On the calculation of activity concentrations and nuclide ratios from measurements of atmospheric radioactivity

A. Axelsson, A. Ringbom *

Swedish Defence Research Agency (FOI), SE-16490 Stockholm, Sweden

HIGHLIGHTS

- We note the need for consistency when reporting measurements of atmospheric radioactivity.
- We discuss various methods for correction of parent–daughter ingrowth.
- We supply expressions for transforming activity concentrations to activity ratios.
- We discuss methods for calculation of uncertainties of nuclide ratios.

ARTICLE INFO

ABSTRACT

Motivated by the need for consistent use of concepts central to the reporting of results from measurements of atmospheric radioactivity, we discuss some properties of the methods commonly used. Different expressions for decay correction of the activity concentration for parent–daughter decay pairs are presented, and it is suggested that this correction should be performed assuming parent–daughter ingrowth in the sample during the entire measurement process. We note that, as has already been suggested by others, activities rather than activity concentrations should be used when nuclide ratios are calculated. In addition, expressions that can be used to transform activity concentrations to activity ratios are presented. Finally we note that statistical uncertainties for nuclide ratios can be properly calculated using the exact solution to the problem of confidence intervals for a ratio of two jointly normally distributed variables, the so-called Fieller's theorem.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

Sensitive isotope-specific measurements of atmospheric radio-activity are performed on a routine basis all over the world. The majority of measurement systems are operated in national networks for the purpose of emergency preparedness. Another important application is verification of the Comprehensive Nuclear Test-Ban Treaty (CTBT) (CTBTO, 2013). At the end of 2013, more than 80% of the planned global 80-station network, constituting the radionuclide part of the International Monitoring System (IMS) for CTBT-verification, was sending data to the CTBTO Preparatory Commission International Data Center (IDC) in Vienna.

The dominating technique for this kind of measurements is aerosol sampling on a particulate filter, followed by sample preparation and activity measurement, normally performed using a high-purity germanium (HPGe) detector. However, triggered by the build-up of the IMS in the last 15 years, the technique to detect

atmospheric radioxenon has undergone considerable development (Auer et al., 2004), and has proven to be a very useful tool for detection of underground nuclear explosions (Ringbom et al., 2009, 2014; Saey et al., 2007). This measurement principle is based on air collection on activated charcoal traps followed by gas chromatographic purification and finally measurement of the gas-sample using a beta-gamma coincidence system or a HPGe detector (Ringbom et al., 2003; Popov et al., 2005; Fontaine et al., 2004; Bowyer et al., 1998).

Two quantities are of special interest when analyzing data from atmospheric radioactivity measurements: *activity concentrations* and *nuclide ratios*. The two quantities are used for different purposes. Activity concentration, normally given in Bq/m^3, is necessary for *e.g.,* dose assessments and for source reconstruction using atmospheric transport modeling (Becker et al., 2007). Nuclide ratios are often used to characterize the source, *e.g.* to distinguish a reactor release from a nuclear explosion (Kalinowski et al., 2010), or to obtain timing information by comparison with scenarios calculated from nuclear data (Nir-El, 2004; Pan and Ungar, 2012). It is important to realize one fundamental difference between these two quantities: the activity concentration at

* Corresponding author. Tel.: +46 8 5550 3449; fax: +46 8 5550 3494.
E-mail address: anders.ringbom@foi.se (A. Ringbom).

a certain location will depend upon the atmospheric transport from the source to this location, while nuclide ratios, given that no fractionation of the two nuclides occurs during transport (*e.g.*, due to wet or dry deposition), are independent of transport mechanisms, dilution, plume shape, and of how the air sample is collected. We find that some confusion exists when nuclide ratios are reported: sometimes the ratios are calculated using activity concentrations, sometimes using activities, and sometimes it is not even clear what has been used. Mixing the two concepts can result in errors in the analysis. If the nuclide ratios are used for *e.g.*, determine the time of a nuclear event, such as time zero for a nuclear explosion, these errors will then propagate and cause errors in the timing. The basis of the misconception lies in the way the activity concentration is normally measured and calculated. The activity concentration can never be measured with better timing precision than the sampling time of the measurement system, since the true time profile of the activity concentration in the sampled air is unknown. This means that when calculating the activity concentration, an assumption has to be made regarding its time profile during sampling. The activity ratio, on the other hand, can be calculated at any given time using information from the activity measurement on the collected sample, since its behavior over time is determined only by the decay (again given that no fractionation occurs).

The purpose of this paper is to discuss the method normally used to calculate the activity concentration (Section 2), including a few examples of modified approaches. Furthermore, we present expressions for calculation of useful nuclide ratios from data produced by atmospheric radioactivity measurements. These equations are presented in Section 3. Commonly, data from atmospheric radioactivity monitoring are reported as activity concentrations only. In Section 4, we show how to convert these data to activity ratios with well-defined time references that can be used for *e.g.*, timing of nuclear events. In all sections we present expressions for both independent and parent–daughter decay.

Finally, in Section 5, we address a few aspects of the calculation of statistical uncertainties of nuclide ratios. The intention is to point to the fact that in cases when small activities are measured, in particular if the denominator has a small value with large uncertainties, the statistical confidence interval of the ratio should not be estimated using straightforward error propagation, *e.g.*, by adding the uncertainties in quadrature. Instead, the confidence interval for the ratio is more properly calculated using the so-called "Fieller's theorem", resulting in asymmetric statistical uncertainties.

2. The activity concentration equation

2.1. Independent decay

A typical measurement of airborne radioactivity consists of three stages: air sampling, sample preparation, and activity measurement. The collected sample is transferred to a detector system, for example by compressing a filter and placing it on a HPGe detector, or by transferring a purified gas sample to a beta-gamma detector system. The most common expression used for calculating the activity concentration C in air (in Bq/m^3) is

$$C = \frac{n}{\varepsilon \cdot B} \frac{\lambda^2}{(1 - e^{-\lambda t_C}) e^{-\lambda t_P} (1 - e^{-\lambda t_A})} \frac{t_C}{V}, \qquad (1)$$

where the quantities are n, the number of counts recorded for a specific decay mode, λ, the nuclide decay constant [s^{-1}], ε, the detection efficiency of the counter used in the measurement, B, the branching ratio of the decay to the radiation counted, V, the sampled air volume [m^3], t_C, the sample collection time [s], t_P, the

sample processing time [s], and t_A, the measurement acquisition time [s].

The applicability of Eq. (1) rests on three assumptions:

- The nuclide decays independently from other nuclides.
- The activity concentration C in air is assumed constant during sampling.
- The sampling rate S [m^3/h] is assumed constant during sampling and can be written $S = V/t_C$.

We are implicitly assuming that corrections have been performed for dead time, as well as for any coincidence summing effects and sample losses. Eq. (1) can be derived starting from the following differential equation (Evans, 1995), describing the dynamic behavior of the activity collected in the sample:

$$\frac{dA}{dt} = CS[1 - u(t - t_C)] - \lambda A, \qquad (2)$$

where $u(t)$ is the unit step function, switching from zero to unity at time t_C. This function describes the fact that the sampling is turned off at time $t = t_C$. It can easily be shown that during collection, the activity will be described by $(CS/\lambda)(1 - e^{-\lambda t})$, during processing/decay by $(CS/\lambda)(1 - e^{-\lambda t_C}) e^{-\lambda(t - t_C)}$, and during activity measurement by $(CS/\lambda)(1 - e^{-\lambda t_C}) e^{-\lambda t_P} e^{-\lambda(t - t_C - t_P)}$. The number of counts registered in the detector is obtained by integrating the last expression over the activity measurement time and multiplying by efficiency and branching ratio. Eq. (1) is then obtained by solving for C. An example of the resulting activity as a function of time is illustrated by the solid curve in Fig. 1.

In the general case, the actual airborne activity concentration at the sampler will vary during sampling according to the meteorological conditions and the time duration of the plume of activity being sampled. The constant activity concentration assumption has nevertheless usually been regarded as reasonable, if only because an activity measurement on an air sample after sampling has concluded cannot in any case yield information on the time variation of the airborne activity. It can only yield an average value during the sampling period.

Obviously, large differences between true and reported C can occur if the plume-shape is very different from the assumed flat distribution. The effect of different plume shapes is illustrated in Fig. 2, showing three different plumes of ^{135}Xe analyzed by a radioxenon system with typical collection-, processing-, and measurement times. The three different plumes would all result in a reported activity concentration of 1 Bq/m^3, while the true activity concentrations in the plumes were 1, 19, and 8 Bq/m^3, respectively.
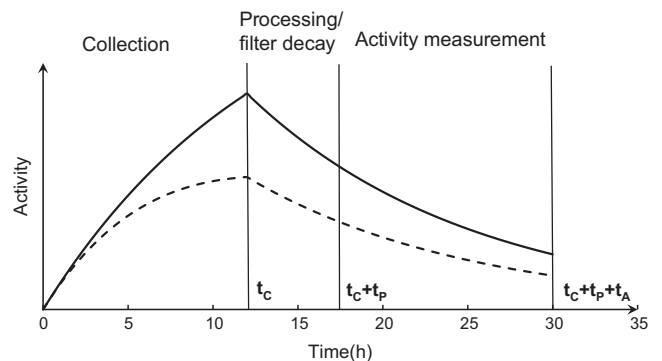

Fig. 1. Collected activity as a function of time for an independently decaying isotope sampled with constant air flow on, *e.g.*, a particulate filter or charcoal trap. The sampling is performed from $t = 0$ to $t = t_C$, followed by sample preparation during a time t_P, after which the activity is measured for a time t_A. The solid curve is calculated according to the assumption resulting in Eq. (1), and the dashed curve is obtained using "decaying air" (Eq. (4)). The assumed nuclide half-life is 9.14 h (the half-life of ^{135}Xe).