

Radioactive Ingrowth of Polonium-210 in Tobacco Plants

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Polonium-210 is an α -emitting radioactive element found in tobacco and thought to be a possible initiator of bronchial cancer in cigarette smokers. An evaluation of the ingrowth of $^{210}_{84}\text{Po}$ in leaves of tobacco showed it to be de-

rived from and in radioactive equilibrium with $^{210}_{82}\text{Pb}$. Linear extrapolations of $^{210}_{84}\text{Po}$ ingrowth obtained by a modified Bateman equation indicated that $^{210}_{82}\text{Pb}$ entered the tobacco leaves during the growing season.

Radioactive ingrowth as used in this discussion is an increase in the concentration of $^{210}_{84}\text{Po}$ in tobacco leaves during storage in a noncontaminating environment. Previous investigators (Radford and Hunt, 1964) have suggested that $^{210}_{84}\text{Po}$ is in radioactive equilibrium with $^{210}_{82}\text{Pb}$ in cigarette tobacco. An attempt to verify such an equilibrium was unsuccessful because the activity of $^{210}_{82}\text{Pb}$ in tobacco tissue could not be measured and no evidence of a $^{210}_{82}\text{Pb} \rightarrow ^{210}_{84}\text{Po}$ radioactive equilibrium in plant tissue is available (Tso *et al.*, 1964).

An investigation was initiated to evaluate the extent of $^{210}_{84}\text{Po}$ ingrowth in tobacco and to determine the possible existence of an equilibrium between $^{210}_{82}\text{Pb}$ and $^{210}_{84}\text{Po}$.

Procedure

The $^{210}_{84}\text{Po}$ was released from plant tissue by the wet ashing procedure described by Jackson (1958). Care was taken to volatilize all HNO_3 to prevent interference in the subsequent plating of $^{210}_{84}\text{Po}$ on Ag disks.

Polonium-210 is the only α -emitter which spontaneously deposits electrochemically on Ag in dilute HCl (Radford *et al.*, 1963). For trace amounts of $^{210}_{84}\text{Po}$, deposition is independent of HCl concentration providing it is in excess of 0.1*N*, and the rate of deposition is increased with increased temperature in the range 25° to 100° C.

The plating of $^{210}_{84}\text{Po}$ on the Ag disks was accomplished by heating at 90° to 100° C. in 0.5*N* HCl solution with continuous stirring for 4 hours. Reduction of Fe^{+3} (which decreases plating efficiency) in the samples was accomplished by the addition of 0.1 gram of ascorbic acid. At the completion of the plating time, the Ag disk was washed with water and air-dried; the α -activity was counted in a Molechem Model 30 solid-state semiconductor α -detector. Counting time was a minimum of 4 hours. Details of methodology, reproducibility, and precision are described by Francis and Chesters (1967).

Results and Discussion

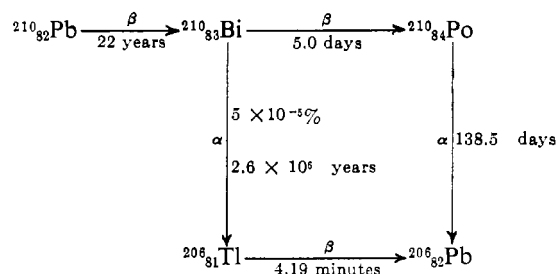
The $^{210}_{84}\text{Po}$ content of cured tobacco leaves was determined at intervals of approximately three months for a period of one year (Table I). All tobacco was grown in Wisconsin during the summer of 1965 and cured under natural conditions in the fall of 1965.

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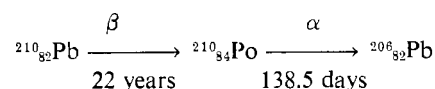
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The tobacco plants were harvested between September 5 and 10; on December 6 they were taken from the tobacco sheds of their respective growers. The leaves were randomly selected from individual plants, dried in a forced-air oven at 80° C., ground to pass an 8-mesh screen, and stored in airtight glass bottles. Increasing concentrations of $^{210}_{84}\text{Po}$ were found after storage, thereby proving the existence of $^{210}_{84}\text{Po}$ ingrowth from some radioactive precursor. To determine the radioactive precursor responsible for such $^{210}_{84}\text{Po}$ ingrowth, one must consider the decay chain of $^{238}_{92}\text{U}$ and establish criteria for the existence of an equilibrium between $^{210}_{84}\text{Po}$ and $^{210}_{82}\text{Pb}$.

In the $^{238}_{92}\text{U}$ radioactive decay series, $^{210}_{83}\text{Bi}$ is formed by the loss of a β particle from $^{210}_{82}\text{Pb}$. Approximately 100% of the $^{210}_{83}\text{Bi}$ decays by β emission forming $^{210}_{84}\text{Po}$ which undergoes the loss of an α particle to form a stable lead isotope, $^{206}_{82}\text{Pb}$.



Since the half-life of $^{210}_{83}\text{Bi}$ is 5.0 days, the activity of $^{210}_{84}\text{Po}$ in tobacco leaves 20 days after the entry of $^{210}_{82}\text{Pb}$ ($^{210}_{83}\text{Bi}$ has reached 15/16 of its steady-state value) will depend on the concentration of $^{210}_{82}\text{Pb}$ and $^{210}_{84}\text{Po}$ and their respective decay rates.



Since

$$\frac{dN_{\text{Pb}}}{dt} = -\lambda_{\text{Pb}}N_{\text{Pb}}, \text{ or } N_{\text{Pb}} = N_{\text{Pb}}^0 e^{-\lambda_{\text{Pb}}t}$$

where N_{Pb}^0 is the initial $^{210}_{82}\text{Pb}$ concentration and λ is the decay rate constant.

Also, $\frac{dN_{\text{Po}}}{dt} = \lambda_{\text{Pb}}N_{\text{Pb}} - \lambda_{\text{Po}}N_{\text{Po}}$ where N_{Po} is the concentration of $^{210}_{84}\text{Po}$; substituting $N_{\text{Pb}}^0 e^{-\lambda_{\text{Pb}}t}$ for N_{Pb}

$$\frac{dN_{\text{Po}}}{dt} = \lambda_{\text{Pb}}N_{\text{Pb}}^0 e^{-\lambda_{\text{Pb}}t} - \lambda_{\text{Po}}N_{\text{Po}}$$

Table I. Ingrowth of $^{210}_{84}\text{Po}$ in Tobacco Leaves

| Sample ^a | $^{210}_{84}\text{Po}$ Contents ^a in Pc./G., Determined in ^b | | | |
|---------------------|--|-------------|-------------|-------------|
| | Dec. 1965 | Mar. 1966 | June 1966 | Aug. 1966 |
| 1A | 0.80 ± 0.09 | 1.56 ± 0.13 | 2.02 ± 0.15 | |
| 1B | 0.76 ± 0.09 | 1.30 ± 0.13 | 1.60 ± 0.13 | 1.86 ± 0.13 |
| 1C | 0.58 ± 0.09 | 1.62 ± 0.13 | 2.02 ± 0.15 | |
| 2A | 0.21 ± 0.06 | 1.71 ± 0.15 | 2.54 ± 0.15 | |
| 2B | 1.16 ± 0.11 | 2.01 ± 0.15 | 2.76 ± 0.15 | 2.76 ± 0.15 |
| 2C | 0.87 ± 0.09 | 1.73 ± 0.15 | 2.01 ± 0.15 | |
| 3A | 0.60 ± 0.09 | 1.25 ± 0.11 | 1.86 ± 0.15 | |
| 3B | 0.52 ± 0.07 | 1.41 ± 0.13 | 1.93 ± 0.15 | 2.18 ± 0.15 |
| 3C | 1.47 ± 0.11 | 1.93 ± 0.15 | 2.10 ± 0.15 | |
| 4A | 0.58 ± 0.07 | 1.34 ± 0.13 | 1.49 ± 0.13 | |
| 4B | 0.75 ± 0.09 | 1.41 ± 0.13 | 1.51 ± 0.13 | 1.76 ± 0.13 |
| 4C | 1.56 ± 0.13 | 1.17 ± 0.11 | 1.51 ± 0.13 | |
| 5A | 0.85 ± 0.09 | 1.32 ± 0.13 | 1.58 ± 0.13 | |
| 5B | 0.82 ± 0.09 | 1.12 ± 0.10 | 1.34 ± 0.13 | 1.27 ± 0.11 |
| 5C | 1.10 ± 0.11 | 1.19 ± 0.11 | 1.81 ± 0.15 | |

^a Samples A, B, and C are different plants taken from the same site.
^b Standard deviation in pc./g. determined from (counts)^{1/2} (time)⁻¹.

rearranged is

$$\frac{dN_{Po}}{dt} + \lambda_{Po}N_{Po} - \lambda_{Pb}N_{Pb}^0 e^{-\lambda_{Pb}t} = 0$$

which is a linear differential equation of the first order in which the final solution is:

$$N_{Po} = \frac{\lambda_{Pb}}{\lambda_{Po} - \lambda_{Pb}} N_{Pb}^0 (e^{-\lambda_{Pb}t} - e^{-\lambda_{Po}t}) + N_{Po}^0 e^{-\lambda_{Po}t}$$

By multiplying this equation by $e^{\lambda_{Po}t}$ one obtains the equation

$$N_{Po} e^{\lambda_{Po}t} = \frac{\lambda_{Pb}}{\lambda_{Po} - \lambda_{Pb}} N_{Pb}^0 [e^{(\lambda_{Po} - \lambda_{Pb})t} - 1] + N_{Po}^0 \dots (1)$$

which is in the form of a linear equation $y = mx + c$ where $y = N_{Po} e^{\lambda_{Po}t}$ expressed as number of atoms of

$^{210}_{84}\text{Po}/\mu\text{g. tobacco}$, $x = \frac{\lambda_{Pb}}{\lambda_{Po} - \lambda_{Pb}} [e^{(\lambda_{Po} - \lambda_{Pb})t} - 1]$.

Substituting $\lambda_{Pb} = 8.6 \times 10^{-5} \text{ day}^{-1}$ and $\lambda_{Po} = 5.0 \times 10^{-3} \text{ day}^{-1}$, $x = 1.76 \times 10^{-2} [e^{4.91 \times 10^{-2}t} - 1]$, which is dimensionless but a function of t . $m = N_{Pb}^0$ and $c =$

N_{Po}^0 . If the major portion (>95%) of the $^{210}_{84}\text{Po}$ in the leaves of tobacco is derived from $^{210}_{82}\text{Pb}$, plotting the ingrowth of $^{210}_{84}\text{Po}$ with the above equation should give a straight line intersecting the x -axis at the latest date of entry of $^{210}_{82}\text{Pb}$ into the tobacco leaves since x is a function of time.

The ingrowth of $^{210}_{84}\text{Po}$ in leaves from five tobacco plants were plotted according to Equation 1 using data from Table II; t is expressed in days and $t = 0$ is taken as July 1, 1965. Figure 1 shows that a linear relationship exists between x and y within the standard deviation of counting for each of the five samples. Thus, $^{210}_{84}\text{Po}$ in the tobacco leaves is derived from and therefore in equilibrium with $^{210}_{82}\text{Pb}$.

The intercepts on the X -axes (Figure 1) indicate that $^{210}_{82}\text{Pb}$ had entered the tobacco leaves before October 1, 1965 in four of the five tobacco samples. Sample 3 indicates that $^{210}_{82}\text{Pb}$ entered the plant before November 1, 1965. Extrapolations of this type are subject to error; however, in four of the five cases the calculated final date of $^{210}_{82}\text{Pb}$ entry into the plant occurred during the growing season, which is in agreement with Tso

Table II. Dates of $^{210}_{84}\text{Po}$ Analysis in Cured Tobacco Leaves

| Sample | Dec. | Time, Days | March | Time, Days | June | Time, Days | Aug. | Time, Days |
|--------|------|------------|-------|------------|------|------------|------|------------|
| 1B | 10 | 163 | 23 | 266 | 23 | 358 | 26 | 422 |
| 2B | 10 | 163 | 5 | 244 | 16 | 351 | 26 | 422 |
| 3B | 10 | 163 | 1 | 240 | 14 | 352 | 26 | 422 |
| 4B | 10 | 163 | 23 | 266 | 23 | 358 | 26 | 422 |
| 5B | 10 | 163 | 10 | 156 | 21 | 356 | 26 | 422 |

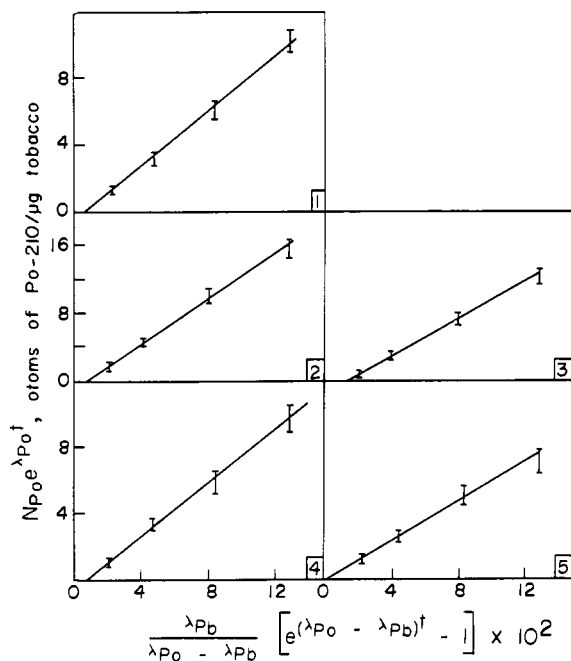


Figure 1. Linear plots of ^{210}Po ingrowth in tobacco leaves

et al. (1966). More importantly, measurements of this type prove conclusively that ^{210}Po does not enter the plant per se but enters as ^{210}Pb either by plant uptake or by deposition in natural radioactive fallout.

Acknowledgment

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