Derivation of the concentration equation assuming parent-daughter pair present in sample

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The common expression used for calculating the air concentration of xenon radioactivity C is

$$C = \frac{n}{\varepsilon\beta\gamma} \frac{\lambda^2}{F_C F_P F_A} \frac{t_c}{V},\tag{1}$$

where n is the background-corrected number of counts in the region of interest (ROI) corresponding to beta-gamma coincidences of the isotope in question, λ is the decay constant of the isotope, ε is the energy-integrated detection efficiency for the ROI, $\beta\gamma$ is the combined beta-gamma branching ratio to the decay, t_c is the sample collection time, and V is the sampled air volume. Decay corrections during sampling, processing and activity measurement are taken care of by the factors F_C , F_P , and F_A , where $F_C = 1 - e^{-\lambda t_c}$, $F_P = e^{-\lambda t_P}$, and $F_A = 1 - e^{-\lambda t_a}$.

The derivation of this equation is based on the following assumptions:

- The concentration C is assumed constant during sampling.
- The sampling rate is assumed constant during sampling and is equal to V/t_c .
- All isotopes decays independently.

The last assumption is actually not true in the case of noble gas sampling, where the isotopes 133 Xe, 133m Xe, 131m Xe, and 135 Xe are studied, since 133m Xe decays to 133 Xe.

This document presents a derivation of a modified version of equation 1, taking the presence of a parent-daughter pair in account when the concentrations are calculated. The derivation is divided into three parts (A,B and C), corresponding to decay during sampling, processing and activity measurement, respectively.

A) Sampling $(0 \le t \le t_c)$

During the sampling phase (*i.e* between t = 0 and $t = t_c$) the activity A varies according to the decay law with a continuous contribution:

$$\frac{dA}{dt} = \frac{VC}{t_c} - \lambda \cdot A \tag{2}$$

If the activity of the sample is assumed to be zero at start of sampling, equation 2 has the solution

$$A = \frac{VC}{t_c \lambda} (1 - e^{-\lambda t}) \tag{3}$$

Let A_1, λ_1 denote the activity and disintegration constant of the parent, and A_2, λ_2 the corresponding values for the daughter nuclide. Equation 3 then gives the activity at time $t = t_c$ for the two nuclides according to

$$A_1(t_c) = \frac{VC_1}{t_c \lambda_1} (1 - e^{-\lambda_1 t_c})$$
(4)

$$A_2(t_c) = \frac{VC_2}{t_c \lambda_2} (1 - e^{-\lambda_2 t_c})$$
(5)

Here we assume that the activity is constant during sampling for both isotopes, a reasonable assumption taking all other uncertainties associated with the time distribution of the airborne activity into account.

B) Processing $(t_c < t \le t_p)$

When the sampling is completed, the number of nuclei for the parent and the daughter isotope $(N_1 \text{ and } N_2)$ varies according to

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \tag{6}$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \tag{7}$$

Equation 6 has the solution

$$N_1(t) = N_1(0)e^{-\lambda_1 t}$$
(8)

and equation 7 results in

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0) (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2(0) e^{-\lambda_2 t}$$
(9)

The first term in 9 describes the contribution to N_2 from the decay of the parent, and the second term is the part describing the decay of the daughter isotope originally present in the sample.

Substituting for activity by inserting $A_2(t) = \lambda_2 N_2$ yields:

$$A_2(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1(0) (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2(0) e^{-\lambda_2 t}$$
(10)

The activity A_1 for the parent isotope during sample processing is simply given by multiplying by a decay factor according to

$$A_1(t) = A_1(0)e^{-\lambda_1 t} = \frac{VC_1}{t_c\lambda_1}(1 - e^{-\lambda_1 t_c})e^{-\lambda_1 t}$$
(11)

The activities at time t_p for the two isotopes are then given by:

$$A_{1}(t_{p}) = \frac{VC_{1}}{t_{c}\lambda_{1}}(1 - e^{-\lambda_{1}t_{c}})e^{-\lambda_{1}t_{p}}$$
(12)

and

$$A_{2}(t_{p}) = \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} \frac{VC_{1}}{t_{c}\lambda_{1}} (1 - e^{-\lambda_{1}t_{c}})(e^{-\lambda_{1}t_{p}} - e^{-\lambda_{2}t_{p}}) + \frac{VC_{2}}{t_{c}\lambda_{2}} (1 - e^{-\lambda_{2}t_{c}})e^{-\lambda_{2}t_{p}}$$
(13)

C) Detector measurement $(t_p < t \le t_a)$

The number of counts n for the daughter nuclide registered by the detector from time t = 0 to $t = t_a$ is given by

$$n = \varepsilon \beta \gamma \int_0^{t_a} A_2(t) dt \tag{14}$$

where A(t) varies according to the same exponential as described in the previous section.

The integral is calculated by inserting the expressions for A_1 and A_2 obtained in the last section (equation 11 and 10):

$$\int_{0}^{t_{a}} A_{2}(t)dt = \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} A_{1}(0) \int_{0}^{t_{a}} (e^{-\lambda_{1}(t)} - e^{-\lambda_{2}t})dt + A_{2}(0) \int_{0}^{t_{a}} e^{-\lambda_{2}t}dt \quad (15)$$

This results in

$$\int_{0}^{t_{a}} A_{2}(t)dt = \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} A_{1}(0) \left(\frac{e^{-\lambda_{1}t_{a}}}{\lambda_{1}} - \frac{1}{\lambda_{1}} - \left(\frac{e^{-\lambda_{2}t_{a}}}{\lambda_{2}} - \frac{1}{\lambda_{2}}\right)\right) + \frac{A_{2}(0)}{\lambda_{2}} (1 - e^{-\lambda_{2}t_{a}})$$
(16)

We now note that equations 12, 13, and 16 can be written

$$A_1(0) = \frac{VC_1}{t_c \lambda_1} F_{C1} F_{P1},$$
(17)

$$A_2(0) = \frac{\lambda_2}{\lambda_2 - \lambda_1} \frac{VC_1}{t_c \lambda_1} F_{C1}(F_{P1} - F_{P2}) + \frac{VC_2}{t_c \lambda_2} F_{C2} F_{P2}$$
(18)

and

$$\int_{0}^{t_{a}} A_{2}(t)dt = \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} A_{1}(0) \left(\frac{F_{A1}}{\lambda_{1}} - \frac{F_{A2}}{\lambda_{2}}\right) + \frac{A_{2}(0)}{\lambda_{2}} F_{A2}$$
(19)

where F_C , F_P , and F_A refers to the decay correction factors in the original concentration equation (1) for the parent (index=1) and daughter (index=2) isotope, respectively.

We now get

$$\frac{nt_c}{\varepsilon\beta\gamma V} = \frac{C_1}{(\lambda_2 - \lambda_1)\lambda_1^2} F_{C1}F_{P1}(\lambda_2 F_{A1} - \lambda_1 F_{A2}) + \frac{F_{A2}C_1}{(\lambda_2 - \lambda_1)\lambda_1} F_{C1}(F_{P1} - F_{P2}) + \frac{F_{A2}C_2}{\lambda_2^2} F_{C2}F_{P2}$$
(20)

A little algebra gives

$$\frac{nt_c}{\varepsilon\beta\gamma V} = \frac{C_1 F_{C1}}{\lambda_1^2 (\lambda_2 - \lambda_1)} (\lambda_2 F_{P1} F_{A1} - \lambda_1 F_{P2} F_{A2}) + \frac{C_2}{\lambda_2^2} F_{C2} F_{P2} F_{A2}$$
(21)

and finally

$$C_{2} = \frac{nt_{c}\lambda_{2}^{2}}{\varepsilon\beta\gamma VF_{C2}F_{P2}F_{A2}} - C_{1} \cdot \frac{\lambda_{2}^{2}}{\lambda_{1}^{2}(\lambda_{2} - \lambda_{1})} \frac{F_{C1}}{F_{C2}} (\lambda_{2}\frac{F_{P1}F_{A1}}{F_{P2}F_{A2}} - \lambda_{1})$$
(22)

where

$$F_{C1} = 1 - e^{-\lambda_1 t_c}$$

$$F_{P1} = e^{-\lambda_1 t_p}$$

$$F_{A1} = 1 - e^{-\lambda_1 t_a}$$

$$F_{C2} = 1 - e^{-\lambda_2 t_c}$$

$$F_{P2} = e^{-\lambda_2 t_p}$$

$$F_{A2} = 1 - e^{-\lambda_2 t_a}$$
(23)

Equation 22 is on the form

$$C_2 = C_2^0 - \xi \cdot C_1 \tag{24}$$

where C_2^0 is the concentration obtained when no correction for the parent decay is performed (equation 1), and ξ is a decay correction parameter, given by

$$\xi = \frac{\lambda_2^2}{\lambda_1^2(\lambda_2 - \lambda_1)} \frac{F_{C1}}{F_{C2}} (\lambda_2 \frac{F_{P1}F_{A1}}{F_{P2}F_{A2}} - \lambda_1)$$
(25)

It is interesting to calculate the parameter ξ in the case of a SAUNA measurement where both ^{133m}Xe and ¹³³Xe are detected. Assuming the standard time parameters for SAUNA ($t_c = 12h$, $t_p = 6$ h, and $t_a = 11$ h) one obtains $\xi = 0.057$. This means that in the case the concentrations for the two isotopes are equal, the measured ¹³³Xe concentration is adjusted down about 6%. In the case of a very long process time, a situation that can occur when the mobile system is used, and the samples are transported for a long time, or if the sample for some other reason has to wait to be analysed at the

laboratory unit, the correction will be larger. A process time of one week (168 h) results in $\xi = 0.50$, and the ratio $C(^{133m}Xe)/C(^{133}Xe)$ could change significantly compared to using the original concentration equation.

Calculation of the critical limit

The criteria wether an isotope is detected or not is governed by the critical limit (L_C) :

$$L_C = k\sigma_0 = k\sqrt{V(n) - n},\tag{26}$$

where k = 1.645 if a 5% risk for false detection is adapted. Now write equation 24 as

$$B_2 n_2 = B_2 n_2^0 - \xi B_1 n_1, \tag{27}$$

where n_2 , n_2^0 , and n_1 are the net counts corresponding to the concentrations C_2 , C_2^0 , and C_1 . Again ignoring the covariance term, the variance of n_2 can be written

$$V(n_2) = V(n_2^0) + \xi^2 \frac{B_1^2}{B_2^2} V(n_1), \qquad (28)$$

and the variance of the zero background in equation 26 becomes

$$\sigma_0^2 = V(n_2^0) + \xi^2 \frac{B_1^2}{B_2^2} V(n_1) - n_2^0 + \frac{B_1}{B_2} \xi n_1.$$
⁽²⁹⁾

Multiply by B_2 to get the L_C in Bq/m^3 :

$$L_C = k\sqrt{B_2^2 V(n_2^0) + \xi^2 B_1^2 V(n_1) - B_2^2 n_2^0 + B_1 B_2 \xi n_1},$$
(30)

where

$$B_{1} = \frac{t_{c}\lambda_{1}^{2}}{\varepsilon_{1}\beta_{1}\gamma_{1}VF_{C1}F_{P1}F_{A1}},$$
(31)

and

$$B_2 = \frac{t_c \lambda_2^2}{\varepsilon_2 \beta_2 \gamma_2 V F_{C2} F_{P2} F_{A2}}.$$
 (32)

By extracting the net counts n_1 and n_2^0 and their corresponding variances, the concentration and L_C for the daughter isotope can be estimated by using equations 22 and 30.