Uranium Aerosol Activity Size Distributions at a Nuclear Fuel Fabrication Plant

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Abstract—Inhalation of uranium aerosols is a concern in nuclear fuel fabrication. Determination of committed effective doses and lung equivalent doses following inhalation intake requires knowledge about aerosol characteristics; e.g., the activity median aerodynamic diameter (AMAD). Cascade impactor sampling of uranium aerosols in the breathing zone of nuclear operators was carried out at a nuclear fuel fabrication plant producing uranium dioxide via ammonium uranyl carbonate. Complementary static sampling was carried out at key process steps. Uranium on impaction substrates was measured using gross alpha counting and alpha spectrometry. Activity size distributions were evaluated for both unimodal and bimodal distributions. When a unimodal distribution was assumed, the average AMAD in the operator breathing zone at the workshops was 12.9–19.3 μm, which is larger than found in previous studies. Certain sampling occasions showed variable isotope ratios ($^{234}$U/$^{238}$U) at different impactor stages, indicating more than one population of particles; i.e., a multimodal activity size distribution. When a bimodal distribution (coarse and fine fraction) was assumed, 75–88% of the activity was associated with an AMAD of 15.2–18.9 μm (coarse fraction). Quantification of the AMAD of the fine fraction was associated with large uncertainties. Values of 1.7–7.1 μm were obtained. Static sampling at key process steps in the workshops showed AMADs of 4.9–17.2 μm, generally lower than obtained by breathing zone sampling, when a unimodal distribution was assumed. When a bimodal distribution was assumed, a smaller fraction of the activity was associated with the coarse fraction compared to breathing zone sampling. This might be due to impactor positioning during sampling and sedimentation of large particles. The average committed effective dose coefficient for breathing zone sampling and a bimodal distribution was 1.6–2.6 μSv Bq$^{-1}$ for $^{234}$U when Type M/S absorption parameters were assumed (5.0 μSv Bq$^{-1}$ for an AMAD of 5 μm). The corresponding lung equivalent dose coefficient was 3.6–10.7 μSv Bq$^{-1}$ (29.9 μSv Bq$^{-1}$ for an AMAD of 5 μm). The predicted urinary excretion level 100 d after inhalation intake was found to be 13-34% of that corresponding to an AMAD of 5 μm. Uranium aerosols generated at a nuclear fuel fabrication plant using ammonium uranyl carbonate route of conversion were associated with larger AMADs compared to previous work, especially when sampling of aerosols was carried out in the operator breathing zone. A bimodal activity size distribution can be used in calculations of committed effective doses and lung equivalent doses, but parameters associated with the fine fraction must be interpreted with care due to large uncertainties.

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INTRODUCTION

In nuclear fuel fabrication, detailed knowledge of the inhalation exposure to uranium aerosols and the corresponding committed effective doses (CED) needs to be gained in order to ensure worker safety and regulatory compliance. Recent work has found evidence for increased lung cancer risk among uranium and plutonium workers (median, mean, and max lung absorbed doses of 2.4, 7.3, and 316 mGy, respectively) following inhalation intake (Grellier et al. 2017). It is thus important to ensure low intake levels and to accurately determine CED following inhalation exposure, which may be challenging due to confounding factors and uncertainties (ICRP 2007; Gilbert 2009; Harrison and Day 2008).

To accurately determine the CED following inhalation intake of uranium aerosols, the deposition of inhaled particles in the airways must be well understood. Particle deposition is commonly modeled using the Human Respiratory Tract Model (HRTM) designed by the International Commission on Radiological Protection (ICRP) (ICRP 1994, 2015). The model assumes a population of particles following a

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log-normal distribution, which can be described by the Activity Median Aerodynamic Diameter (AMAD) and its geometric standard deviation (GSD). The AMAD is the aerodynamic particle size where 50% of the total activity is associated with particles with an aerodynamic diameter greater than the AMAD (\(d_{50}\)). The GSD describes the spread of the activity size distribution. The frequency function is defined (eqn 1) as

\[
dA = \frac{1}{\sqrt{2\pi} \ln(GSD)} \exp \left[ -\frac{(\ln d_{ac} - \ln AMAD)^2}{2(\ln(GSD))^2} \right] d \ln d_{ac},
\]

where \(dA\) is the fraction of radioactivity in the size range between \(\ln d_{ac}\) and \(\ln d_{ac} + d \ln d_{ac}\) where \(d_{ac}\) is the aerodynamic particle diameter (Hinds 1999). The aerodynamic particle diameter of a non-spherical particle is defined as \(d_{ac} = d_e \times \left[ \frac{\rho \times (\rho_0 \chi)^{1/5}}{\rho_0} \right] \), where \(d_e\) is the diameter of a spherical particle with the same volume as the particle considered, \(\rho\) (g cm\(^{-3}\)) is the density of the non-spherical particle, \(\rho_0\) is the reference density (1 g cm\(^{-3}\)), and \(\chi\) is the dynamic shape factor (dimensionless, typically assumed to be 1.5) (Hinds 1999; ICRP 1994, 1997).

A GSD value of 1 describes a monodispersed aerosol, and a large GSD indicates a large spread in the activity size distribution. The equation is applicable for log-normal distributions only, which is typically assumed in the field of radiation protection. In the absence of detailed information, the ICRP recommends a default value of AMAD = 5 \(\mu\)m and GSD = 2.5 (ICRP 1994). These parameters indicate that 95% of the activity is associated with particles in the aerodynamic size range 0.8 \(\mu\)m (5 \(\mu\)m \times 2.5\(^{-2}\) = 0.8 \(\mu\)m) and 31 \(\mu\)m (5 \(\mu\)m \times 2.5\(^2\) \approx 31 \(\mu\)m) (Hinds 1999). The activity size distribution of aerosols is important for modeling deposition in the airways using the HRTM model; e.g., the alveolar deposition is expected to be 3% of the total inhaled activity if the AMAD is 15 \(\mu\)m, whereas the corresponding figure for an AMAD of 5 \(\mu\)m is 10%, thus increasing the lung equivalent dose (\(H_{eq}\)) (ICRP 2015).

The AMAD and GSD can be determined experimentally by sampling aerosols using cascade impactors. A common evaluation method is to plot the accumulated fraction of activity as a function of impactor cut-point in a log-probability graph. This method may give too much weight to early and late impaction stages (Hinds 1999). Alternatively, a nonlinear regression fit to the frequency function (eqn 1) can be applied to determine the AMAD and GSD (Thiel 2002).

The aerosol size distribution will affect the deposition pattern in the respiratory tract and, as a consequence, the biological fate of inhaled radioactive aerosols. Thus, the rate of excretion (e.g., via urine) and CED per inhaled unit of activity will vary with particle size (AMAD and GSD) in addition to the chemical composition/solubility class affecting particle dissolution rate in the respiratory tract.

Early studies have reported cascade impactor measurements of uranium aerosol AMADs in nuclear fuel fabrication. Schieferdecker et al. reported an AMAD of 8.2 \(\mu\)m (Schieferdecker et al. 1985). Thind reported an AMAD of 6.1 \(\mu\)m and GSD of 2.1 (Thind 1986). A more recent review paper stated an average AMAD of 6.2 \(\mu\)m with a GSD of 2.3 (Davesne and Blanchardon 2014). An earlier review paper stated that results below 1 \(\mu\)m were inconsistent, suggesting a bimodal size distribution (Dorrian and Bailey 1995).

Available data are based on static sampling and might not be representative of the breathing zone of operators (Davesne and Blanchardon 2014). To the best of our knowledge, AMADs determined from cascade impactor sampling in the breathing zone of workers in nuclear fuel fabrication have not been published by open sources.

Uranium dioxide (UO\(_2\)) powder for nuclear fuel and light-water reactors is typically produced from uranium hexafluoride (UF\(_6\)) using integrated dry route (IDR) conversion or wet-route conversion via ammonium diuranate (ADU), alternatively via ammonium uranyl carbonate (AUC) (IAEA 2006). These production methods yield UO\(_2\) powder with different properties; e.g., the AUC route of conversion produces a UO\(_2\) powder consisting of coarser particles compared to the other methods (IAEA 2006). From a radiation protection perspective, process history is important since it affects the physicochemical properties of aerosols associated with a certain compound (Ansoborlo et al. 1999). From published data on uranium aerosols in nuclear fuel fabrication, it can be difficult to distinguish which production method was used. To the best of our knowledge, only one study has covered activity size distributions at a site using wet-route conversion via AUC and reported an average AMAD of 8.2 \(\mu\)m (Schieferdecker et al. 1985).

The aim of the present work is to evaluate uranium aerosol activity size distributions (corresponding AMADs and GSDs) in the breathing zone of operators at a nuclear fuel fabrication plant using wet-route conversion via AUC. Portable cascade impactors were used to sample uranium aerosols in the operator breathing zone. In addition, static sampling was carried out at certain process steps. Activity size distributions were evaluated by assuming unimodal as well as bimodal distributions, and the impact on dosimetry calculations was evaluated based on the obtained results. The information in the present work is important in order to improve internal dosimetry, worker safety, and ensure regulatory compliance. Improved internal dosimetry might add to the overall knowledge about health effects associated with low-level exposure to ionizing radiation.

**MATERIALS AND METHODS**

**Impactor sampling**

Cascade impactors (Marple 298, Prod. No. SE298; Thermo Fisher Scientific, Waltham, MA), which operate
at 2.0 L min\(^{-1}\) were used for aerosol sampling. The impactor used in the present work has eight impaction stages (A–H) with corresponding cut-points of 21.3, 14.8, 9.8, 6.0, 3.5, 1.6, 0.9, and 0.5 \(\mu m\), respectively. The cut-point is defined as the aerodynamic diameter of particles having 50% probability of impaction at the given stage. The last stage is followed by a final filter to collect remaining particles. Gilian 5000 sampling pumps (Sensidyne, St. Petersburg, FL) were used with the impactors. The pumps were calibrated and flowrates checked using the methodology described in previous work, with flow rates showing variations typically <1% (Hansson et al. 2017). The small size of the impactor allows for sampling in the breathing zone, which was carried out by personnel carrying the impactor attached to the collar of the overall throughout the work day. Static sampling was carried out by positioning the impactor as close as practically possible to work stations of particular interest.

Mixed cellulose ester membrane (MCE) (Thermo Scientific, SEC-290-MCE) was used as impaction substrate, and polyvinyl chloride (PVC) (SEF-290-P5) with a 5 \(\mu m\) pore size was used as final collection filter unless otherwise stated. Other authors have used an identical setup (Cheng et al. 2009). In the present work, impactor substrates were not coated to prevent particle bounce due to difficulties in reproducing application of thin, even layers of coating material. Furthermore, coating of substrates would distort comparison with planned in vitro dissolution rate experiments using the same sampling methodology. However, an attempt to investigate the presence of particle bounce was carried out by parallel sampling using cascade impactors with coated and uncoated substrates. The coating material (PRF Industrial Line Silicon Oil, Taerosol Oy, Kangasala, Finland) was sprayed onto each MCE substrate in a sweeping motion for 1-2 s at a distance of approximately 15 cm. Parallel sampling was carried out twice (three impactors per occasion) at a work station where scrap pellets are oxidized to triuranium octoxide (\(U_3O_8\)). Impaction stages G and H, as well as the final collection filters, were examined using scanning electron microscopy with energy-dispersive x-ray spectrometry (Phenom ProX, Phenom-World BV, Eindhoven, The Netherlands).

Sampling was carried out in the operator breathing zone at the four major workshops at the site: conversion, powder preparation, pelletizing, and burnable absorber (BA) pelletizing. At the conversion workshop, \(UO_2\) is formed from UF\(_6\) via AUC. At the powder preparation workshop, powder milling, powder blending, and oxidizing of waste materials is carried out. The pelletizing workshop produces \(UO_2\) pellets by pressing, sintering, and grinding. The BA pelletizing workshop is similar but uses milled \(UO_2\) powder, which is blended with \(Gd_2O_3\). An overview of the main uranium flows is presented in Fig. 1. A more detailed description of the processes and typical operations has been described in previous work (Hansson et al. 2017).

Uranium aerosol activity size distributions

Uranium material at the site is handled in batches, where the enrichment levels (mass-percent \(^{235}U\)) vary between 0.71% and 4.95% (average enrichment level 3.8%). The activity ratio \(^{234}U/^{238}U\) increases approximately linearly with enrichment level. As a consequence, enrichment levels at the workshops may vary from day to day.

Sampling of the breathing zone was carried out on 18 occasions (five occasions at each workshop except for the BA pelletizing workshop, where only three occasions could be completed due to production-related circumstances). In addition to the breathing zone sampling, static sampling was carried out at 13 key locations at the different workshops:

- **Conversion workshop**: Static sampling at the transportation of AUC to the fluidizing bed furnaces and of the general workshop air;
- **Powder preparation workshop**: Static sampling at the stations for powder milling station, oxidizing of discarded pellets, oxidizing of waste from pellet grinding, humidity check (for criticality safety reasons) of \(UO_2\) powder and of the general workshop air;
- **Pelletizing workshop**: Static sampling at one of the pellet pressing stations, emptying of sintered pellets and of the general workshop air; and
- **BA pelletizing workshop**: Static sampling at the stations for \(UO_2/Gd_2O_3\) blending and oxidizing of waste from pellet grinding.

Sampling times were based on previous knowledge of airborne activity concentration levels and determined so as to collect sufficient amounts of radioactivity for reliable measurements but avoiding particle overload on the impaction substrates.

**Radioactivity measurements**

The total amount of alpha activity at all impactor substrates and final collection filters were measured for 20–24 h using an LB 790 10-Channel \(\alpha-\beta\) Low-Level Counter.
Table 1. Uranium activity concentrations and activity ratios (breathing zone sampling) based on radiometric data following alpha spectrometry. Uranium activity corresponds to the sum of $^{234}$U, $^{235}$U, and $^{238}$U activity. Uncertainties correspond to ± 1 standard deviation due to counting statistics and flow rate measurements (2.00 ± 0.05 L min$^{-1}$).

| Sampling-ID | Uranium activity concentration (Bq m$^{-3}$) and Activity ratio $^{234}$U/$^{238}$U | Total concentration (Bq m$^{-3}$) |
|-------------|-------------------------------------------------|-------------------------------|
| Conversion workshop | | |
| Sampling 1 | 1.29 (0.04) | 0.70 (0.2) |
| Sampling 2 | 0.132 (0.007) | 5.9 (0.3) |
| Sampling 3 | 0.192 (0.008) | 5.9 (0.3) |
| Sampling 4 | 0.46 (0.02) | 6.2 (0.2) |
| Sampling 5 | 1.17 (0.04) | 6.3 (0.1) |
| Powder preparation workshop | | |
| Sampling 1 | 0.57 (0.02) | 6.6 (0.2) |
| Sampling 2 | 0.76 (0.02) | 5.6 (0.2) |
| Sampling 3 | 0.35 (0.02) | 5.3 (0.3) |
| Sampling 4 | 2.25 (0.07) | 6.3 (0.3) |
| Sampling 5 | 1.34 (0.04) | 6.6 (0.2) |
| Pelletizing workshop | | |
| Sampling 1 | 0.20 (0.001) | 6.1 (0.3) |
| Sampling 2 | 0.138 (0.004) | 5.8 (0.1) |
| Sampling 3 | 0.130 (0.004) | 3.9 (0.1) |
| Sampling 4 | 0.243 (0.007) | 6.2 (0.2) |
| Sampling 5 | 0.090 (0.003) | 7.1 (0.3) |

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Background levels were corrected for and frequently evaluated. The breathing zone samples were also analyzed using alpha spectrometry in order to obtain uranium isotopic data. Samples were acid dissolved in concentrated HNO₃ followed by aqua regia after addition of traceable amounts of ²³²U yield determinant (Isotrak, AEA Technology, PLC, Didcot, UK). Uranium was then extracted from 8 M HNO₃ by tributyl phosphate (TBP) and back-extracted into demineralized water (Holm 1984) and finally electro-deposited onto stainless steel planchets (Hallstadius 1984). Measurements were carried out using passivated implanted planar silicon detectors (Canberra PIPS, Mirion Technologies Inc, San Ramon, CA), ORTEC (Oak Ridge, TN) Octètè R MCA system, and ORTEC Maestro R software. Counting times varied between 1–4 d.

The activity ratio from measurements by alpha spectrometry compared to total alpha activity followed a normal distribution with a mean of 1.01 and a standard deviation of 0.21. Two data points with very low activities deviated from the mean with more than 3 standard deviations and were considered outliers, resulting in a mean ratio of 0.99 with a standard deviation of 0.13. The agreement was considered satisfactory, and thus the remaining samples (static sampling) were not analyzed using alpha spectrometry.

Data reduction

Activity size distributions were evaluated using a unimodal approach (eqn 1). However, alpha spectrometry measurements revealed variable isotope ratios (²³⁴U/²³⁸U) during some sampling occasions, suggesting a multi-modal activity size distribution. Thus a bimodal activity size distribution was evaluated for each sampling occasion (eqn 2). A data reduction protocol developed by O’Shaughnessy and Raabe for sampling carried out with the Marple 298 impactor was used to evaluate activity size distributions (O’Shaughnessy and Raabe 2003). The protocol, which assumes a unimodal log-normal distribution (eqn 1), was modified as to model a bimodal activity size distribution:

\[ dA = \begin{cases} 
\frac{f}{\sqrt{2\pi(\ln(GSD1))}} \exp \left[ -\frac{(\ln d_{w1} - \ln AMAD1)^2}{2(\ln(GSD1))^2} \right] \\
\frac{1-f}{\sqrt{2\pi(\ln(GSD2))}} \exp \left[ -\frac{(\ln d_{w2} - \ln AMAD2)^2}{2(\ln(GSD2))^2} \right] 
\end{cases} \cdot \ln d_{w}, \quad (2) \]

where \( f \) and \( 1-f \) is the fraction of activity in the coarse and fine fraction, respectively. The same approach has been used by other authors investigating bimodal activity size distributions (Cheng et al. 2009).

The measured amount of radioactivity at each impactor substrate was corrected for sampling efficiency by applying a stage impaction efficiency of 0.52, 0.61, 0.78, 0.89, 0.95, 0.96, 0.97, and 0.99 for impactor stages A-H,

| Sampling | f | 1-f | f/0.52 | 1-f/0.52 |
|----------|---|-----|--------|---------|
| Sampling 1 | 0.025 (0.003) | 0.033 (0.002) | 0.066 (0.004) | 0.047 (0.003) |
| Sampling 2 | 0.015 (0.001) | 0.033 (0.002) | 0.046 (0.003) | 0.029 (0.002) |
| Sampling 3 | 0.010 (0.001) | 0.023 (0.002) | 0.020 (0.002) | 0.017 (0.001) |
respectively, as recommended by the manufacturer (Thermo Fisher 2009). Lower and upper bounds considered in the data reduction were 0.1 μm and 50 μm, respectively, as in previous studies (Cheng et al. 2009).

The AMADs, GSDs, and $f$ for each sampling occasion were evaluated by applying a nonlinear least squares regression fit (Microsoft Excel 2010 Problem Solver) to the frequency function (eqn 1 and 2, respectively). Simulations were repeated in R (version 3.5.1., 2018-07-02) and included standard errors of the derived parameters.

Internal dosimetry

Dose coefficients (CED per unit of inhalation intake) and predicted urinary excretion following inhalation of 1 Bq of $^{234}$U were evaluated. The software Integrated Modules for Bioassay Analysis (IMBA) Professional Plus (v. 4.1) was used (Birchall et al. 2007).

The HRTM has recently been revised by the ICRP; thus, new recommendations are available. For example, the particle transport model has been revised (ICRP 2015). Default absorption parameters for UO$_2$ and U$_3$O$_8$ are now considered intermediate Type M/S (rapid fraction, $f_r = 0.03$; rapid rate, $s_r = 1 \text{ d}^{-1}$; slow rate $s_s = 5 \times 10^{-3} \text{ d}^{-1}$) and fractional absorption in the alimentary tract, $f_A = 6 \times 10^{-6}$). Uranyl nitrate [UO$_2$(NO$_3$)$_2$], uranium peroxide hydrate (UO$_4$), ADU [(NH$_4$)$_2$U$_2$O$_7$] and uranium trioxide (UO$_3$) are now considered intermediate Type F/M ($f_r = 0.8$; $s_r = 1 \text{ d}^{-1}$; $s_s = 0.01 \text{ d}^{-1}$, and $f_A = 0.016$) (ICRP 2017). AUC [(NH$_4$)$_4$UO$_2$(CO$_3$)$_3$] is not included in ICRP Publication 137.

An add-on to IMBA has previously been used to model urinary excretion according to the ICRP 130 recommendations (Birchall et al. 2017). This add-on was acquired and used in the present work to model the urinary excretion following inhalation of 1 Bq of $^{234}$U according to the ICRP 130 particle transport model. Activity size distributions derived in the present work were used in combination with default absorption parameters, aerosol density (3 g cm$^{-3}$), and shape factor (1.5) (ICRP 1994, 2015). For the conversion workshop Type F/M and Type M/S materials were considered. For remaining workshops, only Type M/S material was considered.

To the best of our knowledge, as of today, no software exists that allows for calculation of dose coefficients according to the ICRP 130 particle transport recommendations. Thus, dose coefficients were evaluated using the ICRP 66 particle transport model but with the revised ICRP 137 absorption parameters.

RESULTS AND DISCUSSION

Radioactivity measurements and activity size distributions

Radiometric data following alpha spectrometry of breathing zone samples are presented in Table 1, and radiometric data following alpha counting of static sampling are presented
It is evident from Table 1 that isotope ratios ($^{234}\text{U}/^{238}\text{U}$) might vary across impactor stages during a single round of sampling (e.g., sampling 5 at the conversion workshop, sampling 1 at the powder preparation workshop, sampling 3 at the pelletizing workshop). This implies that at least two populations of particles with different size distributions and enrichment levels (i.e., originating from different batches of uranium material) might be present during a sampling occasion. Thus, a bimodal approach might be preferable to describe the data. Many sampling occasions showed uniform isotope ratios (e.g., sampling 1 at the pelletizing workshop). This does not contradict a bimodal distribution, as different process steps might be associated with different size distributions but identical enrichment levels (depending on material batch). It is also noteworthy from Table 1 that the concentration of sampled activity varies with up to an order of magnitude. This implies that day-to-day variations in exposure might be large, presumably due to variations in production and work tasks carried out.

Data from Table 1 are presented as fractions in Fig. 2 for easy comparison between the different workshops. Activity sampled at the conversion workshop was collected at the early impaction stages to a greater extent compared to the other workshops.

### Activity size distributions

Each sampling occasion was evaluated by assuming both a unimodal and a bimodal activity size distribution. Results are presented in Table 2 (breathing zone sampling) and Table 3 (static sampling). The average parameters (Table 2) for each workshop were used to visually compare activity size distributions (Fig. 3).

From Tables 2 and 3 and Fig. 3, it is evident that a typical AMAD at the site is larger than the ICRP default distribution (AMAD = 5 μm, GSD = 2.5). Sampling in the breathing zone of the operators tended to generate more predominant coarse fractions (larger $f$) compared to static sampling. Static sampling can be difficult to carry out in a way

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### Table 2. Breathing zone sampling—estimated AMAD, GSD and $f$ for coarse and fine fractions assuming unimodal and bimodal activity size distributions. Numbers in brackets indicate the standard error from the curve-fitting procedure unless stated otherwise.

| Sampling-ID | Unimodal fit | Coarse fraction | Fine fraction | Bimodal fit |
|-------------|--------------|-----------------|---------------|-------------|
| Conversion workshop | | | | |
| Sampling 1 | 17.7 (0.9) | 1.8 (0.07) | – | – |
| Sampling 2 | 17.0 (1.6) | 2.3 (0.2) | – | – |
| Sampling 3 | 19.1 (0.6) | 1.8 (0.05) | 0.90 (0.23) | 20.1 (0.5) |
| Sampling 4 | 16.9 (0.3) | 1.7 (0.02) | 0.85 (1.12) | 17.7 (1.1) |
| Sampling 5 | 22.0 (0.4) | 1.5 (0.03) | – | – |
| Average (1 SD) | 19.3 (1.8) | 1.6 (0.2) | 0.88 (0.03) | 18.9 (1.2) |
| Powder preparation workshop | | | | |
| Sampling 1 | 15.2 (0.6) | 1.8 (0.05) | 0.77 (0.52) | 16.1 (0.5) |
| Sampling 2 | 12.0 (1.6) | 2.3 (0.2) | – | – |
| Sampling 3 | 14.7 (0.6) | 1.8 (0.05) | 0.73 (0.51) | 15.7 (0.6) |
| Sampling 4 | 6.5 (1.0) | 2.9 (0.4) | 0.56 (0.06) | 13.2 (1.1) |
| Sampling 5 | 16.1 (1.4) | 2.1 (0.1) | 0.94 (0.09) | 15.9 (1.6) |
| Average (1 SD) | 12.9 (3.5) | 2.2 (0.4) | 0.75 (0.13) | 15.2 (1.2) |
| Pelletizing workshop | | | | |
| Sampling 1 | 16.4 (0.4) | 1.9 (0.04) | 0.90 (0.12) | 16.5 (0.3) |
| Sampling 2 | 16.5 (0.8) | 1.7 (0.06) | 0.76 (0.19) | 17.0 (0.5) |
| Sampling 3 | 16.8 (0.8) | 2.1 (0.07) | 0.90 (0.08) | 17.0 (0.9) |
| Sampling 4 | 17.5 (0.4) | 1.9 (0.04) | 0.97 (0.04) | 17.5 (0.5) |
| Sampling 5 | 14.2 (0.7) | 1.8 (0.06) | 0.74 (0.10) | 14.1 (0.4) |
| Average (1 SD) | 16.3 (1.1) | 1.9 (0.1) | 0.85 (0.09) | 16.4 (1.2) |
| BA pelletizing workshop | | | | |
| Sampling 1 | 15.1 (0.8) | 2.1 (0.08) | 0.89 (0.09) | 15.0 (0.8) |
| Sampling 2 | 15.2 (0.6) | 1.8 (0.05) | 0.84 (0.12) | 15.3 (0.4) |
| Sampling 3 | 14.0 (0.9) | 2.2 (0.1) | – | – |
| Average (1 SD) | 14.8 (0.5) | 2.0 (0.2) | 0.87 (0.02) | 15.2 (0.1) |

*No solution for bimodal activity size distribution, suggesting weak fine fraction or unimodal distribution.

*Fine and coarse fractions indistinguishable. Derived parameters not included in average parameter values.

*Peaks for fine and coarse fraction well separated (visual inspection of fit).
that is representative of worker exposure due to positioning and the fact that operators carry out work at multiple positions during a work period. The distance to the source of particle dispersion is likely important since large particles settle more rapidly than small particles (Hinds 1999). Our results suggest that static sampling might underestimate the amount of activity present in the breathing zone and that the derived AMAD might be underestimated.

A bimodal activity size distribution appears to give a better curve fit compared to a unimodal fit (Fig. 3). This is in accordance with the observed variable isotope ratios for breathing zone sampling (Table 1). However, the AMAD and GSD for the fine fraction were difficult to quantify and were frequently not statistically significant (Tables 2 and 3). This was particularly obvious for the conversion workshop, where the fine fraction appeared to be very indistinct in comparison to the coarse fraction.

One could consider weighting the measurement points in the curve-fitting process (Kemmer and Keller 2010). An attempt was made using relative weighting for each of the samplings carried out at the pelletizing workshop. For the fine fraction, the mean AMAD increased from 3.2 μm to 5.0 μm and the mean GSD from 3.5 to 9.1, compared to the unweighted curve-fitting procedure. That large a GSD indicates a very broad peak. We believe that this is a case of overfitting data to the model, which might be misleading, in particular if the late impactor stages are affected by particle bounce. The effect on the coarse fraction was less prominent.

The derived AMADs in the present work are generally considerably higher than previously published data (Davesne and Blanchardon 2014). This might be due to aerosol characteristics associated with the particular production method. The wet-chemical AUC conversion process is known to generate a coarser UO2 powder compared to wet-chemical ADU conversion (Bergqvist and Sahle 2010; Palheiros et al. 2009). The sampling method (i.e., breathing zone vs. static sampling) can also play an important role.

### Dose coefficients

The calculated dose coefficients (CED per unit of 234U inhalation intake) for breathing zone sampling at the four workshops are presented in Table 4. The dose coefficient associated with the ICRP default assumption (AMAD = 5 μm, GSD = 2.5) is included for comparison. Potential exposure at the workshops is associated with CED and Hlung coefficients being 21–51% and 12–29%, respectively, of those obtained for the default 5 μm and Type M/S assumption. Lower dose coefficients are expected since a high AMAD results in a greater fraction of activity depositing in the upper respiratory tract, resulting in a lower dose to the lung.
Urinary excretion

The resulting urinary excretion rate following inhalation intake of 1 Bq \(^{234}\)U is presented in Fig. 4. The urinary excretion rate associated with the ICRP default assumption (AMAD = 5 \(\mu\)m, GSD = 2.5) is included for comparison.

The expected urinary excretion per unit of inhalation intake 100 d after exposure was found to be 13–34% of that corresponding to the default assumption of AMAD 5 \(\mu\)m and Type M/S material. Lower excretion rates are expected since a high AMAD results in a smaller fraction of activity reaching the alveoli and thus the blood.

Uncertainties

Although variable isotope ratios (Table 1) are a strong indicator of bimodal/multimodal activity size distributions, the parameters associated with the fine fractions proved

Table 4. Dose coefficients (CED and \(H_{lung}\)) at the different workshops, following inhalation intake of \(^{234}\)U. ICRP default AMAD (5 \(\mu\)m) for both solubility classes (Type F/M and Type M/S) are included for comparison. Doses are based on an alpha radiation weighting factor of 20 (ICRP 2007). Absorption parameters from ICRP 137 were used.

| Workshop | Coarse fraction | Fine fraction | Weighted dose coefficient |
|----------|----------------|---------------|---------------------------|
|          | f | CED (\(\mu\)Sv Bq\(^{-1}\)) | \(H_{lung}\) (\(\mu\)Sv Bq\(^{-1}\)) | 1-f | CED (\(\mu\)Sv Bq\(^{-1}\)) | \(H_{lung}\) (\(\mu\)Sv Bq\(^{-1}\)) | CED (\(\mu\)Sv Bq\(^{-1}\)) | \(H_{lung}\) (\(\mu\)Sv Bq\(^{-1}\)) |
| Conversion (Type F/M) | 0.88 | 0.1 | 0.2 | 0.12 | 0.4 | 2.7 | 0.1 | 0.5 |
| Conversion (Type M/S) | 0.88 | 1.3 | 1.1 | 0.12 | 4.0 | 21.9 | 1.6 | 3.6 |
| Powder preparation (Type M/S) | 0.75 | 1.6 | 3.0 | 0.25 | 5.5 | 33.6 | 2.6 | 10.7 |
| Pelletizing (Type M/S) | 0.85 | 1.5 | 2.4 | 0.15 | 5.5 | 36.0 | 2.1 | 7.5 |
| BA pelletizing (Type M/S) | 0.87 | 1.6 | 3.0 | 0.13 | 5.6 | 46.0 | 2.1 | 8.6 |
| ICRP 5 \(\mu\)m, Type F/M | – | – | – | – | – | – | – | – |
| ICRP 5 \(\mu\)m, Type M/S | – | – | – | – | – | – | – | – |

\(^{1}\)Calculated by using ICRP Publication 137 absorption data in the ICRP 66 particle transport model. Dose coefficient presented in the ICRP 137 publication is 5.5 \(\mu\)Sv Bq\(^{-1}\).
difficult to quantify as indicated by large uncertainties (Tables 2 and 3). Dose coefficients and excretion patterns are based on these derived parameters and must thus be interpreted with care.

Total analytical uncertainties due to counting statistics, detector efficiency (gross alpha counting only), tracer uncertainty, etc., were estimated to be <10%. The analytical uncertainties were considered lower than sampling-related uncertainties, including sample representativeness, particle bounce/roll-off, and impactor stage collection efficiency. Particle bounce/roll-off can be reduced by coating impaction substrates; e.g., by applying a thin layer of silicone oil. It is important to consider coating material, thickness, and homogeneity. To improve reproducibility, simplify sampling, and allow for easy comparison with planned in vitro dissolution rate studies, coating of the impaction substrates was not carried out in the present work except for a comparative measurement (Appendix B). Previous studies using the same type of impactor have concluded that some roll-off can be expected at impaction stages G and H (Rubow et al. 1987). Cheng et al. used the same type of impactor with coating applied on Stage A only. Stages G and H were uncoated, but the authors reported that bounce did not appear to be significant based on deposition patterns (Cheng et al. 2009). Another study concluded that measurements with uncoated impaction substrates tended to generate lower AMADs, although the difference was not statistically significant (Kirychuk et al. 2009). In the present work, the effect of coated and uncoated substrates was evaluated by parallel rounds of stationary sampling (Appendix B, Table B1 and B2, Fig. B1). Coating of substrates generated a somewhat lower fraction of activity on late impactor stages, in particular stages H and G, as well as the final collection filter. This might indicate that some sampling artefact, e.g., particle bounce, occurs. The presence of particles larger than expected compared to impactor stage cut-point (stages G-H and final collection filter) was verified using scanning electron microscopy for both coated and uncoated rounds of sampling. This might introduce a bias toward higher 1-f and GSD, and lower AMAD, for the fine fraction. However, modeled AMADs and GSDs remained similar for coated and uncoated rounds of sampling, regardless of whether a unimodal or bimodal approach was used (Fig. B1). We consider the effect of coating difficult to quantify due to variations between different rounds of samplings and uncertainties introduced in the coating procedure. It should be mentioned that particle bounce/roll-off is most pronounced at high particle loads (Fujitani et al. 2006). Particle loads in the present work were lower (< 0.1 mg assuming a specific activity of approximately 90 Bq mg$^{-1}$) compared to other studies (approximately 0.2–0.9 mg) (Cheng et al. 2009).

Fig. 4. Predicted urinary excretion rate following inhalation of 1 Bq$^{234}U$ based on derived AMADs and GSDs at the different workshops. The ICRP 130 particle transport and ICRP 137 absorption parameters for Type F/M and Type M/S were used. Type F/M was considered for the conversion workshop only.
Impactor stage collection efficiencies were determined by Rubow et al. (1987) using monodisperse aerosols. Early impaction stages are associated with lower collection efficiencies; thus, the impact on the overall uncertainty is greater when a large fraction of the activity is collected on early impaction stages. In the present work, at least 50% of the measured activity was typically collected at impaction stages A–C (collection efficiencies of 0.52, 0.61, and 0.78, respectively). The impact on derived AMADs, GSDs, and $f$ for the pelletizing workshop was tested by modifying the collection efficiencies in the data reduction protocol with ± 20% for stages A–C. The impact on the derived $f$, AMAD, and GSD for the coarse fraction was less than 5%. The impact on the AMAD and GSD for the fine fraction was within uncertainties presented in Table 2.

CONCLUSION

In the present work, cascade impactors were used to evaluate activity size distributions at a nuclear fuel fabrication plant using wet-route AUC conversion. Sampling was carried out in the operator breathing zone at the four main workshops and at certain process steps. Variable $^{234}\text{U}/^{238}\text{U}$ isotope ratios indicated a multimodal rather than unimodal activity size distribution for breathing zone sampling. A bi-modal distribution (coarse and fine fraction) was assumed. Most activity (75–88%) was associated with the coarse fraction (AMAD 15.2–18.9 μm). The AMAD of the fine fraction was 1.7–7.1 μm, but uncertainties were substantial. When static sampling was carried out, the coarse fraction consisted of a smaller fraction of the activity, and the AMAD was lower.

We conclude that although the parameters associated with the fine fractions were difficult to quantify, the presence of a fine fraction is nonetheless very important to consider in CED assessments. The predicted CED per unit of inhalation intake ($^{234}\text{U}$) at the four main workshops was estimated to 1.6–2.6 μSv Bq$^{-1}$ for the Type M/S solubility class, compared to 5.0 μSv Bq$^{-1}$ when assuming an AMAD of 5 μm. The predicted urinary excretion of $^{234}\text{U}$ per unit of inhaled activity at the four workshops at 100 d after intake was 13–34% of the excretion rate when an AMAD of 5 μm was assumed.

The findings in the present work allow for more realistic assumptions regarding activity size distributions, improving CED evaluations at the site. It also shows that internal dosimetry preferably should be based on breathing zone data rather than static sampling. Improved dosimetry can aid in radiation protection optimization and in future epidemiological studies. Future work includes determination of absorption parameters and fractional uptake to the alimentary tract.

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Table A1. Uranium activity concentrations (static sampling) based on gross alpha counting. Uncertainties correspond to ± 1 standard deviation due to counting statistics and flow rate measurements (2.00 ± 0.05 L min\(^{-1}\)).

| Stage | Uranium activity concentration (Bq m\(^{-3}\)) | Sampling time (min) | Total concentration (Bq m\(^{-3}\)) |
|-------|---------------------------------------------|---------------------|-----------------------------------|
| A     | 21.3 µm                                     |                      |                                   |
| B     | 14.8 µm                                     |                      |                                   |
| C     | 9.8 µm                                      |                      |                                   |
| D     | 6.0 µm                                      |                      |                                   |
| E     | 3.5 µm                                      |                      |                                   |
| F     | 1.6 µm                                      |                      |                                   |
| G     | 0.9 µm                                      |                      |                                   |
| H     | 0.5 µm                                      |                      |                                   |
| Final filter |                                    |                      |                                   |

| Sample Location                           | Uranium activity concentration (Bq m\(^{-3}\)) | Sampling time (min) | Total concentration (Bq m\(^{-3}\)) |
|-------------------------------------------|---------------------------------------------|---------------------|-----------------------------------|
| Conversion workshop                        |                                             |                     |                                   |
| Fluidizing bed furnace                    | 0.0013 (0.0002) 0.0035 (0.0003) 0.0062 (0.0004) 0.0052 (0.0004) 0.0039 (0.0003) 0.0041 (0.0002) 0.0011 (0.0001) 0.0004 (0.0001) 0.0047 (0.0003) | 2682                | 0.03 (0.01)                        |
| General ventilation exhaust                | 0.0008 (0.0002) 0.0013 (0.0003) 0.0030 (0.0002) 0.0030 (0.0004) 0.0018 (0.0002) 0.0015 (0.0003) 0.0003 (0.0002) 0.0003 (0.0003) 0.0015 (0.0003) | 1983                | 0.01 (0.02)                        |
| Powder preparation workshop                |                                             |                     |                                   |
| Powder milling                            | 0.034 (0.003) 0.051 (0.004) 0.133 (0.008) 0.16 (0.01) 0.136 (0.008) 0.30 (0.02) 0.17 (0.01) 0.0404 (0.0003) 0.034 (0.003) 0.396 1.0 (0.2) |                     |                                   |
| Humidity check                            | 0.52 (0.03) 0.39 (0.02) 0.71 (0.04) 0.66 (0.04) 0.50 (0.03) 0.55 (0.03) 0.118 (0.007) 0.075 (0.004) 0.122 (0.007) 1382 3.6 (0.6) |                     |                                   |
| Oxidizing of discarded pellets             | 0.23 (0.03) 0.35 (0.01) 0.84 (0.04) 0.68 (0.03) 0.21 (0.04) 0.09 (0.03) 0.07 (0.02) 0.05 (0.02) 0.11 (0.02) 1335 2.64 (0.09) |                     |                                   |
| Oxidizing of grinding waste                | 0.141 (0.008) 0.22 (0.01) 0.40 (0.02) 0.46 (0.03) 0.40 (0.02) 0.42 (0.02) 0.118 (0.007) 0.053 (0.003) 0.037 (0.002) 942 2.3 (0.4) |                     |                                   |
| General workshop air                       | 0.042 (0.003) 0.066 (0.004) 0.075 (0.004) 0.059 (0.003) 0.028 (0.002) 0.018 (0.001) 0.0072 (0.0005) 0.0056 (0.0004) 0.0052 (0.0004) 1255 0.30 (0.06) |                     |                                   |
| Pelletizing workshop                       |                                             |                     |                                   |
| Pellet press                               | 0.081 (0.005) 0.128 (0.007) 0.25 (0.01) 0.30 (0.02) 0.19 (0.01) 0.099 (0.006) 0.026 (0.002) 0.0158 (0.001) 0.17 (0.01) 1425 1.2 (0.2) |                     |                                   |
| Emptying of sintered pellets               | 0.30 (0.02) 0.47 (0.03) 0.83 (0.05) 1.54 (0.09) 1.20 (0.07) 0.66 (0.04) 0.070 (0.004) 0.026 (0.002) 0.126 (0.007) 5596 5.2 (0.9) |                     |                                   |
| Pellet inspection                          | 0.022 (0.001) 0.027 (0.002) 0.033 (0.002) 0.032 (0.002) 0.018 (0.001) 0.0135 (0.0008) 0.0052 (0.0004) 0.0026 (0.0002) 0.0043 (0.0003) 2887 0.16 (0.03) |                     |                                   |
| General ventilation exhaust                | 0.0077 (0.0006) 0.0169 (0.001) 0.022 (0.001) 0.026 (0.002) 0.0156 (0.0009) 0.0105 (0.0007) 0.0038 (0.0003) 0.0023 (0.0002) 0.0025 (0.0002) 3046 0.11 (0.02) |                     |                                   |
| BA pelletizing workshop                    |                                             |                     |                                   |
| Mixing with Gd\(_2\)O\(_3\)                 | 0.0119 (0.0009) 0.0188 (0.0013) 0.0627 (0.0037) 0.0893 (0.0051) 0.0784 (0.0045) 0.0588 (0.0034) 0.0129 (0.0009) 0.0005 (0.0002) 0.0077 (0.0005) 1938 0.3 (0.1) |                     |                                   |
| Oxidizing of discarded pellets and grinding waste | 0.22 (0.01) 0.26 (0.01) 0.30 (0.02) 0.19 (0.01) 0.079 (0.005) 0.052 (0.003) 0.021 (0.001) 0.013 (0.001) 0.022 (0.001) 1117 1.2 (0.2) |                     |                                   |
Table B1. Fraction of activity at each impactor stage, based on radiometric data following total alpha counting of samples collected during parallel sampling to investigate the effect from particles bounce. Sampling was carried out at the powder preparation workshop, where oxidizing of waste pellets is carried out, at two different occasions with three impactors running in parallel. Uncertainties correspond to ±1 standard deviation due to counting statistics and flow rate measurements (2.00 ± 0.05 L min⁻¹).

|          | Stage A (213 µm) | Stage B (14.8 µm) | Stage C (9.8 µm) | Stage D (6.0 µm) | Stage E (3.5 µm) | Stage F (1.6 µm) | Stage G (0.9 µm) | Stage H (0.5 µm) | Final filter |
|----------|------------------|-------------------|------------------|------------------|------------------|------------------|------------------|------------------|--------------|
| Occasion 1 |                  |                   |                  |                  |                  |                  |                  |                  |              |
| Impactor 1 – no coating applied | 0.105 (0.006)    | 0.140 (0.008)    | 0.32 (0.02)    | 0.26 (0.01)    | 0.067 (0.004)    | 0.022 (0.001)    | 0.018 (0.001)    | 0.0160 (0.0009)  | 0.057 (0.003)  |
| Impactor 2 – coating applied | 0.103 (0.006)    | 0.165 (0.009)    | 0.34 (0.02)    | 0.25 (0.01)    | 0.072 (0.004)    | 0.027 (0.002)    | 0.0100 (0.0006)  | 0.0060 (0.0004)  | 0.035 (0.002)  |
| Impactor 3 – coating applied | 0.107 (0.006)    | 0.163 (0.009)    | 0.34 (0.02)    | 0.24 (0.01)    | 0.084 (0.005)    | 0.027 (0.002)    | 0.0080 (0.0005)  | 0.0050 (0.0003)  | 0.019 (0.001)  |
| Occasion 2 |                  |                   |                  |                  |                  |                  |                  |                  |              |
| Impactor 1 – no coating applied | 0.081 (0.005)    | 0.121 (0.007)    | 0.32 (0.02)    | 0.26 (0.01)    | 0.097 (0.006)    | 0.043 (0.003)    | 0.030 (0.002)    | 0.0160 (0.0009)  | 0.035 (0.002)  |
| Impactor 2 – no coating applied | 0.071 (0.004)    | 0.139 (0.008)    | 0.33 (0.02)    | 0.25 (0.01)    | 0.076 (0.004)    | 0.041 (0.002)    | 0.025 (0.002)    | 0.030 (0.002)    | 0.038 (0.002)  |
| Impactor 3 – coating applied | 0.098 (0.006)    | 0.147 (0.008)    | 0.32 (0.02)    | 0.26 (0.01)    | 0.101 (0.006)    | 0.033 (0.002)    | 0.0120 (0.0008)  | 0.0060 (0.0004)  | 0.031 (0.002)  |
| Average  |                  |                   |                  |                  |                  |                  |                  |                  |              |
| Coating applied | 0.10 (0.01)     | 0.16 (0.02)     | 0.33 (0.03)    | 0.25 (0.02)    | 0.086 (0.008)    | 0.029 (0.003)    | 0.010 (0.001)    | 0.0060 (0.0007)  | 0.028 (0.003)  |
| No coating applied | 0.086 (0.008)   | 0.13 (0.01)     | 0.32 (0.03)    | 0.26 (0.03)    | 0.080 (0.008)    | 0.035 (0.004)    | 0.024 (0.003)    | 0.020 (0.002)    | 0.043 (0.004)  |
| Ratio coating/no coating | 1.2 (0.2)       | 1.2 (0.2)       | 1.0 (0.1)      | 1.0 (0.1)      | 1.1 (0.1)        | 0.8 (0.1)        | 0.41 (0.06)      | 0.28 (0.04)      | 0.66 (0.09)   |

Fig. B1. Comparison of activity size distributions from sampling with and without coated impactor substrates.
Table B2. Derived average (1 SD) parameters for coarse and fine fractions based on data in Table B1.

|                | Unimodal fit |                          | Coarse fraction |                          | Bimodal fit |                          | Fine fraction |
|----------------|--------------|--------------------------|-----------------|--------------------------|-------------|--------------------------|---------------|
|                | AMAD (µm)   | GSD                      | AMAD 1 (µm)     | GSD 1                    | f           | AMAD 2 (µm)     | GSD 2         | 1-f          |
| Coating applied| 13.1 (0.3)  | 1.7 (0.03)               | 14.2 (0.6)      | 2.1 (0.1)                | 0.7 (0.1)   | 12.1 (0.1)     | 1.4 (0.03)    | 0.3 (0.1)    |
| No coating applied | 12.4 (0.3)  | 1.8 (0.03)               | 15.0 (1.9)      | 2.9 (0.4)                | 0.5 (0.1)   | 11.8 (0.2)     | 1.4 (0.02)    | 0.5 (0.1)    |