

How to derive the concentrations of  $^{222}\text{Rn}$  in soil air [English version]\*

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To my gentle reader

This contribution will describe how to derive the concentrations of  $^{222}\text{Rn}$  in soil air, i. e. the solutions of differential equations presented at the Fourth European IRPA Congress (IRPA Europe 2014) which was held in Geneva, Switzerland from 23<sup>th</sup> to 27<sup>th</sup> June, 2014 and the Fifth European IRPA Congress (IRPA Europe 2018) which was held in The Hague, The Netherlands from 4<sup>th</sup> to 8<sup>th</sup> June, 2018.

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**How to derive concentrations of  $^{222}\text{Rn}$  in soil air, namely Equations (4) and (5) in Kataoka (2018) (the same as Equations (8) and (9) in Kataoka and Kigoshi (2014), respectively)**

Let us consider a system which consists of a layer of uranium residue (uranium residue will hereafter be designated as residue) and a single cover layer (a layer of soil) on the surface of the residue, as shown in Figure 1. The system is assumed to be homogeneous horizontally (in the x- and y-directions). The thickness of the layer of the residue is greater than 15 m which is considered to be infinitely deep in respect of the diffusion of  $^{222}\text{Rn}$ . As for the soil, the properties such as bulk dry density, porosity and volumetric water content are known, and the radioactivity of the soil is known to be about the same level as those of ordinary soils, but the contents of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  and the escape-to-production ratio of  $^{222}\text{Rn}$  are unknown.

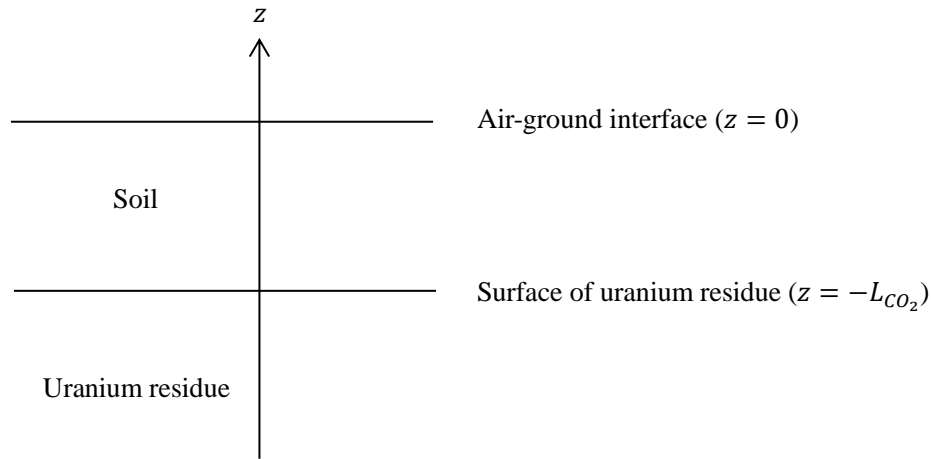


Figure 1. Single cover layer system on the surface of uranium residue.

It is assumed that soil microorganisms and plant roots exist densely in the soil, and that the flow of  $\text{CO}_2$  is accompanied by  $^{222}\text{Rn}$  in a region of high  $\text{CO}_2$  production. Therefore, it is assumed that the flow of  $\text{CO}_2$  is accompanied by  $^{222}\text{Rn}$  in the soil air, namely it is assumed that  $^{222}\text{Rn}$  behaves in the same way as  $\text{CO}_2$  does there. Since this means that, in the soil air, the diffusion coefficient of  $\text{CO}_2$  may be used instead of that of  $^{222}\text{Rn}$ ,  $^{222}\text{Rn}$  transport in the soil is represented by Fick's first law:

$$F_{Rn,s} = -D^{CO_2} \frac{\partial(n_a C_{Rn,a})}{\partial z} \quad (0 \geq z \geq -L_{CO_2}), \quad (1)$$

where  $F_{Rn,s}$  (atoms  $\text{m}^{-2}$  of soil  $\text{s}^{-1}$ ) is the flux density of  $^{222}\text{Rn}$  across the geometric area in the soil,

$D^{CO_2}$  ( $m^2 s^{-1}$ ) is the diffusion coefficient of  $CO_2$  in the soil air,  $C_{Rn,a}$  (atoms  $m^{-3}$  of soil air) is the concentration of  $^{222}Rn$  in the soil air,  $z$  (m) is the depth from the air-ground interface and taken as negative in the downward direction,  $L_{CO_2}$  (m) is the thickness of the soil layer which is on the surface of the residue, and  $n_a$  is the air ratio of the soil defined as the ratio of the air volume to the total volume of the soil, and is expressed as  $n_a = n - n_w$ , where  $n$  is the porosity of the soil and  $n_w$  is the volumetric water content of the soil.

It is assumed that the soil microorganisms and the plant roots do not exist densely in the residue, and therefore  $^{222}Rn$  transport in the residue is represented by Fick's first law:

$$F_{Rn,r} = -D^{Rn,r} \frac{\partial(n_{a,r} C_{Rn,a,r})}{\partial z} \quad (-L_{CO_2} \geq z \geq -\infty), \quad (2)$$

where  $F_{Rn,r}$  (atoms  $m^{-2}$  of residue  $s^{-1}$ ) is the flux density of  $^{222}Rn$  across the geometric area in the residue,  $D^{Rn,r}$  ( $m^2 s^{-1}$ ) is the diffusion coefficient of  $^{222}Rn$  in the residue air (air contained in the residue),  $C_{Rn,a,r}$  (atoms  $m^{-3}$  of residue air) is the concentration of  $^{222}Rn$  in the residue air, and  $n_{a,r}$  is the air ratio of the residue defined as the ratio of the air volume to the total volume of the residue, and is expressed as  $n_{a,r} = n_r - n_{w,r}$ , where  $n_r$  is the porosity of the residue and  $n_{w,r}$  is the volumetric water content of the residue.

The diffusion coefficient of  $CO_2$  in the soil air,  $D^{CO_2}$ , and the diffusion coefficient of  $^{222}Rn$  in the residue air,  $D^{Rn,r}$ , are given by

$$D^{CO_2} = \frac{D_0^{CO_2}}{k} \quad (3)$$

and

$$D^{Rn,r} = \frac{D_0^{Rn}}{k_r}, \quad (4)$$

respectively, where  $k$  and  $k_r$  are the tortuosities of the soil and the residue, respectively, and  $D_0^{CO_2}$  ( $m^2 s^{-1}$ ) and  $D_0^{Rn}$  ( $m^2 s^{-1}$ ) are the molecular diffusion coefficients of  $CO_2$  and  $^{222}Rn$  in air, respectively.

The changes of the concentration of  $^{222}Rn$  in the soil air and the concentration of  $^{222}Rn$  in the residue air with depth  $z$  and time  $t$  (s) are given by Fick's second laws:

$$\frac{\partial(n_a C_{Rn,a})}{\partial t} = -\frac{\partial F_{Rn,s}}{\partial z} - \lambda(n_a C_{Rn,a}) + \rho X \quad (0 \geq z \geq -L_{CO_2}), \quad (5)$$

and

$$\frac{\partial(n_{a,r}C_{Rn,a,r})}{\partial t} = -\frac{\partial F_{Rn,r}}{\partial z} - \lambda(n_{a,r}C_{Rn,a,r}) + \delta_r \varrho_r A_{Ra,r} \quad (-L_{CO_2} \geq z \geq -\infty), \quad (6)$$

respectively, where  $\lambda$  ( $s^{-1}$ ) is the decay constant of  $^{222}\text{Rn}$ ,  $\varrho$  ( $\text{kg m}^{-3}$ ) and  $\varrho_r$  ( $\text{kg m}^{-3}$ ) are the bulk dry densities of the soil and the residue, respectively;  $X$  (atoms  $\text{kg}^{-1}$  of dry soil  $s^{-1}$ ) is the amount of  $^{222}\text{Rn}$  that is produced in the soil and that escapes into the air filled pore space at equilibrium state, and the amount is unknown;  $\delta_r$  is the  $^{222}\text{Rn}$  escape-to-production ratio of the residue, which is defined as the ratio of the amount of  $^{222}\text{Rn}$  that escapes into the air-filled pore space relative to the amount produced in the residue at equilibrium state, and  $A_{Ra,r}$  ( $\text{Bq kg}^{-1}$  of dry residue) is the concentration of  $^{226}\text{Ra}$  in the solid material of the residue.

Substituting Equations (1) and (3) into Equation (5) and assuming that  $n_a$  and  $k$  are constant, and substituting Equations (2) and (4) into Equation (6) and assuming that  $n_{a,r}$  and  $k_r$  are constant, we obtain

$$\frac{\partial C_{Rn,a}}{\partial t} = \frac{D_0^{CO_2}}{k} \frac{\partial^2 C_{Rn,a}}{\partial z^2} - \lambda C_{Rn,a} + \frac{\varrho X}{n_a} \quad (0 \geq z \geq -L_{CO_2}), \quad (7)$$

and

$$\frac{\partial C_{Rn,a,r}}{\partial t} = \frac{D_0^{Rn}}{k_r} \frac{\partial^2 C_{Rn,a,r}}{\partial z^2} - \lambda C_{Rn,a,r} + \frac{\delta_r \varrho_r A_{Ra,r}}{n_{a,r}} \quad (-L_{CO_2} \geq z \geq -\infty), \quad (8)$$

respectively.

First, assuming steady state  $\frac{\partial C_{Rn,a,r}}{\partial t} = 0$  and solving Equation (8) with the boundary condition  $C_{Rn,a,r}(z = -\infty) = \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}}$ , we have

$$C_{Rn,a,r} = A \exp\left(\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} z\right) + \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} \quad (-L_{CO_2} \geq z \geq -\infty), \quad (9)$$

where  $A$  is a constant to be determined. Substituting Equations (4) and (9) into Equation (2), we obtain

$$F_{Rn,r} = -n_{a,r} \sqrt{\frac{D_0^{Rn} \lambda}{k_r}} A \exp\left(\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} z\right) \quad (-L_{CO_2} \geq z \geq -\infty). \quad (10)$$

Next, assuming steady state  $\frac{\partial C_{Rn,a}}{\partial t} = 0$  and solving Equation (7), we have

$$C_{Rn,a} = B \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) + C \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) + D \quad (0 \geq z \geq -L_{CO_2}), \quad (11)$$

where  $B$ ,  $C$  and  $D$  are constants to be determined, and  $D = \frac{\rho X}{\lambda n_a}$ . Since  $z$  is equal to 0 at the air-ground interface, from Equation (11) we get

$$C_{Rn,a,0} = B + C + D, \quad (12)$$

where  $C_{Rn,a,0}$  (atoms  $m^{-3}$  of soil air) is the concentration of  $^{222}\text{Rn}$  in the soil air at the air-ground interface, which is equal to the concentration of  $^{222}\text{Rn}$  in the atmosphere at the air-ground interface. Rewriting Equation (12), we have

$$D = C_{Rn,a,0} - B - C. \quad (13)$$

Substitution of Equation (13) into Equation (11) gives

$$C_{Rn,a} = B \left\{ \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - 1 \right\} + C \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L_{CO_2}). \quad (14)$$

Substitution of Equations (3) and (14) into Equation (1) gives

$$F_{Rn,s} = -n_a \sqrt{\frac{D_0^{CO_2} \lambda}{k}} \left\{ B \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - C \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) \right\} \quad (0 \geq z \geq -L_{CO_2}). \quad (15)$$

Since  $z$  is equal to 0 at the air-ground interface, from Equation (15) we obtain

$$F_{Rn,s,0} = n_a \sqrt{\frac{D_0^{CO_2} \lambda}{k}} (C - B), \quad (16)$$

where  $F_{Rn,s,0}$  (atoms  $m^{-2}$  of soil  $s^{-1}$ ) is the flux density of  $^{222}\text{Rn}$  across the geometric area at the air-ground interface, namely exhalation rate of  $^{222}\text{Rn}$ . Rewriting Equation (16), we have

$$C = \frac{F_{Rn,s,0}}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} + B. \quad (17)$$

Substituting Equation (17) into Equation (15), we have

$$F_{Rn,s} = -n_a \sqrt{\frac{D_0^{CO_2}}{k}} \left[ B \left\{ \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) \right\} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) \right] \quad (0 \geq z \geq -L_{CO_2}). \quad (18)$$

Also, substituting Equation (17) into Equation (14), we obtain

$$C_{Rn,a} = B \left\{ \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) + \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L_{CO_2}). \quad (19)$$

The continuity of the concentration of  $^{222}\text{Rn}$  at the boundary between the layer of the soil and the layer of the residue requires that  $C_{Rn,a} = C_{Rn,a,r}$  at  $z = -L_{CO_2}$ . Thus we obtain the following relation from Equations (9) and (19):

$$A \exp\left(-\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} L_{CO_2}\right) + \frac{\delta_r Q_r A_{Ra,r}}{\lambda n_{a,r}} = B \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1 \right\} + C_{Rn,a,0}. \quad (20)$$

In addition, the continuity of the flux density of  $^{222}\text{Rn}$  exists across the boundary between these two layers,  $(F_{Rn,s})_{z=-L_{CO_2}} = (F_{Rn,r})_{z=-L_{CO_2}}$ . This leads to the following equation:

$$An_{a,r} \sqrt{\frac{D_0^{Rn}}{k_r}} \exp\left(-\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} L_{CO_2}\right) = n_a \sqrt{\frac{D_0^{CO_2}}{k}} \left[ B \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) \right\} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) \right]. \quad (21)$$

Combining Equation (21) with Equation (20), we get

$$\begin{aligned} & \frac{\delta_r Q_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1 \right\} \\ & = B \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) \right\} \end{aligned} \quad (22)$$

Therefore, the unknown  $B$  can be determined from Equation (22):

$$B = \frac{\frac{\delta_r Q_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1 \right\}}{\left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) \right\}}. \quad (23)$$

The unknown  $A$  can be determined from Equations (20) and (23):

$$\begin{aligned} A = & \frac{\frac{\delta_r Q_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1 \right\}}{\left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) \right\}} \\ & \times \left\{ \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2 \right\} \\ & + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1 \right\} + C_{Rn,a,0} - \frac{\delta_r Q_r A_{Ra,r}}{\lambda n_{a,r}} \exp\left(\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} L_{CO_2}\right). \end{aligned} \quad (24)$$



Substituting the determined constant  $B$  into Equation (19), we have

$$C_{Rn,a} = \frac{\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 1 \right\}}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) \right\}} \times \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} z} \right) + \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} z} \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} z} \right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L_{CO_2}). \quad (25)$$

Substituting the determined constant  $A$  into Equation (9), we have

$$C_{Rn,a,r} = \left[ \frac{\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 1 \right\}}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) \right\}} \right. \\ \times \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}} L_{CO_2}} \right) - 1 \right\} \\ \left. + C_{Rn,a,0} - \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} \right] \exp \left( \sqrt{\frac{k_r \lambda}{D_0^{Rn}} L_{CO_2}} \right) \exp \left( \sqrt{\frac{k_r \lambda}{D_0^{Rn}} z} \right) + \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} \quad (-L_{CO_2} \geq z \geq -\infty). \quad (26)$$

Now, the residue being replaced by the same soil as that on the residue, we get

$$\begin{aligned} \varrho_r &= \varrho, \\ n_{a,r} &= n_a, \\ k_r &= k, \\ \delta_r A_{Ra,r} &= X = \delta A_{Ra}, \end{aligned} \quad (27)$$

where  $\delta$  is the  $^{222}\text{Rn}$  escape-to-production ratio of the soil, which is defined as the ratio of the amount of  $^{222}\text{Rn}$  that escapes into the air-filled pore space relative to the amount produced in the soil at equilibrium state, and  $A_{Ra}$  ( $\text{Bq kg}^{-1}$  of dry soil) is the concentration of  $^{226}\text{Ra}$  in the solid material of the soil. In addition, it is assumed that the soil microorganisms and the plant roots do not exist densely in the replaced soil. Therefore,  $\varrho_r$ ,  $n_{a,r}$ ,  $k_r$ , and  $\delta_r A_{Ra,r}$  in Equations (25) and (26) may be replaced with  $\varrho$ ,  $n_a$ ,  $k$ , and  $\delta A_{Ra}$ , respectively, and we get

$$\begin{aligned}
C_{Rn,a} = & \frac{\frac{\delta \varrho A_{Ra}}{\lambda n_a} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \sqrt{\frac{D_0^{CO_2}}{D_0^{Rn}}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 1 \right\}}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \sqrt{\frac{D_0^{CO_2}}{D_0^{Rn}}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \\
& \times \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) + \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L_{CO_2}) \quad (28)
\end{aligned}$$

and

$$\begin{aligned}
C_{Rn,a} = & \left[ \frac{\frac{\delta \varrho A_{Ra}}{\lambda n_a} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 + \sqrt{\frac{D_0^{CO_2}}{D_0^{Rn}}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 1 \right\}}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \sqrt{\frac{D_0^{CO_2}}{D_0^{Rn}}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \right. \\
& \times \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 1 \right\} \\
& \left. + C_{Rn,a,0} - \frac{\delta \varrho A_{Ra}}{\lambda n_a} \right] \exp \left( \sqrt{\frac{k\lambda}{D_0^{Rn}}} L_{CO_2} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{Rn}}} z \right) + \frac{\delta \varrho A_{Ra}}{\lambda n_a} \quad (-L_{CO_2} \geq z \geq -\infty), \quad (29)
\end{aligned}$$

respectively. Rewriting Equations (28) and (29), we have

$$\begin{aligned}
C_{Rn,a} = & \frac{\sqrt{D_0^{Rn}} \left( \frac{\delta \varrho A_{Ra}}{\lambda n_a} - C_{Rn,a,0} \right) - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( \sqrt{D_0^{Rn}} + \sqrt{D_0^{CO_2}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \sqrt{D_0^{Rn}} \right\}}{\sqrt{D_0^{Rn}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \sqrt{D_0^{CO_2}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \\
& \times \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) + \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L_{CO_2}), \quad (30)
\end{aligned}$$

$$\begin{aligned}
< C_{Rn,a} = & \frac{\sqrt{D_0^{Rn}} \frac{\delta \varrho A_{Ra}}{\lambda n_a} - \sqrt{D_0^{Rn}} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 1 \right\} - \sqrt{D_0^{Rn}} C_{Rn,a,0} - \sqrt{\frac{D_0^{CO_2}}{n_a}} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right)}{\sqrt{D_0^{Rn}} \left\{ \exp \left( -L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) + \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 2 \right\} - \sqrt{D_0^{CO_2}} \left\{ \exp \left( -L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) \right\}} \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) + \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 2 \right\} \\
& + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z \right) - 1 \right\} + C_{Rn,a,0} \quad (0 \geq z \geq -L): \text{Equation (8) in Kataoka and Kigoshi (2014)} >
\end{aligned}$$

and

$$\begin{aligned}
C_{Rn,a} = & \left[ \frac{\sqrt{D_0^{Rn}} \left( \frac{\delta \varrho A_{Ra}}{\lambda n_a} - C_{Rn,a,0} \right) - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( \sqrt{D_0^{Rn}} + \sqrt{D_0^{CO_2}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \sqrt{D_0^{Rn}} \right\}}{\sqrt{D_0^{Rn}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \sqrt{D_0^{CO_2}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \right] \\
& \times \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 1 \right\} \\
& + C_{Rn,a,0} - \frac{\delta \varrho A_{Ra}}{\lambda n_a} \exp \left( \sqrt{\frac{k\lambda}{D_0^{Rn}}} L_{CO_2} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{Rn}}} z \right) + \frac{\delta \varrho A_{Ra}}{\lambda n_a} \quad (-L_{CO_2} \geq z \geq -\infty), \tag{31}
\end{aligned}$$

$$\begin{aligned}
< C_{Rn,a} = & \left[ \frac{\sqrt{D_0^{Rn}} \frac{\delta \varrho A_{Ra}}{\lambda n_a} - \frac{\sqrt{D_0^{Rn}}}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 1 \right\} - \sqrt{D_0^{Rn}} C_{Rn,a,0} - \frac{\sqrt{D_0^{CO_2}}}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right)}{\sqrt{D_0^{Rn}} \left\{ \exp \left( -L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) + \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 2 \right\}} \right] \\
& \times \left\{ \exp \left( -L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) + \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 2 \right\} \\
& + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( L \sqrt{\frac{k\lambda}{D_0^{CO_2}}} \right) - 1 \right\} + C_{Rn,a,0} - \frac{\delta \varrho A_{Ra}}{\lambda n_a} \exp \left( L \sqrt{\frac{k\lambda}{D_0^{Rn}}} \right) \exp \left( \sqrt{\frac{k\lambda}{D_0^{Rn}}} z \right) + \frac{\delta \varrho A_{Ra}}{\lambda n_a} \quad (-L \geq z \geq -\infty) : \text{Equation (9) in Kataoka and Kigoshi (2014)} >
\end{aligned}$$

respectively. Equations (30) and (31) are the same as Equations (4) and (5) in Kataoka (2018), respectively, and are also the same as Equations (8) and (9) in Kataoka and Kigoshi (2014), respectively.

Let us do the following in order to make Equations (30) and (31) easier to understand.

Rewriting Equation (24), we have

$$\begin{aligned}
A = & \frac{\frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left[ \left( \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} \right) \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} \right] \exp \left( \sqrt{\frac{k_r \lambda}{D_0^{Rn}}} L_{CO_2} \right)}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \tag{32}
\end{aligned}$$

The unknown  $C$  can be determined from Equations (16) and (23):

$$\begin{aligned}
C = & \frac{\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{ \left( 1 - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \right) \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 1 \right\}}{\left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) + \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - 2 \right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{ \exp \left( -\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) - \exp \left( \sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2} \right) \right\}} \tag{33}
\end{aligned}$$

The unknown  $D$  can be determined from Equations (12), (23), and (33):

$$D = \frac{-2 \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} + \left(1 - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \left(C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0}\right) \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \left(1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \left(C_{Rn,a,0} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0}\right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)}{\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}} \quad (34)$$

Substituting Equations (23), (33) and (34) into Equation (11), we have

$$C_{Rn,a} = \frac{\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{\left(1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1\right\}}{\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}} \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) \\ + \frac{\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{\left(1 - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 1\right\}}{\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}} \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} z\right) \\ + \frac{-2 \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} + \left(1 - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \left(C_{Rn,a,0} - \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0}\right) \exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \left(1 + \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}}\right) \left(C_{Rn,a,0} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0}\right) \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)}{\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}} \quad (0 \geq z \geq -L_{CO_2}). \quad (35)$$

Also, substituting Equation (32) into Equation (9), we get

$$C_{Rn,a,r} = \frac{\frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left[ \left(\frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} - C_{Rn,a,0}\right) \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\}\right] \exp\left(\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} L_{CO_2}\right)}{\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \frac{n_a}{n_{a,r}} \sqrt{\frac{k_r D_0^{CO_2}}{D_0^{Rn} k}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}} \\ \times \exp\left(\sqrt{\frac{k_r \lambda}{D_0^{Rn}}} z\right) + \frac{\delta_r \varrho_r A_{Ra,r}}{\lambda n_{a,r}} \quad (-L_{CO_2} \geq z \geq -\infty). \quad (36)$$

Replacing  $\varrho_r$ ,  $n_{a,r}$ ,  $k_r$ , and  $\delta_r A_{Ra,r}$  in Equations (32), (23), (33), (34), (35), and (36) with  $\varrho$ ,  $n_a$ ,  $k$ , and  $\delta A_{Ra}$ , respectively, and rewriting them, we have

$$A = \frac{\sqrt{D_0^{CO_2}} \left[ \left(\frac{\delta \varrho A_{Ra}}{\lambda n_a} - C_{Rn,a,0}\right) \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\} + \frac{1}{n_a} \sqrt{\frac{k}{D_0^{CO_2} \lambda}} F_{Rn,s,0} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\}\right] \exp\left(\sqrt{\frac{k\lambda}{D_0^{Rn}}} L_{CO_2}\right)}{\sqrt{D_0^{Rn}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - 2\right\} - \sqrt{D_0^{CO_2}} \left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}} L_{CO_2}\right)\right\}}, \quad (37)$$



respectively.

Finally, we describe that Equation (41) (i. e. Equation (30)) and Equation (42) (i. e. Equation (31)) are equal to the steady state solutions of Equations (5) and (6) (differential equations) in Kataoka and Kigoshi (2014) (see Appendix), respectively. In order for these relations to be established,  $D$  (Equation (40)) must be equal to  $\frac{\delta Q A_{Ra}}{\lambda n_a}$ . Hence

$$\frac{-2\sqrt{D_0^{Rn}}\frac{\delta Q A_{Ra}}{\lambda n_a} + \left(\sqrt{D_0^{Rn}} - \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} - \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) + \left(\sqrt{D_0^{Rn}} + \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} + \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right)}{\sqrt{D_0^{Rn}}\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) - 2\right\} - \sqrt{D_0^{CO_2}}\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right)\right\}} = \frac{\delta Q A_{Ra}}{\lambda n_a}. \quad (43)$$

Rewriting Equation (43), we have

$$\frac{\left(\sqrt{D_0^{Rn}} - \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} - \frac{\delta Q A_{Ra}}{\lambda n_a} - \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) + \left(\sqrt{D_0^{Rn}} + \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} - \frac{\delta Q A_{Ra}}{\lambda n_a} + \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right)}{\sqrt{D_0^{Rn}}\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) + \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) - 2\right\} - \sqrt{D_0^{CO_2}}\left\{\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) - \exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right)\right\}} = 0. \quad (44)$$

Equations (5) and (6) in Kataoka and Kigoshi (2014) being solved in the steady state, the condition under which the concentration of  $^{222}\text{Rn}$  in the soil air and the flux density of  $^{222}\text{Rn}$  in the soil are both continuous at  $z = -L_{CO_2}$  is given by

$$\left(\sqrt{D_0^{Rn}} - \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} - \frac{\delta Q A_{Ra}}{\lambda n_a} - \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(-\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) + \left(\sqrt{D_0^{Rn}} + \sqrt{D_0^{CO_2}}\right)\left(C_{Rn,a,0} - \frac{\delta Q A_{Ra}}{\lambda n_a} + \frac{1}{n_a}\sqrt{\frac{k}{D_0^{CO_2}\lambda}}F_{Rn,s,0}\right)\exp\left(\sqrt{\frac{k\lambda}{D_0^{CO_2}}}L_{CO_2}\right) = 0. \quad (45)$$

Equation (45) indicates that Equation (44) is established, and that therefore Equations (41) (i. e. Equation (30)) and Equation (42) (i. e. Equation (31)) become the steady state solutions of Equations (5) and (6) in Kataoka and Kigoshi (2014), respectively.

## References

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## Appendix

Equations (5) and (6) in Kataoka and Kigoshi (2014) are

$$\frac{\partial(n_a C_{Rn,a})}{\partial t} = -\frac{\partial}{\partial z} \left\{ \frac{D_0^{CO_2}}{k} \frac{\partial(n_a C_{Rn,a})}{\partial z} \right\} - \lambda(n_a C_{Rn,a}) + \delta \varrho A_{Ra} \quad (0 \geq z \geq -L) \quad (A-1)$$

and

$$\frac{\partial(n_a C_{Rn,a})}{\partial t} = -\frac{\partial}{\partial z} \left\{ \frac{D_0^{Rn}}{k} \frac{\partial(n_a C_{Rn,a})}{\partial z} \right\} - \lambda(n_a C_{Rn,a}) + \delta \varrho A_{Ra} \quad (-L \geq z \geq -\infty), \quad (A-2)$$

respectively, and it is assumed that  $n_a$ ,  $k$ ,  $\delta$ ,  $\varrho$ , and  $A_{Ra}$  are constant under the ground.  $L$ ,  $L_{CO_2}$  in this contribution, is the depth (positive) to which the soil microorganisms and the plant roots exist densely.