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A method for estimating ^{41}Ar, ^{85,88}Kr and ^{131m,133}Xe doses to non-human biota

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This paper is dedicated to the memory of Dr Clive Williams.

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ABSTRACT

A method is presented for estimating ^{41}Ar, ^{85,88}Kr and ^{131m,133}Xe dose rates to terrestrial wildlife without having to resort to comparisons with analogue radionuclides. The approach can be used to calculate the dose rates arising from external exposures to given ambient air concentrations of these isotopes. Dose conversion coefficient (DCC) values for a range of representative organisms are calculated, using a Monte Carlo approach to generate absorbed fractions based on representing animals as reference ellipsoid geometries. Plume immersion is the main component of the total DCC.

DCC values calculated for a human-sized organism are compared with human dose conversion factors from ICRP Publication 119, demonstrating the consistency of the biota approach with that for humans. An example of application is provided for hypothetical nuclear power plant atmospheric discharges with associated exposures to birds and insects. In this example, the dose rates appear to be dominated by ^{133}Xe and ^{88}Kr, respectively. The biota considered would be protected from the effects of noble gas radiation from a population protection perspective.

1. Introduction

The isotopes ^{41}Ar, ^{85,88}Kr and ^{131m,133}Xe are radioactive noble gases produced in uranium and plutonium nuclear reactors. ^{85,88}Kr and ^{131m,133}Xe are direct fission products and ^{41}Ar is produced indirectly by the activation of natural ^{40}Ar in air in the reactor containment. About 30 cm^3 of fission gas are produced in reactor fuel per MWd thermal (Streit et al., 2006). ^{133}Xe is the dominant gas because of its large yield as a fission product. For example, ^{133}Xe is produced 6.6 (7.0)% per thermal fission of ^{235}U (^{239}Pu) compared with 0.031 (0.041)% for ^{131m}Xe and 0.29 (0.14)% for ^{85}Kr (Nichols et al., 2008). The fission gases can then be released from spent fuel operations due to defective fuel and seal failures, and they become routinely discharged to the atmosphere (Copplestone et al., 2010; GE, 2010).

The proportions of Ar, Kr and Xe isotopes in nuclear reactor releases are variable depending on the type of reactor. In general, they represent a large contribution in terms of activity, but they are generally not considered to be a major contribution to the human dose.

In atmospheric discharges calculated for the generic design assessment of UK Westinghouse reactors, it was estimated that ^{41}Ar, ^{85}Kr, and ^{133}Xe constitute together 76.7% of the total radioactivity emitted to the atmosphere (Westinghouse, 2011). The reactors at AECL Chalk River Laboratories, which are not used for generating power, also have emissions of mixed noble gases, particularly ^{41}Ar (Pilgrim and Audet, 2013). At the Idaho National Engineering and Environmental Laboratory in the US, ^{85}Kr accounted for approximately 75% of the total releases, followed by tritium at 14%, and ^{41}Ar at 10% of the total (INEL, 2003).

Nuclear installations with particularly significant ^{85}Kr contributions include the new nuclear facility at the Darlington nuclear site, Canada (SENES&AMEC, 2009) and the Sellafield Site in the UK (Sellafield, 2013), which in 2012 had airborne ^{85}Kr discharges of 3.8×10^4 TBq from an authorised limit of 4.4×10^5 TBq (compared with 10^2 TBq of an authorised 1.1×10^3 TBq for tritium). It is estimated that annual average discharges of noble gases from AP1000 Westinghouse reactors to the atmosphere will include 3.17 GBq y^{-1} for krypton radionuclides (EA, 2011).

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The Idaho National Laboratory, US, reported that ^{133}Xe and ^{135}Xe each contributed 0.2% to their releases, followed by ^{88}Kr at 0.1% (INEL, 2003). These varied examples show that although the quantities and proportions of noble gases released by nuclear-related industries are variable, significant amounts of Ar, Kr, and Xe are released into the environment.

The noble gases Ar, Kr and Xe are not taken up significantly into the body of living organisms due to their low chemical reactivity. Although they can be present in physical solution, chiefly in the body water and fat, they do not participate at normal pressures in the biochemical reactions of the human body (Tobias et al., 1949). The biological half-life of ^{85}Kr is estimated at 30 s (Turkin, 1973). ICRP publication 119 (ICRP, 2012) classifies these inert gases as insoluble and non-reactive, whereupon human DCCs only take account of external irradiation from submersion in the cloud of gas and internal irradiation from gas within the respiratory tract. For this reason, noble gases in a power reactor context are thought to contribute only a small amount to the human dose (Smith, 2013). For non-human biota, we assume that the radiation dose rates to living organisms will be dominated by external irradiation. For practical purposes, there is no internal dose rate to biota associated with Ar, Kr, and Xe emissions.

Ar, Kr and Xe isotopes are not routinely included in wildlife assessments. Yet, it is clear from the foregoing that a case for assessing them can be made, based on the importance of these radionuclides in the total discharge. As part of the general drive to ensure that the environment is protected from radiation, there is a need to demonstrate the limited impact that these noble gas discharges may have in current and new reactor designs.

Guidance has been published on the requirement to calculate the external dose of noble gases from air immersion in environmental risk assessments (CSA, 2012 clause 7.3.4.1.4). There are some studies reporting explicit biota dose calculations for ^{41}Ar and ^{85}Kr in their environmental risk assessments (Hart et al., 2005). Precedent for calculation of doses to non-human species from the noble gases ^{41}Ar and ^{85}Kr exist for UK EPR nuclear power plant designs (EA, 2011) and the Environmental Application For Hinkley Point C in the UK (NNB, 2011), for example. These studies consider the wildlife of interest to be located away from the source point, typically at a 500-m distance downwind from a single release stack (Copplestone et al., 2010; IAEA, 2001).

Consequently, for compliance and for addressing public perception, there is a clear justification to perform wildlife assessments for Ar, Kr, and Xe.

In 2001, the EA R&D 128 method for assessment of dose rates to wildlife was developed in the UK (Copplestone et al., 2001). Initially, ^{137}Cs had been suggested as an analogue for use in ^{41}Ar and ^{85}Kr dose rate estimations (EA, 2002). However, this led to highly conservative estimates (Beresford et al., 2004) and thus many realistic routine releases containing ^{41}Ar or ^{85}Kr exceeded the screening value of 40 μGy h^{-1} for the terrestrial animals used in the R&D 128 method. Therefore, a new version of the EA R&D 128 assessment tool with an ability to calculate directly ^{41}Ar and ^{85}Kr dose rates was released as a CD (Vives i Batlle and Jones, 2003). This tool still could not calculate ^{88}Kr, ^{131m}Xe and ^{133}Xe dose rates, so the advice was given to use ^{85}Kr as an analogue for these isotopes. In this paper, we will prove that this is not always appropriate.

The R&D 128 approach has now been superseded by the ERICA assessment tool (Beresford et al., 2007; Brown et al., 2008). However, this tool does not have the capability to assess for noble gases and, therefore, the R&D 128 method has remained in use for this specific purpose. To our knowledge, no other independent assessment approach for ^{41}Ar, ^{85,88}Kr, and ^{131m,133}Xe in wildlife has been published in the literature before the present study. Here, the developments of the original R&D 128 method are brought to their natural conclusion, as we are now able to calculate dose rates for all the environmentally relevant Ar, Kr, and Xe isotopes, for a wide range of reference organisms. In publishing this information, we respond to a stated need to give due scientific reference to the method currently in use, but given the fact that the R&D 128 method is superseded by the ERICA approach (and we certainly endorse the general move in this direction) we go further by providing the dosimetry for the ERICA reference organisms whose geometries coincide with the RAPs as defined by the ICRP (ICRP, 2008), hoping that this will be increasingly adopted in future assessments.

2. Materials and methods

To conduct an assessment of ^{41}Ar, ^{85,88}Kr and ^{131m,133}Xe exposure to terrestrial biota, it is necessary to calculate first the average concentrations of radionuclides in surrounding air at ground level. This requires modelling of the atmospheric dispersion of these gaseous radionuclides arising from a given source term to the ground location inhabited by the target organisms. The simplest way to do this is to use Gaussian plume models, as in the approach given in IAEA SRS publication 19 (IAEA, 2001). Such models require very simple inputs: meteorological conditions, location and height of stack and stack emission rates.

Next, one needs to calculate appropriate dose conversion coefficients (DCCs) for the organisms, defined in terms of a specific geometric shape with associated dimensions. The external DCCs for noble gases are formulated relative to activity concentration in the ambient air (in units of μGy h^{-1} Bq^{-1} m^3), consistent with the standard definitions adopted in the ICRP dosimetry for non-human biota (ICRP, 2008).

Multiplying the radionuclide concentration in air by the relevant DCCs enables the derivation of the cloud immersion dose rates for the organisms. Specific steps in this method are described below.

2.1. Semi-infinite cloud approximation

External doses from airborne material can be computed using semi-infinite and finite cloud approximations. When the dimensions of the plume become large in comparison with the mean free paths of γ-radiation emitted from radionuclides within, the simpler semi-infinite cloud model is preferable because for the finite cloud model a spatial integration over the plume volume is performed and this is computationally demanding. The assumption of a semi-infinite plume is justified for long downwind distances. At shorter distances from the source (typically, less than 1 km) external γ-doses are no longer linearly related to the surface level air concentration, hence a correction factor needs to be applied; otherwise doses need to be calculated using a finite cloud approach (Bevelacqua, 2009).

The β-dose contribution is, however, always calculated using a semi-infinite cloud model because the range of β-particles in air is short compared with the dimensions of the plume. The range of gamma rays is longer but for low energies and large plume dimensions the semi-infinite calculation is still acceptable, and will over-estimate the dose, rather than under-estimating it (Bevelacqua, 2009).

In calculating doses to humans, correction factors are usually applied to the calculated cloud dose from ^{41}Ar to account for over-estimation by the semi-infinite plume approximation at shorter distances (Bevelacqua, 2009). For γ-radiation from a plume, a building shielding factor (typically 0.2) also needs to be applied for individuals resident within buildings (Simmonds et al., 1995). A generic building occupancy of 66% tends also to be assumed and a further conservative assumption of 100% site occupancy is also made. These adjustments are not necessary in calculation of doses to biota for the obvious reason that wildlife does not inhabit buildings.