cs, 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 ¹³⁷Cs, we focus our risk assessment exercise on this element, and assess its residence-time and distribution in a river basin after fallout of ¹³⁷Cs 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

47 made to develop tools for understanding and predicting the behavior of radionuclides in
48 environment (Monte et al. 2004). There are usually two types of modelling approaches, namely
49 ‘holistic’ and ‘reductionistic’. The holistic approaches tend to describe the processes based on
50 empirical equations by first order compartment systems, for instance, the MARTE model (Monte
51 2001) and the ECOPRAQ model (Hakanson et al. 2002). While the reductionistic approaches are
52 instead aimed at describing the processes in great details according to primary laws from
53 fundamental disciplines such as physics and chemistry, for instance BIOMOVs II (Konoplev et al.
54 1996). Although the reductionistic models are good tools for the understanding of the overall
55 migration process but it can be difficult for practical purposes due its high requirements for site
56 specific data. The combination of both approaches usually stands as a good compromise between
57 fast computation time and good process-oriented understanding. We thus introduce the semi-
58 distributed processes-oriented model based on the already established catchment model platform –
59 INtegrated CATCHment model (INCA) (Whitehead et al. 1998a; Whitehead et al. 1998b), the
60 purpose of the study is to provide a tool for fast evaluation of the potential risk for a freshwater
61 ecosystem under a hypothetical nuclear accident.

62 **2. Materials and methods**

63 **2.1. Study site receiving ^{137}Cs fallout after an accident at Sellafield Nuclear Facility**

64 It is not straightforward to formulate a set of objective criteria for the definition of hypothetical
65 accidents. Considering the problem from the environmental perspective, maximum deposition over
66 Norwegian territory has been used as the main criterion, to represent a worst case scenario. Western
67 Norway is identified as the most seriously affected region under the hypothetical accident
68 (Thørring et al. 2010; Ytre-Eide et al. 2009). The Vikedal River Catchment in Western Norway
69 discharging into the North Sea (Fig. S2) was chosen for calibrating and testing the model. Vikedal
70 is an important salmon river in Norway (Hesthagen et al. 1999), the coastal area which receives
71 runoff from Vikedal River is also an important aquaculture area. This site is relevant because a
72 potential nuclear accident and subsequent fallout could seriously damage the local environment
73 and harm the local economy. The Vikedal Catchment has a total surface area of 118 km². 30% is
74 covered by forests, 7% is cropland and 63 % is mountain shrub. The model describes a simplified
75 river network structure which is then divided into 5 reaches (Fig. S2). The five reaches represent
76 the five divisions of the river which drains the five corresponding sub-catchments (VK1, VK2,
77 VK3, VKT and VKM).

78 **2.2. Modelling methods**

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

$$88 \quad K_D = \frac{A_i}{C_i} \text{ m}^3 \text{ kg}^{-1} \quad (1)$$

89 where K_D = partition coefficient, m³ kg⁻¹

90 C_i = total dissolved adsorbate remaining in water at equilibrium, Bq m⁻³

91 A_i = amount of adsorbate on the solid at equilibrium, Bq kg⁻¹

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

93
$$k_{\text{decay}} = \frac{\ln(2)}{\tau} \text{ d}^{-1} = 6.2 \times 10^{-5} \text{ d}^{-1} \text{ (2)}$$

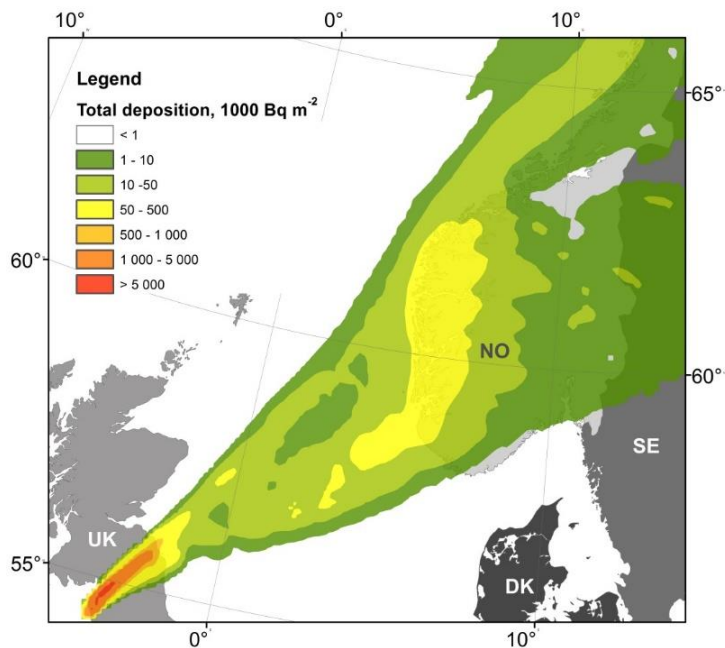
94 **2.3. Source term development**

95 Sellafield is a nuclear fuel reprocessing and nuclear decommissioning site located in Northwest
96 UK (Fig. S2). There have been 21 major incidents in the past half a century which resulted in off-
97 site radiological releases with a rating on the International Nuclear Event Scale (INES) above level
98 3 (Webb et al. 2006). Total activity of ^{137}Cs is assumed to be between $1.9 \times 10^6 - 3.0 \times 10^6$ TBq
99 (Supplementary Material).

100 **3. Results and discussion**

101 **3.1. Atmospheric deposition of ^{137}Cs**

102 ^{137}Cs is assumed to be in aqueous state during the transport as the source tank contains liquid
103 wastes. Fig. 1 shows the total predicted deposition during transport of ^{137}Cs from Sellafield to
104 Norway. It is shown that the highest deposition ($>500 \text{ kBq m}^{-2}$) happens within UK. The
105 Norwegian west coast receives deposition between 50 and 500 kBq m^{-2} (yellow areas in Fig. 1).
106 Specifically, the study sites Vikedal catchment received a total of 127 kBq m^{-2} during the 48-hour
107 period, which is of similar magnitude to what was received in Norway after the Chernobyl accident
108 ($50\text{-}200 \text{ kBq m}^{-2}$ deposition over Norway). After the Chernobyl accident, central Norway and
109 especially mountainous regions were affected by relatively high levels of ^{137}Cs deposition. The
110 maximum, above 50 kBq m^{-2} ^{137}Cs , was observed in the Valdres and Jotunheimen areas (Baranwal
111 et al. 2011). The helicopter measurements made in 2011 over Jotunheimen have revealed that the
112 deposition in 1986 was above 200 kBq m^{-2} in the most contaminated areas (Skuterud et al. 2014).

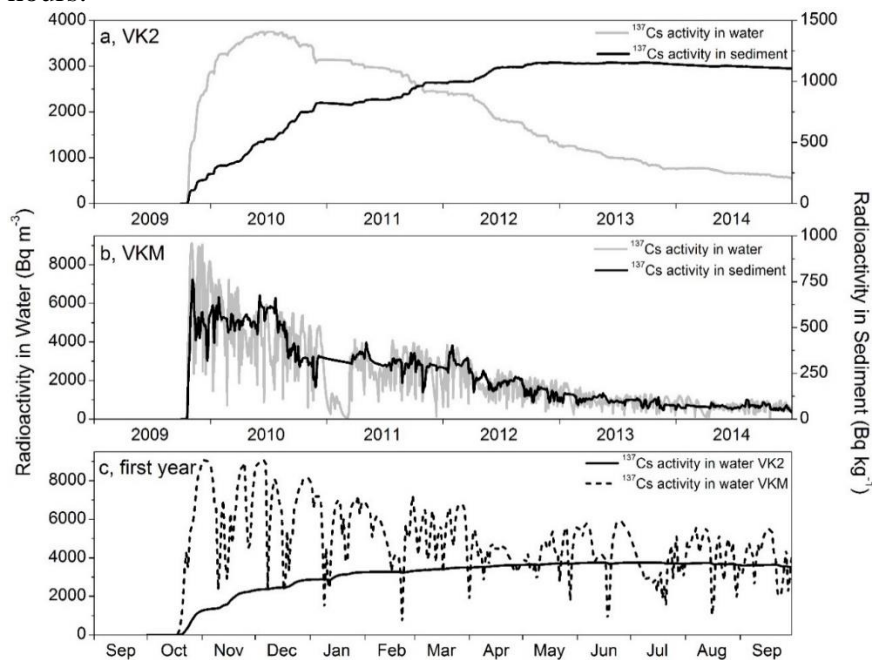


113
114 **Fig. 1.** Total deposition of ^{137}Cs under worst case scenario in Bq m^{-2} simulated by the SNAP
115 model

116 **3.2. ^{137}Cs transport in the aquatic compartments**

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

120 ^{137}Cs activity of both water and sediment of VK2. The specific activity in water of VK2 reached ~
 121 3800 Bq m^{-3} three months after the accident, and then decreased gradually to $< 500 \text{ Bq m}^{-3}$ after
 122 five years. Meanwhile, the specific activity in the sediment of VK2 reached 1100 Bq kg^{-1} after four
 123 years then progressively decreased. Fig. 2b shows the ^{137}Cs activity in both water and sediment of
 124 VKM. Compared with the lake reach, activity in both water and sediment of the river reach shows
 125 a fast response to ^{137}Cs deposition. The specific activity in water of VKM increased to its top about
 126 9000 Bq m^{-3} only 10 days after the accident, and the activity decreased by half only one year after
 127 the accident. The specific activity in sediment of VKM also shows a similar pattern as that in river
 128 water of VKM. In general, the activity in river reaches shows more dynamic fluctuations than that
 129 of the lake reach, consistent with the lake residence (~ 200 days) time buffering inflowing ^{137}Cs .
 130 At the same time, activity in river reaches is heavily affected by the precipitation, as heavy rainfall
 131 generates high surface flow which carries much of particle-bound ^{137}Cs into the river within a few
 132 hours.



133
 134 **Fig. 2.** Simulated specific activity of ^{137}Cs in sediment (black lines) and water (gray lines) in
 135 Reach VK2 (panel a) and VKM (panel b), as well as comparison between ^{137}Cs in water in reach
 136 VK2 (solid line) and VKM (dashed line) during the first year after the accident (panel c)
 137

3.3. Sensitivity analysis

138 From the modelling results, the main key processes controlling the ^{137}Cs transport in a watershed
 139 includes the partitioning of ^{137}Cs between aqueous and solid phases, soil particles erosion, sediment
 140 transport in river and hydrologic processes. A sensitivity analysis exercise was therefore carried
 141 out to quantify the relative importance of the above-mentioned processes which translate into nine
 142 parameters (Table S1) in INCA model. A range of upper and lower boundaries of the parameters
 143 were given based on literature or previous INCA model experiences. For example, the most
 144 sensitive parameter is the partition coefficient between water and suspended particles (K_D). Cs
 145 forms few stable complexes and is likely to exist in water as the free Cs^+ ion, which adsorbs rather
 146 strongly to most minerals, especially clays (USEPA 1999). In the simulation, the K_D value is
 147 determined based on geometric mean of 219 field experiments (IAEA 2010), the maximum and

148 minimum of the reported values were used to define the upper and lower boundary for the K_D in
 149 sensitivity analysis to examine the variance of the results given the range of a selected parameter..
 150 The model results are also very sensitive to easily accessible fraction. This parameter describes
 151 how much percentage of particle complexation sites is easily accessible which is usually on the
 152 surface of particles, and the rest of sites are in the inner part of the particles which are less
 153 accessible. Bigger easily accessible fraction could result in the fast equilibrium of partitioning.
 154 Hydrologic residence time, which determines how fast the rainfall becomes surface and sub-surface
 155 flow, also influence model results. The current model is a generic tool to quickly evaluate the
 156 potential risks related to a hypothetical accident, it has limited information on site specific data,
 157 however better knowledge of mineralogy of the sediment grains, the organic matter content, Fe
 158 content in sediment and solute composition in water at the study site could further improve
 159 modelling results to be more accurate.

160 3.4. Comparison with past accidents

161 Table 1 summarizes some of the reported ^{137}Cs activity in lakes that were affected by the Chernobyl
 162 or Fukushima accident. The simulated results of this study show similar scale of water ^{137}Cs activity
 163 to that of the Finnish lakes, at a comparable spatial scale. Four months after the Chernobyl accident,
 164 ^{137}Cs in Lake Päijänne water reached 1650 Bq m^{-3} (Vetikko and Saxen 2010). Here we predict
 165 3800 Bq m^{-3} in Lake Vikedal, which is of comparable magnitude. However, ^{137}Cs activity in the
 166 sediments is predicted to be one order of magnitude lower than that observed in the Finnish lakes
 167 (Table 1). Our results are close to that from Japanese studies (Table 1), specifically those at Lake
 168 Akimoto (Matsuda et al. 2015), Lake Hibara, Lake Agari-Onuma, Lake Teganuma and Lake
 169 Inbanuma (Fukushima and Arai 2014). The reason for the different activity levels of ^{137}Cs between
 170 our study and those on Finish lakes is likely caused by two main reasons. First of all, the Vikedal
 171 river is a very clear river where the SPM concentrations at most of the time are around 1 mg/L
 172 (<https://vanmiljo.miljodirektoratet.no>), the lack of SPM greatly limits the transport of ^{137}Cs into
 173 sediment. Secondly, Lake Vikedal is located at relatively upper reach of the Vikedal River, which
 174 means that the lake doesn't have a big catchment area. Therefore, the particle output from the
 175 catchment to Lake Vikedal is also relatively small compared with Finish Lake Päijänne.

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

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

| Lake | Time elapsed (yr) | Water (Bq m^{-3}) | Sediment ($\text{Bq kg}^{-1} \text{ d.w.}$) | 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 Fukushima 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) |

187 4. Conclusions

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

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

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 205 building at the river-basin scale”.

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