

## **Radiation Physics Note No. 158**

## Review of Control of Occupational Exposure to Airborne Radioactivity at Fermilab

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# **‡** Fermilab

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## Introduction

The purpose of this Radiation Physics (RP) Note is to document the technical basis of the program for protecting against occupational exposure to airborne radioactivity at Fermilab. It includes a discussion of the nature of airborne radioactivity at particle accelerators, a review of the application of standards to Fermilab and their evolution with time, an assessment of methodologies used to identify and quantify airborne radioactive hazard, and a review of control measures. Extensive references to other materials, including publications and other RP Notes, will be made throughout this document. This Note is intended to be complementary to and not duplicative of RP Note No. 7, "Fermilab Internal Dosimetry Technical Basis Document" (McG13).

In this revision a new Appendix has been added to discuss the calculation of Derived Air Concentrations (DACs) for accelerator-produced radionuclides.

## Nature of Airborne Radioactivity at High Energy Accelerators

The topic of airborne radioactivity has been well-studied by specialists in accelerator radiation protection throughout the historical development of accelerators. Among prominent early works are those of Kase [(Ka67), (Ka68)], Höfert (Ho69), Warren et al. (Wa69), and Rindi and Charalambus (Ri67). Excellent discussions have been given by Patterson and Thomas (Pa73) and by Sullivan (Su92). The International Atomic Energy Agency (IAEA) in two reports also gives very useful summaries [(IA79), (IA88)]. The National Council on Radiation Protection and Measurements (NCRP) has reviewed this topic in its Report No. 144 (NC03). A rather detailed discussion is presented by Cossairt (Co16) that covers in detail the production of airborne radionuclides and their reduction via ventilation and other means.

Moreover, five decades of operational experience at Fermilab as documented in numerous references, a sample of which are cited here, confirms the conclusions of the body of the published literature [(Bu89), (RPN)].

Based on this large volume of knowledge and body of professional experience, the main features of airborne radioactivity at accelerators are as follows:

• Work on activated components can generate dust particles and, rarely, gaseous materials that present an exposure hazard (Ma95). Typical contamination encountered in the accelerator environment is not of high enough activity to present an airborne radioactivity concern. A reasonable value for the resuspension factor<sup>1</sup> of radioactivated dust can be taken to be 10<sup>-6</sup> m<sup>-1</sup> (Ce09). Most contamination wipes taken in the accelerator enclosures are of radioactivated dust and are on the order of a few hundred net counts minute<sup>-1</sup> (ncpm) on a 100 cm<sup>2</sup> wipe sample. 500 ncpm is 2.25 nCi/100 cm<sup>2</sup>, or 22.5 pCi cm<sup>-2</sup>, if one assumes a typical value of 10% counting efficiency<sup>2</sup>. In rare cases, certain contamination wipes may

<sup>&</sup>lt;sup>1</sup> The resuspension factor is the ratio of the airborne concentration to the surface contamination of a contaminant. It is expressed in units of inverse length, commonly in m<sup>-1</sup>.

<sup>&</sup>lt;sup>2</sup> In field work, the assumption of a 10% counting efficiency is typically assumed. More accurate measurements using the automatic sample changer in place at the Fermilab Radionuclide Analysis Facility are provided in FRCM Chapter 5 and documented in detail in RP Note No. 96 as updated April 23, 1996.

be as large as approximately 1000 ncpm, representing a surface contamination of 45 pCi cm<sup>-2</sup>. Applying resuspension factor of  $10^{-6}$  m<sup>-1</sup> (= $10^{-8}$  cm<sup>-1</sup>) to such a "high" value as 50 pCi cm<sup>2</sup> results in an airborne concentration of order 5.0 x  $10^{-13}$  µCi ml<sup>-1</sup>, far less than any tabulated value of Derived Air Concentration (DAC) for accelerator-produced radionuclides. There are commonly two different sets of DAC-values, one related to uptakes of radionuclides (inhalation) and the other for immersion in an atmosphere. Chapter 3 of the Fermilab Radiological Control Manual (FRCM)<sup>3</sup> documents the mitigation measures used at Fermilab to control this hazard. Furthermore, the hazard from this source and its mitigation has been discussed in depth by McGimpsey (McG13). RP Form 25, "Field Analysis of Particulate Air Sample<sup>4</sup>" is used in rare cases to analyze such particulate radionuclides. The proper use of this form results in the determination of the "gross- $\beta$ " activity of the area. Should a somewhat arbitrary but conservative threshold of  $1 \times 10^{-2}$  pCi ml<sup>-1</sup> be exceeded, the assigned Radiation Safety Officer (RSO) is to be contacted. Indeed, reverse application of a resuspension factor of 10<sup>-8</sup> cm<sup>-1</sup> to 1.0x10<sup>-2</sup> pCi ml<sup>-1</sup> results in a wipe activity of (100  $\mu$ Ci) on 100 cm<sup>2</sup>, which corresponds to a count rate of about 3.7x10<sup>5</sup> cpm on the standard Geiger-Mueller (GM) counter in use at Fermilab that is welldocumented to have an approximately 10% efficiency for detecting the radionuclides encountered (see footnote 3). Such an activity level is readily detectable on wipes, even when counted in field conditions with portable instruments. Thus, this threshold is less, in most cases far less, than the DAC-values for all radionuclides that could be present and represents a reasonable threshold for involvement of professional radiation protection experience and judgment. Thus, such airborne contaminants in particulate form would be readily detected by means of the surface wipes at levels far below the DAC-values.

• The dominant stable nuclides in the atmosphere and the isotopic abundances are given in Table 1 using the U. S. Standard Atmosphere (Li00). From these results, it is apparent that three stable target nuclides dominate; <sup>14</sup>N, <sup>16</sup>O, and <sup>40</sup>Ar.

Element or	% By	% Isotopic	$N_j$ (Atoms cm <sup>-3</sup> )
Isotope	Volume	Abundance (Tu05)	
Nitrogen (N <sub>2</sub> )	78.084		4.1959x10 <sup>19</sup>
$^{14}$ N		99.636	4.1806x10 <sup>19</sup>
$^{15}N$		0.364	1.5273x10 <sup>17</sup>
Oxygen (O <sub>2</sub> )	20.9476		1.1256x10 <sup>19</sup>
<sup>16</sup> O		99.757	1.1229x10 <sup>19</sup>
<sup>17</sup> O		0.038	4.2774x10 <sup>15</sup>
$^{18}O$		0.205	2.3075x10 <sup>16</sup>
Argon (Ar)	0.934		2.5094x10 <sup>17</sup>
<sup>36</sup> Ar		0.3336	8.3715x10 <sup>14</sup>
<sup>38</sup> Ar		0.0629	$1.5784 \times 10^{14}$
<sup>40</sup> Ar		99.6035	2.4995x10 <sup>17</sup>

Table 1 Atomic number densities of the most prominent elements and stable nuclides in the atmosphere at sea level at STP (273.15 °K, 760 mm Hg).

<sup>&</sup>lt;sup>3</sup> In this Note the current version of the FRCM at the time of this revision is implicit.

<sup>&</sup>lt;sup>4</sup> See also FRCM Chapter 5 Appendix 5E.

The activation of air proceeds according to Eq. (1), producing an activity concentration *a*(*t<sub>irrad</sub>*) (decays s<sup>-1</sup>, Bq) of a particular radionuclide as a function of irradiation time *t<sub>irrad</sub>* (s) (Co16),

$$a(t_{irrad}) = \frac{\lambda N \sigma \phi}{\lambda + r} \left\{ 1 - \exp\left[ -(\lambda + r)t_{irrad} \right] \right\}.$$
 (1)

In Eq. (1) uniform mixing of the air and irradiation with a constant flux density  $\phi$  (particles s<sup>-1</sup> cm<sup>-2</sup>), of *N* target atoms cm<sup>-3</sup> is assumed. The production cross section for a specific radionuclide is  $\sigma$  (cm<sup>-2</sup>). If the half-life of the radionuclide of interest is  $t_{1/2}$ , the decay constant  $\lambda = \ln 2/t_{1/2}$ . In Eq. (1) *r* takes into account the ventilation of the space and represents the number of air changes per unit time (s<sup>-1</sup>). If there were no ventilation, the product  $N\sigma\phi$  is the activity concentration that would be reached at saturation, when production equals decay of the given radionuclide. With Eq. (1), after a long irradiation time one reaches an "equilibrium" or "saturation" concentration with the quantity in the curly {} brackets approaching a value of approximately unity. Obviously flux densities and particle spectra and composition will differ widely within a given beam enclosure volume. Thus, in practical work this equation must be averaged over the volume, particle types, etc. Upon completion of operation and commencement of a cool-down period,  $t_{decay}$ , Eq. (2) applies,

$$a(t_{irrad}, t_{decay}) = \frac{\lambda N \sigma \phi}{\lambda + r} \left\{ 1 - \exp\left[ -(\lambda + r) t_{irrad} \right] \right\} \exp\left\{ -\lambda t_{decay} \right\},$$
(2)

where  $t_{decay}$  (s) is the time after cessation of operation during which decay takes place.

- From the references listed above, the radionuclides produced from these airborne target nuclides that are of concern for radiation protection purposes are, in order of atomic mass number, <sup>3</sup>H, <sup>7</sup>Be, <sup>11</sup>C, <sup>13</sup>N, <sup>15</sup>O, <sup>38</sup>Cl, <sup>39</sup>Cl, and <sup>41</sup>Ar. This is true for both occupational radiation protection considerations and for environmental protection considerations pertaining to radionuclide releases. It is possible to produce the extremely long-lived <sup>14</sup>C in air at accelerator by means of thermal neutron capture with the  $(n_{thermal},p)$  reaction. While the cross section for this reaction is somewhat large, 1848 mb at 0.025 eV, the extremely long half-life of 5700 years compared with plausible irradiation times and ventilation rates coupled with the rapid decline of cross section with neutron energy precludes the development of measurable concentrations of this radionuclide [see Eq. (1)]. Other shorterlived radionuclides with mass numbers below that of <sup>41</sup>Ar are produced at lesser rates within the beam enclosures. These do not present a potential for personnel exposure within the beam enclosures due to the fact that personnel are precluded from access to these enclosures during beam operations. The short half-lives mean that these short-lived radionuclides decay to negligible levels prior to delayed access or migration of the air from the enclosures due to leakage or engineered ventilation systems.
- Nearly always at accelerators, the radiological hazard from airborne radioactivity is of considerably less significance for worker protection than that presented by the activated components, and of course of complete insignificance compared with the prompt radiation hazards.

- As with all nuclear reactions, the reaction product nuclei are initially produced fully stripped, with all atomic electrons missing. Amplified by the intense levels of ionizing radiation present, it is likely that the nuclei become neutral atoms or become bound in neutral molecules rather rapidly. The physical state of the radionuclides is that expected for the given chemical element for room temperature/pressure conditions. Individual discussions pertinent to each of the significant radionuclides will be provided in the next section.
- Airborne radioactivity hazards can be classified as either inhalation hazards or immersion<sup>5</sup> hazards. Inhalation hazards apply to those radionuclides that will be taken into the body and incorporated, at least for a finite period of time, into body tissues. Non-gaseous components typically represent inhalation hazards, with tritium as a notable exception of a gaseous inhalation hazard. Immersion hazards are presented by gaseous radionuclides that expose the body with photons from gamma-ray emission and/or annihilation radiation along with any  $\beta$ -particle exposure from the decay of the material. The gaseous materials are in general not taken up by the body and retained by bodily organs when the immersion hazard is applicable.
- Ventilation control is used to control both releases to the environment and air quality within beam enclosures. Commonly, ventilation rates are reduced during operations to keep the environmental releases ALARA. During accesses, the ventilation rates are commonly increased to provide the necessary air quality for workers. Thus, upon cessation of operations, the concentrations of the individual radionuclides will be reduced by decay and through dispersal by the ventilation system as discussed in the references [e.g., (Su92), (Co16)]. This implies that the composition of the radionuclides within enclosures during periods when workers are present may differ from that measured during operational conditions. Under certain circumstances this variability of the concentrations can be important in determining the applicability of standards.
- At Fermilab, the radionuclide compositions of individual ventilation streams for the major beam targets, those where the radiological hazards are the most significant, are measured and characterized as part of the program of monitoring environmental releases. This is accomplished using essentially full-time monitoring of the gross activity of the ventilation streams at the ventilation release points commonly called "stacks". The radionuclide compositions are characterized by means of gamma-ray spectroscopy and analyses of the radionuclide decay time using the multi-channel scaling technique. The composition of the released radionuclides within the enclosures may be inferred from these results at the release points by correcting for the radioactive decay during transport to the ventilation stack. This is discussed further below and also in chapter 8 of (Co16). The ventilation stacks are monitored using isokinetic sampling following U.S. EPA guidance (ES17).
- Following standard practice at U.S. Department of Energy accelerators, and others elsewhere, beam enclosures to which entry of workers is denied during accelerator

<sup>&</sup>lt;sup>5</sup> Some references refer to the "immersion" scenario of exposure to airborne radionuclides as "submersion". "Immersion" appears to be a better description of the situation and is preferred in this Note. There is no known technical difference between the two terms.

operations is precluded by hard-wired radiation safety interlocks managed in accordance with FRCM Chapter 10 and are inaccessible to personnel. For these work spaces, occupational radiation protection standards including the posting requirements and the DACs are applicable only when personnel can be present in these spaces.

## **Discussion of Specific Radionuclides**

In this section, the properties of specific radionuclides are discussed. These are the radionuclides found to dominate the radiation field due to airborne radioactivity at large accelerators. They have sufficiently long half-lives to be present in accelerator enclosures during accesses or are released to the environment. For convenience, the half-lives are given in Table 2. The references, for example (Co16), provide values of the production cross sections.

 ${}^{3}H$  – The body of experience at accelerators shows that  ${}^{3}H$  ("T") is in the form of tritiated water (HTO) due to the lack of free hydrogen in the atmosphere needed to produce HT molecules. In beamline enclosures and in airborne releases it is likely suspended as water vapor. The <sup>3</sup>H can be produced directly from spallation reactions on the constituents of air. It is also produced from spallation on the oxygen in cooling water systems and can subsequently enter the enclosure atmospheres by means of evaporation, leaks, and spills. While near-equilibrium concentrations are reachable in water systems, following Eq. (1) the long half-life of <sup>3</sup>H renders reaching a concentration in equilibrium between production in air and decay nearly impossible. This is a far different situation than for the short-lived radionuclides since the production cross section for <sup>3</sup>H is of same order of magnitude as those of the other radionuclides present as discussed below, according to the references cited in (Co16), a maximum of approximately 30 mb (millibarns). Thus the activity concentration of <sup>3</sup>H is much less than those found for other common radionuclides found in the air at accelerators due to the much smaller decay constant (reciprocal mean-life) for this long-lived radionuclide. Due to its low  $\beta$ -decay end point energy, tritium is difficult to detect. A method, albeit retrospective in nature, has been devised to measure the tritium concentrations found in the condensate collected in dehumidification systems based upon refrigeration technology to assess tritiated water concentrations in accelerator atmospheres. This approach has been discussed in detail by Lauten et al. (La13). For radiation protection purposes, tritium is taken to be an inhalation and skin absorption hazard rather than an immersion hazard.

 $^{7}$ Be – This radionuclide is also of significantly long half-life so that, like tritium, it is not expected to reach equilibrium concentrations comparable to those found for the short-lived radionuclides. According to the references cited such as (Co16), the production cross section has a maximum value of 10 mb. At production this radionuclide is expected to appear as a solid particulate and quickly become neutral atoms. Because of its chemical properties, it is readily accumulated on the filters used in particulate air samplers. If it travels with any net positive electrical charge as an ion, the accumulation on particulate filters is enhanced. Its presence can be identified and quantified by use of Geiger-Mueller detectors and gamma-ray spectroscopy, respectively. For radiation protection purposes it is considered to be an inhalation hazard rather than an immersion hazard.

 ${}^{11}C$  – This short-lived positron-emitter is primarily produced by spallation reactions on the constituents of air. Its main detection signature is the two 0.511 MeV photons produced in the

annihilation of the positron associated with each decay. As tabulated in (Co16), the maximum production cross section is about 10 mb. For this radionuclide and for <sup>13</sup>N and <sup>15</sup>O, also positronemitters that decay in the same manner, "identification" amounts to measuring the sum of the concentrations of the trio <sup>11</sup>C, <sup>13</sup>N, and <sup>15</sup>O. From this total concentration, the quantification of the individual radionuclides is done by capturing a grab sample of the air using isokinetic techniques and measuring the decay of the activity as a function of time. The decay curve is fit to the sum of the exponential terms representing the individual decays of all radionuclides present in accordance with Eq. (2). <sup>11</sup>C is found in the gaseous state because when it is produced in the air, it readily combines with oxygen to from CO<sub>2</sub>. For radiation protection purposes, due to the gaseous form, the immersion hazard is the applicable consideration.

 $^{13}$ N – This is another short-lived positron-emitter with characteristics similar to those of  $^{11}$ C. From the references cited in (Co16) the maximum production cross section is 10 mb. For radiation protection purposes, due to the gaseous form of nitrogen at room temperature, the immersion hazard is the applicable consideration. The concentration is determined along with that of  $^{11}$ C and  $^{15}$ O.

 ${}^{15}$ O – This is the third short-lived positron-emitter with characteristics similar to those of  ${}^{11}$ C. From the references cited in (Co16) the maximum production cross section is 40 mb. For radiation protection purposes, due to the gaseous form of oxygen at room temperature, the immersion hazard is the applicable consideration. The concentration is determined along with that of  ${}^{11}$ C and  ${}^{13}$ N.

 ${}^{38}$ Cl – This radionuclide is produced by the  ${}^{40}$ Ar( $\gamma$ ,pn) ${}^{38}$ Cl reaction. It has a reaction threshold of 20.6 MeV (NN14) and, from the references cited in (Co16), a maximum production cross section of 4 mb.  ${}^{38}$ Cl is a gamma-emitter that is readily detectable using gamma ray spectroscopy. Given the chemical reactivity, it can readily combine with other materials to become solids capable of precipitating out or it can be in gaseous form. For radiation protection purposes, it is an inhalation hazard rather than an immersion hazard.

<sup>39</sup>Cl – This radionuclide is produced by the <sup>40</sup>Ar( $\gamma$ ,p)<sup>39</sup>Cl reaction. It has a reaction threshold of 12.5 MeV (NN14) and, from the references cited in (Co16), a maximum production cross section of 7 mb. Like <sup>38</sup>Cl, <sup>39</sup>Cl is a gamma-emitter that is readily detectable using gamma ray spectroscopy. Given the chemical reactivity, it can readily combine with other materials to become solids capable of precipitating out or it can be in gaseous form. For radiation protection purposes, it is an inhalation consideration rather than an immersion hazard.

<sup>41</sup>Ar – This radionuclide is produced by the capture of thermal neutron by <sup>40</sup>Ar by means of the ( $n_{thermal},\gamma$ ) reaction. At a thermal neutron energy of 0.025 eV, this reaction has a cross section of 660 mb according to the references cited in (Co16). Given the extreme variability of thermal neutron flux densities within accelerator beam enclosures, the production of <sup>41</sup>Ar is highly variable. The produced <sup>41</sup>Ar will be gaseous given its status as a noble gas. Measurements of the composition of the airborne radioactivity at Fermilab have found a wide variety of relative concentrations of <sup>41</sup>Ar attributed to this variability of thermal neutron flux densities. A "rule of thumb" based on experience at Fermilab is that the concentration of the <sup>41</sup>Ar can be taken to have a median value of about 2.5% of the total activity concentration. This is clearly an approximation that, while useful for purposes of estimation, commonly needs measurements for verification. With beta-gamma sensitive radiation detectors, the presence of <sup>41</sup>Ar in a grab

sample will be evident due to the half-life involved and the spectroscopic signature of a 1.293 MeV gamma ray photon. For radiation protection purposes, due to the gaseous form of the noble gas argon at room temperature, the immersion hazard is the applicable consideration.

## Protection Standards for Radiological Work Involving Airborne Radioactivity

Radiation Protection Programs at U.S. Department of Energy Facilities including Fermilab are required to be in compliance with the provisions of 10 CFR 835. The Fermilab primary implementation document is the DOE-approved Radiation Protection Program (RPP), most recently revised in October 2018. 10 CFR  $835^6$  provides values for the DACs for various radionuclides, many specifically listed in the Regulation. A worker breathing air at an average concentration of 1.0 DAC, as specified by DOE, of a given radionuclide, assuming no other radionuclides to be present, for an entire working year of 2000 hours would receive 5000 mrem (0.05 Sv) of committed effective dose from that pathway. When mixtures of *n* different radionuclides are encountered, Eq. (3) must be satisfied,

$$\sum_{i}^{n} \frac{C_{i}}{DAC_{i}} < 1, \tag{3}$$

where  $C_i$  is the concentration of the *i*<sup>th</sup> radionuclide compared with its individual DAC-value  $DAC_i$ . The quantity now known as the DAC is associated with specific definitions of organ weighting factors established by the International Commission on Radiation Protection (ICRP)<sup>7</sup>. 10 CFR 835 tabulates two different sets of DAC-values, one related to inhalation of radionuclides, 10 CFR 835 Appendix A, and the other for immersion, 10 CFR 835 Appendix C. Furthermore, 10 CFR 835 defines an "airborne radioactivity area" as an area accessible to individuals in which the applicable DAC, for inhalation or immersion conditions, is likely to be exceeded or in which an individual present in the area (presumably an occupational worker), could receive an intake exceeding 12 DAC-hours in a week. 12 DAC-hours corresponds to a committed effective dose of 30 mrem (300 µSv).

A quantity analogous to the DAC used in former years under other systems of radiation protection, that are now considered to be obsolete, was called the Maximum Permissible Concentration in Air (MPC<sub>a</sub>). As with the present system, a worker breathing air at an average concentration of 1.0 MPC<sub>a</sub> for a working year of 2000 hours would receive 5000 mrem (0.05 Sv) of dose equivalent from that pathway and mixtures would satisfy an equation analogous to Eq. (3). For the eight airborne radionuclides that dominate the airborne radionuclide composition at accelerators, Table 2 gives the current values of DACs along with those of the now obsolete MPC<sub>a</sub>s. The former values established by ICRP Committee II (IC60) are included for ready comparison with the published literature. All values listed are referenced to a 5000 mrem (0.05 Sv) annual limit on committed effective dose, or in former system, a whole body dose equivalent of 5000 mrem (0.05 Sv) in a year.

<sup>&</sup>lt;sup>6</sup> In this Note, unless otherwise noted 10 CFR 835, "Occupational Radiation Protection" (i.e., at U.S. Department of Energy facilities) always refers to the current version in place at the time of writing.

<sup>&</sup>lt;sup>7</sup> One must use caution in referencing ICRP publications as the ICRP for some exposure pathways now recommends a 2000 mrem (0.02 Sv) annual limit on committed effective dose while DOE currently uses a 5000 mrem (0.05 Sv) annual limit on committed effective dose.

The regulatory tables commonly give different values for certain chemical forms of the material. Since the exercise of determining these chemical forms is far from trivial, the smallest value, and hence the most restrictive or "conservative", is tabulated here and also in the Fermilab Radiological Control Manual (FRCM).

In comparing these standards, it is generally clear that, for the most part, the DAC-values for the radionuclides of concern at accelerators, have remained remarkably constant for decades. It is also of interest that the variability of these most conservative values from one radionuclide to another does not span a very large domain. The Appendix to this Note discusses the origin of the values listed in Table 2 in more detail.

Nuclide	Half-Life	DA	AC	DA	С	DAC	DAC	<b>MPC</b> <sub>a</sub>
		Inhale	ed Air	Immer	sion	Inhaled Air	Immersion	(IC60)
		(Cur	rent	(Current		(Pre-2007	(Pre-2007	
		10 CF	R 835)	10 CFR 835) <sup>D</sup>		10 CFR 835)	10 CFR 835)	
		Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>	Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>	pCi ml <sup>-1</sup>	pCi ml <sup>-1</sup>	pCi ml <sup>-1</sup>
$^{3}H(H_{2}O)$	12.32 y	7.0E+05	20			20		5
<sup>7</sup> Be	53.22 d	4.0E+05	10			8		6
<sup>11</sup> C	20.3 min	6.0E+06	100	7.0E+04 <sup>B</sup>	1.0 <sup>B</sup>	200	4	2.6 <sup>C</sup>
<sup>13</sup> N	9.96 min			7.0E+04 <sup>B</sup>	1.0 <sup>B</sup>		4	2.3 <sup>c</sup>
<sup>15</sup> O	1.18 min			7.0E+04 <sup>B</sup>	1.0 <sup>B</sup>		4	2.0 <sup>C</sup>
<sup>38</sup> Cl	37.24 min	2.0E+05	5	7.0E+04 <sup>B</sup>	1.0 <sup>B</sup>	20	3	3
<sup>39</sup> Cl	55.6 min.	1.0E+05	2	7.0E+04 <sup>B</sup>	1.0 <sup>B</sup>	20		
<sup>41</sup> Ar	1.83 h			1.0E+05	3.0		3	2

Table 2 Standards on Concentrations of Airborne Radionuclides A

<sup>A</sup>All tabulated values were obtained from the referenced version of 10 CFR 835 for DAC and (IC60) for MPC<sub>a</sub> unless otherwise noted. For different inhalation conditions, the most conservative (lowest) value is shown. For the DACs, "Inhaled" and "Immersion" values come from Appendices A and C, respectively, unless otherwise noted. As discussed in the Appendix, the SI units (Bq m<sup>-3</sup>) are considered "primary" due to their origin while the customary (pCi ml<sup>-1</sup>) values are calculated from the SI values. 1.0 pCi ml<sup>-1</sup> = 3.7E04 Bq m<sup>-3</sup>.

<sup>B</sup> See the Appendix to this Note for possible working values for these radionuclides. Individual values for these radionuclides are not specified in the current version of 10 CFR 835, unlike in the version of 10 CFR 835 in place prior to the revisions that were set-forth in 2007 listed in the above table. The assumption implicit in the Regulation is exposure within a <u>semi-infinite</u> cloud.

<sup>C</sup>These values, not available in (IC60), were calculated for the whole body as the critical organ within a <u>semi-infinite</u> cloud by Höfert (Hö69).

<sup>D</sup>For any single radionuclide not listed above with decay mode other than alpha emission or spontaneous fission and with radioactive half-life less than two hours, the DAC value shall be 7E+04 Bq m<sup>-3</sup> (1.0 pCi ml<sup>-1</sup>)

### **Immersion Exposure in Enclosures of Finite Size**

The role of the DAC for those radionuclides where the immersion pathway dominates has long been known to be problematic due to the fact that most occupational work occurs indoors, where the workers are not found within a cloud of radionuclides of "infinite" or even "semi-infinite" radius. Within a cloud of radionuclides found within a structure of finite radius, at a given level of concentration, the dose received will be less than that experienced within an infinite cloud because the individual cannot be exposed to  $\beta$ -particles and photons emitted by decaying nuclei from beyond the dimensional boundary set by the enclosure walls. This is fundamentally different from the inhalation pathway where the radionuclide taken into the body is, of necessity, present in the breathing zone of the exposed individual. 10 CFR 835 Appendix C

recognizes this situation and specifically permits modification of the DACs to allow for immersion in a cloud of finite, rather than infinite dimensions.

This situation was also recognized by many of the early workers as discussed in most of the older references cited here [e.g., (Pa73), (Ri67), (Su92), (Wa69), (IA79), (Ia88)]. In particular, Kase (Ka67), in following the methodology discussed in (IC60), calculated a value of MPC<sub>a</sub> for <sup>13</sup>N of 1.3 pCi ml<sup>-1</sup> for a work-year dose of 5000 mrem. In a follow-up article, (Ka68), found that within a finite sphere having a radius of 2.0 m, a value of 6.7 pCi ml<sup>-1</sup> would be more appropriate. Furthermore, in a small immersion cloud, equivalent to being in an enclosure of finite rather than infinite radius, the fraction of the committed effective dose due to the  $\beta$ -particles will be larger than that due to the photons compared with the situation in larger clouds. This is also well-described by Sullivan (Su92).

A more extensive calculation that discusses the MPC<sub>a</sub> values for the important "immersion exposure" radionuclides is that of Höfert (Hö69). Höfert calculated values of MPC<sub>a</sub> for the four radionuclides of greatest concern where immersion is the exposure pathway (<sup>11</sup>C, <sup>13</sup>N, <sup>15</sup>O, and <sup>41</sup>Ar) as a function of cloud radius *R* taking into account both the  $\beta^{\pm}$  and photon components of the dose delivery to the whole body. In effect, the value of *R* may just as well be considered the radius of a particular beam enclosure or room. While the MPC<sub>a</sub> values themselves have been superseded by DAC-values (see above) assigned for regulatory purposes, they may be used to provide multiplicative scaling factors to adjust DAC-values tabulated for semi-infinite clouds to those more appropriate for rooms of finite size, as specifically suggested in 10 CFR 835 Appendix C. Figure 1 provides such scaling factors.

The variability of the three curves in the regions of intermediate values of *R* for the  $\beta^+$ -emitters <sup>11</sup>C, <sup>13</sup>N, and <sup>15</sup>O is tied to their differing  $\beta$ -decay mean energies of 0.386, 0.492, and 0.735 MeV, respectively (NN14). On the other hand, the radiation associated with the decay of <sup>41</sup>Ar consists of a  $\beta^-$  decay of mean energy 0.459 MeV and a gamma-ray of 1.293 MeV (NN14). Since the energy of these photons is significantly more than the 0.511 MeV annihilation photons seen with the other radionuclides and the photon mean free path increases as a function of energy in this energy domain (NI14), the more energetic photons emitted in the <sup>41</sup>Ar decay contributed to the dose at larger distances than do the annihilation photons seen with the  $\beta^+$ -emitters. In plotting Fig. 1, the choice of the value of *R* to assign to the "infinite" value was arbitrarily taken to be 1000 meters, a length much larger than those of particle accelerator enclosures. From (NI14) for 0.511 MeV photons, the mean free path is 11.54 g cm<sup>-2</sup>, corresponding to 95.78 m at room temperature while for 1.293 MeV photons from <sup>41</sup>Ar, the mean free path is 18.02 g cm<sup>-2</sup> corresponding to 149.5 m at room temperature. Thus, *R* = 1000 m represents 10.4 mean free paths for the annihilation photons and 6.6 mean free paths for the <sup>41</sup>Ar photons.

In using Fig. 1, a conservative approach should be taken for the use of these factors dependent upon the detailed available knowledge of the radionuclide composition present. To reiterate, these correction factors only apply to the radionuclides where immersion dose is the exposure pathway. They are not applicable to inhalation conditions where the intake volume is the time-integrated breathing rate.



Figure 1 Multiplicative factor to correct tabulated DAC-values to those appropriate for a finite cloud of radionuclides of radius R using the results of Höfert (Hö69). The "data points" on these curves are the values inferred from Höfert's results. The lines between them are intended to guide the eye.

While the subject of this Note is primarily related to *occupational* radiation protection, it is worthwhile to note that there are similar standards set forth by the Department of Energy for *environmental* protection of the public and the environment. Presently, these are called Derived Concentration Standards (DCSs) and are set forth in a technical standard referenced by DOE Order 458.1 (DO11). For a given radionuclide, the DCS-values are much smaller than are the DAC-values. This is the result of the fact that a member of the public living full time for 8766 hours per year in an airborne radionuclide concentration having the equivalent of 1.0 DCS will receive a committed effective dose of 100 mrem while a worker spending their working year of 2000 hours in an airborne radionuclide concentration having the equivalent of 1.0 will receive a committed effective dose of 5000 mrem. The scaling is not an exact one due to "round-

off" employed in regulatory tables and somewhat different exposure modeling for the environmental protection scenario. For environmental protection purposes the effects of a finite size of the volume in which the individual is exposed is generally inapplicable since, conservatively, one must assume the exposure to be received outdoors. The Appendix to this Note has further discussion of these quantities.

# **Operational Approach to the Management of Accelerator-Produced Airborne Radioactivity**

Lauten and Leveling (La96) have extensively documented the process of monitoring airborne radioactivity in Fermilab accelerator enclosures where operational experience guided by targeted beam power indicates the potential for significant production of airborne radionuclides<sup>8</sup>. The basic methodology is discussed in other documents that support ongoing operations, notably that by Lauten and Leveling (La96, La04).

This selection of significant sources naturally leads to a strong connection with measurements made in association with the ESH&Q Section Radiation Physics Science Department to characterize and monitor the airborne radionuclide releases in accordance with the National Emissions Standards for Hazardous Air Pollutants (NESHAPs) for releases of airborne radionuclides from U.S. DOE facilities as set forth in 40 CFR 61 Subpart H. The "characterization" consists of measurements of the composition of the ventilation stream as the air leaves the ventilation "stack" in terms of individual radionuclides. 40 CFR 61 Subpart H and associated guidance documents pertaining to measurement techniques are followed in the course of these measurements. Specific provisions relate to the use of isokinetic sampling techniques (ES17). The results are then correlated with the "gross-beta" measurement of stack monitors supplied by the ESH&Q Section that are utilized for routine monitoring of the effluent of the ventilation stacks. The detector used in the permanently mounted stack monitors is a thin window pancake Geiger-Mueller tube technology that is sensitive to  $\beta$ -particles and to lesser extent photons. These monitors view the airborne particle releases continually. Their output data are logged on the ESH&Q Section's "MUX" system and, more conveniently for occupational radiation protection purposes, in real time to the Accelerator Division's Accelerator Control Network (ACNET) system.

Documentation maintained by the Radiation Physics Operations Department assigns action levels to readings recorded by the ACNET system for specific enclosures that are monitored based on operating experience. The airborne activity levels are correlated with percentages of the DAC-values estimated to be present within the enclosure. If DAC thresholds are exceeded, a "wait time" is enforced before access is permitted by the Accelerator Division Operations Department. The goal is to preclude personnel entry to areas that would require posting as airborne radioactivity areas as defined by 10 CFR 835. The DAC-values are applied conservatively, in full recognition that the composition of the airborne radioactivity field inside

<sup>&</sup>lt;sup>8</sup> (La96) was written in the context of the "pre-2007" values of DACs found in 10 CFR 835. This reference remains in need of revision to incorporate the newer values of DACs found in Table 2 and also to incorporate more recent technical notes documenting revisions to the basic methodology expressed in this reference. Such a revision is anticipated to result in insignificant differences given the small changes in appropriate DAC values.

the enclosure will be different from that found at the exit of the stack<sup>9</sup>. This technique has led to complete avoidance of the need to have posted airborne radioactivity for the accelerator enclosures at Fermilab.

The composition at the stack exit will intrinsically be "richer" in long-lived radionuclides and "poorer" in short-lived radionuclides compared with that present inside the enclosure. Commonly this fact is not of great importance due to the frequent, but not universal, dominance of <sup>11</sup>C (see references). Since knowledge of the travel time through the ventilation system is readily available, in principle one can correct the composition at the point of release for the decay in transit to estimate the composition inside the enclosures by simply correcting for the decay of the individual radionuclides for the time taken to travel from the enclosure to the exit of the stack. However, this would remain only an estimate since <u>some</u> place-to-place variations in composition would still exist within enclosures of reasonable volumes. Perhaps fortuitously, as seen in Table 2, the domain of DAC-values encountered is not large. Thus, the setting of action levels based on the DAC of <sup>11</sup>C and to a lesser extent <sup>13</sup>N is <u>conservative</u> for mixtures richer in the shorter-lived radionuclides to be found within the enclosure. Here "being conservative" implies <u>overestimating</u> the potential committed effective dose.

The general experience is that in view of these controls of entry, the exposure of occupational workers entering beam-line enclosures in terms of committed effective dose received is completely dominated by photons emitted by activated components. Occupational doses due to airborne radioactivity are essentially of no significance related to the airborne monitoring requirements of 10 CFR 835. Furthermore, if personnel were to inadvertently receive significant doses from this pathway the doses would be recorded by the DOE Laboratory Accreditation Program (DOELAP)-accredited personnel radiation dosimetry monitoring badges separately as equivalent dose to the whole body (largely due to photons) and equivalent dose to the skin (largely due to the  $\beta$ -particles). In fact, significant occupational exposures to accelerator-produced airborne radioactivity within structures (enclosures, etc.) of finite size would be identifiable in a background of dose from photons emitted by activated components by an unusually large equivalent dose to the skin from the  $\beta$ -particles. This enhancement of the equivalent dose to the skin over that to the whole body has not been seen at Fermilab.

# Conclusion

Fermilab has implemented appropriate measures to assure adequate identification, control, and mitigation of occupational airborne radionuclide hazards. Continued vigilance is a key element in this program.

<sup>&</sup>lt;sup>9</sup> The determination of <u>offsite</u> composition of the radionuclide release under the NESHAP of 41 CFR 61 Subpart H is accounted for in the application of the CAP-88 computer model required by the NESHAP Regulation.

#### References

- Certain references in this list are maintained on ESH&Q Docdb at: <u>https://esh-docdbcert.fnal.gov/cgi-bin/cert/ShowDocument?docid=2656</u>.
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- (ES17) Fermilab Radionuclide Air Emissions Manual, Fermilab ESH&Q Docdb No. 712 (Revision 5, February 2017). This manual is based upon 40 CFR Part 61, Appendix A, Method 2. Found on Fermilab ESH&Q Docdb <u>https://esh-docdbcert.fnal.gov/cgi-bin/cert/ShowDocument?docid=712</u>.
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- (IC95) ICRP Publication 68, "Dose coefficients for intakes of radionuclides by workers", International Commission on Radiological Protection (Elsevier Science Ltd, Tarrytown, NY, 1995).

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- (Su92) A. H. Sullivan, A guide to radiation and radioactivity levels near high energy particle accelerators (Nuclear Technology Publishing, Ashford, Kent, United Kingdom, 1992).
- (Wa69) G. J. Warren, D. D. Busick, and R. C. McCall, Radioactivity produced and released from water at high energies", *Proceedings of the second international conference on accelerator radiation dosimetry and experience* (Stanford, CA 1969, U. S. Technical Information Service CONF-691191), pp.99-110.

### APPENDIX

#### **Discussion of Air Concentration Standards**

#### Introduction

This Appendix documents results of a review of the values of 10 CFR 835 Derived Air Concentrations (DACs) designed for worker protection and also the Derived Concentration Standards (DCSs) designed for environmental protection expressed in DOE-STD-1196-2011 (DO11). This Appendix provides a brief survey of how the values for both DACs and DCSs are calculated, perhaps useful for instructional purposes. At the outset, it should be noted that in the calculations of the values of DACs and DCS, the values are always determined first in SI units (Sv, Bq, m<sup>3</sup>, etc.) reflective of the primary references of international origin employed by DOE to determine these standards, the Publications of the International Commission on Radiological Protection (ICRP). For more than 30 years the ICRP has worked exclusively in SI units. It is clear that the calculation of 10 CFR 835 Appendix A values, for the inhalation pathway, can be complicated by the need to apply biokinetic information to fully implement the ICRP recommendations. These values are for inhalation conditions in which the material is absorbed into the body. Further discussion of these DAC-values will be provided later in this Note. The footnotes to 10 CFR 835 Appendix A provide some limited additional information concerning DOE's determination of the DAC-values. It is clear that the calculation from primary reference sources for immersion conditions, those intended to be covered by the DACs set forth in 10 CFR 835 Appendix C, of working values of the concentration of individual radionuclides that would result in an effective dose of 5000 mrem in a year to a worker exposed to 2000 hours of such radioactivity, is much more straightforward than are the inhalation DACs of Appendix A.

### Definitions

10 CFR 835 provides the primary definitions of Derived Air Concentration and Annual Limit on Intake:

<u>Derived Air Concentration</u> (DAC) means, for the radionuclides listed in appendix A of this part<sup>10</sup> the airborne concentration that equals the ALI [Annual Limit on Intake] divided by the volume of air breathed by an average worker for a working year of 2000 hours (assuming a volume of 2400 m<sup>3</sup>). For radionuclides listed in appendix C of this part, the air immersion DACS were calculated for a continuous, non-shielded exposures via immersion in a semi-infinite cloud of radioactive material."

<u>Annual limit on intake</u> (ALI) means the derived limit for the amount of radioactive material taken into the body of an adult worker by inhalation or ingestion in a year. ALI is the smaller value of intake of a given radionuclide in a year by the reference man (ICRP Publication 23) that would result in a committed effective dose of 5 rems (0.05 sieverts (Sv)) (1 rem = 0.01 Sv) or a committed equivalent dose of 50 rems (0.5 Sv) to any individual organ or tissue. ALI values for intake by ingestion and inhalation of selected radionuclides are based on International Commission on Radiological Protection Publication 68, <u>Dose Coefficients for</u>

<sup>&</sup>lt;sup>10</sup> "part" in this context means "10 CFR Part 835".

Intakes of Radionuclides by Workers, published July, 1994 (ISBN 0 08 042651 4). This document is available from Elsevier Science Inc., Tarrytown, NY.

Thus, the DAC-values, ultimately, are derived from the cited international references.

# 10 CFR 835 Tabulated DAC-values

Table A1 contains the DAC-values for the airborne radionuclides encountered at particle accelerators as set forth in 10 CFR 835 Appendices A and C for radionuclides specifically listed in those Appendices, current as of time of Revision 2 of this Note. Blank entries reflect lack of listing of the radionuclide in question. From the definitions cited above and the practices of the International Commission on Radiological Protection (ICRP), the fundamental quantities involved in calculating the DACs are provided in SI units with results in Bq m<sup>-3</sup>. As discussed above, these are taken here to be the *primary* values to be used for occupational worker protection. Values in "customary" units of pCi ml<sup>-1</sup> are thus are intrinsically *secondary* units. 1.0 pCi ml<sup>-1</sup> = 3.7E+04 Bq m<sup>-3</sup>.

In Table A1, "tabulated" means those values taken as listed in the 10 CFR 835 Appendices. There are generally values of the inhalation DAC for three different inhalation Conditions listed for the radionuclides in appendix A; Type F (fast), Type M (moderate), and Type S (slow). The absorption types (F, M, and S) have been established to describe the absorption type of the materials from the respiratory tract into the blood. The range of half-times for the absorption types correspond to: Type F, 100% at 10 minutes; Type M, 10% at 10 minutes and 90% at 140 days; and Type S 0.1% at 10 minutes and 99.9% at 7000 days. The determination of the absorption types requires detailed biokinetic calculations and consultation of advanced textbooks and references on internal exposure by the airborne pathway. In view of the generally incomplete knowledge of inhalation "types" encountered by workers at Fermilab for the purposes of this Note, the smallest, and most restrictive values are used. "Calculated" values in pCi ml<sup>-1</sup> are those determined from the tabulated DAC-values in units of Bq m<sup>-3</sup> by simple unit conversion. They are given to three significant figures to show the "round off" discrepancies, some rather large, in these comparisons as the 10 CFR 835 Appendices follow the standard regulatory practice of using only one significant figure and somewhat inscrutable rounding practices. Of the radionuclides of importance for purposes of this note listed in Appendix C, only <sup>41</sup>Ar does not require a choice of inhalation type to be made due to the nature of pathway as being immersion, not inhalation.

As one can see, taking the SI values (Bq m<sup>-3</sup>) to be the primary DAC values in view of their origin from ICRP Publications, the "calculated" values in customary units (pCi ml<sup>-1</sup>) are reasonably close and within regulatory "round off".

Nuclide	Half-Life	835 Appendix A			8	35 Appendix (	С
		Worker Inhalation			We	orker Immersi	ion
		pCi ml-1Bq m-3pCi ml-1tabulatedtabulatedcalculated		pCi ml <sup>-1</sup> tabulated	Bq m <sup>-3</sup> tabulated	pCi ml <sup>-1</sup> calculated	
<sup>3</sup> H <sup>A</sup>	12.32 y	2.00E+01	7.00E+05	1.89E+01			
<sup>7</sup> Be	53.22 d	1.00E+01	4.00E+05	1.08E+01			
<sup>11</sup> C <sup>B</sup>	20.33 min	1.00E+02	6.00E+06	1.62E+02			
$^{13}$ N	9.96 min						
<sup>15</sup> O	1.18 min						
<sup>38</sup> Cl	37.24 min	5.00E+00	2.00E+05	5.41E+00			
<sup>39</sup> Cl	55.6 min	2.00E+00	1.00E+05	2.70E+00			
<sup>41</sup> Ar	1.83 h				3.00E+00	1.00E+05	2.70E+00

 Table A1 Values of Derived Air Concentrations from 10 CFR 835

<sup>A</sup>for HTO, tritiated water

<sup>B</sup>The "vapor" form was chosen, conservatively, for <sup>11</sup>C as being the most restrictive compared with CO or  $CO_2$  since we have no feasible methodology to determine the chemical form.

# Calculation of Working Values for Immersion Conditions for Accelerator-Produced Radionuclides

The list of specific radionuclides for which values of DACs under immersion conditions in Appendix C of 10 CFR 835 exist is limited to isotopes of the noble gases argon, xenon, and krypton. In fact, perusal of (IC95) includes the same list of radionuclides in its "Annexe D" that provides the effective dose rate per unit air concentration (Sv d<sup>-1</sup> Bq<sup>-1</sup> m<sup>3</sup>) from whence the 10 CFR 835 Appendix C values were derived. Aside from <sup>41</sup>Ar, none of these noble gas isotopes are found at particle accelerators, as discussed in detail in the body of this Note. Yet, it is well known that immersion dose is an important consideration at accelerators where one finds themselves inside enclosures immersed in finite-sized clouds of photon-emitting radionuclides in addition to <sup>41</sup>Ar such as the other radionuclides considered here, excluding, of course, <sup>3</sup>H with its lack of an emitted photon and extremely low decay energy, and <sup>7</sup>Be because of its small branching ratio for photon emission. Most fortunately, the introductory material for 10 CFR 835 Appendix C permits adjustments to the immersion DAC-values to allow for submersion in a cloud of finite dimensions, as was discussed in detail in the main body of this Note.

DOE on August 11, 2017 revised the values to be  $7.0 \times 10^4$  Bq m<sup>-3</sup> (1.0 pCi ml<sup>-1</sup>) for radionuclides not listed in Appendix C that have a decay mode other than alpha emission or spontaneous fissions and have radioactive half-life less than two hours. Notice also that in this instance the regulatory "round-off" to 1.0 pCi ml<sup>-1</sup> differs greatly from the proper unit conversion to 1.9 pCi ml<sup>-1</sup>≈2.0 pCi ml<sup>-1</sup>. As with general regulatory practice, such values for unlisted radionuclides are prudently taken to be quite conservative for protective reasons to address unlisted radionuclides of exceptionally high radio-toxicity, notably alpha-particle emitters. These values are far less than the Appendix A inhalation values for the airborne radionuclides encountered and, as will be shown here, much less than working values that can be calculated using credible methods for the airborne radionuclides produced at particle accelerators in accordance with the 10 CFR 835 definition of the DAC quantity.

It is clear that in order to calculate accurate values of the effective dose for actual exposures to such airborne radioactivity, realistic working values for the specific radionuclides present at accelerators are needed for comparison with the 10 CFR 835 Appendix C values for immersion exposures to determine the limiting values for worker protection consistent with the annual limit on effective dose explicit in the definition of the DAC in 10 CFR 835. Fortunately, (DO11) provides a reliable methodology to calculate such values because it specifically addresses radionuclides encountered at particle accelerator facilities in its presentation of Derived Concentration Standards (DCSs) for both environmental air inhalation and air immersion. Prior to the amendments to 10 CFR 835 promulgated in 2007, its Appendix C also listed specific values for these radionuclides in Appendix C.

The air immersion DCS value,  $DCS_{im}$ , is that concentration which in a semi-infinite cloud of a given radionuclide would result in an effective dose  $H_{eff}$  of 0.001 Sv (100 mrem) for a so-called "reference person". It can be determined from:

$$DCS_{im} = \frac{H_{eff}}{tK_{im}} = \frac{0.001 \text{ (Sv)}}{tK_{im}},$$
(A)

where *t* is the duration of exposure. For the environmental protection purpose of (DO11) *t* is an entire year of 3.156 x  $10^7$  sec (365.25 days).  $K_{im}$  (Sv s<sup>-1</sup> Bq<sup>-1</sup> m<sup>3</sup>) is the effective dose rate coefficient under immersion conditions that converts the concentration of a particular radionuclide in the air to dose rate received from the immersion scenario by the exposed person.

(DO11) provides tabulated values<sup>11</sup> of  $K_{im}$  for a wide variety of radionuclides and gives the primary references from whence they originated. Since the immersion scenario is primarily one of external exposure, unlike for the inhalation pathway discussed later in this RP Note, there are no gender or age specific dependences for  $K_{im}$  reflective of its application in (DO11) to exposures to members of the general public.

Since the  $K_{im}$  values taken from (DO11) have no age or gender dependence, Eq. (A) can be adapted to directly calculate a working value of air concentration standard that would result in an effective dose of 0.05 Sv to a worker exposed to such air for the working year of 2000 hours,  $WV_{im}$ :

$$WV_{im} = \frac{H_{eff}}{tK_{im}} = \frac{0.05 \text{ (Sv)}}{tK_{im}},$$
 (B)

Where  $H_{eff}$  now has the value of the 0.05 Sv (5000 mrem) annual limit applied to workers, *t* is the duration of exposure of 2000 hours y<sup>-1</sup> expressed in seconds for worker protection purposes as specified in the 10 CFR 835 definitions (see above).

Following Tables 6 and A-3 in (DO11) and Eqs. (A) and (B) one can construct Table A2. To further demonstrate the validity of this approach for other radionuclides specifically listed in

<sup>&</sup>lt;sup>11</sup> The values of  $K_{im}$  in (DO11) are calculated from United States Environmental Protection Agency and ICRP documents referenced within that DOE Technical Standard.

Appendix C in addition to <sup>41</sup>Ar, value of three isotopes of krypton listed, not found at accelerators, also in 10 CFR 835 Appendix C, <sup>74</sup>Kr ( $t_{1/2} = 11.5$  min), <sup>76</sup>Kr ( $t_{1/2} = 14.8$  h), and <sup>87</sup>Kr ( $t_{1/2} = 76.3$  min), are added to this table. These are included to illustrate the validity of the methodology for more radionuclides than just the singular relevant one, <sup>41</sup>Ar. The two rightmost columns of Table A2 give the listed values in 10 CFR 835 Appendix C illustrating the relatively good consistency between the latter values and those deduced from (DO11) using this simple methodology. For illustration purpose, three significant figures are displayed Table A2.

Nuclide	DCSim		Air	DCS <sub>im</sub> Direct		Immersio	n Working	DAC <sub>im</sub> Values as	
	As Tabı	ilated in	Immersion	Calcul	ation	Values, W	Vim Inferred	Tabulated	in 10 CFR
	(DO11)	Table 6	Dose Rate	(100 m	rem/y)	fr	om	835 App	endix C
	Coefficients		Coefficients,			(5000 mrei	n y <sup>-1</sup> , 2000 h		
			Kim, from			y <sup>-1</sup> work	ting time)		
			Table A-3 of						
		1	(DO11)						-
	Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>	Sv s <sup>-1</sup> Bq <sup>-1</sup> m <sup>3</sup>	Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>	Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>	Bq m <sup>-3</sup>	pCi ml <sup>-1</sup>
<sup>11</sup> C	6.90E+02	1.90E-02	4.56E-14	6.88E+02	1.86E-02	1.52E+05	4.12E+00		
<sup>13</sup> N	6.90E+02	1.90E-02	4.57E-14	6.87E+02	1.86E-02	1.52E+05	4.11E+00		
<sup>15</sup> O	6.90E+02	1.90E-02	4.60E-14	6.82E+02	1.84E-02	1.51E+05	4.08E+00		
<sup>38</sup> Cl	4.30E+02	1.20E-02	7.36E-14	4.26E+02	1.15E-02	9.44E+04	2.55E+00		
<sup>39</sup> Cl	4.50E+02	1.20E-02	6.97E-14	4.50E+02	1.22E-02	9.96E+04	2.69E+00		
<sup>41</sup> Ar	5.20E+02	1.40E-02	6.15E-14	5.10E+02	1.38E-02	1.13E+05	3.05E+00	1.00E+05	3.00E+00
<sup>74</sup> Kr <sup>A</sup>	6.70E+02	1.80E-02	4.70E-14	6.68E+02	1.80E-02	1.48E+05	3.99E+00	1.00E+05	3.00E+00
<sup>76</sup> Kr <sup>A</sup>	1.70E+03	4.70E-02	1.83E-14	1.71E+03	4.63E-02	3.79E+05	1.03E+01	3.00E+05	1.00E+01
<sup>87</sup> Kr <sup>A</sup>	8.00E+02	2.20E-02	3.97E-14	7.91E+02	2.14E-02	1.75E+05	4.73E+00	1.00E+05	4.00E+00

	Table A2 Calculation o	f Worker	Immersion	<b>DAC-values</b>	from	( <b>DO11</b> )	) Information
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<sup>A</sup>This radionuclide is not found in air at particle accelerators. It is included in this list for the sole purpose of providing a cross-check on the methodology in addition to that afforded by the <sup>41</sup>Ar.

As expected, the DCS<sub>im</sub> values calculated here (the DCS<sub>im</sub> Direct Calculation values) verify quite well, within round-off, those tabulated in (DO11), as seen in the 2<sup>nd</sup> and 3<sup>rd</sup> columns from the left. Immersion conditions do not apply to exposures to <sup>3</sup>H and <sup>7</sup>Be so they are not included in Table A2. For the radionuclides in this set, the values of  $DAC_{im}$  and  $WV_{im}$  are smaller, and thus more protective, than are the 10 CFR 835 Appendix A inhalation values so that without further knowledge of exposure pathways, those values are the ones that should be used for the practical task of determining effective doses in the airborne radioactivity fields produced by the Fermilab accelerators. The value of WV<sub>im</sub> calculated for <sup>41</sup>Ar also agrees within round-off to that tabulated in 10 CFR 835 Appendix C, thus providing a check point for our calculation. It is noted that the conversions between the primary Bq m<sup>-3</sup> values and the secondary pCi ml<sup>-1</sup> values is performed in a more mathematically correct manner in (DO11) than in 10 CFR 835 Appendix C. Furthermore, the values calculated for the three positron emitters <sup>11</sup>C, <sup>13</sup>N, and <sup>15</sup>O are essentially identical with the "pre-2007" 10 CFR 835 values and not far off for that <sup>38</sup>Cl listed in Table 2. It is also quite interesting that these more "modern" values are essentially the same as those published in 1960. This is reflective of the long-term stability of radiation protection standards.

# Immersion Modality Conclusion

Given this complete understanding of their origin, the tabulated values of DACs in SI units (Bq m<sup>-3</sup>) should be taken as the primary values with the customary units (pCi ml<sup>-1</sup>) calculated from them as the secondary units. The working values determined using this methodology provide a possible method for calculation of effective doses to exposed individuals with an equivalent level of protection to those incorporated into the DAC values tabulated in 10 CFR 835 Appendix C. Postings of Airborne Radioactivity Areas, however, shall be consistent with the immersion DACs set forth in 10 CFR 835 Appendix C understanding that the Regulation clearly allows for corrections consistent with immersion in a cloud of finite, rather than semi-infinite conditions. These corrections are discussed in the main body of this Note.

# Attempt at Direct Calculation of Worker Inhalation DAC-values

Given the above success in calculating immersion  $WV_{im}$  one is motivated to also verify the worker inhalation  $DAC_{inh}$  likewise from (DO11) at least for purposes of comparison as this reference also sets forth inhalation values for Derived Concentration Standards,  $DCS_{inh}$  in its Table 5. The revision of 10 CFR 835 in August 2017 made no changes to the Appendix A inhalation DAC-values. To do this calculation is somewhat complicated because the values of  $DCS_{inh}$  in (DO11) utilize population age distribution and gender weighting factors due to its primary application to protection of members of the general public. Table A3 reproduces such factors as directly taken from Table 3 of (DO11). For our present purpose, the only values needed are the population fractions for male and female adults, acknowledging the dearth of minors in the workplace and assuming that the (DO11) population and gender weighting fractions for males and females as "weights", one obtains an average air intake of  $V_{day=}$  19.88 m<sup>3</sup> d<sup>-1</sup> for this "reference worker". Note that this represents the air breathed for all hours not just working hours.

		Population	Fraction <sup>12</sup>	Daily Intake Air (m <sup>3</sup> )		
Ref Group	Age x, (y)	Male	Female	Male	Female	
Newborn	0 to 1	0.00693	0.00660	4.15	4.15	
1-y	1 to 3	0.01383	0.01321	5.89	5.89	
5-у	3 to 7	0.02864	0.02731	9.00	9.08	
10-y	7 to 12	0.03814	0.03632	15.20	15.00	
15-у	12 to 17	0.03672	0.03482	20.00	15.80	
Adult	<u>&gt;</u> 17	0.36630	0.39118	22.20	17.70	

Table A3 U. S. Population and Usage Data from Table 3 of (Do11)

Table A4 provides calculations of DCS and DAC-values referenced effective dose coefficients with those tabulated in (DO11) and 10 CFR 835 Appendix A, respectively.

<sup>&</sup>lt;sup>12</sup> As a consistency check, the population fractions do, indeed, sum to a value of 1.0, as they should!

	Comparison Inhalation D the Public Bas 365.25 d	n of Tabulated and CS-values for Adul ed on 0.001 Sv y <sup>-1</sup> y <sup>-1</sup> , 8366 Hours y <sup>-1</sup>	l Calculated lt Members of (100 mrem y <sup>-1</sup> ), Exposure	Comparison of Tabulated and Calculate Inhalation DAC-values for Adult Worke Based on 0.05 Sv y <sup>-1</sup> (5000 mrem y <sup>-1</sup> ), 2000 Hours y <sup>-1</sup> Exposur			
Nuclide	(DO11) Tabulated Inhalation DCS <sub>inh</sub> Values	Inhalation Adult Effective Dose Coefficients from (Do11) Kinh	Calculated Inhalation <i>DCS</i> inh Values	10CFR835 Appendix A Tabulated Inhalation DAC <sub>inh</sub> Values	Calculated Inhalation DAC <sub>inh</sub> Values from ICRP Dose Coefficients CFR 835 Appendix A		
	Bq m <sup>-3</sup>	Sv Bq <sup>-1</sup>	Bq m <sup>-3</sup>	Bq m <sup>-3</sup> Sv Bq <sup>-1</sup> Bq			
<sup>3</sup> H	7.80E+03	1.83E-11 <sup>A</sup>	7.53E+03	7.00E+05	1.80E-11 <sup>A</sup>	5.79E+05	
<sup>7</sup> Be	2.40E+03	5.58E-11	2.47E+03	4.00E+05	5.20E-11	4.01E+05	
<sup>11</sup> C	6.90E+03	1.85E-11	7.45E+03	6.00E+06	3.20E-12	6.51E+06	
<sup>13</sup> N							
<sup>15</sup> O							
<sup>38</sup> C1	2.60E+03	4.75E-11	2.90E+03	2.00E+05	7.30E-11	2.85E+05	
CI							
<sup>39</sup> Cl	2.60E+03	4.87E-11	2.83E+03	1.00E+05	7.60E-11	2.74E+05	
<sup>39</sup> Cl <sup>41</sup> Ar	2.60E+03	4.87E-11	2.83E+03	1.00E+05	7.60E-11	2.74E+05	

<sup>A</sup>This value is for tritiated water vapor, the most plausible airborne exposure pathway for HTO.

It is advantageous to explain this table column-by-column starting from the left. Only results in SI units are tabulated, given the ease of the unit conversion and the origin of the dose coefficients in the SI system.

Columns 2, 3, and 4 compare inhalation DCS values calculated from the original sources with those listed in (DO11)

<u>Column 2</u>: This gives the tabulated, and hence "official", values of the inhalation Derived Concentration Standard,  $DCS_{inh}$ , for those radionuclides of interest for purposes of this Note for which they are provided for members of the general population, with the age and gender distribution taken into account using the values found in Table 5 of (DO11).

<u>Column 3</u>: This lists the adult values of the inhalation dose coefficients,  $K_{inh}$  provided in Table A-2 of (DO11). (DO11) describes in detail the origin of these dose coefficients that ultimately are largely derived from ICRP Publications. The values chosen are listed here are the most restrictive, in this case the numerically <u>largest</u>, values of  $K_{inh}$  for adults are used.

<u>Column 4</u>: This lists the values of *DCS*<sub>inh</sub> calculated according to the following:

$$DCS_{inh} = \frac{H_{eff}}{tK_{inh}V_{day}},$$
(C)

where  $H_{eff}$  for this purpose is the annual dose limit to members of the public of 0.001 Sv,  $V_{day}$  is the volume of air breathed per day by the average adult as determined above and *t* is the time duration of the exposure (365.25 d).

The results compare quite well with those tabulated in (DO11) for the age and gender-weighted members of the general population. (The values of  $K_{inh}$  have no gender-dependence aside from that of the daily air intake volume accounted for by the weighted average.) Exact agreement should <u>not</u> be expected since the calculation was performed only for adults including selection of the adult values of  $K_{inh}$  and the weighted average over gender used for  $V_{day}$ . The main point here is to verify the integrity of the methodology and the conclusion is that it is sound. This is the apparent result.

Columns 5, 6, and 7 of Table A4 show the results of attempting to calculate inhalation DACs for workers from original sources. Again, the presentation of the results will be column-by-column.

<u>Column 5</u>: This column lists the most restrictive inhalation DAC-values from 10 CFR 835 Appendix A for those radionuclides that are of interest for this Note.

<u>Column 6</u>: In this column a different set of Inhalation Effective Dose Coefficients,  $K_{inh}$ , is listed that originate from (IC95) and are reportedly used as part of the development of the inhalation DACs provided in 10 CFR 835 Appendix A. It is noted that in some cases there are significant differences between these coefficients and those in Column 3. A similar, but not identical, set of dose coefficients are published in (IC11).

<u>Column 7</u>: This column lists the calculated values of  $DAC_{inh}$  using the Column 6 dose coefficients using the following:

$$DAC_{inh} = \frac{H_{eff}}{K_{inh}V_{workyear}} = \frac{0.05 \text{ (Sv)}}{K_{inh}2400 \text{ (m}^3)},$$
 (D)

where the 10 CFR 835 value for air volume breathed in a year by a worker has been utilized. For tritium water vapor, the footnotes for 10 CFR 835 appendix A are adjusted downward to make allowance for skin absorption. This special reduction of 50% in the value of  $DAC_{inh}$  for tritium water vapor was applied here, also for purpose of comparison with the tabulated values. Here it should be noted that this volume of air is larger than the volume one gets by the product  $V_{day}$ =19.88 m<sup>3</sup>d<sup>-1</sup> = 0.792 m<sup>3</sup>h<sup>-1</sup>x2000 working h yr<sup>-1</sup>=1656 m<sup>3</sup>yr<sup>-1</sup>. The larger annual worker breathing volume may be representative of higher respiration rates during work compared with during sleep.

## Inhalation DAC Conclusion

The differences between Columns 5 and 7 are largely reflective of differences between the dose coefficients. In general, the values found in Column 7 agree generally well, but not "exactly" with the tabulated "official" values of Column 5. Differences are to be expected as the footnotes of 10 CFR 835 Appendix A allow for further calculations beyond these simple ones to have been made by DOE and are beyond the scope of this note. Also, for some of the radionuclides, the tabulated limiting value are *deterministic* doses for specific organs and tissues not further addressed here.