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Reconstructing the deposition environment and long-term fate of Chernobyl ^{137}Cs at the floodplain scale through mobile gamma spectrometry☆

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ABSTRACT

Cs-137 is considered to be the most significant anthropogenic contributor to human dose and presents a particularly difficult remediation challenge after a dispersal following nuclear incident. The Chernobyl Nuclear Power Plant meltdown in April 1986 represents the largest nuclear accident in history and released over 80 PBq of ^{137}Cs into the environment. As a result, much of the land in close proximity to Chernobyl, which includes the Polessie State Radioecology Reserve in Belarus, remains highly contaminated with ^{137}Cs to such an extent they remain uninhabitable. Whilst there is a broad scale understanding of the depositional patterns within and beyond the exclusion zone, detailed mapping of the distribution is often limited. New developments in mobile gamma spectrometry provide the opportunity to map the fallout of ^{137}Cs and begin to reconstruct the depositional environment and the long-term behaviour of ^{137}Cs in the environment. Here, full gamma spectrum analysis using algorithms based on the peak-valley ratio derived from Monte Carlo simulations are used to estimate the total ^{137}Cs deposition and its depth distribution in the soil. The results revealed a pattern of ^{137}Cs distribution consistent with the deposition occurring at a time of flooding, which is validated by review of satellite imagery acquired at similar times of the year. The results were also consistent with systematic burial of the fallout ^{137}Cs by annual flooding events. These results were validated by sediment cores collected along a transect across the flood plain. The true merit of the approach was confirmed by exposing new insights into the spatial distribution and long term fate of ^{137}Cs across the floodplain. Such systematic patterns of behaviour are likely to be fundamental to the understanding of the radioecological behaviour of ^{137}Cs whilst also providing a tracer for quantifying the ecological controls on sediment movement and deposition at a landscape scale.

1. Introduction

Cs-137 is considered the one of the most radiologically significant anthropogenic radionuclide within the environment (Miller, 2007). The majority of present day global inventory of ^{137}Cs is derived from atmospheric weapons testing (1945–1963). More recently, further more-localised contamination has been introduced into the environment through reactor meltdowns occurring at Chernobyl Nuclear Power Plant (ChNPP) in 1986 and Fukishima Diachii Nuclear Power Plant (FDNPP) in 2011. The widespread nature of ^{137}Cs and the risk it poses to human health are as a result of a combination of physical and chemical properties. Foremost, it is one of the more abundant fission products generated during a nuclear chain reaction and it has a relatively low melting point (670.8 °C) allowing it to readily disperse into the atmosphere after a meltdown or nuclear bomb detonation (Isaksson and Raaf, 2017). Once in the environment, it can exhibit complex biogeochemical behaviour given it is highly soluble, has a tendency to react with environmental media in particular clay minerals and can be readily mobilised in the presence of organic matter. It can also easily enter

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the food chain as it is readily taken up into plant material substituting for naturally occurring potassium (Penrose et al., 2016). In terms of human exposure, the combination of 30.1 year half-life, high energy beta decay and the 662 keV gamma-ray from the short-lived daughter product ^{137m}Ba make ^{137}Cs both an internal and external radiation hazard. As a result, following any nuclear incident one of the fundamental tasks is to establish, using high quality data, the extent and intensity of ^{137}Cs deposition to inform decision making. This need has been exemplified by, for example, the largescale data collection efforts initiated by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) within a month of the FDNPP accident (Povinec et al., 2013) and the multinational efforts over a number of decades to confidently characterise the spatial distribution of fallout across Europe after the ChNPP accident (Izrael et al., 1996).

In the longer term, continued measurement of ^{137}Cs is also important, not only to update population dose assessments (Beresford et al., 2016; Konoplev et al., 2017), but to monitor redistribution of the radiocontaminant within the environment occurring as a function soil movement, weathering and erosion (Rawlins et al., 2011). These are the same process that have enabled soil scientists to regularly utilise ^{137}Cs as a medium-term tracer for soil redistribution since the 1960's (Mabit and Dercon, 2014; Ritchie and McHenry, 1990).

Cs-137 is relatively straightforward to measure through gamma-ray spectrometry of the 662 keV photon released by its daughter product ^{137m}Ba within which it is always in secular equilibrium (Povinec et al., 2003). However, difficulties often arise when trying to account for depth variation of ^{137}Cs contamination given that significant penetration into the soil column can occur over time making precise estimates of the total inventory (Bq m^2) problematic (Tyler, 2004). The oldest and most direct way to measure the depth distribution is by extracting cores from a site and measuring small increments of the core (typically less than 5 cm slices) within the laboratory using low background High Purity Germanium (HPGe) detectors; thus building a profile. Collection, preparation and analysis of environmental samples can nonetheless make this procedure time consuming and expensive often yielding limited spatial representivity, which can become a major issue when significant spatial heterogeneity of contamination is encountered (Golosov et al., 2000). Possibly a more appropriate method that can address spatial resolution issues more effectively is the deployment of field-based mobile gamma-ray spectrometry (Tyler, 2008). Utilising this technique, measurements can be made directly in the field using a variety of gamma-ray detectors mounted to a specific platform such as static surveyor stands (*in situ*) (Gering et al., 1998), vehicles (carborne) (Aage et al., 2006), airborne (helicopters and fixed wing) (Rawlins et al., 2011) and unmanned aerial vehicle (UAVs) (Okuyama et al., 2008). The height of a detector primarily determines the spatial quality of data given that the field of view (FOV) of the detector will be increased with increasing height (Tyler et al., 1996a). Alongside time and cost restraints this factor is crucial in determining the choice of platform to characterise the underlying spatial distribution of ^{137}Cs contamination.

In situ technologies, utilising highly sensitive HPGe detectors, represent the standard for characterisation of gamma-ray emitting radionuclides (ICRU, 1994). Nonetheless, HPGe detectors generally have low efficiency, require heavy cooling systems and are as a result bulky and not suited for mapping large areas in high spatial resolution. Therefore portable systems, known as mobile gamma-ray spectrometry, tend to use more robust sodium iodide (NaI:Tl) detectors with larger detection volumes (and hence efficiency) in order to improve statistical accuracy (Tyler, 2008). Airborne survey systems as an example can carry hundreds of kilos of NaI:Tl providing very high detection efficiencies that are ideal to rapidly characterise ^{137}Cs contamination on a national scale. However, such surveys come at considerable cost and for safety reasons tend to be conducted at altitudes above 100 m resulting in a large FOV, making resolution of localised contamination difficult. Carborne systems on the other hand can provide a higher spatial quality of data for significantly less cost whilst still being able to carry significant detection volumes, such surveys tending to be restricted to roads or open fields. UAVs offer enormous potential in the field of gamma-ray spectrometry, however current affordable technologies cannot provide the payloads to carry detectors with sufficient detection efficiency, for long enough periods, to characterise large contaminated areas in high spatial resolution (Martin et al., 2016).

Lightweight detection systems originally designed for nuclear security missions are now commonly being implemented in the field as backpack or handheld devices (Fig. 1) to characterise ^{137}Cs in relatively heavily contaminated environments (Cresswell et al., 2013; Kock and Samuelsson, 2011; Nilsson et al., 2014; Plamboeck et al., 2006; Sanderson et al., 2013). Typically, a large number of georeferenced spectra are taken during a survey and, conventionally, the count rate in the full energy peak is used to determine the amount of ^{137}Cs contamination in the soil. However, large uncertainties in inventory estimates (Bq m^{-2}) can be encountered when significant spatial heterogeneities in burial depth are encountered. Recently, Varley et al. (2017) demonstrated that through the use of the peak-to-valley ratio (PVR) improved inventory estimates could be made for aged Chernobyl deposits in Belarus. The benefit of utilising the PVR to account for burial depth was first realised by Zombori et al. (1992) and has since been used widely to estimate the burial depth of ^{137}Cs (Feng et al., 2012; Gering et al., 2002, 1998; Kastlander and Bargholtz, 2005; Tyler, 2004). In essence, the PVR is the ratio of the full energy photopeak and the forward scattered photons (Fig. 1). Forward scattered photons are those photons that have lost a small amount of energy on transition out of the soil and are represented by the “valley” region of the gamma spectrum that may be observed between the full energy photopeak at 662 keV and the Compton edge occurring at 480 keV. The greater the depth within the soil at which the photon originates, the more probable it is that the photon will undergo forward scattering and generate a count within the valley region of the spectrum. This leads to an increase of the valley “height” and a concomitant decrease in the PVR.

Following the calibration and validation work of assessing the PVR method for mobile handheld gamma spectrometry (Varley et al., 2017) it was apparent that one calibration site exhibited a systematic change in the deposited ^{137}Cs activity and depth distribution towards a tributary of the Pripyat River. The primary aim was to follow up on this observation and to complete a high resolution (<1 m) mobile gamma spectrometry survey across the flood plain to characterise and explain the systematic distribution in a landscape that had been highly impacted by the 1986 Chernobyl accident and that had hitherto been considered to be highly heterogenous with regards to the ^{137}Cs distribution.

Large areas of the watershed of the Pripyat River, the main watercourse of the impacted region and including feeder streams, lakes, marshes and drainages, are within the 30-km exclusion zone that was ultimately imposed around the ChNPP. Since the accident, high concentrations of radionuclides have been observed within stagnant and slow moving bodies of water associated with the Pripyat River. Such radionuclides accumulate primarily in relation to sediments although accumulations of contaminants can also be observed within the water itself and associated biota (Gudkov et al., 2010). Floodplains have been shown to represent relatively complex post-depositional environments (Golosov et al., 2013; Iwasaki et al., 2015) due to the influence of flood events on a number of processes that govern the environmental behaviour of ^{137}Cs and the