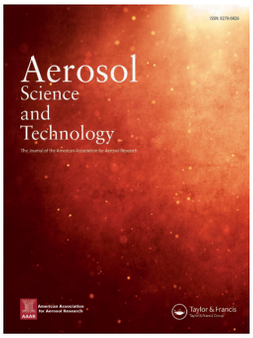

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Potential aeolian transfer of radioactive dusts from contaminated saltmarshes to coastal residential areas

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Abstract

Dust samples from a residential area next to the Dee estuary saltmarsh, north-west England, UK, were collected at two heights and analyzed to investigate the potential for transporting radioactive dusts inland from the saltmarsh via an aeolian pathway. ^{137}Cs activities were found in the range from 20 ± 4 to 794 ± 113 Bq kg^{-1}, with mean of 193 Bq kg^{-1} and from 15 ± 4 to 252 ± 8 Bq kg^{-1}, with mean of 109 Bq kg^{-1} in dusts at 2 m and 0.3 m heights, respectively. The total annual effective doses were found as 1-41 µSv y^{-1} and 2–95 µSv y^{-1} for minimum and maximum occupancy, respectively. The maximum value of 41 µSv y^{-1} for the minimum outdoor occupancy falls below the world average value of 70 µSv y^{-1} (UNSCEAR, 2000) but the maximum value of 95 µSv y^{-1} for the maximum occupancy is 36% higher than the world average value and is non-negligible. The collected dust samples were about 75% organic and 25% mineral particles. During sampling period at 2 m height, mean and maximum wind speeds were found 5-8 m s^{-1}, and 16-22 m s^{-1}, respectively. Detailed examination of radionuclide, grain size and wind data during dust collection period reveal that the estimated threshold wind speeds (5-23 m s^{-1}) over the saltmarsh are sufficient to entrain radioactive dust particles (63-4 µm diameter) to be transported and deposited in inland, and hence poses potential health risks to coastal populations.

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1. Introduction

Humans can be exposed to anthropogenic radionuclides discharged to the sea from nuclear installations through various routes (McKay and Pattenden, 1990). Previous studies have shown that offshore Irish Sea sediment containing radionuclides discharged historically from the Sellafield nuclear waste reprocessing plant in Cumbria, NW England, represents a potentially significant source of radionuclides to the surrounding coastal environment (Aston and Stanners, 1982; Aston et al., 1981). Research has revealed spatial and temporal patterns of radionuclides concentrations in offshore, intertidal and saltmarsh sediments in the Irish Sea, and the onshore movement of these contaminated sediments through tidal currents is the dominant transport mechanism of Sellafield-derived radionuclides, leading to high ^{241}Am and ^{137}Cs activities in the coastal region (Bonnett et al., 1988; Charlesworth et al., 2006; Fridlington et al., 1995; Horrill et al., 1992; Hutchinson, 1994; MacKenzie and Scott, 1993; MacKenzie et al., 1998; McDonald et al., 1990; Oldfield et al., 1993).

It has also been discovered that radionuclides have a high affinity for fine saltmarsh sediments and the activity levels of more soluble ^{137}Cs in far distant Dee estuary saltmarshes are quite significant and comparable to the levels of activity in sites much closer to Sellafield (Bonnett et al., 1988; Clifton, 1998; Oldfield et al., 1993; Rahman, 2010). Based on previous findings, it is assumed that the high radionuclide activities stored within these saltmarsh sediment records may pose significant potential for radiation exposures via different exposure pathways to saltmarsh users and people in close proximity to these contaminated lands. This is especially the case for locations along the eastern (English) coast of the Dee estuary where settlements are located downwind of the

predominant winds from the west and northwest (cf. Elliot, 2004), and thus serve as receptors for dusts derived from the coast. Moreover, it is apparent that no detailed investigation has been made to identify the potential health risks from these saltmarsh radionuclides to the general public via direct exposure and inhalation of radioactive saltmarsh generated aeolian dusts.

Humans can be irradiated from saltmarshes contaminated with radionuclides as it is observed that well developed upper saltmarshes are often used for agricultural and recreational purposes, like picnic spots, golf courses, country parks, caravan parks, nature conservation, wildfowling, grazing etc., and these saltmarshes are in close proximity to coastal populations. Hence there is considerable potential for human exposure to these anthropogenic radionuclides in saltmarshes. Due to the long half-lives of ^{137}Cs (30.2 y) and ^{241}Am (432.7 y), exposures from ^{137}Cs and ^{241}Am are considered more important than other Sellafield-derived radionuclides with short half-lives. Humans residing close to radioactive contaminated areas may therefore have been exposed to higher doses of radiation from ^{137}Cs or ^{241}Am, both internally and externally, than the general population. External exposure from saltmarsh radiation to members of the public in close proximity to coastal areas depends on the occupancy time. In 1989, it was reported by MAFF that external gamma dose rates observed in British saltmarshes are mainly due to ^{137}Cs. In contrast, internal exposure continues until the radioactive material is flushed from the body by natural processes, or when it decays. When a person inhales or ingests a radioisotope, it is distributed to different organs and stays there for days, months or years until it decays or is excreted (Maekawa, 2002).

Policy for radioactive materials in England and Wales (the countries concerned in this study) comes under Environmental Permitting (England and Wales) Regulations 2016 (EPR16), although this does not specifically apply to private individuals, nor to naturally-occurring radionuclides in their normal setting (BEIS, 2018). However, legislation provisions are not intended to exclude radionuclides which are present in the environment as a result of permitted or exempted discharges. Further, it explicitly covers naturally occurring radioactive materials (NORM) used in 'NORM industrial activities' which include: a. "the remediation of contamination from any type 1 NORM industrial activity", and b. materials containing artificial radionuclides that exceed threshold values (0.1 Bq g^{-1} in the case of ^{241}Am and 1 Bq g^{-1} in the case of ^{137}Cs). Indeed, "a substance is radioactive material or radioactive waste where, following its disposal, a process occurs which was not foreseen at the time of disposal, and results in a substantial increase in radiation exposure to the public or radioactive contamination in the environment." (BEIS 2018, p.19). As such, this applies to the condition being examined in this study. In such circumstances, radioactive materials are exempt from waste regulation if they do not exceed either threshold maximum quantities or activities concentrations: 10^4 Bq and 10Bq g^{-1}, respectively, for ^{137}Cs, and 10^4 Bq and 1Bq g^{-1} for ^{241}Am. Otherwise, a disposer is required to make a radiological assessment of the 'reasonably foreseeable' pathways for the exposure of the public to radiation and be satisfied that the radiation dose assessment does not exceed 300 µSv y^{-1} to the public. In the case of this study, we examine the aeolian transfer of fine dust as a possible pathway.

Several previous studies established that the Dee estuary saltmarshes have significant amount of ^{241}Am and ^{137}Cs activities in their core records and exhibit a gradual trend of increasing concentrations with decreasing grain size (Clifton, 1998; Clifton et al., 1997; Hutchinson, 1994; Hutchinson and Prandle, 1994; Oldfield et al., 1993; Rahman, 2010). Moreover, a thorough study completed by Rahman [2010] on the Dee estuary saltmarshes revealed significant ^{241}Am and ^{137}Cs activities in their core records, again with a gradual trend of increasing concentrations with decreasing grain size, and with a high affinity between fine grain sizes (silt to clay particles, i.e., < 63 µm) and radionuclide activities. The total radioactivity does not depend solely on particle size distribution but on the availability of radionuclides that become attached to particles (Rahman et al., 2013).

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Consequently, it can be assumed that there is a potential for these radionuclides to be physically remobilised with any resuspended dry sediments as aeolian dust particles (silt to clay particles, i.e., below 63 μm) resulting from wind erosion. In assessing the potential health hazard from the aeolian transport of dusts contaminated by legacy discharges of radionuclides into the coastal environment, this study translates the findings from Rahman [2010] on the relationships between ^{137}Cs and ^{241}Am activity and grain size determined for the Dee estuary saltmarsh sediments, and goes beyond the assessment undertaken by Rahman et al. [2013] on the health hazard posed by the aqueous redistribution of sediment-bound radionuclides due to coastal erosion of saltmarshes. Here, assessment of the aeolian pathway is especially important for pollutants that are known to pose significant health risks for coastal residents who might inhale radionuclides attached to aeolian dusts or may ingest radionuclides in the foodchain (cf. Maekawa, 2002), or may directly be exposed during outdoor activities in their yard.

The present study focuses on the Parkgate saltmarshes in the Dee estuary, NW England (Fig. 1), where the examined areas possess high levels of radioactivity and are in close proximity to residential areas. Hence, there is the potential for transferring radioactive dusts from contaminated saltmarshes to inland residential areas via the aeolian pathway. With this view, aeolian dusts were collected on two Frisbee passive dust collectors filled with round and polished marbles, following the principles described in Sow et al. (2006), to observe seasonal as well as height-related variations in the accumulated dusts. These dusts were analysed for their ^{137}Cs and ^{241}Am activities and their grain size distribution, thus providing a radionuclide flux, a budget expressed as a proportion of the saltmarsh radioactivity reservoir, an analysis of the likely source of the radionuclide pollution, and an assessment of the potential health risk to the population of the adjacent coastal area. Together with seasonal and height parameters, radionuclide data were also examined in relation to meteorological data from the nearest weather station, which is situated in the outer part of the Dee estuary at Hilbre Island, approximately 14 km NW from the study site (location: 53˚22.956′N 3˚13.680′W). The wind data were supplied by the Proudman Oceanographic Laboratory (now the National Oceanography Centre), Liverpool. The particle size analysis (PSA) of dusts was undertaken using an optical microscopic method at × 600 magnification to identify grains of < 10 μm size ranges, which are the dust particles of respirable size - called PM_{10} (HPA, 2010).

2. Research methodology

2.1 Study site description

The Dee is a funnel-shaped macro-tidal, coastal plain type estuary (53°10′ N; 3°10′W) located in the eastern Irish Sea between the Wirral peninsula, England and the Flintshire coastline of North Wales (Huckle et al., 2004; Moore et al., 2009). It is ca. 130 km south of Sellafield (calculated from 1:10 000 Ordnance survey map of UK supplied by EDINA) and is one of the largest estuaries in NW England, extending from the estuary mouth far up to the city of Chester (Fig. 1). Most of the estuary is occupied by intertidal mudflat and saltmarsh habitats, resulting from progressive silting of the estuary since the 11th century. Natural processes of silt deposition have been accelerated by human interventions to reclaim land and to maintain the main tidal channel for navigation (Marker, 1967). By the early 18th century, canalisation of the river Dee at the head of estuary in association with the construction of a sea wall, as well as associated land drainage and reclamation, drastically changed the hydrodynamic regime. The ‘new cut’ canalisation thus forced a shift of the main navigation channel from the eastern English shore towards the Welsh shore. As a result, heavy siltation and a high level of accretion on the eastern English shore led to the estuary becoming a very shallow system due to the ensuing low energy conditions, and hence huge areas became colonised by saltmarsh, pioneered initially by *Salicornia* and then dominated by *Spartina anglica* in the 1930s (Marker, 1967; Moore et al., 2009). The modern Dee estuary has an effective length of 30 km, with a maximum width of 8.5 km at the estuary mouth and occupying an area of 16,101 ha (Davidson 1996;

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Moore et al., 2009). Its tidal range varies between 5.33 m at the landward end at Connah's Quay and 9.72 m at the seaward end at Hilbre Island (Moore et al., 2009).

2.2 Establishment of dust collector

Two Frisbee dust collectors were deployed on May 2008 over a time period of fourteen months in a resident's garden nearby the contaminated Parkgate site of Dee estuary saltmarsh, at 2 m and 0.3 m heights from the ground (Fig. 2a-2c) and at approximately ca. 3-4 m and ca. 1-2 m above the saltmarsh surface, respectively. The aims of deploying the dust collectors were to investigate differences in radionuclide flux as a function of boundary layer transport processes and the samples were collected at intervals to investigate any seasonal variability in radionuclide flux. The design of each Frizbee dust collector (FDCO) follows a combination of Marble dust collector (MDCO) and Frizbee dust collector that has been modified after a study completed by Sow et al. [2006].

Each dust collector consists of a single-piece Frizbee plastic disc that was inverted and fixed with a large plastic funnel (20 cm) using glue, and mounted on a plastic water pipe (3.2 cm diameter) at a given height above the ground. To trap sufficient dusts for gamma analyses, a circular piece of 0.2 cm plastic mesh was placed inside the Frizbee disc and glass marbles were filled on top of the mesh (Fig. 2a-2c). Glass marbles were used to prevent dust that had not yet filtered or washed (by rain water) into the bottom of the disc from being blown away i.e., to trap all dusts deposited on the collectors. There was a hole of ca. 1.7 cm at the centre of the disc and another small plastic funnel (6 cm) was fixed using glue in the middle of the underside of the Frizbee disc, and fitted with a 105 cm-long clear plastic tube that goes into a pre-washed plastic collection container so that rain-washed dust collected by the Frizbee disc can pass through tube into the collection container. The only difference between the two collectors was their mounting heights, with consequent different lengths of water pipe. For additional protection both dust collectors were covered with wire meshes along with some spikes to protect the collectors from contamination by birds. The two dust collectors are defined as LH (for long height dust collector at 2.0 m) and SH (for short height dust collector at 0.3 m) in the present study.

2.3 Sample collection and preparation

All particles within the dust samples are expected to be a mixture of particles (polluted and unpolluted) originating either from the saltmarsh, roadside and inland areas as well as from wet deposition of atmospheric dust via rain water and sampling includes dusts and radionuclides (if any) that have been desorbed into the rain water. Dusts and rain waters captured by the collectors were sampled at approximately 2-month intervals over a fourteen-month period, and results were sub-divided into two hydrological seasons: summer (April-September) and winter (October-March). During each collection period, the dusts and rain water were transferred via a collection pipe (plastic) into two plastic containers placed underneath the dust collectors on the ground. After returning samples to the laboratory, they were kept in the fridge before further chemical treatment. The plastic containers used for collection were thoroughly pre-washed, acidified with 10% nitric acid (HNO_3) and washed again with double distilled water before placing in the field each time.

2.4 Chemical treatment of rain water and gamma measurement

Before chemical treatment, each dust-washed rain water sample was transferred into a pre-washed 2 litre plastic bottle with proper labelling which was acidified with 10% nitric acid (HNO_3) and double distilled water. Each collection from two dust collectors was treated for chemical separation individually with proper identifications under a fume cupboard. Radionuclides were removed from the dust washed rain water samples (i.e., solution) by co-precipitation with manganese dioxide (Short, 1999), separated from the supernatant liquid by filtration through a 0.45 μm membrane filter

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(Cellulose Nitrate filter paper supplied by Whatman International Limited) with the help of a vacuum pump. When the whole solution was filtered, the filter paper with its residues, i.e., the dust sample was dried at 40°C in an oven and then placed onto a pre-weighed piece of cling film. After that, the filter paper along with the cling film was wrapped up, rolled like a cigarette and carefully placed inside the low volume plastic sample holder with proper identification for gamma measurements using a well-type High Purity Germanium (HPGe) detector.

2.5 Gamma measurements of aeolian dusts

Gamma measurements of all dust samples were made using a well-type HPGe detector, operated with an electric field of 3000 V with negative bias and with a resolution of 1.89 and 2.45 at 59.7 keV and 1332 keV, respectively. The detector had a crystal length of 6.08 cm, diameter of 5.52 cm, effective well depth of 4.42 cm and well diameter of 1.65 cm. The detector configuration was set in the 40-2000 keV range, for detecting photons and the liquid nitrogen reservoir of the HPGe detector was regularly filled with liquid nitrogen at 77°K. To avoid back-scattering and reduce background activity, the detector was well-shielded by lead and copper sheets. The sample holder employed was of 0.5 cm diameter and 5 cm height.

The energy calibration of the detector was done using NPRL ^{137}Cs and ^{241}Am point sources and used to check regularly before starting of the sample counts and the HPGe (well-type) detectors have been calibrated using NPL standard sources adsorbed onto a powdered resin base. The calibration of these secondary standards was checked against the University of Liverpool Environmental Radioactivity Research Centre's (ERRC) HPGe coaxial detector. The absolute efficiencies of the detectors were determined using calibrated sources and sediment samples of known activity. Corrections were made for the effect of self-absorption of low energy γ-rays within the sample (Appleby et al., 1992). Radioactivities of dust samples were calculated using ERLIB software that includes corrections for the effect of self-absorption of low energy γ-rays within the sample (Appleby et al., 1992).

2.6 Microscopic particle size analysis

As the amounts of dust particles were insufficient to perform a standard laboratory particle size analysis (PSA) using a laser granulometer, optical microscopic grain size analyses were performed using a Nikon optical microscopic at × 600 magnification for twelve microscope slides of dust samples from both the long and short height dust collectors (six from each collector). A total of 50 'field-of-view' observations were made on each slide to reduce possible analytical uncertainty. All observations were made starting from an end point of the slide and viewing it in same direction and continuing the observations in successive parallel rows to avoid any repetition of same image during grain count analyses. Percent volume was calculated based on the relative number of grains in each size class from a total number of 50 grains in each sample. For example, if 15 of the 50 grains observed were in the size range 8-4 µm, this would be an equivalent grain number volume of 30%. The same metric is applied to the relative proportions of organic and mineral particles. The total number of grains (both organic and mineral) observed were counted according to nine grain size classes: 500-250, 250-125, 125-63, 63-31, 31-16, 16-8, 8-4, 4-2 and <2 µm (medium sand, fine sand, very fine sand, coarse silt, medium silt, fine silt, very fine silt, clay/mud size range particles). In addition to determining the grain size category, each particle was classified, according to its appearance, as organic (translucent, orange to brown coloured with rounded edges or internal structure) or mineral (clear to opaque, light or no colour, straight or jagged edges), excluding any residual particles of MnO_2 from the co-precipitation analysis (Fig. 3a).

3. Results

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3.1 Radionuclide data and total annual effective dose estimations

Table 1 summarises the radionuclide analytical data of aeolian dust samples from six dust collections. The dust collections, dating roughly between April to early September are considered as 'summer' and from September to early April are considered as 'winter'. No ^{241}Am could be detected above the minimum detectable activity limits whereas ^{137}Cs could be detected for all dust samples. For the long height collector, the ^{137}Cs activity concentrations varied from 20 ± 4 to 794 ± 113 Bq kg^{-1} with an average of 193 Bq kg^{-1}; whereas for the short height collector, these values varied from 15 ± 4 to 252 ± 8 Bq kg^{-1} with an average of 109 Bq kg^{-1}. The highest activities for the long and short height collectors were found to be 794 ± 113 and 295 ± 8 Bq kg^{-1}, respectively.

Following detailed calculation procedures stated in Rahman et al. [2013], considering the minimum and maximum outdoor occupancy hours of 312 h and 730 h, the measured ^{137}Cs activity concentration ranged (15–794) Bq kg^{-1} in investigated dust samples of this study correspond to the estimated minimum and maximum total annual effective dose range (including external and inhalation) of (1-41) µSv y^{-1} and (2–95) µSv y^{-1}, respectively. It is seen that for minimum outdoor occupancy hours, the maximum estimated total annual effective dose of 41 µSv y^{-1}, falls within the world average value of 70 µSv y^{-1} (UNSCEAR, 2000), whereas for the maximum outdoor occupancy hour the total annual effective dose of 95 µSv y^{-1} is 36% higher than the world average value, which is significant. Other studies have also revealed higher ^{137}Cs activity concentration data from the Dee estuary compared with other saltmarshes in the UK (Bonnet et al., 1988; Hutchinson and Prandle, 1994; Oldfield et al., 1993; Plater and Appleby, 2004; Rahman, 2010; Rahman et al. 2013). This exposure pathway indicates a non-negligible concern and further in-depth investigation on the health impact is recommended.

The average mass of dust per day for the long height collector varies from 0.004 to 0.021 g day^{-1} whereas it varies from 0.002 to 0.037 g day^{-1}, for the short height collector (Table 3). Table 1 also reveals that there is an overall increasing trend of activity levels of ^{137}Cs for both collectors in winter, with the exception of the activity level of the second dust sample collection from the short collector, which is unexpectedly high. Moreover, data of Table 3 reveals that there is no particular increasing trend of dust accumulation. From first principles, it is reasonable to expect comparatively higher wind erosion and dust production in winter than in summer as the saltmarsh surface is covered less by vegetation compared to summer and winds are also generally higher in winter (e.g. Poirier, 2014). However, the saltmarsh surface is unlikely to dry out in winter and this probably reduces the potential for wind erosion during winter.

3.2 Microscopic particle size analytical data

A detailed study on radionuclide association with sized particle done by Rahman (2010) revealed that higher radionuclide activities are observed in fine silt to clay particles (< 2 µm), i.e., there is an increasing trend with decreasing particle size (Table 2). Hence, microscopic analysis was undertaken to identify % volume of particle size fractions of < 63 µm within the collected dust samples.

Figure 3b illustrates the % volume against grain size (from 500 to > 2 µm size ranges) distribution patterns for both long and short height dust collectors. It needs to be mentioned here that particles less than 4 µm could not be identified due to limitations of the magnification at × 600. It is seen that there is a gradual increase in % volume of grains from coarse silts to clay particles (i.e., from 4 µm to > 2 µm) for all dust samples from both collectors. It is seen that the highest percentage volume of grains was found for the grains in range of 2 to > 2 µm. On average, approximately 75% of grains were fine organic particles. Indeed, there may be some contribution from the MnO_2 precipitate arising from the chemical treatment of rain water to collect the radionuclides. Figure 3a shows an example digital photograph of the common view microscopic slides of dust samples. The

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microscopic view of representative slides of all dust samples in this study revealed that there are mainly two types of particles: organic and mineral. The optical density of the organic particles is high compared to the mineral particles. The particles with light brown or orange colours are the organic matter whereas particles with clear to opaque, light or no colour, straight or jagged edges are the mineral particles (mainly feldspar type). Particles with opaque type are assumed as the MnO_2 precipitate.

3.3 Possible reasons for the absence of ^{241}Am in dust samples

Though ^{241}Am is present in the Dee estuary saltmarsh surface sediments, it is absent in the windblown dust samples. A better explanation of relatively high concentration of ^{137}Cs in organic matter compared to ^{241}Am can also be understood from analysing characteristics of these two radionuclides, as various radionuclides behave differently in the environment and their chemical form, distribution coefficient (TF), mobility and other factors of radionuclides have an influence on their transfer to plants (Golmakani, 2008). As such, ^{137}Cs is environmentally mobile, acting like potassium, and can exist in both solution or particulate forms. It is thus readily taken up by plants through absorption/adsorption process and also be attached with mineral particles and be attached on to the saltmarsh plants during tidal inundation. In contrast, ^{241}Am exists more in particulate form in environment, and is relatively immobile due to its low solubility characteristic. It is less readily taken up by saltmarsh plants, mainly adsorbs on to mineral matter and, thus, remains largely in the soil or sediment (Duursma, 1972; Evans et al., 1983; Comans et al., 1989; EGE, 1994; Appleby, 1997).

Therefore, it is assumed in this study that the high % volume of organic (ca. 75%) and lower % volume of mineral (ca. 25%) dust particles are the main reasons for the relatively high activity levels of ^{137}Cs and below minimum detectable activity levels of ^{241}Am. However, opaque black particles (i.e., MnO_2 precipitate) were counted as opaque 'minerals' and thus contributed to the % volume of 'minerals'. Hence, the mineral contents may slightly be overestimated and the % volume of organic contents are likely higher than the estimated value (i.e., >75%). In addition, although rain-derived atmospheric ^{137}Cs (i.e., from fallout) is also included within the dust samples in this study due to the sampling method, our literature survey confirmed that this contribution is insignificant at the time of the sampling (Mitchell, 2010; RIFE, 2017).

4. Interpretation and discussion

For a better understanding, the radionuclide analytical data from the dust samples can be interpreted and discussed following three steps. In the first step, the radionuclide data from collected dust samples will be examined to validate the source of origin of measured ^{137}Cs; i.e., to investigate whether it originated from the saltmarsh or from fallout. In the second step, microscopic data from all dust samples will be examined to identify % volume of grains present within the dust samples in the size range of < 63 µm and also the types of particles (i.e., organic and mineral). ^{137}Cs activity data from the dust and size-fractioned samples (i.e., < 63 µm grains) will be compared to observed radioactivity levels in saltmarsh sediments to identify possible reasons for activity variations of ^{137}Cs in the dust samples. In the third step, radionuclide data of dust samples will be examined to identify the seasonal variations of dust accumulation rate and associated radionuclide activity. Moreover, to justify the potential of having saltmarsh-derived ^{137}Cs in dust samples collected from nearby residential area, meteorological data of the Hilbre Island weather station will also be considered.

4.1 Validation of the source of origin of ^{137}Cs

Dusts accumulate from both dry and wet deposition of dust from atmosphere. Dry deposition may come from roadside soil, inland soil and from nearby saltmarsh areas, wet deposition is via rainfall. Fallout-derived ^{137}Cs can be found in both inland and in coastal areas but the Sellafield-derived ^{137}Cs

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are found in estuary saltmarshes (Oldfield, et. al., 1993; Bonnet et al., 1989; Hutchinson, 1994). It is therefore important to consider the origin of the measured ^{137}Cs within the collected dust samples. Atmospheric fallout of ^{137}Cs has arisen from nuclear weapons testing since 1945 and from the Chernobyl accident (1986) in Russia. It was shown by Peirson and Salmon (1959) that the deposition of weapon test ^{137}Cs is a linear function of rainfall and the accumulation of the fallout ^{137}Cs activity is generally found to be higher in upland regions of the United Kingdom where the annual rainfall is high (Chandler and Gregory, 1976, Clark and Smith, 1988, Smith and Clark, 1989). Lake sediments also form an important component of any study of fallout deposition as these records incorporate direct fallout onto the lake together with a proportion of fallout on to the catchment and subsequently transferred to the lake via runoff.

As rain-derived atmospheric ^{137}Cs (i.e., from fallout) was included within the dust samples due to the sampling method, recent radioactivity data from air, rain water and lake sediments from different parts of UK along with North West England have been investigated to identify and confirm the source of measured ^{137}Cs in dust samples. The literature survey confirmed that this contribution is insignificant at the time of the study (Mitchell, 2010; RIFE, 2017). As reported by Mitchell [2010], ^{137}Cs concentrations in air and rain water samples collected during 2010 from seven sites of all around UK found below the analytical detection limit and were comparable to the previous 10 year's data. Also, the results showed a continuing decline of anthropogenic radioactivity in the environment. In another report, RIFE [2017] showed that the ^{137}Cs concentrations in air and rain water collected from nine stations around UK were found in the range between $< 0.5.3 \times 10^{-7}$ to $< 1.0 \times 10^{-7}$ Bq kg^{-1} and < 0.0074 to < 0.017 Bq l^{-1}, respectively, which are negligible. The results were also less than 0.01% of those observed in 1986, the year of the Chernobyl accident. Hence, it is apparent that rain-derived atmospheric fallout contribution of ^{137}Cs included within the dust samples in present study is insignificant. Moreover, the records of fallout ^{137}Cs activity in rain water from NW England found it to be below the analytical detection limit (personal communication, Peter Appleby) and past decay-corrected (to 2008, i.e., the time of sampling) lake sediment records of ^{137}Cs fallout activities (Appleby et al., 2003, Bonnett et al., 1989; Rahman, 2010) also revealed that the ^{137}Cs fallout radioactivity is negligible in NW England at the time of sampling. Hence, the available data indicates that the ^{137}Cs present in dust samples of this study originated mainly from the Dee estuary saltmarsh.

4.2 Particle size data and associations

The particle size data from six sediment cores (Fig. 4; after study of Rahman, 2010) from the Dee estuary saltmarsh reveal that the mean grain size of the top 20 cm sediments (i.e., where the peak activities of Sellafield-derived ^{137}Cs are located (Rahman, 2010; Rahman and Plater, 2014; Rahman et al., 2013) is < 10 µm. This means, the dust samples from the Dee estuary are derived from sediments of the same size. Also, comparison of the ^{137}Cs activity data (Table 2) show that the ^{137}Cs activity levels of dust samples are similar to the existing saltmarsh surface ^{137}Cs activity range, with the exception of one dust sample from the higher collector where a high ^{137}Cs activity was found high (794 Bq kg^{-1}) compared to surface activity range of the marsh sediments (Table 1). The potential reason for this high activity can be explained by observing down-core activity profiles (Fig.4) and the measured surface and peak activity dataset of the saltmarsh sediments (Rahman et al. 2013). Here, it can be seen that the value 794 Bq kg^{-1} falls within the activity profiles of both sides of peak activities for all six cores from the Dee estuary (Fig. 4). Thus, there is every possibility of having high ^{137}Cs activities in dust samples derived either from highly active samples from other positions of Dee estuary surface saltmarsh sediments that have not been measured or from sediments that has been redistributed from depth as a result of physical erosion, e.g., from the cliff of the lower saltmarsh (Rahman., 2010).

Furthermore, it can be seen from the Dee estuary saltmarsh long cores profiles (Fig 4) that the surface sediments are rich in organic content (ca. 30%-40%) and is apparent from the Loss on

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Ignition (LOI) down-core profile which reduces with core depth. Moreover, though there is no data available on ^{137}Cs concentrations from the Dee estuary saltmarsh plants, data on vegetation from another Sellafield-contaminated UK saltmarsh conducted by Beresford [2007] reported high activities of 1500 ± 670 Bq kg^{-1} and 380 ± 116 Bq kg^{-1}, for ^{137}Cs and ^{241}Am, respectively, which are significant. Hence, as dust samples in present study were up to 75% organic particles (and potentially higher), a valid the reason could be due to the uptake of more soluble highly active ^{137}Cs radionuclides by the saltmarsh plants from the saltmarsh sediments which then decomposed after their growth period and later transformed into dust size particles. Thus, it is assumed that the ^{137}Cs concentrations in collected dust samples in present study are saltmarsh generated and ^{137}Cs attached to both the organic and mineral dusts are responsible for the activity concentrations.

4.3 Justification for potential saltmarsh to land transfer of ^{137}Cs

Dry sediments from the saltmarsh surface can be lifted and suspended into air via wind erosion. It is well known that the grain movement is initiated when the wind-induced lift and shear velocity at the bed overcome the force of gravity acting on the particles. Three types of particle movement may occur during wind erosion and these are: suspension, saltation and surface creep. Out of these three movements, saltation is the most common. All movements also depend on the available grain size, and there is a certain wind threshold velocity for each grain size (Bagnold, 1941; Greeley and Iversen, 1985; Pye, 1987).

The wind friction speed threshold curve for the Earth was first derived by Bagnold [1941]. In 1985, Greeley and Iversen presented wind threshold friction speed curves (Fig. 5a) for the particles ranging from 10 to 10 000 µm for Earth, Mars and Venus, where they showed that for Earth, the particle size of ca. 100 µm (fine sand) is most easily be moved by a wind speed of ca. 0.2 m s^{-1} (denoted as V in Fig. 5a), and as grain size reduces the wind threshold friction speed value increases. However, Greeley and Iversen's threshold wind friction speed data (1985) were limited only to 10 µm size particles and no study has been made beyond this size so far (personal communication, Ron Greeley, 2010). Moreover, there are no specific wind friction threshold speed data for the fine particles in size range from 10 to < 2 µm, i.e., for fine silts to clay size particles. However, after study of Greeley and Iversen [1985], another study led by Kjelgaard et al. [2004] mentioned the value of wind friction threshold speed for PM_{10} group particles (i.e., < 10 µm, but not for a wider range of finer particles) based on field experiments from agricultural soils of Columbia Plateau. Kjelgaard et al. [2004] found from their experiment that the emission of PM_{10} from these soils appears to occur at a threshold friction speed of 0.4 m s^{-1} (V) and an approximate threshold wind velocity (V_{2m}) of 8 m s^{-1} at 2 m height. They also found that soil disturbance can result in reduced threshold values, ca. 0.3 at 6 m s^{-1} for V and $V_{2m,}$ respectively.

It is observed that for Earth, the wind threshold friction speed (V) for 10 µm size particles found by Greeley and Iverson [1985] is also ca. 0.45 m s^{-1} which matches very closely with the threshold friction speed (V) for a soil particle size group of <10 µm investigated by Kjelgaard et al. [2004]. Hence, the consistency of threshold friction velocities for 10 and <10 µm size particles encourages extrapolation of the threshold curve of Greeley and Iverson [1985] further to 2 µm in this study for a better estimation of approximate threshold friction velocities for each individual finer grain size fractions from 10-2 µm, as shown in Figure 5b. Hence, from the extrapolated curve, the wind speed friction velocities for particles of 63, 31, 16, 8, 4 and 2 µm were estimated at approximately 0.2, 0.25, 0.35, 0.5, 1.0 and 3 m s^{-1} (Fig. 5b).

Thus, to estimate approximate threshold windspeeds at 2 m height above the saltmarsh surface and to justify the potential aeolian transfer of ^{137}Cs attached to finer grains from the saltmarsh, wind speed data (at 10 m height) of the Hilbre weather station (Table 3) were analysed. Here we note that the wind speed follows a logarithmic profile above the bed. In this coastal location, adjacent to an

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aerodynamically smooth bed (sea surface, tidal flat and saltmarsh), we assume the majority of frictional reduction in wind speed is achieved close to the bed and that the wind speed data for 10 m height is an acceptable approximation for that at 2 m height. Further, that the wind speed at 10 m may exceed that at 2 m provides us with a likely maximum estimate of erosion and transport potential for dusts derived from the saltmarsh.

At first, the logarithmic decrease in wind velocity approaching the bed, i.e., the threshold friction speed (V) of finer particles (< 10 μm) for the saltmarsh surface, was estimated using the values of wind threshold velocity (V_{2m}) of 8 m s^{-1} at 2 m height (Kjelgaard et al., 2004) and a saltmarsh surface roughness value (z_0) of 0.0002 m (Kitwiroon et al., 2002) in the well-known Prandtl- von Karman logarithmic wind profile equation (Priestly, 1959):

$$V_Z = \frac{V}{k} \ln \frac{Z}{Z_0},$$

where V_Z is the wind speed at height z, V is the wind threshold friction speed, k is Karman's constant (0.4) and z_0 is the surface roughness length.

The estimated threshold wind friction speed (V) for saltmarsh surface was found to be 0.35 m s^{-1}, and is in good agreement with the estimated shear wind speeds (V) for 10-< 10 μm diameter particles of Greeley and Iverson [1985] and Kjelgaard et al. [2004]. This agreement further justifies the use of estimated wind friction speeds, V, of finer particles (63-< 2 μm) for an approximation of threshold wind speeds, $V_{Z,}$ for these particles at 2 m height. The estimated values of V_Z for 63, 31, 16, 8, 4 and 2 μm size particles were found to be 5, 6, 8, 12, 23 and 69 m s^{-1}, respectively.

The wind speed data from Hilbre reveal that for the whole dust collection period the wind speed varied from 0 to 22 m s^{-1}, with a mean wind speed variation of 5-8 m s^{-1}. The wind gust values ranged from 0 to 28 m s^{-1} with the mean wind gust variation of 7 to 10 m s^{-1} (Table 3). Thus, comparison of the wind speed data from Hilbre with estimated threshold wind speeds for finer particles confirm that the wind over Parkgate saltmarsh during the whole dust collection period were high enough to entrain fine 63-4 μm (i.e., coarse to fine silts) particles but not clay particles. If this is the case, then it can be assumed that the available ^{137}Cs activities present within the dust samples in this study were generated from contaminated Parkgate saltmarshes and associated mainly with silt size particles. The presence of clay size particles within the dust samples indicates that there may be another source of the finest dusts, e.g., atmospheric dust, the nearby road side and inland areas or, indeed, the MnO_2 precipitate from the analysis. However, we cannot exclude the likely transfer of agglomerated clay particles, which act aerodynamically as coarser (compound) grains and then break down during the laboratory analyses. Furthermore, there is also potential for clay particles to be transferred from the saltmarsh surface via saltation or reptation (ballistic impact) processes. Without data for further examination of this, our interpretation centres on the transport of 'solitary' grains via suspension or intermittent suspension

Table 3 show the mean and maximum wind speed, wind gusts and total mass of dust per day data from both dust collectors for six the collection periods. Whilst we generally observe higher ^{137}Cs activities in the winter season samples (Table 1) this is not accompanied by a higher sediment accumulation or a substantial difference in the wind field characteristics. Further, there is no consistent trend in the recorded grain size distributions (Figure 3b), which is consistent with no overall change in the prevailing wind field. Without clear evidence of causation, we speculate that the ^{137}Cs loading is higher during winter when the saltmarsh is in a phase of annual deterioration, i.e., a higher source availability rather than a higher net flux of particles. The non-linear sampling period along with the summative nature of the sampling (covering extended periods of time of variable wind speed and weather) limits further examination of this phenomenon.

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5. Conclusion

Detailed examination of radionuclide and grain size data from dust samples in this study reveals that as majority of the dust is organic (ca. 75%) then it is likely that the dust does not come only directly from the saltmarsh surface but largely from the decomposition of plants growing on the saltmarsh. Consequently, it is likely that more organic dust particles originated from plants (rich in ^{137}Cs, but poor in ^{241}Am) during summer and autumn at the end at the main plant growth period, which can also be observed from the mass flux data. Also detailed analyses of activity concentration data of ^{137}Cs from the saltmarsh surface, down core sediment (historic peak activity data) and dust samples reveal that there is a potential for the aeolian transport of radioactive dust particles inland from nearby contaminated saltmarshes, and that the estimated wind threshold velocities (5-23 m s^{-1}) for finer particles (63-4 µm, except <4 µm) and available mean and maximum wind speed data range of 5-28 m s $^{-1}$ over the Dee estuary saltmarsh surface (at 2 m height) shows that it is possible to liberate saltmarsh-derived radioactive dust. Radiological risk analysis reveals that although the maximum value of 41 µSv y^{-1} for the total annual outdoor effective dose for minimum occupancy falls below the world average value (70 µSv y^{-1}) the maximum value of 95 µSv y^{-1} for the total annual outdoor effective dose for maximum occupancy is 36% higher than the world average value. Hence, the health risk posed to coastal populations by ^{137}Cs present in saltmarsh-derived airborne dusts is non-negligible.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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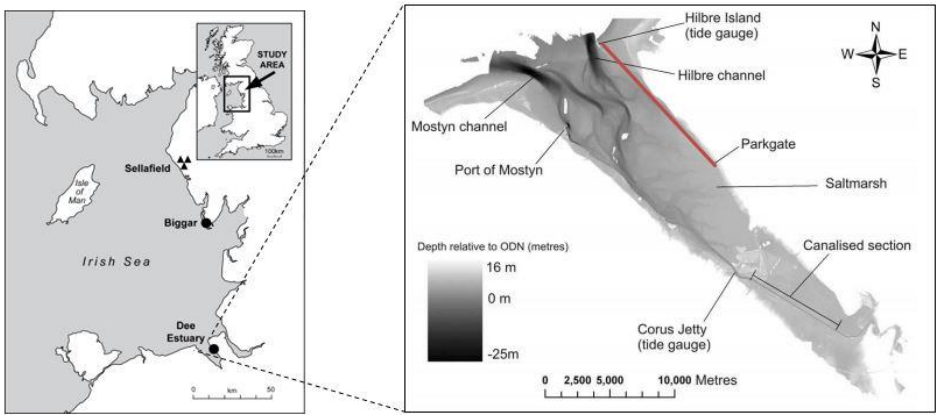

Figure 1. Locations of the Dee estuary saltmarshes in north-west England, UK (after Moore et al., 2009; extended modified figure); solid dark red line: distance between Hilbre Island and Parkgate saltmarsh (14 km); ODN refers to the altitude datum for Great Britain, Ordnance Datum Newlyn.

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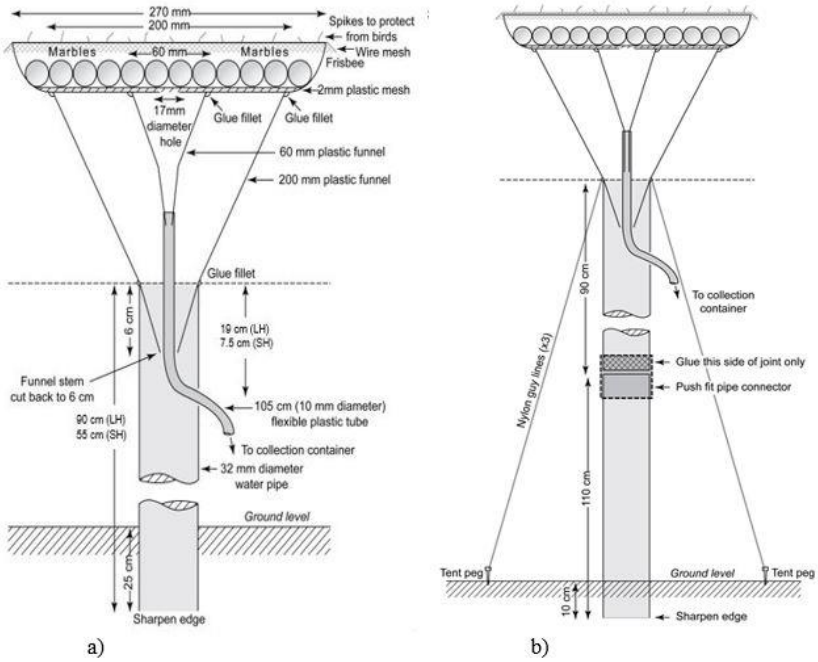

Figure 2. (a) Dust collectors (Long height, LH and Short height, SH) with funnel and top tube; (b) overall view of LH and SH dust collectors.

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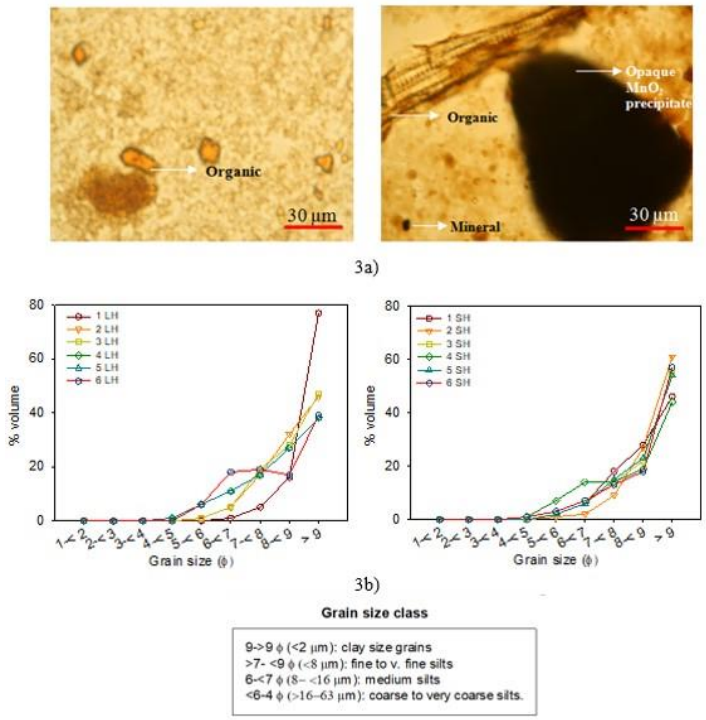

Figure 3 (a) Example microphotographs of representative dust samples. (b) Particle size distributions, by volume, of collected dust samples. Grain size is shown in φ to emphasize the importance of the finer grain sizes in the distribution, where $\varphi = -\log_2$ mm of the grain diameter, i.e., 8-9 φ = 4-2 µm, >9 φ = <2 µm.

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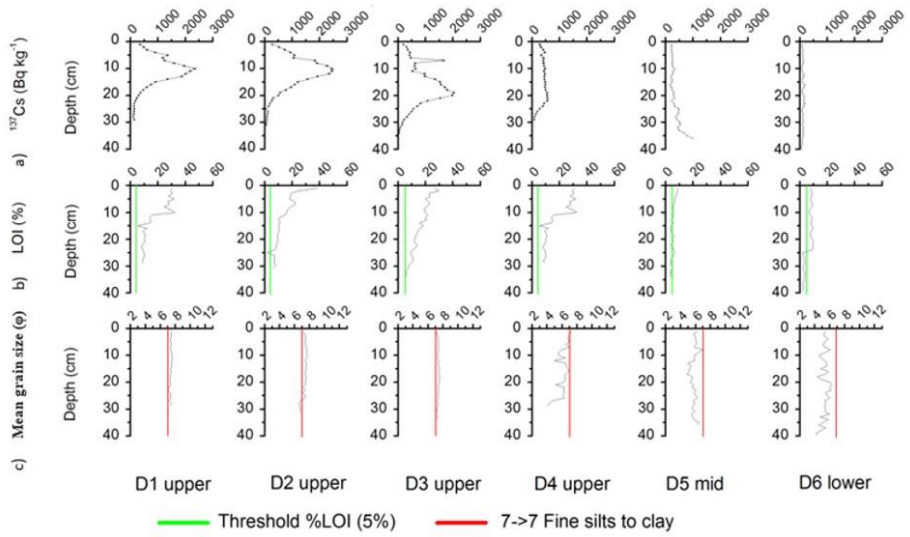

Figure 4. Summary figures of ^{137}Cs (Bq kg^{-1}), LOI (%) and mean grain size (8-9 φ = 4-2 μm, >9 φ = <2 μm) data from six long cores (20 cm) of the Dee estuary upper saltmarshes after study of Rahman [2010].

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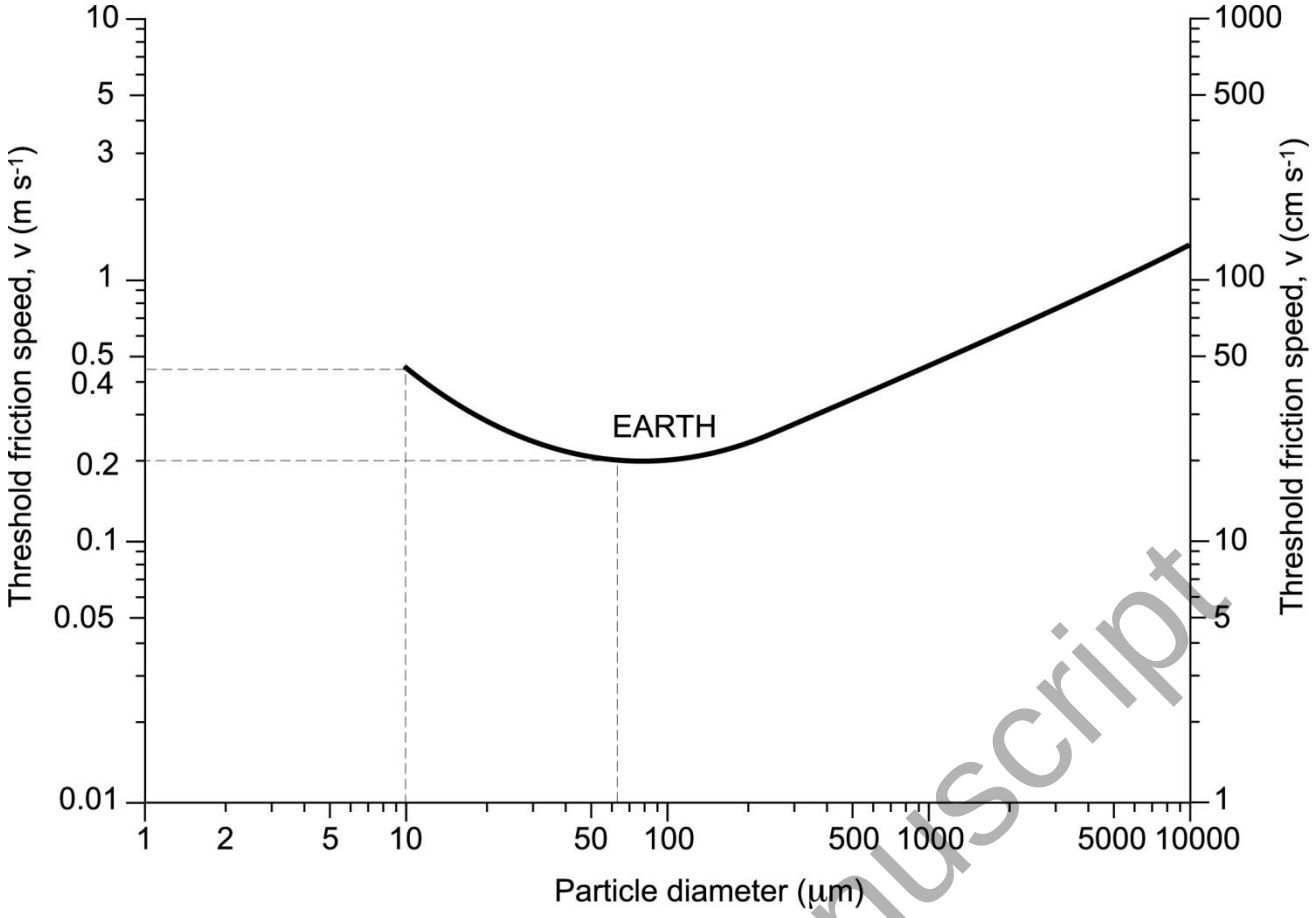

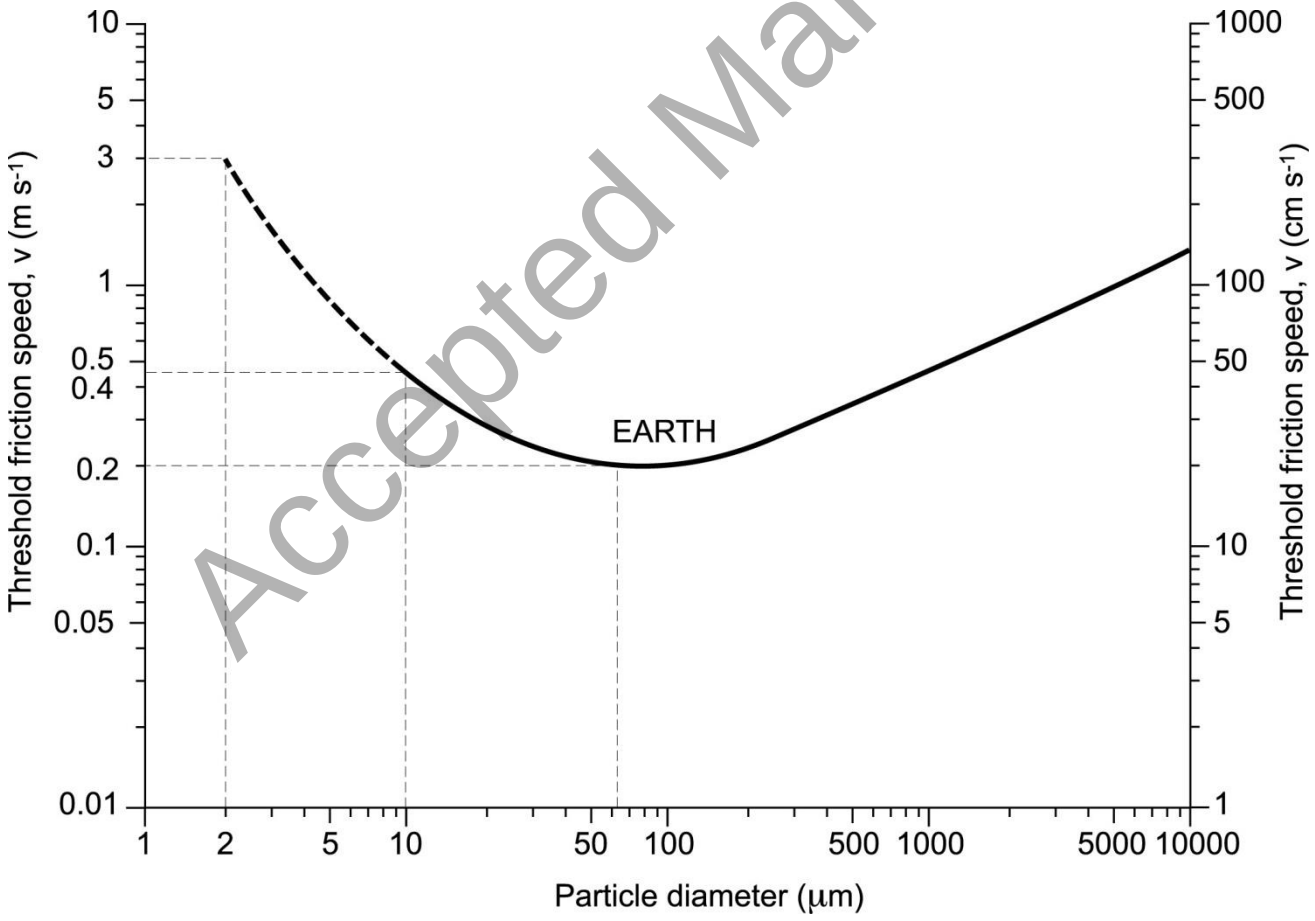

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Figure 5. (a) Threshold friction speed versus particle diameter for the Earth from study of Greeley and Iverson, 1985 (modified form with original curve). (b) Estimated threshold friction speed versus

particle diameter for the Earth; extracted from Greeley and Iverson (1985) study, and extended, modified form reproduced in present study based on practical data of Kjelgaard et al. (2004).

Table 1. Radionuclide activity data for ^{137}Cs and ^{241}Am (Bq kg^{-1}) in aeolian dust samples from long height (LH) and short height (SH) dust collectors.

Sample collection no.	No of days	Season	Sample ID	Net dust weight (g)	Mass per day (g)	^{137}Cs activity (Bq kg^{-1})	^{241}Am activity (Bq kg^{-1})
1.	31	S	1LH	0.511	0.016	27 ± 6	BDL
			1SH	1.149	0.037	15 ± 4	BDL
2.	30	S	2LH	0.227	0.008	80 ± 12	BDL
			2SH	0.190	0.006	252 ± 8	BDL
3.	27	S	3LH	0.570	0.021	20 ± 4	BDL
			3SH	0.163	0.006	74 ± 24	BDL
4.	58	W	4LH	0.128	0.002	207 ± 25	BDL
			4SH	0.086	0.002	233 ± 47	BDL
5.	79	W	5LH	0.027	0.004	794 ± 113	BDL
			5SH	0.645	0.008	57 ± 23	BDL
6.	128	S	6LH	0.682	0.005	31 ± 4	BDL
			6SH	1.617	0.013	24 ± 2	BDL

Note: Symbols S and W, LH and SH, BDL are summer and winter, long height collector (2 m) and short height collector (0.3 m), below detectable limit, respectively.

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Table 2. ^{241}Am and ^{137}Cs surface activities (Bq kg^{-1}, dry wt basis) in surface sediment (non particle size fractioned) and particle size fractioned samples of short core (10 cm) sediments from the Dee estuary saltmarshes of NW England, UK (Rahman, 2010).

Sample no	Marsh Position	Total Radionuclide activity (Bq kg^{-1})		Average ^{241}Am (Bq kg^{-1}) in particle size fractioned samples			Average ^{137}Cs (Bq kg^{-1}) in particle size fractioned samples		
		^{241}Am	^{137}Cs	in coarse silts (63-<31) μm	in medium silts (31-<16) μm	in fine silts to clay (16-<8) μm	in coarse silts (8-<4) μm	in medium silts (4-<2) μm	in fine silts to clay (2->2) μm
1	UO	87 ± 6 (1)	350 ± 19 (1)	-	-	-	-	-	-
2	UO	73 ± 5 (1)	276 ± 15 (1)	-	-	-	-	-	-
3	UM	77 ± 3 (1)	198 ± 8 (1)	-	-	-	-	-	-
4	UM	62 ± 7 (1)	282 ± 24 (1)	-	-	-	-	-	-
5	UO	171 ± 3 (10)	907 ± 9 (10)	69 (10%)	103 (21%)	164 (20%)	407 (10%)	558 (21%)	1279 (20%)
6	UO	269 ± 4 (10)	1309 ± 10 (10)	36 (10%)	224 (20%)	241 (19%)	194 (10%)	1085 (20%)	1636 (19%)
7	UO	103 ± 3 (10)	306 ± 8 (10)	61 (11%)	91 (20%)	108 (17%)	243 (11%)	310 (20%)	417 (17%)
8	UO	88 ± 4 (10)	373 ± 10 (10)	79 (11%)	92 (20%)	122 (18%)	315 (11%)	513 (20%)	633 (18%)
9	UO	136 ± 2 (10)	485 ± 5 (10)	98 (10%)	163 (14%)	212 (16%)	335 (10%)	574 (14%)	976 (16%)
10	UO	130 ± 3 (10)	491 ± 8 (10)	93 (13%)	137 (15%)	182 (14%)	373 (13%)	777 (15%)	1119 (14%)
11	UO	139 ± 3 (10)	568 ± 8 (10)	71 (12%)	138 (19%)	169 (18%)	299 (12%)	654 (19%)	1118 (18%)

Note: UO and UM are upper marsh- organic and -mineral; Errors presented as standard ± 1 σ. Data in () show the mean % abundance of total sample volume of corresponding size-fractions.

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Table 3: Comparison of dust accumulation rate with meteorological data at 2 m height from the Hilbre weather station near the Dee estuary during dust collection periods (wind data supplied by Proudman Oceanographic Laboratory, Liverpool).

Sample no	No of days	Season	Mean wind speed (m s^{-1})	Min-Max wind speed (m s^{-1})	Mean wind gust (m s^{-1})	Min-Max wind gust (m s^{-1})	Mass per day (g)
1	31	S	5	0-16	7	0-20	0.016 (LH) 0.037 (SH)
2	30	S	8	0-22	10	0-27	0.008 (LH) 0.006 (SH)
3	27	S	7	0-19	9	0-21	0.021 (LH) 0.006 (SH)
4	58	W	8	0-20	10	0-28	0.002 (LH) 0.002 (SH)
5	79	W	7	0-20	8	0-24	0.004 (LH) 0.008 (SH)
6	128	S	6	0-21	7	0-23	0.005 (LH) 0.013 (SH)

Note: LH and SH are long and short height collectors.

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