EVALUATION OF DEFAULT ANNUAL LIMIT ON INTAKE (ALI) FOR YELLOWCAKE AND URANIUM ORE

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EXECUTIVE SUMMARY

In October 1994, the Atomic Energy Control Board (AECB)[1] recommended default values for the annual limit on intake (ALI) for uranium ore and two types of yellowcake to the Saskatchewan Labour Working Group on Long-Lived Radioactive Dust. Since then, new solubility data has become available and the International Commission on Radiological Protection (ICRP) has recently evaluated its biokinetic models. Thus, it is appropriate to review the ALIs using the updated solubility data and the current ICRP biokinetic models to determine if changes to the default ALI values are required.

The primary objective of this study was to develop appropriate default ALIs for workers at uranium mines and mills in Canada using the most recent available data on solubility and dosimetric methods for uranium ore, calcined yellowcake and non-calcined yellowcake. It is acknowledged that future uranium mines and mills in Canada could produce ore and yellowcake with somewhat different characteristics than at present. Therefore, this study also provides high level guidance on how to develop appropriate ALIs for new ores and for yellowcake of different origins and/or processes or for future North American technology.

The first step in the study was the completion of a literature review. Upon completion of the literature review, efforts were made to collect the most recent solubility data available for Canadian uranium mining and milling operations and to select solubility data that was considered representative of current uranium mines and mills in Canada. Using this solubility data, absorption parameters were derived and provided as inputs to the dosimetric model. The dosimetric model used for this study is the same as that used by the ICRP to derive dose coefficients for intakes of radionuclides. Finally, the results of the modelling are used in the process for calculating the revised ALI values. It should be noted the proposed ALIs in this report are based on radiological dose; however, in some instances the limiting factor for ALIs is chemical toxicity.

The proposed and current ALIs for a variety of uranium mixtures are provided in Table ES.1.

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[1] In 2000, the Canadian Nuclear Safety Commission (CNSC) was established under the *Nuclear Safety and Control Act* and replaced the AECB.

Table ES.1   Proposed and Current ALIs

Sample Type	Proposed ALI (Bq Total Alpha)		Current ALI (Bq Total Alpha
	5 µm	10 µm	
Leach Feed Ore	4,500	8,000	2,800
Nominal Canadian Ore	4,500	8,000	2,800
Dryer Discharge (dried yellowcake)	48,000	93,000	290,000
Concentrate (calcined yellowcake)	3,100	5,100	3,300
Calciner Product (calcined yellowcake)	4,500	7,300	3,300

As shown in Table ES.1, the ALIs with an activity median aerodynamic diameter (AMAD) of 5 µm particle size are comparable (except for dried yellowcake) to the current ore ALIs. For a 10 µm particle, the ALIs proposed in this report would be about twice as large as the current ALIs (except for dried yellowcake). The solubility of yellowcake is affected by thermal processing with calcined yellowcake having a lower ALI than dried product. For present purposes, we suggest nominal (default) ALIs (Table ES.2) based on the current assumption of 5 µm particles with the alternative of using the ALIs for 10µm particles where supported by measurement data.

Table ES.2   Recommended Default ALIs for Ore and Yellowcake

Material	Proposed Default ALI (Bq Total Alpha)
Ore	4,500
Dried Yellowcake	48,000
Calcined Yellowcake	3,100

These recommended default ALIs are provided under the caveat that companies should be allowed to provide product-specific ALIs when justified.

It should be noted that the ICRP is currently updating its biokinetic and dosimetric models, which will be used to develop dose coefficients in the ICRPs forthcoming Occupational Intake Radionuclides (OIR) document. Although the ICRPs biokinetic model (ICRP 1995d) for uranium will not change, the effective dose coefficients will be higher resulting in lower ALIs. Hence, once the OIR document is published, a comparison of the current and OIR dose conversion factors should be made.

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ACRONYMS

ADU	Ammonium Diuranante
AECB	Atomic Energy Control Board
AI	Alveolar-Interstitial
ALI	Annual Limit on Intake
AMAD	Activity Median Aerodynamic Diameter
BB	Bronchial
bb	Bronchiolar
CED	Committed Effective Dose
CFR	Code of Federal Regulations
CNSC	Canadian Nuclear Safety Commission
DAC	Derived Air Concentration
Fr	Rapid Release of Uranium
GI	Gastrointestinal Tract
HRTM	Human Respiratory Tract Model
ICRP	International Commission on Radiological Protection
ISR	In Situ Recovery
LLRD	Long-Lived Radioactive Dust
LN_{TH}	Lymphatics
NIST	National Institute of Standards and Technology
OIR	Occupational Intake Radionuclides
PHE	Public Health England
SLF	Simulated Lung Fluid
Sr	Time Constant for Fast Release
Ss	Time Constant for Slow Release
TEDE	Total Effective Dose Equivalent
TGLD	Task Group on Lung Dynamics
TGLM	Task Group Lung Model
U.S. NRC	United States Nuclear Regulatory Commission

1.0   INTRODUCTION

1.1   BACKGROUND

In October 1994, the Atomic Energy Control Board (AECB)[2] recommended default values for the annual limit on intake (ALI) for uranium ore and two types of yellowcake to the Saskatchewan Labour Working Group on Long-Lived Radioactive Dust. These default ALIs were developed by AECB staff and were based on solubility data for various types of uranium compounds measured as part of an AECB funded research project. The solubility data were used as inputs into the then current dosimetry software at the time to derive compound-specific ALIs (CNSC 2012). These ALIs are still currently used to assign doses to workers at uranium mines and mills.

New solubility data has become available from the work of Canadian mining companies and the ICRP has recently evaluated its biokinetic models. Thus, it is appropriate to review the ALIs using the recent solubility data and the current ICRP biokinetic models to determine if changes to the default ALI values are required.

1.2   OBJECTIVES

ALIs relate airborne concentrations of radionuclides to dose and are thus useful for managing dose and the intake of radioactive dust at uranium mines and mills. The estimated doses from inhaled radionuclides depend on a number of factors, including the solubility of the materials which in turn is driven by chemical speciation, the dosimetric model formulation and the input parameters for the dose estimation. The primary objective of this study is to develop appropriate default (nominal) ALIs for workers at uranium mines and mills in Canada using the most recent available data on solubility and dosimetric methods for uranium ore, calcined yellowcake[3] and non-calcined yellowcake.

The ALIs developed in this study recognize the variability in ores and the effect of metallurgical processing on solubility within the Canadian uranium mining and milling industry. It is also acknowledged that future uranium mines and mills in Canada could produce ore and yellowcake with somewhat different characteristics than at present. In addition, not all yellowcake processed in Canada is from Canadian mills. Therefore, this study also provides high level guidance on how to develop appropriate ALIs for new ores and for yellowcake of different origins and/or processes or for future North American technology.

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[2] In 2000 the Canadian Nuclear Safety Commission (CNSC) was established under the *Nuclear Safety and Control Act* and replaced the AECB.

[3] A uranium concentrate powder that is produced by drying leach solutions at temperatures of 400°C or more, and is typically less soluble than non-calcined yellowcake.

1.3   APPROACH

The first step in the study was the completion of a literature review. This included reviewing publically available data and participating in discussions with uranium companies, regulatory agencies and scientific authorities. Upon completion of the literature review, efforts were made to collect the most recent solubility data available for Canadian uranium mining and milling operations and to select solubility data that was considered representative of uranium mines and mills in Canada. Using this solubility data, absorption parameters were derived and provided as inputs to the dosimetric model. The dosimetric model used for this study is the same as that used for the ICRP dosimetric calculations. It should be noted that the dosimetric modelling performed for this study was carried out by Public Health England (PHE). Finally, the results of the modelling are used in the process developed for selecting ALI values and the subsequent estimation of revised ALI values.

1.4   OUTLINE OF REPORT

The report is structured as follows:

- Chapter 2 **Literature Review** – Provides the information obtained through review of publically available data, discussions with uranium companies and discussions with regulatory and scientific authorities.

- Chapter 3 **Data Identification and Selection of Uranium Solubility Data** – Provides the data associated with estimation of ALIs.

- Chapter 4 **Derivation of Absorption Parameters** – Provides the derivation of the absorption parameters.

- Chapter 5 **Dosimetric Modelling** – Provides a description and results of the dosimetric modelling.

- Chapter 6 **Estimation of Annual Limit on Intake (ALI)** – Describes the process for estimating ALIs and provides the revised ALIs.

- Chapter 7 **References** – Lists the materials cited in this study.

- Annex A **Activity Ratios for Natural Uranium**

- Annex B **Assumed F_1 Values**

2.0   LITERATURE REVIEW

2.1   APPROACH

The literature review was conducted by gathering the publicly available information on solubility studies of uranium ore, calcined yellowcake and non-calcined yellowcake. This included searching various databases (e.g., Google Scholar, Science Citation Index, Web of Science, etc.), and participating in discussions with uranium companies, regulatory agencies and scientific authorities. The citations for all literature, along with an electronic copy of each document were maintained in a database. The results of this literature review (e.g. results of in vivo and/or in vitro solubility studies) are provided below in the corresponding subsections.

2.2   REGULATORY AGENCIES

2.2.1   Canadian Nuclear Safety Commission

As mentioned previously, AECB recommended default ALIs to the Saskatchewan Labour Working Group on Long-Lived Radioactive Dust for uranium ore and two types of yellowcake (Government of Canada 1994). The information used to develop these default ALIs was presented in a series of reports on the in vitro lung fluid solubility behaviour of workplace aerosols that are of radiological health concern in the Canadian uranium industry (AECB 1988). The radionuclides of interest were uranium-238, thorium-230, radium-226 and lead-210. The study was carried out in five phases over several years. Materials examined for this study included (AECB 1988, 1989, 1993, 1994):

- Low-grade concentrate from Beaverlodge;
- Uranium ore from Elliot Lake;
- Commercially produced unsintered uranium dioxide;
- Medium grade ore from Saskatchewan;
- Fine powder mixture of thorium dioxide and nickel metal; and,
- High grade ore from Saskatchewan.

The resulting ALIs are provided in Table 2.1.

Table 2.1 **ALIs for Non-Calcined Uranium, Calcined Uranium and Uranium Ore in Saskatchewan (Government of Canada 1994)**

Material	Dose Conversion Factor, mSv per Bq U-238 (Inhalation)	ALI, total alpha Bq
Non-Calcined Yellowcake (2 alpha particles)	1.4E-04	290,000
Low Temperature Calcined Yellowcake (2 alpha particles)	1.4E-04	290,000
High Temperature Calcined Yellowcake (2 alpha particles)	1.2E-02	3,300
Uranium in Ore (2 alpha particles)	9.2E-03	4,400
Uranium Ore (7 alpha particles)	5.1E-02	2,800

In 1995, AECB calculated ALIs for long-lived radioactive dust (LLRD) by using the software code LUDEP[4], which implements the ICRP's Human Respiratory Tract Model (HRTM) (1994a) with the ICRP Systemic Biokinetic Model (1979). Input parameter values for in-vitro dissolution rates and activity median aerodynamic diameter (AMAD) obtained from research studies (AECB 1995) were used in the calculations. The AECB stated that these interim ALIs may be used to assess doses received at uranium mining and milling facilities for control purposes until site-specific values are determined. The calculated ALIs were:

- Yellowcake: 290,000 Bq total alpha activity
- Calcined Yellowcake: 3,300 Bq total alpha activity
- Ore: 2,800 Bq total alpha activity

These ALIs are still currently used to assign doses to workers at uranium mines and mills, unless site-specific ALIs have been determined.

2.2.2 United States Nuclear Regulatory Commission

The United States Nuclear Regulatory Commission (U.S. NRC) presents nuclide specific ALIs in the US Code of Federal Regulation, Title 10, Chapter 20 – *Standards for Protection Against Radiation* (10 CFR 20) in Appendix B. For natural uranium, the U.S. NRC reports the following ALIs and associated DACs (Derived Air Concentrations) for each of the three solubility classes (i.e., D, W and Y) in Table 2.2.

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[4] Developed by the U.K. National Radiological Protection Board.

Table 2.2   U.S. NRC ALIs for U-238[a]

Natural Uranium	ALI[b], µCi (Bq) [Inhalation]	Inhalation: DAC[c], µCi/ml (Bq/mL) [Inhalation]
D	1.0 (37,000)	6E-10 (2E-05)
W	0.8 (29,600)	3E-10 (1E-05)
Y	0.05 (1,850)	2E-11 (7E-07)

a) 10CFR20, Appendix B, Table 1.

b) Intake that would result in TEDE in 5 rem/year.

c) Annual average over 2000 working hours that would result in intake of one ALI.

For most radionuclides, the U.S. NRC ALIs are defined for three solubility "Classes" based on ICRP 30 dosimetry (ICRP 1979) and the compound solubility classification system of the ICRP Committee 2, Task Group Lung Model (TGLM – Morrow, 1966). To describe the clearance of radioactive materials from the lungs, the materials were classified as D (most soluble), W (moderate solubility), and Y (most insoluble), referring to relative retention time in the pulmonary region. The retention half times for these classifications are:

- Class D: < 10 days
- Class W: 10 – 100 days
- Class Y: > 100 days

This solubility classification scheme and associated ALIs are still used by U.S. NRC and its licensees must demonstrate compliance to dose and intake limits using this system in accordance with the specific requirements of 10 CFR 20.1204 *Determination of Internal Exposure*. Additionally, the U.S. NRC has a weekly intake limit of 10 mg for "Soluble Uranium" at 10 CFR 20.1201(e).

As a point of contrast, in the classification scheme of the HRTM, the absorption types F (fast), M (moderate) and S (slow) refer to absorption characteristics of the material only, whereas, classes of ICRP 30 (D/W/Y) are based on retention times in the pulmonary region. However, in ICRP Publication 68 (1995a), those compounds in Class D were assigned to Type F, Class W to Type M, and Class Y to Type S. This was also done in ICRP Publication 71 (1995b) in the cases where more specific information was not available.

2.3   URANIUM PRODUCERS

2.3.1   Canada

2.3.1.1   Cameco

Cameco is one of the world's largest uranium producers and is a leading provider of processing services required to produce fuel for nuclear power plants. Within Canada, Cameco operates uranium mines, uranium mills, a uranium refinery, a uranium conversion plant and a fuel fabrication facility.

Solubility Studies on Uranium Mines and Mills

In 2005, Cameco conducted a study to assign absorption type specified by ICRP Publication 71 criteria (Fast, Moderate and Slow) to site-specific radionuclides found at Key Lake, McArthur River and Rabbit Lake facilities (Cameco 2005). In order to produce site-specific solubility data, simulated lung fluid (SLF) solubility experiments were performed for the various types of uranium bearing materials present at the sites. A total of 23 samples were collected at the mine sites and the samples were sieved to obtain particles less than 20µm ("inhalable" particle sizes) before analysis for uranium and other radionuclides. The results showed that radionuclides in all ore samples had low solubilities in SLF, as summarized in Table 2.3 below.

Table 2.3   Absorption Types for Ore Samples

Radionuclide	Mine Site	Absorption Type[a]
Ra-226	Rabbit Lake	M
	McArthur River	S
Po-210	Rabbit Lake	S
	McArthur River	S
Pb-210	Rabbit Lake	S
	McArthur River	S
Th-230	Rabbit Lake	S
	McArthur River	S
Th-234	Rabbit Lake	M
	McArthur River	S
Uranium	McArthur River	S

a)   Cameco 2005

In addition to ore samples, this Cameco study also assigned absorption types for concentrate, leach and calciner samples. The uranium components in the Rabbit Lake concentrate and leach feed samples were assigned to Type F[5] and Type M[6], respectively (Cameco 2005). The solubility of the Key Lake calciner samples in SLF was based on the presence of both the non-calcined component, uranyl sulphate dehydrate and the calcined component U_3O_8. The study found that the calciner samples with a higher non-calcined component had a higher solubility than those with a higher calcined component (U_3O_8) (Cameco 2005).

Precipitating Agent and Drying Temperature at Uranium Mills

Cameco operates the largest (Key Lake) and second largest (Rabbit Lake) uranium mill in the world. These two uranium mills operate in northern Saskatchewan and produce U_3O_8 (yellowcake). The major steps in the milling process include:

- Grinding, which breaks the ore into sand;
- Leaching to dissolve the uranium;
- Separation of uranium solution from waste solids;
- Solvent extraction to produce a purified uranium solution; and,
- Yellowcake precipitation and drying which recovers uranium as crystal (Rabbit Lake) or granular powder (Key Lake).

Through discussions with Cameco, the following information was obtained for the Key Lake and Rabbit Lake mills. The precipitating agent used at the Key Lake site is ammonia, the drying temperature is 840ºC and the free moisture in the final product is approximately 0.02%. At Rabbit Lake, the precipitating agent is hydrogen perioxide, the drying temperature ranges from 120 to 140ºC and the free moisture in the final product is approximately 2%. For yellowcake, the precipitating agent and drying/calcining details are important factors in determining solubility[7].

2.3.1.2   AREVA

AREVA operates the McClean Lake uranium mill in Northern Saskatchewan, which has produced uranium concentrate since 1999. The milling processed used at McClean Lake is broadly similar to that used at Key Lake and Rabbit Lake.

Through discussions with AREVA, it is understood that the McClean Lake mill uses an ammonia precipitating agent and a calcining temperature of 800ºC.

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[5] One sample (within standard deviation) could have been assigned to type F or type M (Cameco 2005).
[6] Values of the parameters were very close to those for type S (Cameco 2005).
[7] See for example Table 3.1.

2.3.2 United States

Cameco utilizes in situ recovery (ISR) mining techniques at two operations in the United States to extract uranium contained in sandstone aquifers. These are the Crow Butte (Nebraska, USA) and Smith Ranch-Highland (Wyoming, USA) ISR plants. ISR involves circulation of solutions through ore-bearing formations to dissolve the uranium in situ and pump the uranium to the surface for recovery. This results in minimal disturbance of the surface and produces no waste rock or mill tailings (Cameco 2013).

Cameco conducted a yellowcake characterization project in 2009 - 2010 to assess the solubility and related characteristics of uranium concentrate from both the Crow Butte and Smith Ranch-Highland uranium ISR plants. The Cameco Innovation and Technology Development Research Centre in Port Hope, Ontario performed this work for purposes of determining dissolution rates in lung fluid simulants and chemical speciation. In addition to dissolution rates, uranium content (%) and molecular speciation via x–ray diffraction was determined. These products are produced by peroxide precipitation processes and dewatered in vacuum dryers at temperatures < 300^{0}C. The results of these studies are reported by Tairova *et al.* (2010) and Cameco (2010 and 2011), see Figures 2.1 and 2.2.

Figure 2.1   Dissolution of Smith Ranch-Highland Samples
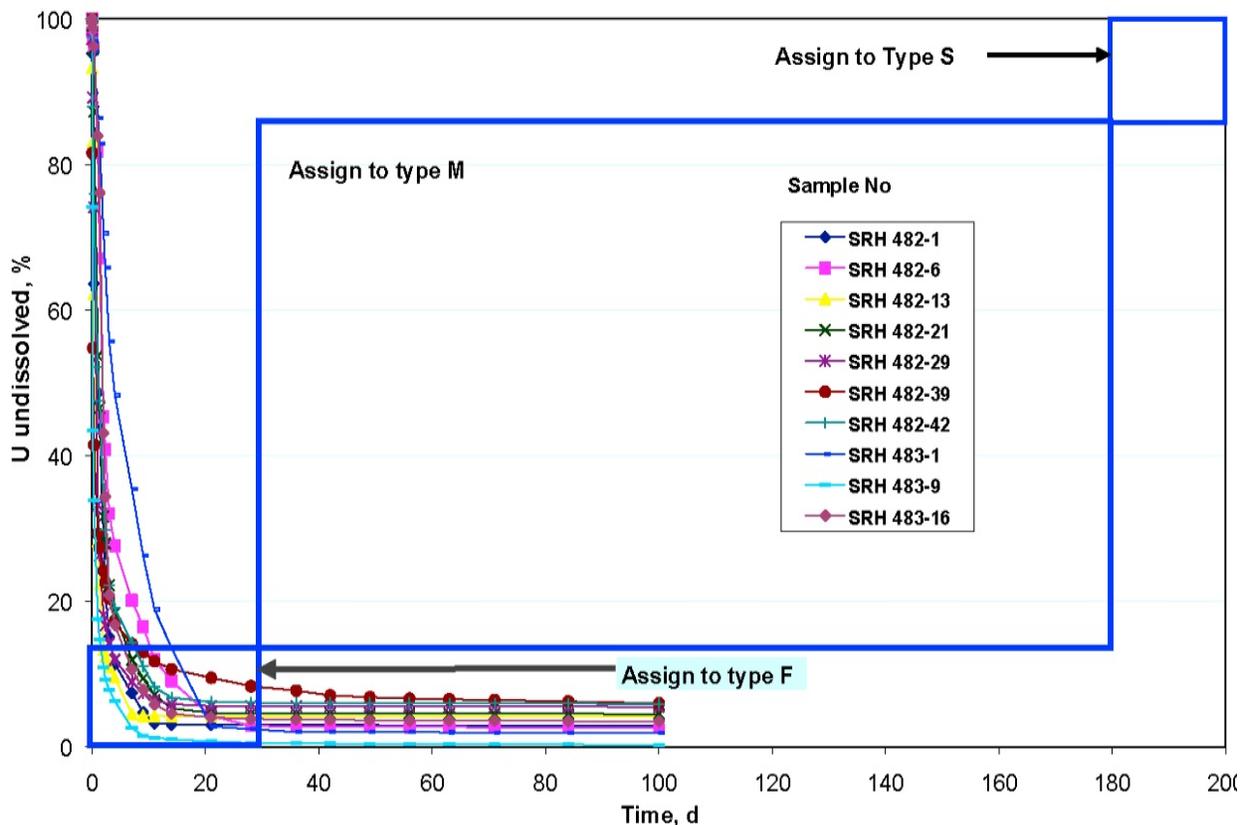
Figure 2.2   Dissolution of Crow Butte Samples
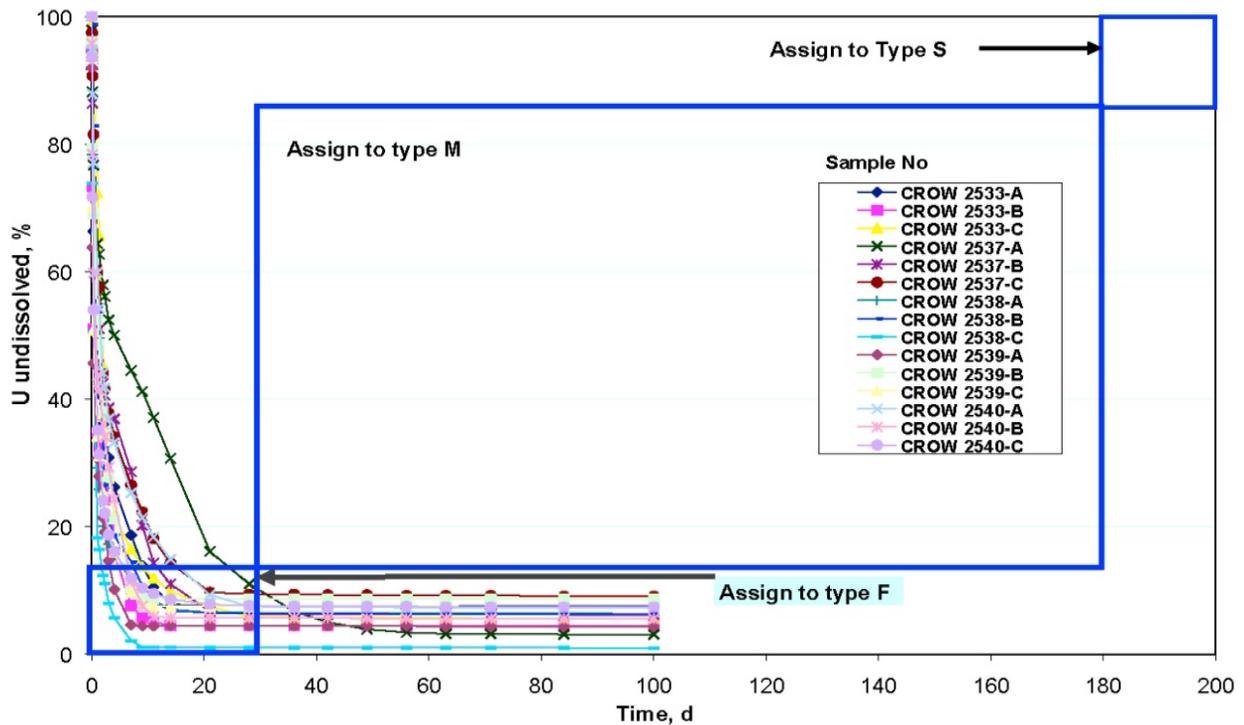
All samples exhibited solubility characteristics that met the definition of absorption Type F as defined in ICRP 71, which considers Type F "generally equivalent" to solubility Class D from ICRP 30 (ICRP 1979), i.e. the most "soluble" category (Note that these facilities are licensed by and under the jurisdiction of the U.S. NRC and therefore the older TGLD (Morrow 1966) classification system and ICRP 30 (ICRP 1979) dosimetry apply – see Section 2.2.2).

2.4   OPEN LITERATURE

Uranium mills and ISRs that operated in North America during the 1960s and 1970s typically used an ammonia precipitation process producing ammonium diuranate (ADU) which was then dried (calcined) at relatively high temperatures, usually > 1000°C. Characterization studies performed on these products, including in vitro lung fluid solubility studies (methods after Moss 1976) and X-Ray diffraction analysis indicated these products were primarily insoluble U_3O_8 and UO_2 (Spitz and Robinson 1981; Kalkwarf, 1983; Eidson and Mewhinney 1983; Eidson 1994). In the historical descriptions of the clearance of radioactive materials from the lungs using ICRP 30 model (ICRP 1979), materials were classified into three Classes (i.e., D, W and Y), referring to retention time in the pulmonary region, as described in Section 2.2.2.

Most products were reported to exhibit multi-phase solubility since they included a combination of several oxides, e.g. Class Y and Class W components in the same product; some had all three (Class D, W and Y) including more soluble UO_3 (Blauer and Brown, 1980; Blauer, Kent and Dennis 1982; Eidson and Griffith, 1984). Differences between individual mill products were attributed to differences in details of precipitation chemistry and thermal exposure, i.e., feed rate and temperature of the calciner (Merrit 1971; Wyoming Mineral Corporation, 1980; Blauer and Brown, 1980; Eidson 1994).

Annex D of ICRP 71 (ICRP 1995b) provides instructions on how to assign material to absorption types based on experimental data (e.g., lung fluid simulant studies) using absorption rates at different times rather than overall retention or clearance rates. Specifically, for an *in vitro* dissolution experiment, classification depends on the amount of undissolved material, or percent retained, at specified time intervals. Excluding particle transport, the classification criteria for absorption types F, M, and S are as follows:

- Type F   < 13% remains at 30 days
- Type M   > 13% at 30 days and < 87% at 180 days
- Type S   > 87% remains at 180 days

Regarding more recent studies on yellowcake solubility, Section 2.3.2 presents information reported by Cameco on yellowcake produced by peroxide precipitation and dried at relatively low temperature at their ISR facilities in the U.S.

Additionally, a study by Metzger (1997) reported on the Irigary ISR (Wyoming US), which also produced yellowcake by peroxide precipitation at drying temperatures lower than that of the high fired calciners described above. This study included the analysis of products from in-situ uranium recovery at the Irigary plant in Wyoming. At that time, the Irigary plant leached uranium in situ via a carbonate/bicarbonate leach solution, precipitated with hydrogen peroxide, dried to produce the final yellowcake at ~540°C (a much lower temperature than used in earlier mills). Product composition was determined via x-ray diffraction techniques and indicated the final product's uranium species content to contain approximately 80% $UO_4 \bullet {}_2H_2O$ and 15% UO_3. Air samplers were located throughout different parts of the plant to represent the majority of the locations for potential exposure throughout the process. Solubility studies using lung fluid simulants were conducted in Gambles Solution (Moss 1976) following methods of Edison and Mewhinney (1983). The dissolution rate was determined over a period of 28 days. Airborne uranium from all worker areas sampled was highly soluble, with 97% dissolving with a 0.25 - 0.30 day half time and 3 % with a half time of 15 -20 days. Accordingly, using the ICRP 30 classification since this was at that time and is still used by their licensing agency (U.S. NRC), the authors suggested that this material should be assigned 97% solubility Class D and 3% Class W.

3.0 DATA IDENTIFICATION AND SELECTION OF URANIUM SOLUBILITY DATA

3.1 URANIUM RETENTION

Cameco and AREVA have conducted studies on the solubility of site-specific uranium concentrate and ore samples at their mines and mills to assign absorption types fast (F), moderate (M) and slow (S), according to ICRP criteria. Samples available to the present study were collected from the following:

- Rabbit Lake Non-Calcined Concentrate, Table 3.1 (Cameco);
- Rabbit Lake Ore Slurry, Table 3.2 (Cameco);
- Key Lake Non-Calcined Concentrate, Table 3.3 (Cameco);
- Key Lake Calcined Concentrate, Table 3.4 (Cameco);
- McArthur River Dried Ore Slurry, Table 3.5 (Cameco); and,
- McLean Lake Concentrate, Table 3.6 (AREVA).

The uranium retention[8] (as a percentage) was provided for each sample of each material over an elapsed time (up to 100 days). The resulting uranium retention for each of the materials identified above are provided in Tables 3.1, 3.2, 3.3, 3.4, 3.5 and 3.6.

Table 3.1   Uranium Retention for Rabbit Lake Non-Calcined Concentrate

Sample No.	RLF 1	RLF 2	RLF 3	RLF 4	RLF 5	RLF 6
Total Uranium in a sample	64.7 mg	69.3 mg	63.2 mg	63.8 mg	67 mg	64.8 mg
Elapsed Time (days)	Uranium Retention (%)					
0	100	100	100	100	100	100
0.2	93.2	90.8	96.3	94.5	88.2	89.5
0.3	91.3	86.3	95.6	92.5	83.9	85.8
0.8	73.3	70.5	95.1	92.3	81.8	82.2
1.1	71.2	66	93.5	91.5	76.9	78.1
1.8	65.1	54	88.4	89.1	68.4	67.9
2	63.1	51.6	86	87.9	61.3	64.7
2.7	58.4	45.1	78.4	84.6	55.5	56.8
9	29.5	18.5	48.5	58.1	25.1	28.1
20	0	0	2.9	21.5	9.9	6.5
30			0	12.1	2.5	1.9
50				6	1.9	1.8
78				5.9	1.7	1.8
100				5.8	1.4	1.8

Not included in model fitting as data was suspect

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[8] Amount of material that was not dissolved.

Table 3.2   Uranium Retention for Rabbit Lake Ore Slurry

Sample No.	RLF 7	RLF 8	RLF 9
Total Uranium in a sample	10.7 mg	8.5 mg	8 mg
Elapsed Time (days)	Uranium Retention (%)		
0	100	100	100
0.17	98	97.7	95
0.3	95.6	95.2	94.4
0.8	94	94.7	93.9
1.08	93.6	94.5	93.8
1.75	92.9	94.1	93.3
2	92.8	93.9	93.2
2.7	92.6	93.7	92.8
10	92.1	93	92.2
20	91.7	92.5	91
30	91.4	92.3	90.8
50	89.7	91.3	89.9
78	87.9	89.1	88.6
100	86.4	86.2	86.2

Table 3.3   Uranium Retention for Key Lake Non-Calcined Concentrate

Sample No.	RLF 13	RLF 15	RLF 17
Total Uranium in a sample	64 mg	58.5 mg	60.5 mg
Elapsed Time (days)	Uranium Retention (%)		
0	100	100	100
0.17	73.3	68.8	76.7
0.3	59.5	58.3	67.1
0.8	42.3	40.1	43.7
1.08	36.3	34.2	36.8
1.75	25.4	22	23.9
2	22.1	18.7	19.9
2.7	15.9	10.8	11.3
10	0	0	0
20			
30			
50			
78			
100			

Not included in model fitting as data was suspect

Table 3.4   Uranium Retention for Key Lake Calcined Concentrate

Sample No.	RLF 14	RLF 16	RLF 18	RLF 19
Total Uranium in a sample	81.2 mg	79.3 mg	75.8 mg	0.6565 mg
Elapsed Time (days)	**Uranium Retention (%)**			
0	100	100	100	100
0.2	87	96.5	73.6	91
0.3	85.5	95.8	73.2	90.5
0.8	82.8	95.3	72.8	90.1
1.1	81.9	95	72.6	90.1
1.8	81	94.8	72.3	90.1
2	80.9	94.8	72.2	90.1
2.7	80.7	94.7	71.9	90
9	80.2	94.4	70.4	90
20	79.7	94	68.6	89.9
30	79.4	93.6	67.2	89.8
50	78.7	93.2	65.2	89.6
78	77.9	93.2	64	89.5
100	77.5	93.1	63.5	89.3

Table 3.5   Uranium Retention for McArthur River Dried Ore Slurry

Sample No.	RLF 10	RLF 11	RLF 12
Total Uranium in a sample	108.5 mg	123.2 mg	110.6 mg
Elapsed Time (days)	**Uranium Retention (%)**		
0	100	100	100
0.17	99.8	100	99.8
0.3	99.8	99.9	99.8
0.8	99.8	99.9	99.8
1.08	99.8	99.9	99.7
1.75	99.8	99.9	99.7
2	99.8	99.9	99.7
2.7	99.8	99.8	99.7
10	99.7	99.8	99.7
20	99.7	99.4	99.7
30	99.7	99.4	99.7
50	99.6	99.4	99.6
78	99.3	99.3	99.4
100	99.2	99.2	99.3

Table 3.6   Uranium Retention for McClean Lake Concentrate

Sample No.	AREV 17	AREV 18	AREV 19	AREV 20	AREV 21	AREV 22	AREV 23	AREV 24	AREV 25	AREV 26	AREV 27	AREV 28	AREV 29	AREV 30	AREV 31	AREV 32
Lot #	A0012	A0021	A0023	A0023	D0013	D0013	A0029	A0029	M0936	M0936	M0938	M0938	M0939	M0939	D0020	D0020
Total Uranium in sample (mg)	41.9	41.9	42.1	42.1	42.2	42.2	42.1	42.1	42.1	42.1	42.1	42.1	42	42	41.8	41.8
Elapsed Time (d)	Uranium Retention (%)															
0	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
0.01	99.15	99.03	99.24	99.9	99.26	99.14	99.33	99.74	99.58	100	99.64	99.43	99.47	99.91	99.4	99.29
0.1	98.51	98.4	98.81	99.46	98.92	98.88	98.91	98.96	99.31	100	99.32	99.19	98.99	99.57	98.9	98.88
0.18	98.17	98.08	98.67	99.22	98.83	98.78	98.8	98.86	99.25	99.99	99.23	99.11	98.89	99.35	98.79	98.75
0.26	97.7	97.89	98.61	99.04	98.78	98.71	98.74	98.81	99.13	99.99	99.18	99.06	98.83	99.19	98.71	98.68
0.35	97.57	97.78	98.56	98.88	98.73	98.66	98.7	98.77	99.1	99.99	99.13	99.01	98.78	99.05	98.66	98.65
1.01	97.29	97.54	98.43	98.66	98.61	98.54	98.6	98.67	99.01	99.98	98.95	98.85	98.61	98.7	98.43	98.52
1.14	97.27	97.52	98.42	98.65	98.6	98.53	98.59	98.66	99	99.98	98.93	98.83	98.59	98.68	98.4	98.47
1.26	97.25	97.5	98.42	98.63	98.59	98.52	98.58	98.65	98.99	99.98	98.91	98.82	98.58	98.67	98.38	98.45
1.35	97.23	97.49	98.41	98.63	98.58	98.51	98.58	98.64	98.99	99.98	98.9	98.81	98.57	98.66	98.36	98.44
2.02	97.14	97.4	98.36	98.57	98.53	98.45	98.52	98.58	98.93	99.95	98.82	98.74	98.49	98.59	98.26	98.37
2.31	97.11	97.37	98.34	98.55	98.51	98.44	98.51	98.56	98.91	99.91	98.79	98.72	98.45	98.56	98.21	98.35
3	97.01	97.3	98.31	98.5	98.47	98.39	98.48	98.51	98.85	99.82	98.73	98.67	98.39	98.5	98.13	98.29
4	96.91	97.22	98.26	98.44	98.42	98.34	98.45	98.48	98.8	99.72	98.66	98.61	98.31	98.44	98.05	98.23
7	96.83	97.13	98.22	98.4	98.38	98.31	98.42	98.45	98.74	99.53	98.61	98.56	98.23	98.4	97.97	98.16
9	96.8	97.1	98.21	98.39	98.37	98.3	98.41	98.45	98.73	99.43	98.59	98.55	98.2	98.39	97.95	98.13
11	96.77	97.09	98.21	98.39	98.36	98.3	98.4	98.44	98.72	99.35	98.59	98.54	98.19	98.38	97.93	98.09
14	96.77	97.07	98.2	98.38	98.36	98.29	98.39	98.44	98.72	99.34	98.47	98.54	98.18	98.37	97.93	98.03
21	96.72	97.04	98.19	98.37	98.35	98.28	98.38	98.43	98.71	99.1	98.46	98.53	98.17	98.37	97.86	97.97
28	96.7	96.98	98.18	98.36	98.34	98.27	98.37	98.42	98.69	98.87	98.45	98.53	98.17	98.36	97.81	97.89
35	96.67	96.95	98.17	98.35	98.33	98.26	98.37	98.41	98.68	98.6	98.44	98.52	98.16	98.35	97.78	97.82
42	96.64	96.92	98.16	98.34	98.32	98.26	98.36	98.4	98.67	98.35	98.43	98.51	98.15	98.34	97.75	97.77
49	96.63	96.9	98.16	98.34	98.32	98.25	98.35	98.4	98.67	98.08	98.43	98.51	98.14	98.34	97.74	97.72
57	96.6	96.85	98.15	98.33	98.31	98.25	98.35	98.39	98.66	97.62	98.43	98.5	98.14	98.34	97.71	97.63
63	96.58	96.83	98.14	98.32	98.31	98.24	98.35	98.39	98.65	97.29	98.43	98.5	98.13	98.34	97.7	97.58
70	96.57	96.8	98.13	98.3	98.3	98.23	98.34	98.38	98.64	96.94	98.42	98.49	98.09	98.33	97.7	97.54
84	96.55	96.76	98.1	98.27	98.28	98.22	98.32	98.36	98.63	96.14	98.4	98.48	98.08	98.31	97.68	97.48
91	96.53	96.74	98.08	98.26	98.26	98.21	98.31	98.35	98.62	95.73	98.39	98.47	98.07	98.3	97.67	97.46

3.2 PARTICLE SIZE

Based on the results of the literature review and from discussions with Canadian operators on measurements in Canadian mines, the AMAD for material/aerosols at Canadian mines and mills is thought to typically be larger than the current default of 5 microns, ranging from 5 to 15 microns. Therefore, for a sensitivity analysis, this study estimated the ALIs assuming an AMAD of 5 μm and 10 μm.

3.3 ISOTOPE ASSUMPTIONS

The estimation of ALIs for this study assumes natural isotopic ratios for U-238, U-235 and U-234 for ores and subsequent decay chain members of the U-238 and 235 decay chains, and assumed that no Th-232 is present. For all non-ore materials (e.g., calcined product), it was assumed only the uranium isotopes were present and at natural isotopic ratios (see annex A). In addition, the analysis assumed that all radon is retained at the time of intake (i.e., no radon escaping) which adds a slight conservatism to the dose estimation.

4.0   DERIVATION OF ABSORPTION PARAMETERS

The absorption parameters derived for this study were the rapid release of uranium (Fr), time constant for fast release (Sr) and the time constant for slow release (Ss) for the simulated lung fluid samples collected at the Canadian mines and mills (i.e., Cameco and AREVA). The data used for this derivation covered a broad range of processes and has been provided in Tables 3.1 through 3.6.

It was assumed that there was the possibility of two solubilities of uranium[9] in the simulated lung fluid:

$$U(d) = Fr * \exp(-Sr * d) + (100 - Fr) * (\exp(-Ss * d))$$

Where

        U(d) is the amount of undissolved uranium after a time of d days.

        Fr is the percentage of rapid release uranium.

        Sr is the time constant for the dissolution of rapid release uranium in units of per day.

        Ss is the time constant for the dissolution of the slow release uranium in units of per day.

The statistical software SYSTAT13 was used with the uranium retention data[10] to estimate the parameters in the double exponential equation above. The Nonlinear Regression procedure with default method Gauss-Newton was used and the range of Fr was set to be between 0 and 100%. Appropriate starting values were determined using parameters estimated from previous studies on the simulated lung fluid concentrations. The number of iterations was set to 200[11].

Table 4.1 shows the results of the Fr (%), Sr and Ss for Cameco operations, while Table 4.2 shows similar data from AREVA's McClean Lake mill. The column titled "Type" was as provided by Cameco and the column titled "R^2" is the correlation coefficient between the observed and predicted values. In some cases, there was very little rapid release uranium present (low Fr value) and the Sr could not be estimated. In these cases, a single decay curve was used to model the decay. The experimental dissolution kinetic curves fitted with model discussed above for Cameco and AREVA data are provided in Figures 4.1, 4.2 and 4.3. In addition, dissolution kinetic curves are also provided for selected samples, Figure 4.4.

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[9]   Historical data (as described in Section 2.4), suggests that yellowcake may have multi-phase solubility.

[10] Percent of uranium retained over time (i.e., 0 to 100 days).

[11] This number of iterations was set as a maximum in order for convergence to occur.

Table 4.1   Estimated Fr (%), Sr and Ss from Cameco Simulated Lung Fluids

Site	Material	Sample Number	Sample Description	Type	Fr (%)	Sr (d^{-1})	Ss (d^{-1})	R^2
Rabbit Lake	Noncalcined	RLF-1	Dryer discharge sample	F	24.0	1.61	1.0E-01	0.99
Rabbit Lake	Noncalcined	RLF-2	Dryer discharge sample	F	36.8	1.15	1.4E-01	1.00
Rabbit Lake	Noncalcined	RLF-3	Dryer discharge sample	F	0.19	unknown	7.9E-02	0.99
Rabbit Lake	Noncalcined	RLF4	Dryer Feed sample	M/F	1.71	unknown	6.6E-02	0.99
Rabbit Lake	Noncalcined	RLF5	Dryer Feed sample	F	12.7	7.54	1.4E-01	1.00
Rabbit Lake	Noncalcined	RLF6	Dryer Feed sample	F	12.0	5.92	1.4E-01	1.00
Key Lake	Calcined	RLF 14	Calciner Packing Area	M	18.8	5.91	5.4E-04	0.98
Key Lake	Calcined	RLF 16	Calciner Packing Area	S	5.26	5.83	2.2E-04	0.97
Key Lake	Calcined	RLF 18	Calciner Product	M	28.0	16.2	1.5E-04	0.99
Key Lake	Calcined	RLF 19	Aerosol Calciner Sample	S	9.90	13.7	9.3E-05	1.00
Key Lake	Noncalcined	RLF 13	Centrifuge Discharge	F	38.0	5.36	5.1E-01	1.00
Key Lake	Noncalcined	RLF 15	Centrifuge Discharge	F	31.9	8.39	6.5E-01	1.00
Key Lake	Noncalcined	RLF 17	Centrifuge Discharge	F	24.2	5.49	6.8E-01	1.00
Rabbit Lake	Ore Slurry	RLF-7	Leach Feed Sample	M	6.22	3.67	9.3E-04	0.99
Rabbit Lake	Ore Slurry	RLF-8	Leach Feed Sample	M	5.38	4.88	9.5E-04	0.98
Rabbit Lake	Ore Slurry	RLF-9	Leach Feed Sample	M	6.49	7.85	9.5E-04	0.98
McArthur River	Dried Ore Slurry	RLF-10	Dried Ore Slurry	S	0.17	unknown	6.3E-05	0.91
McArthur River	Dried Ore Slurry	RLF-11	Dried Ore Slurry	S	0.13	unknown	8.0E-05	0.80
McArthur River	Dried Ore Slurry	RLF-12	Dried Ore Slurry	S	0.22	unknown	4.7E-05	0.79

All Samples	**Minimum**	0.13	1.15	4.7E-05
	Median	9.90	5.87	9.5E-04
	Maximum	38.0	16.2	0.677
Noncalcined Samples	**Minimum**	0.19	1.15	0.066
	Median	24.0	5.49	0.137
	Maximum	38.0	8.39	0.677
Calcined and Ore Samples	**Minimum**	0.13	3.67	4.7E-05
	Median	5.80	5.91	1.9E-04
	Maximum	28.0	16.2	9.5E-04

Table 4.2   Estimated Fr (%), Sr and Ss from AREVA Simulated Lung Fluids Collected from Concentrate Samples

Site	AREVA Lot No.	Sample Number	Type	Fr (%)	Sr (d^{-1})	Ss (d^{-1})	R^2
McClean Lake	A0012	AREV 17	S	2.95	5.93	7.6E-05	0.96
McClean Lake	A0012	AREV 18	S	2.65	7.69	8.6E-05	0.94
McClean Lake	A0023	AREV 19	S	1.64	12.7	3.8E-05	0.91
McClean Lake	A0023	AREV 20	S	1.48	4.00	3.4E-05	0.98
McClean Lake	D0013	AREV 21	S	1.48	12.5	3.7E-05	0.88
McClean Lake	D0013	AREV 22	S	1.54	13.4	3.7E-05	0.86
McClean Lake	A0029	AREV 23	S	1.47	14.1	3.2E-05	0.91
McClean Lake	A0029	AREV 24	S	1.43	10.0	3.1E-05	0.96
McClean Lake	M0936	AREV 25	S	1.12	6.99	3.8E-05	0.91
McClean Lake	M0936	AREV 26[a]	S	0.00 [b]	1.05 [b]	4.5E-04 [b]	- [b]
McClean Lake	M0938	AREV 27	S	1.25	4.81	5.4E-05	0.90
McClean Lake	M0938	AREV 28	S	1.30	6.74	3.5E-05	0.88
McClean Lake	M0939	AREV 29	S	1.58	7.03	5.2E-05	0.90
McClean Lake	M0939	AREV 30	S	1.50	2.74	2.9E-05	0.98
McClean Lake	D0020	AREV 31	S	1.81	6.08	7.8E-05	0.91
McClean Lake	D0020	AREV 32	S	1.65	9.12	1.2E-04	0.93

All Concentrate Samples (Except AREV 26)			
Minimum	1.12	2.74	2.9E-05
Median	1.50	7.03	3.8E-05
Maximum	2.95	14.1	1.2E-04

a) Sample AREV 26 was not included in the calculation of summary statistics as the experimental dissolution kinetics curve was very different from the other sample (AREV 25) that originated from the same uranium concentrate sample (M0936), and all the other samples in this study.
b) The parameter estimates failed to converge after 200 iterations.

Figure 4.1   Experimental Dissolution Kinetic Curves for Non-Calcined Samples (Cameco)
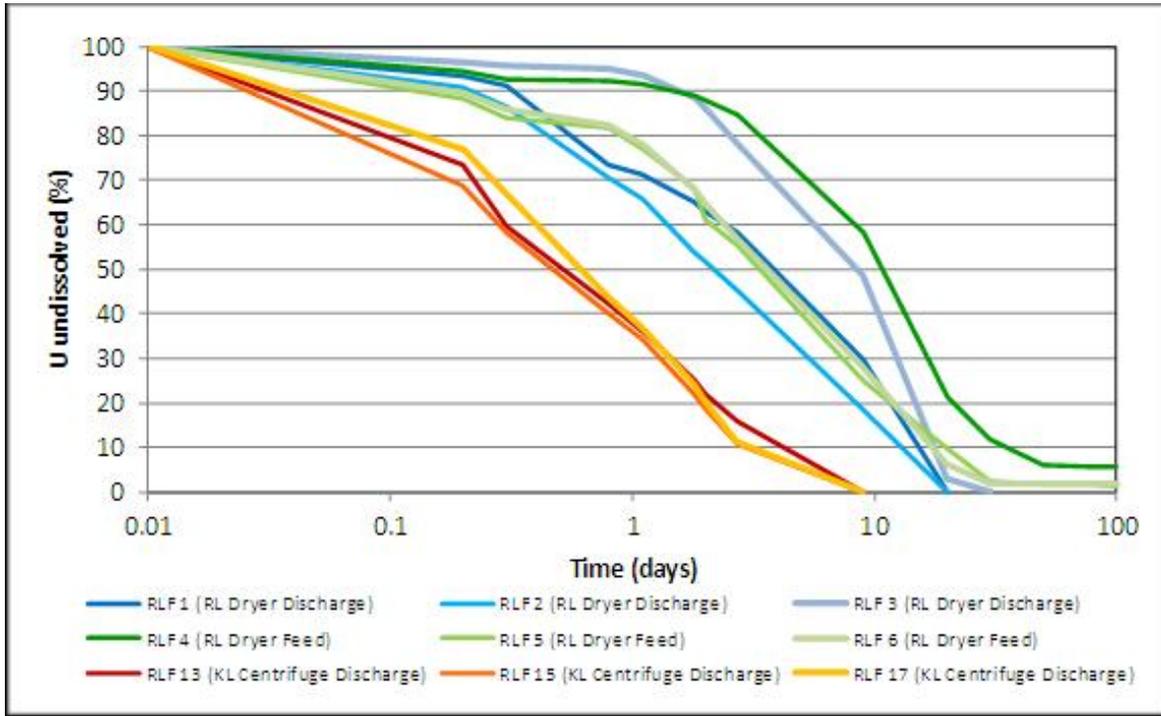

Figure 4.2   Experimental Dissolution Kinetic Curves for Calcined and Ore Samples (Cameco)
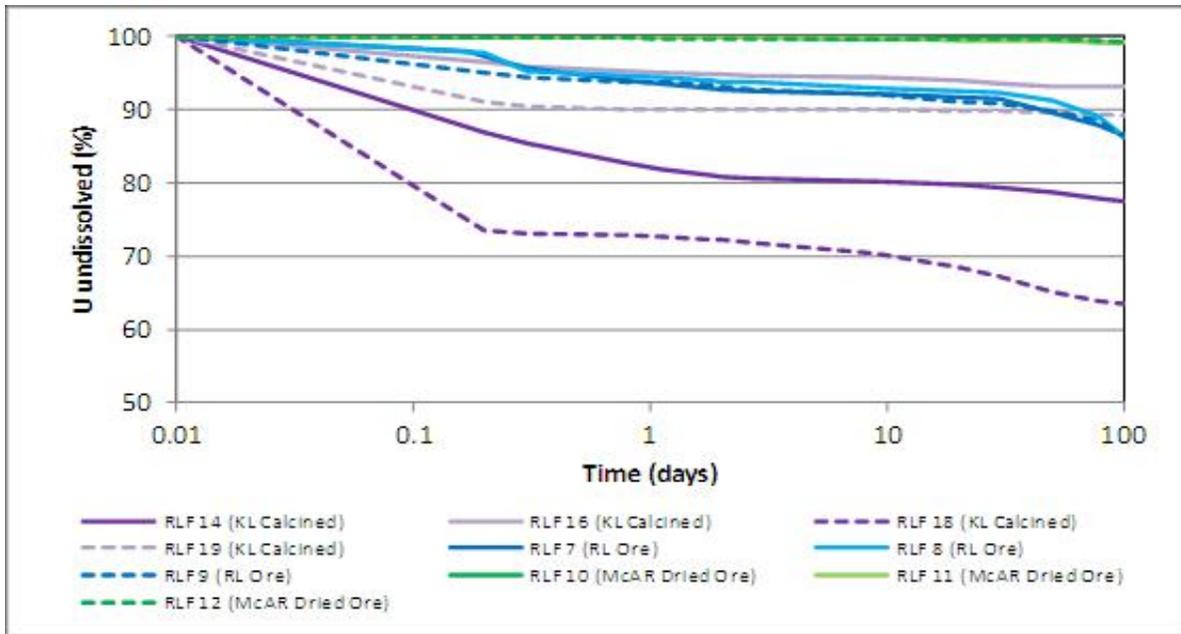
Figure 4.3   Experimental Dissolution Kinetic Curves for McClean Lake Concentrate Samples (AREVA)
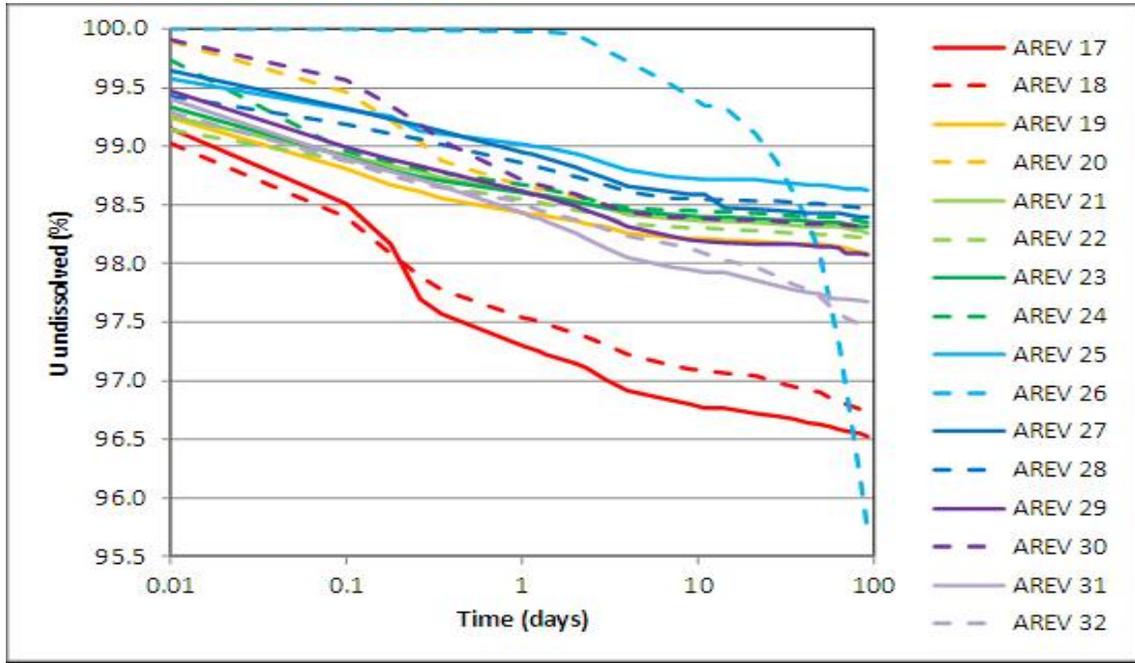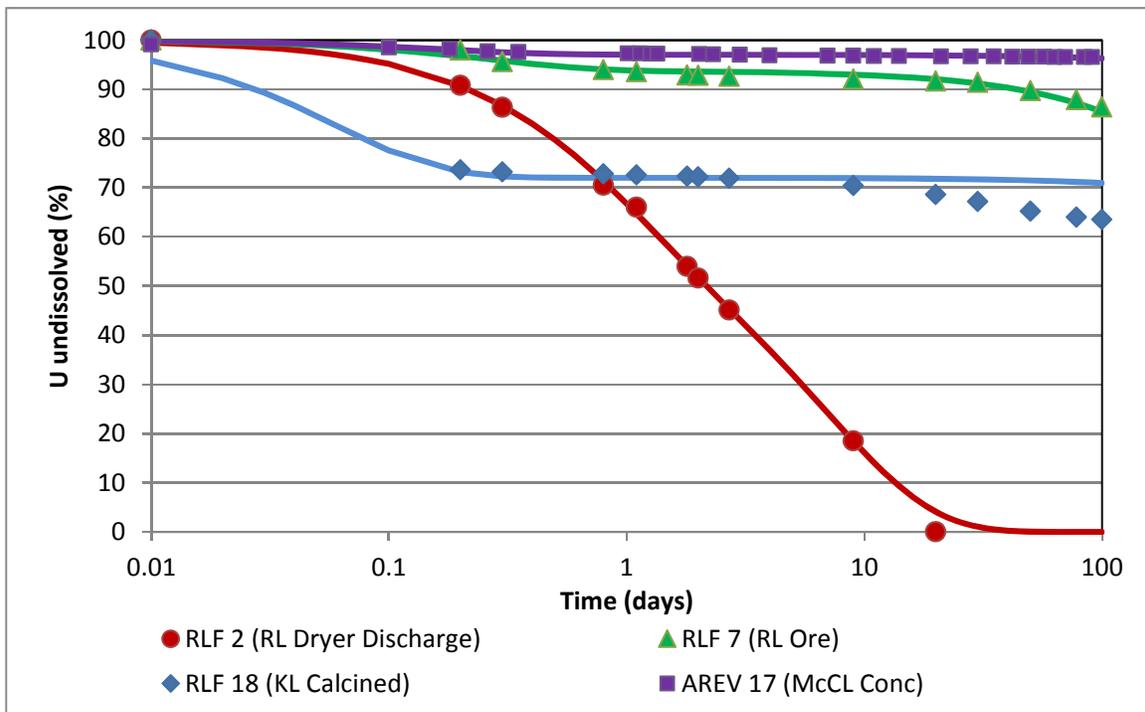

Figure 4.4   Experimental Dissolution Kinetic Curves for a Selection of Samples

The results in Table 4.1 show that the Sr ranges by about an order of magnitude from 1.2 to 16 d^{-1} and the Ss ranges by about four orders of magnitude from 5E-05 to 0.7 d^{-1}. These values vary by the stage of uranium processing and the presence of calcining. The following are the major classifications of the simulated lung fluids:

- Ore Slurries: these typically had low rapid release uranium and had highly reproducible results between the two broad types. The Rabbit Lake leach feed samples were typically about 6% releases with a Sr of 3.6 to 7.9 d^{-1} and a very low Ss of about 1E-03 d^{-1}. The McArthur River dried Ore Slurry had a very low proportion of fast release and due to low amount of this material the Sr could not be calculated and a single decay rate was assumed to determine an Ss of 4.7E-05 to 8.0E-05d^{-1}.

- Dryer Feed and Discharge: there were six samples that are apparently divided into three groups. RLF-1 and RLF-2 (dryer discharge) had a higher fast release proportion (24 to 37%) compared to RLF-5 and RFL-6 (12 to 13%) while the Ss was similar to all four samples. The Sr for RLF-1 and RLF-2 was lower than that for RFL-5 and RLF-6. RLF-3 and RLF-4, although one dryer discharge sample and a dryer feed sample, had very little free release and needed to be fit with single decay model. The Ss for this was lower than the Ss for the other four samples.

- Uncalcined Centrifuge: relatively high free release fraction (24% to 38%) with fairly uniform Sr from 5.4 to 8.4 d^{-1}. The Ss was generally the highest of all samples ranging from 0.6 to 0.7 d^{-1}.

- Calcined Materials: a smaller amount of free release (5% to 28%) and a bit higher Sr of 5.8 to 16 d^{-1} when compared to the uncalcined centrifuge materials (possibly some mixing of calcined and non-calcined material). The largest difference with the uncalcined materials is the very low Ss which approaches the Ss in ore samples.

The results in Table 4.2 indicate that all of the samples from McClean Lake have a low rapid release fraction (less than 3%) and the Sr and Ss values are fairly uniform with about a one order of magnitude range. As noted, AREV 26 was from the same uranium concentrate sample as AREV 25; however, the experimental dissolution kinetics curve was very different from AREV 25 and from the other samples in the study. The reasons for this discrepancy are not known and it is suggested that these values are inappropriate for modelling dose.

The defined mixtures and generic solubility classes that were modelled are provided in Table 4.3 and were selected using professional judgement.

Table 4.3   Solubility Examples with Defined Mixtures

Sample	Sample Type	Fr (%)	Sr	Ss
RLF-7 (Cameco)	Leach Feed Ore	6.22	3.67	9.3E-04
AREV-17 (McLean Lake)	Concentrate	2.95	5.93	7.7E-05
RLF-2 (Cameco)	Dryer Discharge	36.77	1.15	1.4E-01
RLF-18 (Cameco)	Calciner Product	27.96	16.16	1.5E-04
Nominal Canadian Ore				
Reference	Radionuclides	Fr (%)	Sr	Ss
Marsh 2012[a]	Uranium	22	0.78	1.4E-03
Duport 1991 as reported in Marsh 2012	Thorium	14	4.56	6.8E-04
Duport 1991 as reported in Marsh 2012	Radium	11	7.32	4.1E-04
Duport 1991 as reported in Marsh 2012	Lead	26	3.91	1.0E-03
Marsh 2012[b]	All Others	18	4.1	8.9E-04

a) U-238, U-234 and U-235

b) Assumed for other long lived radionuclides in uranium ore dust. These values are the arithmetic mean of the above.

5.0   DOSIMETRIC MODELLING

Dosimetric modelling has been carried out for a standard adult worker exposed to uranium ore, calcined product and dryer discharge product. The effective dose and the absorbed dose to organs were calculated by implementing the following:

- Human Respiratory Tract Model (HRTM), ICRP Publication 66 (ICRP, 1994a);
- Gastrointestinal (GI) Tract Model, ICRP Publication 30 (ICRP, 1979);
- Biokinetic Models for Polonium, Lead and Radium, ICRP Publication 67 (ICRP, 1993);
- Biokinetic Models for Uranium and Thorium, ICRP Publication 69 (ICRP, 1995d);
- Model for actinium and protactinium with assumption of urinary to faecal excretion ratio of 1:1, ICRP Publication 30 (ICRP, 1982); and,
- Methodology to Calculate Effective Dose, ICRP Publication 60 (ICRP, 1995c).

Doses from decay products formed within the body following uptake to blood of radioisotopes of lead, radium and uranium were evaluated based on the biokinetic behaviour of the specific decay product (i.e., independent kinetics). For other elements, it was assumed that the decay products following uptake in the blood had the same kinetics as the parent of the chain (i.e., shared kinetics). These assumptions are in agreement with the ICRP (1995d).

The modelling was performed using the Public Health England's (PHE) internal dosimetry code PLEIADES (Fell *et al.*, 2007). This code has been used to generate dose coefficients for a series of ICRP publications and CDs from ICRP 67 onwards (e.g. ICRP 1993, 1994b, 1995c, 1995d, 1996, 1997, 2002).

For the uranium ore, it was assumed that the decay products of the U-238 series and the U-235 series were in secular equilibrium at the time of intake. The activity ratio of U-235/U-238 was assumed to be 0.046, which is typical for natural uranium. It was also assumed that there was no Th-232 present in the ore (i.e. activity ratio of Th-232/U-238=0). Therefore, the nuclides considered in the calculations were:

- U-238, U-234, Th-230, Ra-226, Pb-210 and Po-210 of the U-238 chain; and,
- U-235, Pa-231 and Ac-227 of the U-235 chain.

It was assumed that radon (Rn-222) does not escape from the uranium ore particle before intake. However, if 25% of the Rn-222 were assumed to escape from the uranium ore dust particles then the effective dose would reduce by less than about 6% (Duport and Edwardson, 1984). For the non-ore material, it was assumed that only the uranium nuclides were present in the material with activity ratios typical of natural uranium (i.e. 48.87% of U-238, 2.26% of U-235 and 48.87% of U-234 by activity; see Annex A).

Modelling was performed assuming the AMAD of the inhaled aerosol was 5 μm, which is the ICRP default value for a worker, and 10 μm as a sensitivity analysis based on information (personal communication) with Cameco and AREVA that the particle size is actually larger (5 μm – 15 μm) than currently assumed.
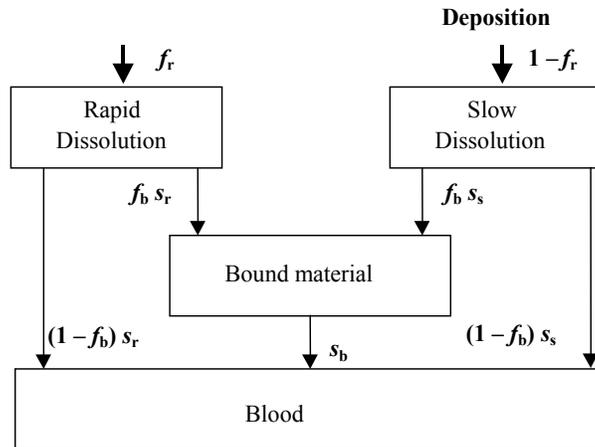
5.1 ABSORPTION PARAMETER VALUES

The HRTM treats clearance of materials from the respiratory tract as a competitive process between absorption into blood and particle transport to the alimentary tract and lymphatic system. It was assumed that particle transport rates were the same for all materials, whereas absorption into blood was assumed to be material-specific. The model assumes that the rate of absorption is the same in all respiratory tract regions except in the anterior nose (region ET_1) where none occurs.

The HRTM treats absorption to blood as a two stage process:

- Dissociation of the particles into a material that can be absorbed into blood. This is termed dissolution.
- Uptake of material dissolved from particles, or material deposited in a soluble form.

Dissolution can be considered as the process in which the deposited material dissolves in the lung fluid. To represent time dependent dissolution, a fraction (Fr) dissolves rapidly at a rate Sr while the remaining fraction (1-Fr) dissolves more slowly at a rate Ss (Figure 5.1). Uptake is usually assumed to be instantaneous but for some elements a fraction of the dissolved material is absorbed more slowly as a result of binding to the respiratory tract components. To represent time dependent uptake, a fraction, f_b, of the dissolved material is assumed to be retained in a bound state, from which it is transferred into blood at a rate s_b, while the remaining fraction (1-f_b) transfers to blood instantaneously (Figure 5.1). Because the bound state is considered to represent the interaction of an element in dissociated (ionic) form with cells forming the lining of the respiratory tract, the bound fraction f_b, and uptake rate s_b, are assumed to be element-specific. The ICRP default absorption types assumes that there is no binding (i.e. the bound fraction, f_b =0).

Figure 5.1   Compartment Model for Time-Dependent Absorption into Blood, Dissolution and Update (ICRP 1994a)

For the present calculations, it was assumed that uptake is instantaneous (i.e. $f_b = 0$) for all the elements considered and that absorption from lung to blood can therefore be represented by dissolution. Parameter values for f_r, s_r, s_s to be used in the calculations are provided in Table 4.3, which were based on the results of solubility measurements of different uranium samples in simulated lung fluid. While there are a range of possible values, professional judgement was used to select notional/nominal data to represent the four different types of uranium samples:

- Leach feed ore (RLF-7, Cameco);
- Concentrate (AREV-17 , McLean Lake);
- Dryer discharge (RLF-2, Cameco); and,
- Calciner product (RLF-18, Cameco).

Calculations were also performed for 'nominal Canadian ore', in which the HRTM absorption parameter values were derived by Marsh, *et al*., 2012 based mainly on the solubility measurements of Duport, *et al*. 1991. The values assumed for f_1, the fraction of activity absorbed to blood from the small intestine, are the ICRP Publication 68 default values given for each element (ICRP, 1994b). [Refer to Table B.1 of Annex B].

It should be noted that the nominal values suggested in this report are useful for generic evaluation for classes of workers. Caution should be taken in applying these values to individual worker exposure scenarios.

5.2   RESULTS OF DOSE CALCULATIONS

For each of the sample types, the committed effective dose (CED), absorbed dose to kidney and absorbed dose to lung per Bq of U-238 inhaled were calculated for the given uranium mixture, as shown in Tables 5.1 and 5.2. The absorbed dose to lung was calculated by calculating the absorbed dose to each region of the lung (bronchial (BB), bronchiolar (bb), alveolar-interstitial (AI), lymphatics (LN$_{TH}$)) and taking a weighted mean using the weighting factors assigned for the partition of radiation detriment among the regions of the lung (ICRP, 1994).

Table 5.1   Committed Effective Dose (CED), Absorbed Dose to Kidney and Absorbed Dose to Lung per Bq of U-238 for Different Uranium Mixtures (5 µm AMAD)

Sample	Sample Type	Absorbed dose to organ (Sv per Bq of U-238)		CED (Sv per Bq of U-238)
		Lung	Kidney	
RLF-7 (Cameco)	Leach Feed Ore	7.0E-06	7.5E-07	3.6E-05
AREV-17 (McLean Lake)	Concentrate	4.4E-06	1.3E-08	1.3E-05
RLF-2 (Cameco)	Dryer Discharge	2.2E-07	1.1E-07	8.3E-07
RLF-18 (Cameco)	Calciner Product	2.9E-06	7.7E-08	8.9E-06
Duport *et al.*, 1991; Marsh *et al.*, 2012	Nominal Canadian Ore	6.3E-06	8.4E-07	3.6E-05

Table 5.2   Committed Effective Dose (CED), Absorbed Dose to Kidney and Absorbed Dose to Lung per Bq of U-238 for Different Uranium Mixtures (10 µm AMAD)

Sample	Sample Type	Absorbed dose to organ (Sv per Bq of U-238)		CED (Sv per Bq of U-238)
		Lung	Kidney	
RLF-7 (Cameco)	Leach Feed Ore	3.4E-06	4.4E-07	2.0E-05
AREV-17 (McLean Lake)	Concentrate	2.1E-06	7.2E-09	7.8E-06
RLF-2 (Cameco)	Dryer Discharge	1.1E-07	5.8E-08	4.3E-07
RLF-18 (Cameco)	Calciner Product	1.4E-06	5.4E-08	5.5E-06
Duport *et al.*, 1991; Marsh *et al.*, 2012	Nominal Canadian Ore	3.1E-06	5.0E-07	2.0E-05

5.3   RESULTS OF BIOASSAY CALCULATIONS

Bioassay calculations of urinary excretion rates and the retention in the lung and kidney following 1 Bq intake of U-235 for the different sample types were completed. Figures 5.2 to 5.4 provide the retention in lung and kidney, and the urinary excretion rates following the intake of 1 Bq of natural uranium assuming an AMAD of 5 µm for each of the sample mixtures given in Table 4.3.

Figure 5.2   **Retention in Lung Following 1 Bq Intake of Natural Uranium for Different Materials**
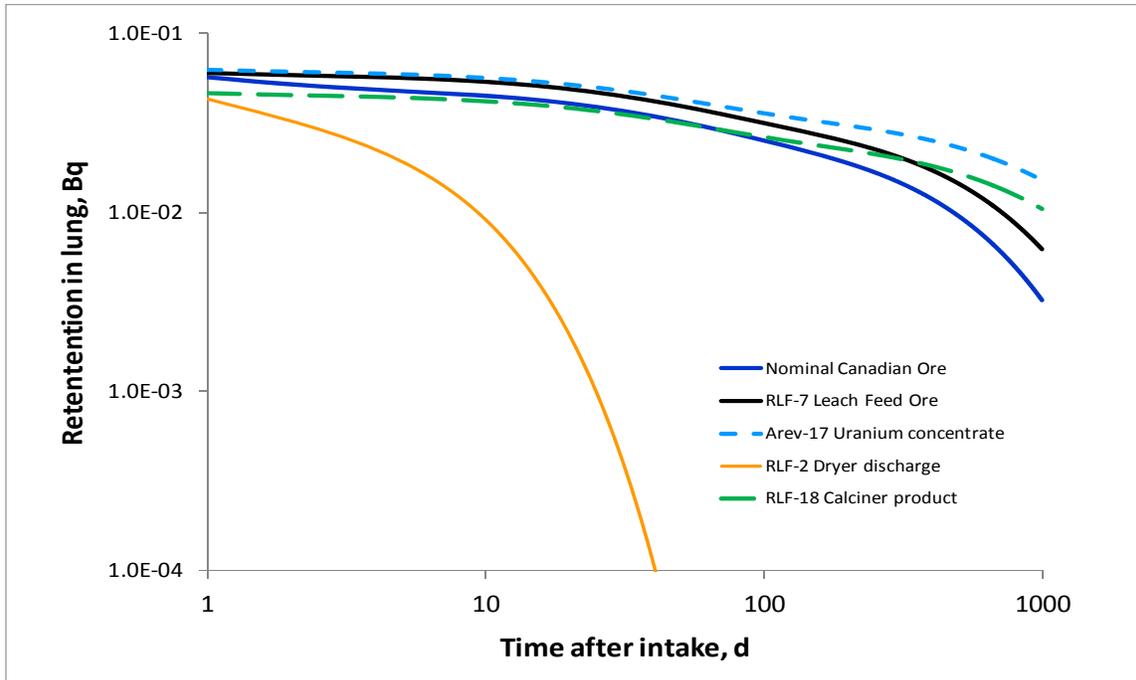
Figure 5.3   **Retention in Kidneys Following 1 Bq Intake of Natural Uranium for Different Materials**
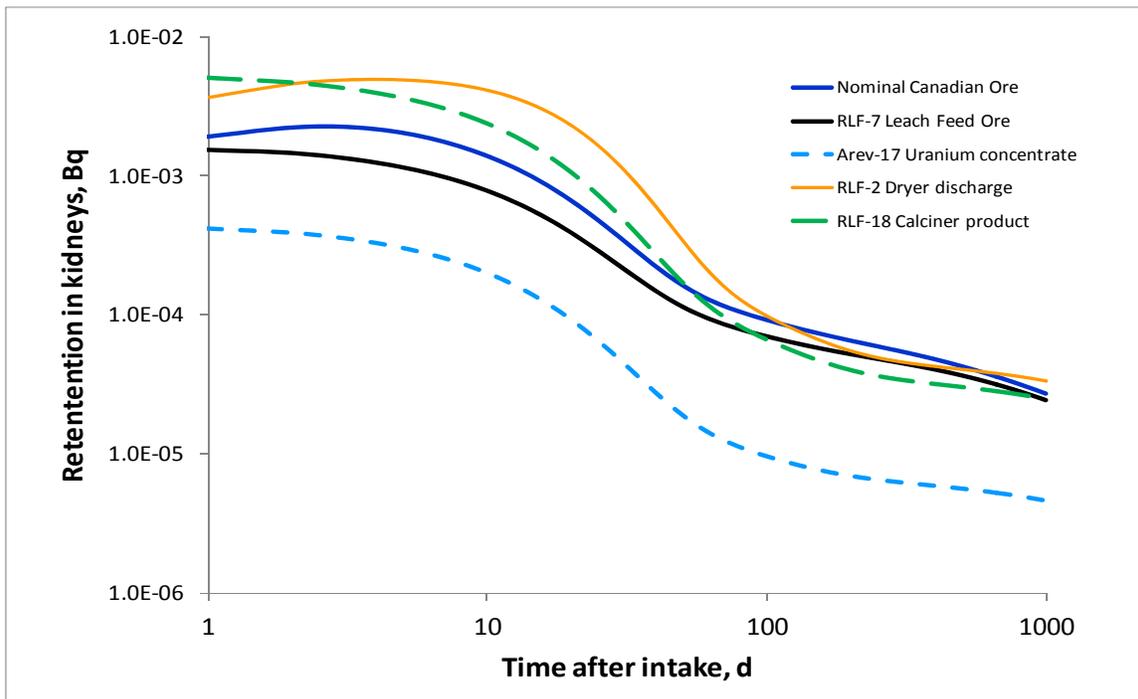
Figure 5.4   Daily Excretion in Urine Following 1 Bq Intake of Natural Uranium for Different Materials
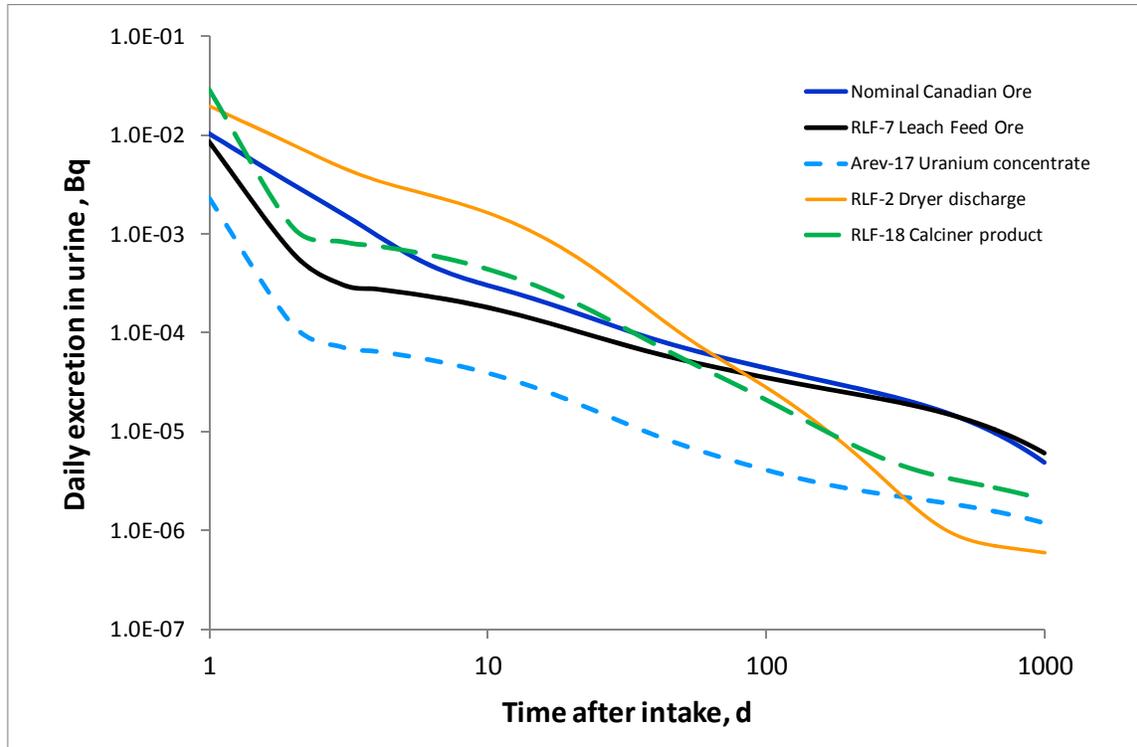
The daily urinary excretion rate is the total activity in a 24- hour urine sample at the end of collection (not at analysis). In other words, the daily excretion rate at time *t* is the activity excreted between times *t* and *t*-1 where *t* is measured in days. If urine samples are collected over periods less than 24 hours, then they should be normalised to an equivalent 24 hour value.

6.0   ESTIMATION OF ANNUAL LIMIT ON INTAKE (ALI)

6.1   DISCUSSION

Based on the assumptions discussed in Chapters 3 and 4, modelling results in Chapter 5, number of alpha particles present in material and assuming a reference effective dose of 20 mSv, the proposed ALIs for different uranium mixtures with two different AMADs (5 µm and 10 µm) are calculated and provided in Table 6.1.

Table 6.1   Proposed and Current ALIs

Sample	Sample Type	Number of Alpha Particles[a]	Proposed ALI[b] (Bq Total Alpha) 5 µm	Proposed ALI[b] (Bq Total Alpha) 10 µm	Current ALI (Bq Total Alpha
RLF-7 (Cameco)	Leach Feed Ore	8[c]	4,500	8,000	2,800
Duport *et al.*, 1991, Marsh *et al.*, 2012	Nominal Canadian Ore	8[c]	4,500	8,000	2,800
RLF-2 (Cameco)	Dryer Discharge (dried yellowcake)	2[d]	48,000	93,000	290,000
AREV-17 (McLean Lake)	Concentrate (calcined yellowcake)	2[d]	3,100	5,100	3,300
RLF-18 (Cameco)	Calciner Product (calcined yellowcake)	2[d]	4,500	7,300	3,300

a)   Number of alpha particles corresponds to both proposed and current ALIs, with exception of Ore (see footnote c).
b)   Proposed ALI in Bq Total Alpha = number of alpha particles * Proposed ALI in Bq U-238.
c)   As indicated in Section 5.0, it was assumed that no Rn-222 escaped from the uranium ore particle before intake; therefore, all 8 alpha particles would be present. However, the current ALI was calculated assuming 25% of the radon escaped; thus, 7 alpha particles were assumed to be present (i.e., 4 alpha particles + 0.75*4 alpha particles = 7 alpha particles).
d)   Assume materials are in equilibrium; therefore, only include U-238 and U-234 alpha particles as U-235 is at 4.6%.

As shown in Table 6.1, the ALIs with a 5 µm particle size are comparable (except for dried yellowcake) to the current ore ALIs. For the 10 µm particle size, the ALIs proposed in this report are about twice as large as the current ALIs (except for dried yellowcake). For yellowcake, as previously discussed, solubility is affected by thermal processing with calcined yellowcake having a lower ALI than dried product.

For present purposes, we suggest nominal (default) ALIs (Table 6.2) based on the current assumption of 5 µm particles with the alternative of using the ALIs for 10 µm particles where supported by measurement. It should be noted the proposed ALIs in this report are based on radiological dose; however, in some instances the limiting factor for ALIs is chemical toxicity[12].

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[12] The amount of uranium retained in the kidney and the reference primary guidance level of 1.0 $\mu gU/g_{kidney}$ (ORNL 2012) are used to estimate the ALI based on chemical toxicity. This reference primary guidance level is used to derive immediate action levels, while the equilibrium value of 0.3 $\mu gU/g_{kidney}$ is used to derive investigation levels (ORNL 2012).

Table 6.2   Recommended Default ALIs for Ore and Yellowcake

Material	Proposed Default ALI (Bq Total Alpha)
Ore	4,500
Dried Yellowcake	48,000
Calcined Yellowcake	3,100

It should be noted that the recommended ALIs are based on the caveat that companies should be allowed, perhaps encouraged, to provide product specific ALIs when justified.   It is recommended that the product specific ALIs be calculated using the same general methodology that is described in this report. Therefore, the following steps should be taken when estimating product specific ALIs:

- obtain representative solubility data for all materials of interest;
- use solubility data to derive absorption parameters for each material of interest;
- determine appropriate particle size, radionuclide distribution and f_1 values for all materials of interest;
- use material specific data (e.g., absorption parameters, particle size, etc.) as inputs into dosimetric modelling (e.g., PLEIADES, IMBA, etc.);
- outputs of dosimetric modelling should include committed effective dose factors (in units of Sv/Bq) for each material of interest; and,
- using committed effective dose factors (Sv/Bq) and a reference dose level (e.g., 20 mSv), calculate ALI for each material of interest.

7.0   REFERENCES

Atomic Energy Control Board (AECB) 1995. *Atomic Energy Control Board Interim Annual Limits on Intake for Long-Lived Radioactive Dust.* January 2.

Atomic Energy Control Board (AECB) 1994. *Phase I Extension: Physical Characteristics and Solubility of Long-Lived Airborne Particulates in Uranium Producing and Manufacturing Facilities.* AECB Project No. 4.103.2, INFO-0363-1/Rev.1.

Atomic Energy Control Board (AECB) 1993. *Physical Characteristics and Solubility of Long-Lived Airborne Particulates in Uranium Producing and Manufacturing Facilities – Phase II.* AECB Project No. 4.103.2, INFO-0363-2.

Atomic Energy Control Board (AECB) 1989. *Physical Characteristics and Solubility of Long-Lived Airborne Particulates in Uranium Producing and Manufacturing Facilities.* A report upon completion of the Extension to Phase I of the study, INFO-0363-1.

Atomic Energy Control Board (AECB) 1988. *Physical Characteristics and Solubility of Long-Lived Airborne Particulates in Uranium Producing and Manufacturing Facilities.* A report upon completion of Phase I of the study, INFO-0363.

Blauer M. and S. Brown 1980. *Physical and Chemical Parameters Affecting the Dissolution of Yellowcake in Simulated Lung Fluids*. Abstracts of the 25th Annual Meeting of Health Physics Society, Seattle, Paper # 177. Pergamon Press.

Blauer, M., J. Kent and N. Dennis 1982. *Dissolution Fractions and Half Times of Single Source Yellowcake in Simulated Lung Fluids.* Health Physics, 42, 469-477.

Cameco 2013. http://www.cameco.com/mining/experience/. Accessed on January 16, 2013.

Cameco 2011. US NRC ADAMS, Ascension # ML110760153, J Schmuck, Cameco Resources to R Burrows, U.S. NRC, March 15.

Cameco 2010. US NRC ADAMS, Ascension # M102640195, T. Young, Cameco Resources to R. Burrows, U.S. NRC, September 17.

Cameco 2005. *Solubility of Radionuclides in Simulated Lung Fluid.* PHR·P7665, Report No. TD 05-09, November.

Canadian Nuclear Safety Commission (CNSC) 2012. *Evaluation of Default Annual Limits on Intake (ALI) for Yellowcake and Uranium Ore*. Contract No. 87055-11-0636.

Duport P., R. Robertson, K. Ho and F. Horvath 1991. *Flow-Through Dissolution of Uranium-Thorium Ore Dust, Uranium Concentrate, Uranium Dioxide, and Thorium Alloy in Simulated Lung Fluid*. Radiat. Prot. Dosim. 38(1/3):121-133.

Eidson, A.F. and J.A. Mewhinney 1983. *In Vitro Dissolution of Respirable Aerosols of Industrial Uranium and Plutonium Mixed Oxide Nuclear Fuels*. Health Physics, 45:1023-1037.

Eidson, A.F. and W.C. Griffith 1984. *Techniques for Yellowcake Dissolution Studies In Vitro and Their Use in Bioassay Interpretation*. Health Physics, 46: 151-163.

Eidson, A.F. 1994. *The Effect of Solubility on Inhaled Uranium Compound Clearance: A Review*. Health Physics, 67: 1-14.

Fell, T.P., A.W. Phipps and T.J. Smith 2007. *The Internal Dosimetry Code PLEIADES*. Radiat. Prot. Dosim. 124(4): 327-338.

Government of Canada 1994. Memo to R. McCabe: *SAWR #3043 ALIs for non-calcined Uranium, Calcined Uranium and Uranium Ore Dust in Saskatchewan*. Memo from F. Horvath, cc: R. Chatterjee and K. Bundy. October 28.

International Commission on Radiological Protection (ICRP) 2002. ICRP CD1: Database of Dose Coefficients: Workers and Members of the Public. Elsevier.

International Commission on Radiological Protection (ICRP) 1997. *Individual Monitoring for Internal Exposure of Workers*. ICRP Publication 78. Annals of the ICRP, Vol. 27 (3–4).

International Commission on Radiological Protection (ICRP) 1996. *Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 5. Compilation of Ingestion and Inhalation Dose Coefficients*. ICRP Publication 72. Annals of the ICRP, Vol. 26 (1).

International Commission on Radiological Protection (ICRP) 1995a. *Dose Coefficients for Intakes of Radionuclides by Workers*. ICRP Publication 68. Annals of the ICRP, Vol. 24 (4).

International Commission on Radiological Protection (ICRP) 1995b. *Age-Dependent Doses to Members of the Public from Intake of Radionuclides Part 4: Inhalation Dose Coefficients*. ICRP Publication 71. Annals of the ICRP, Vol. 25 (3-4).

International Commission on Radiological Protection (ICRP) 1995c. *1990 Recommendations of the International Commission on Radiological Protection.* ICRP Publication 60. Annals of the ICRP, Vol. 21(1-3).

International Commission on Radiological Protection (ICRP) 1995d. *Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 3 Ingestion Dose Coefficients.* ICRP Publication 69. Annals of the ICRP, Vol. 25 (1).

International Commission on Radiological Protection (ICRP) 1994a. *Human Respiratory Tract Model for Radiological Protection.* ICRP Publication 66. Annals of the ICRP, Vol. 24 (1-3).

International Commission on Radiological Protection (ICRP) 1994b. *Dose Coefficients for Intakes of Radionuclides by Workers.* ICRP Publication 68. Annals of the ICRP, Vol. 24 (4).

International Commission on Radiological Protection (ICRP) 1993. *Age-Dependent Dose to Members of the Public from Intake of Radionuclides: Part 2 Ingestion Dose Coefficients.* ICRP Publication 67. Annals of the ICRP, Vol. 23 (3-4).

International Commission on Radiological Protection (ICRP) 1982. *Limits for Intakes of Radionuclides by Workers.* ICRP Publication 30, Part 3. Annals of the ICRP, Vol. 6 (2/3).

International Commission on Radiological Protection (ICRP) 1979. *Limits for Intakes of Radionuclides by Workers*. ICRP Publication 30, Part 1. Annals of the ICRP, Vol. 2 (3-4).

Kalkwarf, D.R. 1983. *Dissolution Rates of Uranium Compounds in Simulated Lung Fluid*. Sci. Total Environ. 28:405-414.

Marsh J.W., E. Blanchardon, D. Gregoratto, W. Hofmann, K. Karcher, D. Nosske, and L. Tomasek 2012. *Dosimetric Calculations for Uranium Miners for Epidemiological Studies*. Radiat Prot Dosimetry, 149 (4): 371-383.

Merritt, R.C. 1971. *The Extractive Metallurgy of Uranium*. Colorado School of Mines Golden.

Metzger, R., D. Wichers, J. Vaselin and P. Velasquex 1997. *Solubility Characterization of Airborne Uranium from an In Situ Uranium Processing Plant*. Health Physics, 72:41-422.

Morrow 1966. *Deposition and retention models for internal dosimetry of the human respiratory tract.* Task Group on Lung Dynamics. Health Phys.12, 173-207.

Moss, O.R. 1976. *Simulants of Lung Interstitial Fluid.* Health Physics, 36:447-448.

Oak Ridge National Laboratory (ORNL) 2012. *Controlling Intake of Uranium in the Workplace: Applications of Biokinetic Modelling and Occupational Monitoring Data.* ORNL/TM-2-12/14. Prepared by R.W. Leggett, K.F. Eckerman, C.W. McGinn and R.A. Meck.

Spitz, H. and B. Robinson 1981. *Measurements to Determine the Solubility of Uranium Yellowcake Materials in the Lung of Millworkers Following Acute Exposure.* DOE Contract, DE - ACO6-76RLO 1830. October.

Tairova, G., M. Boucher, K. Toews, M. Ioffe, J. Takala, M. Murchie and E. Ozberk 2010. *Solubility of Radionuclides in Simulated Lung Fluid.* Proceedings of the 3rd International Conference on Uranium and 40th Annual Hydrometallurgy Meeting. Saskatoon, Saskatchewan, Canada.

Wyoming Mineral Corporation (Westinghouse Electric Corporation) 1980. *Yellowcake Solubility Studies 1979 -1980.* September.

ANNEX A

ACTIVITY RATIOS FOR NATURAL URANIUM

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The activity ratios were calculated from nuclear half-lives (ICRP 2008), atomic mass and isotopic compositions for uranium taken from National Institute of Standards and Technology (NIST) website (NIST 2013). The resulting activity ratios are provided in Table A.1.

Table A.1   Activity Ratio for Natural Uranium

Isotope	Atomic mass	Isotopic Composition (*IC*)[a]	Half life (y)	Activity (Bq/g of U-nat)[b]	Activity ratios	Adjusted Isotopic Composition[c]	Equilibrium Activity Ratio
U-234	234.0410	0.00540%	2.457E+05	12223	48.62%	0.00545%	48.87%
U-235	235.0439	0.72040%	7.037E+08	569	2.26%	0.72040%	2.26%
U-238	238.0508	99.27420%	4.468E+09	12347	49.12%	99.27415%	48.87%

a) Isotopic Composition is the mole fraction of the various isotopes
b) Activity of each isotope per gram of U-mix = $IC \times N_A / A_m \times \ln(2)$ / half-life(s) [1 year = 365.25 days]
   Avogadro constant (N_A) = 6.022 141 29 x 10^{23} mol^{-1} (http://physics.nist.gov/cuu/Constants/index.html).
   Relative atomic mass of U-mix, (A_m) = 238.0289.
c) Isotopic composition of U-234 and U-238 was adjusted to obtain activity equilibrium between these isotopes.

REFERENCES

International Commission on Radiological Protection (ICRP) 2008. *Nuclear Decay Data for Dosimetric Calculations.* ICRP Publication 107. Annals of the ICRP, Vol. 38 (3).

NIST Physical Measurement Laboratory 2013. *Atomic Weights and Isotopic Compositions.* (http://www.nist.gov/pml/data/comp.cfm)

ANNEX B

ASSUMED F_1 VALUES

ANNEX B ASSUMED F1 VALUES

The f_1 values assumed in these calculations are the ICRP Publication 68 (ICRP 1994) default values given for each element. As default values are given for different compounds of an element, the default value associated with the most appropriate absorption type for a given element was assumed. The f_1 values used in the modelling are provided in Table B.1.

Table B.1   Assumed f_1 Values

Sample	Absorption Type[a]	U	Th	Ra	Pb	Po	Pa	Ac
RLF-7 (Cameco)	M	0.02	0.0005	0.2	0.2	0.1	0.0005	0.0005
AREV-17 (McLean Lake)	S	0.002	0.0002	0.2	0.2	0.1	0.0005	0.0005
RLF-2 (Cameco)	F	0.02	0.0005	0.2	0.2	0.1	0.0005	0.0005
RLF-18 (Cameco)	M	0.02	0.0005	0.2	0.2	0.1	0.0005	0.0005
Nominal Canadian Ore	b	0.02	0.0005	0.2	0.2	0.1	0.0005	0.0005

a) ICRP Publication 71 (1995) criteria were used to assign the absorption type for the given samples based on the solubility characteristics given in Table 4.3.
b) Values given for unspecified compounds were assumed to be ICRP default values (ICRP 1994b).

REFERENCES

International Commission on Radiological Protection (ICRP) 1994. *Dose Coefficients for Intakes of Radionuclides by Workers.* ICRP Publication 68. Annals of the ICRP, Vol. 24 (4).

International Commission on Radiological Protection (ICRP) 1995. *1990 Recommendations of the International Commission on Radiological Protection.* ICRP Publication 60. Annals of the ICRP, Vol. 21(1-3).