Characterising radium-226 particles from legacy contamination to support radiation dose assessments

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Highlights

- Particle characterisation data required to support dose assessments is presented
- Radiological, physical and chemical characterisation of Ra-226 particles is ongoing
- Characterisation data for one Ra-226 particle is presented as an example
- Plan to use Ra-226 particle characterisation data in dose assessments is discussed
- A new radioactive particle definition is needed to support dose assessments

Abstract

Radioactive particles are physically discrete sources of radioactivity that have been released into the environment as a result of past emergencies, events and practices. As the release of radioactive particles is often unplanned, the source term has not been characterised, and the potential radiation doses have not been prospectively assessed. If a plausible exposure pathway exists, radioactive particles in the environment may present a hazard to the public depending on their radiological, physical and chemical characteristics. Given their physically discrete nature, standard assessment approaches such as dispersion and transfer modelling of liquid and gaseous radioactive releases, are not appropriate for radioactive particles. The challenge for national regulatory authorities is to calculate potential radiation doses from unplanned releases of radioactive particles into the environment, assess whether the doses are relevant to radiological protection and decide whether actions are required to reduce potential doses. To address this challenge, this paper presents the approach being adopted to radiologically, physically and chemically characterise Ra-226 particles from a contaminated legacy site using gamma spectrometry, optical macroscopy and SEM-EDS. The use of particle characterisation data to support radiation dose assessments is discussed and consideration is given to radioactive particles in the context of radiological protection.

1. Introduction

Radioactive particles are physically discrete sources of radioactivity that have been released into the environment as a result of past emergencies, events and practices including nuclear power reactor accidents, accidents involving nuclear weapons, nuclear weapons testing, the use of depleted uranium in military operations and legacy contamination from past practices (IAEA, 2011). As the release of radioactive particles is often unplanned, the source term has not been characterised and the potential radiation doses have not been prospectively assessed. If a plausible exposure pathway exists, radioactive particles in the environment may present a hazard to the public depending on their radiological, physical and chemical characteristics. Given their physically discrete nature, standard assessment approaches such as dispersion and transfer modelling of liquid and gaseous radioactive releases, are not appropriate for radioactive particles (Dale et al., 2008). The challenge for national regulatory authorities is to calculate potential radiation doses from unplanned releases of radioactive particles into the environment, assess whether the doses are relevant to radiological protection and decide whether actions are required to reduce potential doses.

1.1. Radioactive particles in radiological protection

The International System of Radiological Protection, as published and maintained by the International Commission on Radiological Protection (ICRP), considers three different types of radiation exposure situation intended to encompass all possible circumstances where radiation exposure could occur: planned, emergency and existing. Existing exposure situations, relevant to unplanned releases of radioactive particles, are exposure situations that already exist when a decision on control must be taken. That decision could be to take action to reduce potential radiation doses, such as implementation of remediation strategies or site management controls, or to do nothing if such action is not warranted (ICRP, 2007). This should be done in accordance with a graded approach to ensure any decisions or actions taken are commensurate with the radiation risks associated with the exposure situation (IAEA, 2014).

Existing exposure situations include exposures that exist due to past emergencies, events or practices as well as naturally occurring exposures. The level of radiological protection in existing exposures situations is defined by the reference level, which is the level of dose or risk, above which it is judged to be inappropriate to plan to allow exposures to occur, and below which optimisation of protection should be implemented. The reference level is not prescribed by the ICRP but should be set by national regulatory authorities taking into account the prevailing circumstances of the existing exposure situation under consideration (ICRP, 2007).

Unlike planned exposure situations where the radiation source term is already characterised, and the potential radiation doses prospectively assessed against dose limits and constraints, the source term in an existing exposure situation is often not well characterised due to its unplanned nature (Dale et al., 2008). In these circumstances, data on the source term characteristics, including any radioactive particles present, need to be obtained to undertake an assessment of potential radiation doses against the reference level and take appropriate actions, if necessary.

A significant amount of research has been undertaken to characterise radioactive particles from a number of existing exposure situations that have arisen due to past emergencies, events and practices: nuclear power reactor accidents at Chernobyl, Ukraine (Pöml and Burakov, 2017; Shiryaev et al., 2018) and Fukushima, Japan (Kaltofen and Gundersen, 2017; Martin et al., 2016; Yamaguchi et al., 2016); accidents involving nuclear weapons at Palomares, Spain (Aragón et al., 2008; Jimenez-Ramos et al., 2010; Jiménez-Ramos et al., 2012, 2008, 2006; Lind et al., 2007; López et al., 2007; Pöllänen et al., 2006) and Thule, Greenland (Eriksson et al., 2005; Lind et al., 2005); nuclear weapons testing (Burns et al., 1995; Conway et al., 2009; Jernström et al., 2006); the use of depleted uranium in military operations (Lind et al., 2009; Sajih et al., 2010; Salbu et al., 2005, 2003; Török et al., 2004); and legacy contamination from past practices at Sellafield, England (Clacher, 2011, 2010; Cowper, 2009), Dounreay (Aydarous et al., 2008; J. Darley et al., 2003) and Dalgety Bay, Scotland (Wilson et al., 2013).

The next step is to use the data from radioactive particle characterisation studies to calculate potential radiation doses, which will allow the assessment of the existing exposure situations against the reference level and, if necessary, implementation of remediation strategies or site management controls to reduce potential radiation doses in accordance with a graded approach.

1.2. Existing exposure situation at Dalgety Bay, Scotland

A significant amount of work has been undertaken by, and on behalf of, the Scottish Environment Protection Agency (SEPA) to address the existing exposure situation at Dalgety Bay, which is an estuarine bay located on the north bank of the Firth of Forth estuary in Scotland (Figure 1). Radioactive particles, containing Ra-226 and associated alpha-, beta- and gamma-emitting daughter radionuclides, were first discovered on the beach at Dalgety Bay in 1990 and originate from past practices undertaken on the land adjacent to the bay by the UK Ministry of Defence (MoD). The land was host to MoD air force activities (RNAS Donibristle and HMS Merlin) between 1917 and 1959, a time when Ra-226 was used in paint to luminise aircraft components (Patton, 2013). Activities on the land left a legacy of radioactive contamination and SEPA has been undertaking regular radiological monitoring surveys that have collectively recovered >1000 radioactive particles, varying greatly in physical size and Ra-226 activity (approx. 1kBq - 76MBq).

The existing exposure situation at Dalgety Bay was assessed by SEPA in 2013 (Dale, 2013). The assessment concluded there is a significant possibility of members of the public receiving radiation doses via skin contact and inadvertent ingestion above the relevant reference levels as defined in The Radioactive Contaminated Land (Scotland) Regulations 2007 Statutory Guidance (Scottish Government, 2010). As a result, the MoD is implementing a remediation strategy, meanwhile site management controls, such as signage and site demarcation, are being maintained by SEPA to protect the public.

As part of this assessment, SEPA commissioned a number of ad hoc research studies to provide the underpinning data for the skin contact and inadvertent ingestion dose calculations, as well as highlighting limitations and knowledge gaps requiring further research. Skin dose modelling was undertaken to estimate potential skin doses but, due to the lack of particle characterisation data, a point source geometry was assumed. This approach neglected the impact of self-absorption of radiation within the particles leading to an overestimation of skin dose, particularly for the alpha and beta emissions (Charles, 2008). Consequently, direct measurements of potential skin doses were undertaken using radiochromic film (RCF) dosimetry for ten particles. Improved skin dose modelling would have required details of the size, shape, density, chemical composition and radionuclide content, which were not available (Charles and Gow, 2010). The direct measurements demonstrated the impact of self-absorption, as the measured doses were significantly lower than the previously modelled doses. The beta emissions were the dominant contributor to skin dose, with a potential contribution from the alpha emissions at shallow skin depths, although the RCF was not specifically calibrated for alpha emissions (Charles and Gow, 2010). Additionally, the particle activity measurements reported in both studies are subject to significant uncertainty (15 - 30%), contributing to the overall uncertainty of the skin dose estimates.

Simulated gastrointestinal digestion was undertaken using sixty particles for calculating doses from inadvertent ingestion and found a wide range of gastrointestinal solubility (1 – 35% of particle activity) but, due to the lack of particle characterisation data, the cause of such a wide range could not be investigated. The percentage solubilities reported are also subject to significant uncertainty due to the 25% uncertainty on the particle activity measurements (Tyler et al., 2013).

A sample of nine particles was radiologically, physically and chemically characterised with the aim of investigating their origin, deposition and transport within the local environment at Dalgety Bay (Wilson et al., 2013). The analyses reported a range of activities, sizes, shapes and chemical composition, and alluded to the presence of distinct sub-populations, but the limited sample size did not allow for statistically robust conclusions. Additionally, these particles were different to those used in the skin and ingestion dose studies (Charles, 2008; Charles and Gow, 2010; Tyler et al., 2013), therefore the influence of particle characteristics on skin and ingestion doses could not be investigated.

An improved understanding of the hazard presented by the Ra-226 particles is not only important for the work ongoing at Dalgety Bay but also other sites potentially contaminated due to past practices using radium as well as existing exposure situations involving radioactive particles from other sources. It is important to understand the particle characteristics that influence radiation doses and to have a suitable level of confidence in radiation dose assessments for ensuring that existing exposure situations are assessed appropriately, and for implementing appropriate remediation strategies or site management controls.

1.3. Research Aim & Objectives

The aim of this paper is to highlight the research currently underway to radiologically, physically and chemically characterise a representative sample of Ra-226 particles from Dalgety Bay, creating particle profiles containing the necessary data to support radiation dose assessments. Research objectives address the limitations and knowledge gaps identified in previous research, specifically to:

i. Radiologically, physically and chemically characterise a larger sample of Ra-226 particles;
ii. Improve particle activity estimates by reducing the uncertainty in activity measurements;
iii. Assess potential skin contact doses through improved measurement and modelling, and investigate the particle characteristics having the greatest influence on skin doses;
iv. Assess potential inadvertent ingestion doses and investigate the particle characteristics influencing gastrointestinal solubility; and
v. Determine whether there are any distinct sub-populations of particles that have common characteristics, consider their provenance and distribution in the environment and any implications for the development of remedial measures and the management controls currently in place.

The research introduced in this paper addresses research objectives (i) and (ii) and methods are currently under development to address research objectives (iii), (iv) and (v).

More broadly, this paper builds on the significant amount of research already published on radioactive particle characterisation and discusses the next step of using the data generated from characterisation studies to calculate potential radiation doses. Taking this next step allows for the assessment of existing exposure situations against the reference level and, if necessary, implementation of remediation strategies or site management controls to reduce potential radiation doses in accordance with a graded approach.

2. Materials and Methods

Radiological, physical and chemical characterisation of a representative sample of Ra-226 particles is underway using a variety of analytical techniques. The particles have been selected from those recovered during the monitoring surveys undertaken by SEPA based on in-field estimates of Ra-226 activity and physical size. Each particle is undergoing the same suite of analyses to produce particle profiles containing the necessary data to support radiation dose assessments.

2.1. Radiological Characterisation

The radioactivity content of each particle is analysed by gamma spectrometry. The analysis is using an Ortec Gamma-X (GMX) N-Type High Purity Germanium (HPGe) Coaxial Photon Detector linked to Ortec Gamma Vision Software for spectrum analysis. Due to the irregular geometries of the Ra-226 particles, efficiency calibrations associated with standard sample geometries, such as Marinelli beakers, are not appropriate. To minimise the impact of the irregular geometries, an efficiency calibration has been derived that is as independent from sample geometry as possible (Tyler et al., 2013). Using two gamma reference point sources, Ra-226 (100kBq, UR371, Eckert & Ziegler) and Pb-210 (231kBq, KU654, AEA Technology), a particle-specific efficiency calibration has been derived at distance from the detector (20cm), allowing the Ra-226 particles to behave as point sources, minimising the effect of sample geometry. The particle-specific efficiency calibration has been quality checked using a different gamma reference point source, Eu-152 (43.8kBq, AF4331, Eckert & Ziegler).

Using the particle-specific efficiency calibration, each particle is counted at 20cm above the detector until the counting uncertainty (2σ) is <5% to determine the activity of Ra-226 and its gamma-emitting daughter radionuclides Pb-214, Bi-214 and Pb-210. The particles are counted in plastic pots, which are closed with a lid but not hermetically sealed.

2.2. Physical Characterisation

The size and shape of each particle is analysed by optical macroscopy. The analysis is using a Leica M420 optical macroscope fitted with an Olympus ColourView III digital camera linked to the Olympus Stream Image Analysis Software for image acquisition, processing and measurement. The macroscope is calibrated for a range of magnifications (5.8x, 8x, 10x, 12.5x, 16x, 20x, 25x and 35x) in order to analyse a range of particle sizes. Each magnification is calibrated using a stage micrometer and a quality check is performed before and after each batch of particles is analysed using a reference object. The dimensions of the reference object have been accurately measured using a set of calibrated Vernier callipers.

Each particle is placed on the macroscope stage beneath the objective lens under oblique incident illumination from two opposing directions to minimise shadows. The most appropriate magnification is selected, and adjustments made to the white balance and exposure. Due to the irregular geometry of the particles, images are acquired using the manual Extended Focal Imaging (EFI) function in Stream, which acquires multiple images at different focal points and combines them into a single image with the whole particle in focus. Once the EFI is captured, the image is segmented using HSV thresholding to separate the particle from the image background, allowing the software to make the required size and shape measurements. Each particle is imaged in multiple orientations, at least three if possible, in order to calculate average size and shape parameters for each particle. However, due to the nature of their production and time spent in the environment, the shape of the particles varies considerably. Depending on their shape, some particles are more amenable to imaging in different orientations than others, and may have images taken in greater, or fewer, than three orientations. A total of 10 different size (radius, diameter, equivalent circular diameter, area and perimeter) and shape (shape factor, sphericity, aspect ratio, elongation, convexity) measurements are taken for each orientation. The individual measurements for each orientation are recorded as well as the mean and standard deviation of all orientations of the particle. Qualitative observations are also made such as the presence of void spaces and particle friability.

The mass of each particle is measured using an Oxford GM-2505D 5-figure laboratory balance. The balance is calibrated annually by a UKAS accredited calibration service and the calibration is checked before and after each batch of particles is measured. The check is performed using a set of calibration weights (1, 10 and 100mg; Kern & Sohn) manufactured to International Organisation of Legal Metrology (OIML) standards. Each calibration weight is measured in triplicate to ensure the balance is performing correctly. The mass of each particle is measured once, and the result recorded when the balance has stabilised.

2.3. Chemical Characterisation

The surface chemical composition of each particle is analysed by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS). The analysis is using a Zeiss EVO MA-15 variable pressure SEM fitted with a backscattered electron (BSE) detector, a secondary electron (SE) detector, a variable pressure secondary electron (VPSE) detector and an Oxford Instruments X-Max 80mm^2 SDD EDS detector. The BSE detector is positioned directly above the microscope stage whereas the SE, VPSE and EDS detectors are positioned at an angle (approx. 30°) relative to the stage. The SEM is using Zeiss SmartSEM Software for BSE, SE and VPSE image acquisition and Oxford Instruments AZtec Software for EDS analysis. The SEM-EDS is calibrated annually by the manufacturer using a multi-element standard and a quality check is performed before and after each batch of particles is analysed using a cobalt standard.

The particles are mounted on standard 12.5mm diameter aluminium SEM pin stubs using carbon adhesive disks. The particles are not subject to any sample preparation, such as the application of a conductive coating or polishing, in order to preserve their original characteristics for the subsequent analyses required for the assessment of potential radiation doses. As the particles are uncoated, the analysis was performed under low vacuum conditions to help prevent the accumulation of electrostatic charge on the particle surface (Wilson et al., 2013).

Each particle is imaged in a single orientation using the BSE detector to gather qualitative information on surface chemical composition based on atomic number (Z), where areas composed of high-Z elements appear brighter than areas composed of low-Z elements. SE and VPSE images are acquired to gather qualitative information on surface topography, revealing surface features and areas of the particle surface that are in shadow relative to the SE, VPSE and EDS detector positions. The EDS is used to gather information on surface chemical composition, identifying individual elements to produce element distribution maps and calculating element abundance (weight %). Carbon is excluded from the EDS analysis due to interference from the carbon adhesive disks.

Due to the irregular geometry of the particles creating shadows, and to allow inter-particle comparison, the surface chemical composition data is collected semi-quantitatively to show the relative abundance of elements on the surface of the particles. Three different types of spectra are acquired for each particle: a particle spectrum acquired from the entire surface of the particle facing the detector; phase spectra acquired from areas on the particle surface where elements appear to be heterogeneously distributed, forming a distinct phase; and point spectra acquired from areas on the particle surface where elements appear to be highly localised. Each spectrum provides element abundance data (weight %) for the part of the particle surface from which the spectrum is acquired.

3. Results

Whilst the analysis is underway for the full representative sample of Ra-226 particles, the complete particle profile for one sample, DBP-03-07, is presented as an example.

3.1. Radiological Characterisation

The results of the gamma spectrometry analysis for DBP-03-07 show the presence of Ra-226 (5.97kBq ± 6.77%) and its gamma-emitting daughter radionuclides Pb-214 (5.65kBq ± 6.04%), Bi-214 (5.57kBq

± 6.06%) and Pb-210 (4.77kBq ± 8.39%). The particle-specific calibration has significantly reduced the total uncertainties (2σ) to <7%, compared to previous studies (Charles, 2008; Charles and Gow, 2010; Tyler et al., 2013). Pb-210 is the only exception due to the higher activity uncertainty of the Pb-210 source used in the efficiency calibration. The radionuclide with the greatest activity is Ra-226, closely followed by Pb-214 and Bi-214, with the lowest activity attributable to Pb-210. Considering the total uncertainties, the activities of Ra-226, Pb-214 and Bi-214 could be in secular equilibrium, whereas the Pb-210 is not due to its activity being significantly lower than the other radionuclides.

3.2. Physical Characterisation

The results of the optical macroscopy analysis for DBP-03-07 are presented in Figure 2, showing the images generated from the EFI, and in Tables 1 and 2, showing the size and shape measurements obtained from the image analysis. DBP-03-07 had EFI's taken in five different orientations at 25x magnification. The images reveal the irregular shape of the particle, which is different in all five particle orientations. All images show the presence of void spaces within the particle and there is evidence of its deposition in a coastal environment with what appears to be sand grains embedded in some of the voids in images 1 and 5. The particle did not show a tendency to be friable and appeared to be physically robust, although this was under the careful handling of laboratory conditions.

The size measurements indicate that the particle has a mean diameter of 2.5mm. The size measurement showing the greatest variation is the perimeter, reflecting the irregular geometry, which is supported by the low shape factor. The sphericity and convexity factors are <1 indicating the extent to which the particle deviates from a perfect sphere, which is supported by the aspect ratio and elongation factor both being >1. The particle mass was measured as 0.01232g.

3.3. Chemical Characterisation

The results of the SEM-EDS analysis for DBP-03-07 are presented in Figure 3, showing the BSE, SE and VPSE images, Figure 4, showing the EDS element maps and Table 3, showing the relative abundance of each element in the particle (weight %). The BSE image reveals distinct high-Z and low-Z regions across the particle surface and the SE and VPSE images reveal a rough surface topography. The EDS element maps reveal the presence of several different elements, some of which appear to be homogenous (excluding shadows), whereas others appear to be heterogeneous by either forming distinct phases or being highly localised. The particle orientation in the SEM-EDS analysis appears to be most closely aligned with the orientation in image 2 from the optical macroscopy, but notably not the same.

Oxygen (O) appears to be homogeneous, which is confirmed in the relative abundance data where it is consistently high across all spectra and is always the most abundant. Silicon (Si) also appears to be homogeneous across all spectra except for a few significantly brighter, highly localised areas in the Si map. These were investigated using point spectra and, based on their chemical composition, are likely to be embedded sand grains, although this is not as obvious in image 2 of Figure 2 compared to images 1 and 5. Iron (Fe) is another element that appears to be present across the particle as it is present in all spectra but it is also forming a distinct phase as can be seen from the Fe map and the high relative abundance of 26.9% in the Fe phase spectrum compared to all other spectra. The map for aluminium (Al) also appears to be homogeneous and this is reflected in the relative abundance data with the exception of the absence of Al in the copper (Cu) and chlorine (Cl) phases.

All other significant elements appear to be heterogeneous by either forming distinct phases or being highly localised. The Cu and Cl maps are interesting as these elements appear to be at least partly co-located, likely as copper (II) chloride, which is supported by the optical macroscopy image where a blue/green colouration can be seen. However, the Cu is more widely distributed than the Cl. Calcium (Ca) is another element that appears to be present throughout the particle but also forming a distinct phase, as supported by the element abundance data. The tin (Sn) map is particularly interesting as the Sn is present in a distinct phase that can be easily identified as the high-*Z* area in the BSE image. The Sn is very localised, only appearing in one other spectrum (excluding the particle spectrum). Titanium (Ti) is even more highly localised appearing as a distinct point on the element distribution map.

4. Discussion

Although the results presented are for one particle as an example, there are some initial observations with potential implications for the planned future work to support radiation dose assessments. The significance of the following observations will be clearer when the full set of particle characterisation data is available and will be considered in the subsequent dose assessment work, as appropriate.

4.1. Radiological Characterisation

In the gamma spectrometry results, it was noted that the activity of Pb-210 is significantly lower than the other radionuclides; there are a number of reasons why this might be the case. Firstly, due to the physical half-life of Pb-210 (22.3 years) and the potential age of the Ra-226 present in the particles, the Pb-210 will have not yet reached secular equilibrium. For example, if it were assumed that the Ra-226 was pure when used to make the paint and the particles are 70 years old, the Pb-210 would have only reached approximately 90% of its secular equilibrium activity at the time of analysis.

Secondly, the 46.54keV Pb-210 photon is the lowest energy of all photons included in the analysis meaning it is the most susceptible to self-absorption within the particle and may never reach the detector. This is due to the photoelectric effect, which is highly dependent on the effective atomic number of the absorbing material. The impact of this is mitigated to some extent by the Pb-210 reference source used in the efficiency calibration where the Pb-210 is sealed in a Perspex disk and due to the particle having a small physical size. However, differences in chemical composition and density of the Pb-210 source and the particle will result in different photoelectric cross-sections for the 46.54keV photons.

Lastly, the daughter radionuclide immediately after Ra-226 in the decay chain is Rn-222, which is gaseous. If some Rn-222 is able to escape from the particle this would result in all subsequent daughter radionuclides, including Pb-210, having a lower activity and never reaching secular equilibrium with Ra-226. The activities of Pb-214 and Bi-214 are both slightly lower than Ra-226, which could suggest a slight disequilibrium due to a small amount of Rn-222 loss. However, considering the total uncertainties, the activities of Pb-214 and Bi-214 could be in secular equilibrium with Ra-226. Either way, the fact that the measured activities of Pb-214 and Bi-214 are high indicates that even if there is some loss of Rn-222 it will be a small amount and is unlikely to be a significant contributor to the lower Pb-210 activity.

In addition to Rn-222, it is important to note that there will be other Ra-226 daughter radionuclides present in the particles not identified in the gamma spectrum; Po-218, Po-214, Bi-210 and Po-210. Due to the short physical half-lives of Po-218 and Po-214, they will also be in secular equilibrium with Ra-226 (or Rn-222 if some is being lost). However, as Bi-210 and Po-210 are after Pb-210 in the decay chain, these radionuclides will be in secular equilibrium with the Pb-210 rather than the Ra-226. It is important to consider this in a dose assessment of the Ra-226 particles, which must take account of the full decay chain, as all the radionuclides will contribute to the dose to a greater or lesser extent.

4.2. Physical Characterisation

In the optical macroscopy results, the irregular shape and the presence of void spaces in the particle was noted. Although the particle did not show a tendency to be friable under laboratory conditions, due to its irregular shape and multiple void spaces, it could be friable if it were left to persist in the environment where it would be subject to physical and chemical weathering processes. Particle friability is a potentially significant consideration when deciding whether to undertake remedial actions and the subsequent development of remediation strategies. Friable particles may not present a plausible exposure pathway in their current form due to their physical size but may do so in future if left to persist in the environment. Weathering processes can break down particles into smaller fragments, potentially altering the exposure pathways by creating particles that may more easily adhere to the skin, may be more easily inadvertently ingested or, if sufficiently small, inhalable. Additionally, the presence of void spaces could have an impact on inadvertent ingestion doses by providing an increased surface area for interaction with gastrointestinal fluids.

4.3. Chemical Characterisation

An important observation from the SEM-EDS results is that the EDS did not detect Ra-226 or any of its daughter radionuclides in DBP-03-07. This was also reported in Wilson et.al (2013) for all particles included in the analysis and was attributed to the detection limit of the EDS being too high, which is also likely to be the case here. However, despite not detecting it, the heterogeneous distribution of some of the other elements on the particle surface could be an indication that the Ra-226, and associated daughter radionuclides, are also heterogeneously distributed. Intra-particle activity heterogeneity could have important implications for radiation dose assessments as skin contact doses could be more localised than suggested by particle size. Furthermore, this could be important for friable particles where the particle activity would not be equally distributed between the separate fragments after particle breakdown. In terms of inadvertent ingestion dose, the activity could be more, or less, available for gastrointestinal absorption depending on whether the activity is localised on the surface or distributed throughout the particle. Skin contact doses will also be affected by this whereby self-absorption of the emissions would be greater for activity distributed throughout the particle compared to activity localised on the particle surface.

4.4. Future work to support radiation dose assessments

Once the planned programme of Ra-226 particle characterisation is complete, the radiological, physical and chemical characterisation data will be used in the assessment of potential radiation doses to the public via skin contact and inadvertent ingestion; the most significant exposure pathways at Dalgety Bay.

To address research objective (iii), a skin contact dose model for the Ra-226 particles is being developed using the Monte Carlo N-Particle (MCNP) radiation transport code. The mass, size, and shape data from the physical characterisation as well as the elemental composition data from the chemical characterisation are being used to parameterise the model. Direct measurements of skin contact doses will be undertaken using radiochromic film (RCF) dosimetry. Once validated by the direct measurements, the skin dose model will be used to explore the influence of different particle characteristics on skin doses. The reduced uncertainty on the particle activity measurements will reduce the overall uncertainty of the skin contact dose calculations providing greater confidence in skin dose assessments.

To address research objective (iv), the particles are being analysed by simulated gastrointestinal digestion to determine the gastrointestinal solubility of the particles, allowing for the calculation of potential inadvertent ingestion doses. The mass, size, and shape data from the physical characterisation as well as the elemental composition data from the chemical characterisation will be used to explore the influence of different particle characteristics on gastrointestinal solubility. The reduced uncertainty on the particle activity measurements will reduce the overall uncertainty of the gastrointestinal solubility measurements providing greater confidence in inadvertent ingestion dose assessments.

To address research objective (v), the full suite of particle characterisation data will be used to investigate whether there are any distinct sub-populations with common characteristics, as alluded to by Wilson et al. (2013), consider their provenance and distribution in the environment and any implications for the implementation of remediation strategies or site management controls. An improved understanding of the hazard presented by the Ra-226 particles is not only important for the work ongoing at Dalgety Bay but also other sites potentially contaminated due to past practices using radium as well as existing exposure situations involving radioactive particles from other sources.

The particle characterisation data and its planned use in radiation dose assessment introduced here for the Ra-226 particles could serve as an example of how it could be applied to particles in other existing exposures situations. To facilitate this, a definition of radioactive particles in the context of radiological protection and in accordance with a graded approach is needed. The International Atomic Energy Agency (IAEA) currently defines radioactive particles as *"…a localized aggregation of radioactive atoms that give rise to an inhomogeneous distribution of radionuclides significantly different from that of the matrix background."* (IAEA, 2011). However, this is neither in the context of radiological protection nor in accordance with a graded approach.

As detailed earlier, the level of radiological protection in existing exposures situations is defined by the reference level, which is the level of dose or risk above which it is judged to be inappropriate to plan to allow exposures to occur, and below which optimisation of protection should be implemented. Consequently, for a definition of radioactive particles to be in the context of radiological protection, it must focus on the hazard presented by radioactive particles and the potential radiation doses, rather than their inhomogeneity relative to the matrix background. As the current IAEA definition is focused on the matrix background, it is not in the context of radiological protection and is driving the consideration of all radioactive particles, regardless of their hazard, which is not in accordance with a graded approach.

A new definition of radioactive particles is needed to ensure that the management of existing exposure situations involving radioactive particles is relevant to radiological protection and in accordance with a graded approach. Such a definition could be set in relation to the reference level as this would address the needs of national regulatory authorities and allow for the proper implementation of the international system of radiological protection. If individual radioactive particles could deliver doses above the reference level, these particles would require a particle-specific dose assessment to be undertaken. If individual particles could not deliver doses above the reference level, these particles could be considered as particulate contamination, of which consideration is only needed regarding the rate of release of the radioactivity from the particles. These particles do not require a particle-specific dose assessment as, once radioactivity is released from the particles, standard distribution coefficients and/or concentration ratios would apply. Such a definition would ensure that the management of existing exposure situations involving radioactive particles is relevant to radiological protection and in accordance with a graded approach.

5. Conclusions

The particle profile for one Ra-226 particle from Dalgety Bay has been presented and discussed to highlight the radiological, physical and chemical characterisation underway and how these data will be used in radiation dose assessments. Once the particle profiles are complete for the full sample of Ra-226 particles, the characterisation data will be used to improve radiation dose calculations and understand the particle characteristics that influence skin contact and inadvertent ingestion doses. An improved understanding of the hazard presented by the Ra-226 particles is not only important for the work ongoing at Dalgety Bay but also other sites potentially contaminated due to past practices using radium as well as existing exposure situations involving radioactive particles from other sources.

More broadly, there is a significant body of work already published characterising radioactive particles from a variety of sources and the next step is to use the data from radioactive particle characterisation studies to calculate potential radiation doses. This will allow for the assessment of existing exposure situations against the reference level and, if necessary, implementation of remediation strategies or site management controls to reduce potential radiation doses in accordance with a graded approach. The programme of research discussed in this paper on the Ra-226 particles from Dalgety Bay provides an example of how to take this next step and the same approach could be applied to other existing exposure situations. However, a new definition of radioactive particles is needed to ensure that the management of existing exposure situations involving radioactive particles is relevant to radiological protection and in accordance with a graded approach.

Declaration of competing 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: Dalgety Bay showing length of coastline where Ra-226 particles are found (red line = ~850m)

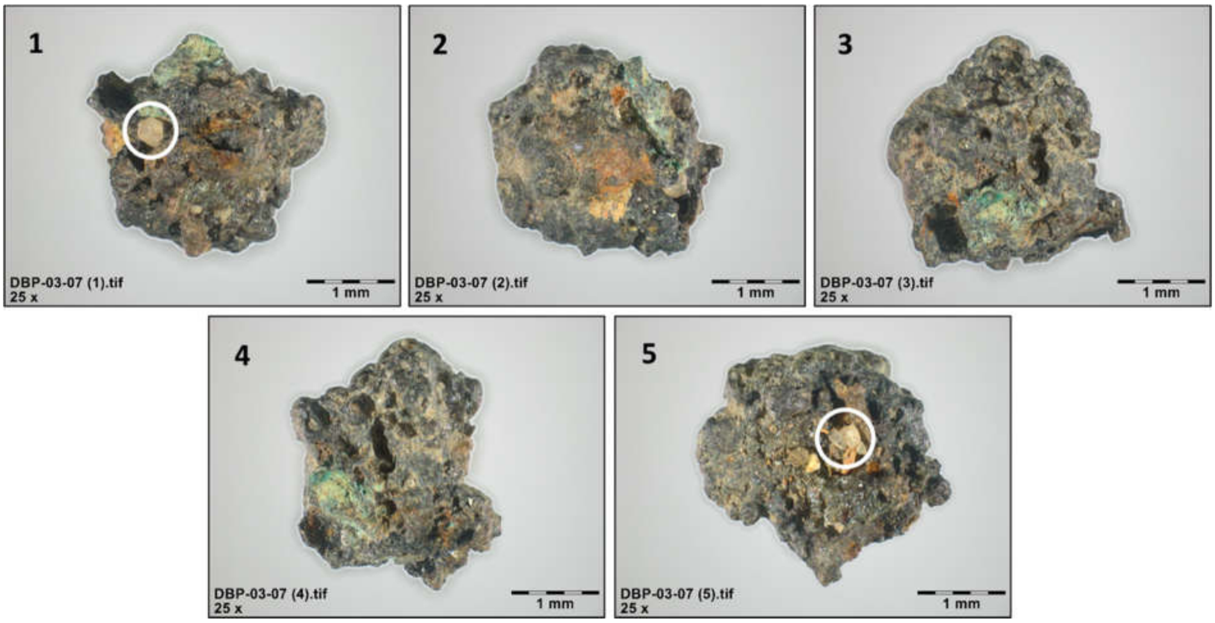

Figure 2: Optical macroscopy images of Dalgety Bay Ra-226 particle, DBP-03-07, in five different orientations (White circles = sand grains) (Scale bar = 1mm)

Table 1: Size measurements for Dalgety Bay Ra-226 particle, DBP-03-07

Particle	Radius (mm)	Diameter (mm)	Equivalent Circular Diameter (mm)	Area (mm²)	Perimeter (mm)
DBP-03-07 (1)	1.189	2.378	2.382	4.457	18.513
DBP-03-07 (2)	1.219	2.438	2.447	4.703	15.41
DBP-03-07 (3)	1.254	2.508	2.519	4.984	17.345
DBP-03-07 (4)	1.199	2.397	2.434	4.653	17.812
DBP-03-07 (5)	1.279	2.558	2.56	5.146	16.755
Mean	1.228	2.456	2.468	4.789	17.167
Standard Deviation	0.038	0.076	0.071	0.275	1.174

Table 2: Shape measurements for Dalgety Bay Ra-226 particle, DBP-03-07

Particle	Shape Factor	Sphericity	Aspect Ratio	Elongation	Convexity
DBP-03-07 (1)	0.163	0.862	1.126	1.077	0.893
DBP-03-07 (2)	0.249	0.741	1.16	1.161	0.927
DBP-03-07 (3)	0.208	0.937	1.1	1.033	0.929
DBP-03-07 (4)	0.184	0.688	1.25	1.206	0.868
DBP-03-07 (5)	0.23	0.771	1.149	1.139	0.92
Mean	0.207	0.8	1.157	1.123	0.908
Standard Deviation	0.034	0.099	0.057	0.069	0.026

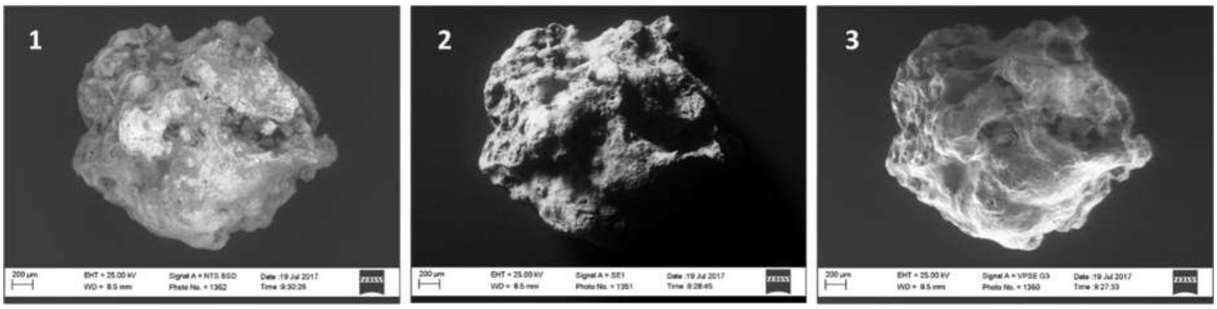

Figure 3: (1) Backscattered Electron (BSE), (2) Secondary Electron (SE) and (3) Variable Pressure Secondary Electron (VPSE) images of Dalgety Bay Ra-226 particle, DBP-03-07

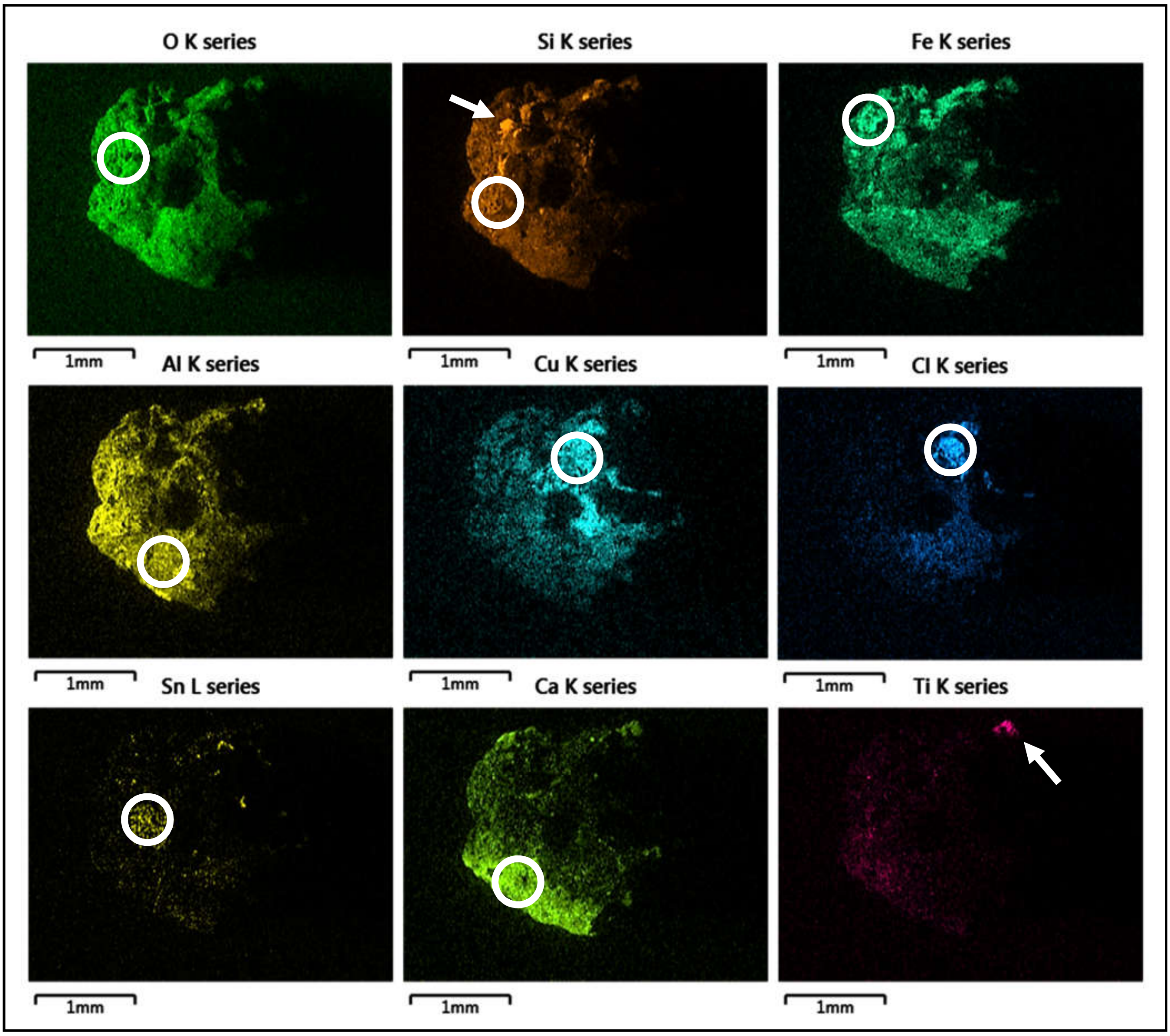

Figure 4: Energy dispersive X-ray spectroscopy (EDS) element distribution maps of Dalgety Bay Ra-226 particle, DBP-03-07 (White circles = phase spectra; Arrows = point spectra)

Table 3: Surface chemical composition (weight %) of Dalgety Bay Ra-226 particle, DBP-03-07 (The highest value for each element is italicised and in bold. If an element is not present in a spectrum it is shown as zero.)

Element	**Whole Particle**	**Al Phase**	**Cu Phase**	**Fe Phase**	**Ca Phase**	**Sn Phase**	**Cl Phase**	**Si Point**	**Ti Point**
O	42.0	49.0	33.7	31.6	51.6	39.6	42.2	57.2	49.6
Fe	12.4	9.1	5.9	***26.9***	9.8	7.8	3.8	3.0	11.5
Si	12.2	18.4	7.2	7.7	13.9	11.1	7.3	***34.8***	8.6
Cu	11.1	2.6	***41.4***	11.3	3.0	4.6	35.2	1.1	3.3
Al	6.9	***7.5***	0.0	6.1	6.9	6.1	0.0	1.9	5.3
Ca	3.1	4.0	1.1	3.6	***9.2***	1.7	0.6	0.4	4.7
Mg	2.6	1.8	6.8	3.9	1.8	1.3	0.0	0.5	1.4
Sn	2.0	0.0	0.0	0.0	0.0	***11.9***	0.0	0.0	1.0
Cl	1.3	0.4	2.2	2.2	0.3	0.2	***9.9***	0.1	0.0
Na	1.2	2.5	0.0	1.4	0.0	1.9	0.0	0.6	0.8
Zn	0.8	0.3	0.8	2.0	0.9	1.0	0.0	0.0	0.3
Pb	0.8	0.0	0.0	1.4	0.8	0.9	0.0	0.0	0.0
K	0.7	1.1	0.3	0.0	0.0	1.0	0.5	0.2	0.4
Ti	0.6	1.0	0.2	0.0	0.7	1.0	0.1	0.2	***10.7***
Ta	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
P	0.5	0.5	0.0	0.4	0.7	0.4	0.0	0.0	2.0
Ni	0.5	0.2	0.5	0.5	0.2	0.2	0.4	0.0	0.1
Mn	0.5	0.7	0.0	0.3	0.0	0.6	0.0	0.0	0.3
S	0.4	0.2	0.0	0.5	0.3	0.0	0.0	0.0	0.0
Sb	0.0	0.0	0.0	0.0	0.0	8.8	0.0	0.0	0.0
Ba	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total	**100**	**100**	**100**	**100**	**100**	**100**	**100**	**100**	**100**