Polonium-210: A Volatile Radioelement in Cigarettes

Abstract. *Polonium-210, which emits alpha particles, is a natural contaminant of tobacco. For an individual smoking two packages of cigarettes a day, the radiation dose to bronchial epithelium from Po^{210} inhaled in cigarette smoke probably is at least seven times that from background sources, and in localized areas may be up to 1000 rem or more in 25 years. Radiation from this source may, therefore, be significant in the genesis of bronchial cancer in smokers.*

Although it is well known that ionizing radiation is carcinogenic in man (*1*), there has been no evidence that radioisotopes in cigarettes are implicated in the production of lung cancer. Measurements of potassium-40 and radium isotopes have been made in tobacco (*2, 3*) but these elements are not volatile at the temperature of a burning cigarette—600°C to 800°C (*4*)—and therefore cannot deliver any significant radiation dose. Turner and Radley (*3*) measured total alpha-particle activity in raw tobacco and cigarette ashes, and concluded that the dose derived from radon and its daughter products would be less than 1 percent of that of the normal background to the bronchial epithelium. Our studies were undertaken to reevaluate the concentration of alpha-emitting radioelements in cigarettes. In our experiments we have found in tobacco significant amounts of Po^{210}, which we believe to be in equilibrium with its parent Pb^{210}. A substantial part of this polonium appears in cigarette smoke, and on the basis of certain assumptions we calculate that polonium may constitute a significant initiator of neoplasia in the bronchial epithelium of a cigarette smoker.

The methods we have used for separating radium isotopes and Po^{210} have been described previously (*5*). Briefly, the sample is treated with hot concentrated HCl (wet-ashing), the polonium is plated on silver, and the radium isotopes are then coprecipitated with lead sulfate and barium sulfate. We have not found it necessary to modify our routine procedure in separating polonium or radium from cigarettes, although with digestion by hot concentrated HCl, solution of cigarettes or ash is incomplete and a considerable amount of undigested material remains. Nevertheless, we have recovered nearly 100 percent of polonium added to cigarettes during digestion, the same recovery as that for teeth and bones (*5*). Polonium samples were counted in gas-flow proportional counters with background counts of alpha particles in the range of 0.5 to 1.5 count/hr. We have confirmed that the radioactivity plated on silver with the initial separation is Po^{210} by observing its decay, which has closely conformed to the Po^{210} half-life of 138 days.

This report deals only with the polonium content of cigarettes and cigarette smoke. The cigarettes we used were four of the regular-sized American brands purchased in local stores. The smoke from cigarettes puffed artificially was obtained by drawing air through cigarettes connected to a filter holder. This holder contained a fiberglass prefilter (*6*) backed by a Millipore HA filter that retains 100 percent of the particles in the size range of tobacco-smoke particles (*7*). The fiberglass prefilter actually collected nearly all of the smoke and was used to prevent rapid clogging of the HA filter. From the filter holder, gas from the smoking cigarette was led through a trap containing 50 ml of 0.5 normal HCl, then to a rotameter connected to a vacuum line. When the cigarette was smoked artificially it was in a nearly vertical position. The vacuum line was clamped off between puffs, and during each puff the flow rate was maintained at 15 ml/sec for 2 to 3 seconds. The puffs were carried out every 50 seconds for 6 minutes (or eight puffs); this puffing pattern was the average of that, observed without their knowledge, for a number of smokers in our laboratory, and is similar to that reported by Hilding (*8*). It resulted in consumption of about 60 to 70 percent of the cigarette, or somewhat less than that observed in cigarettes smoked by human subjects. Side stream smoke (smoke not drawn through the cigarette) was captured by placing a liter bottle over the burning cigarette. Air and smoke in the bottle were continuously drawn through the same filter system as the mainstream smoke, but the tube from the bottle was clamped only during puffing, when the tube from the cigarette was open. In this way both side stream and mainstream smoke were trapped together on the filters.

In these artificial smoking experiments, the following portions of the cigarette and smoke were analyzed for polonium: the ash and butt of the cigarette; smoke condensate on the fiberglass and HA filters; condensate that formed on the tubing and metal parts of the filter holder and on the walls of the liter bottle, all of which were carefully wiped after the cigarette was smoked; and the HCl trap through which the filtered air had been drawn. Blank analyses on filters, trap solution, and wipings of the tubing, filter holder, and bottle showed no polonium activity with the exception of the fiberglass filters, which had approximately 0.015 pc of Po^{210}. The HA filter and HCl trap showed little or no polonium after smoking, and in the results the content observed on the fiberglass (usually about 90 percent of the total) was combined with that from the condensate and HA filter.

Table 1 shows the polonium content of whole cigarettes of four brands. Also given are the contents of the ash, butt, and total smoke when all of the smoke, including the side stream, was trapped. Included in Table 1 are the results for mainstream smoke alone collected in different experiments. Table 1 indicates that only about half the polonium in the total smoke was in the mainstream when cigarettes were puffed by our technique. When all the smoke was captured, about 80 percent of the polonium disappearing in the smoking process could be accounted for. These results suggest that most of the volatile polonium is rapidly adsorbed on the smoke particles, although a small part could be gaseous and escape our trapping procedure, including the HCl trap. Because of the nature of the smoking pattern that was used, we believe that the mainstream smoke is probably low in these experiments, compared to actual smoking conditions. For example, the higher concentrations in the butt of brand D, Table 1, indicate that these cigarettes were not as completely smoked as the others. For this reason no significance can be attached to differences in mainstream content between filter and nonfilter brands. When cigarettes were smoked by human subjects the polonium content of the butt and ash was lower than that of the artificially smoked cigarettes, and the proportion of polonium lost from the cigarette was higher for filter cigarettes. We suspect that the amount of polon-

Table 1. Polonium content of American cigarettes and smoke from cigarettes artificially puffed. Figures in parentheses are number of analyses.

Po^{210} content (pc)					Recovery* (%)	Po^{210} in mainstream smoke (pc)	Ratio of mainstream to total smoke (%)
Whole cigarette	Ash	Butt	Total smoke	Total in ash, butt, and smoke			
			Brand A, nonfilter				
0.43(4)	0.031(2)	0.13(2)	0.19(2)	0.35	81	0.10(3)	52
			Brand B, nonfilter				
0.48(5)	0.053(2)	0.12(2)	0.26(2)	0.43	90	0.12(2)	46
			Brand C, filter				
0.39(4)	0.035(2)	0.094(2)	0.19(2)	0.32	82	0.088(2)	47
			Brand D, filter				
0.40(4)	0.033(2)	0.15(2)	0.17(2)	0.35	88	0.070(3)	41

* Ratio of total in ash, butt, and smoke to total in whole cigarette.

ium absorbed by an individual may be dependent on the mode of puffing as well as on the fraction of total smoke inhaled.

The alpha-emitting isotopes we have investigated all occur naturally, and presumably have always been present in tobacco. Absorption of Pb^{210}, the parent of Po^{210}, by the plant roots may be supplemented by foliar absorption from "natural fallout" from decay of Rn^{222} in the atmosphere (*9*). From analysis of 5-year-old cigarettes, we conclude that the amount of polonium in fresh whole cigarettes (Table 1) is in equilibrium with the Pb^{210} parent. With respect to radium and polonium content, tobacco appears to be typical of plants generally, as judged by the total alpha-particle activity found in plants by Mayneord *et al.* (*10*). The radiation hazard from Po^{210} arises primarily because polonium is known to be completely volatile above 500°C (*11*), or well below the temperature of a burning cigarette. In addition, polonium binds rapidly and strongly to surfaces, and hence attaches readily to smoke particles. Finally, its intermediate half-life of 138 days assures ample time for translocation of particles to the bronchi to take place.

The basic question which arises from our measurements concerns the radiation dose to the bronchial epithelium from Po^{210} present in tobacco smoke. To put the following calculations in perspective, we estimate that the background dose to the bronchial epithelium is approximately 200 mrem per year, or about 5 rem per 25 years—similar to estimates by Chamberlain and Dyson (*12*) and Shapiro (*13*). In this estimate we have used a relative biological effectiveness (RBE) of 10 for alpha particles, in accord with recommendations of the International Commission for Radiation Protection (*14*).

The radiation dose delivered by polonium inhaled from cigarettes can be analyzed for two conditions. The first condition defines approximately the minimum radiation dose to be expected; in this case the dose arises from particles carried across the bronchial epithelium in the process of excretion by mucus flow up the bronchial tree. The smoke particles are assumed to be deposited by diffusion, largely on the alveolar epithelium, from where they are phagocytosed and carried up the bronchial epithelium (*15*). In this case we estimate that the minimum dose delivered by these particles for an individual smoking two packages of cigarettes a day for 25 years would be about 36 rem (*16*) or seven times the background exposure. This estimate does not take account of the radiation dose arising from Pb^{210} absorption in smoke, either from the beta particles emitted by Pb^{210} and Bi^{210}, or from the polonium daughter which would arise in the lungs from lead absorption. In addition, this calculation neglects the slowing effect of smoke on ciliary action (*17*), which would prolong the exposure time and increase the dose. For these reasons we believe that this estimate is probably conservative, and the dose could be 100 rem or more from this process.

The second condition defines the dose which might arise if local concentration of polonium occurs in various regions of the bronchial tree. The radiation dose delivered by local concentrations of polonium from particles in the bronchial epithelium wall itself, depends on the fraction of particles accumulated in these regions, and on the mean residence time occurring in such cases. There is no quantitative basis on which to estimate these two factors in human lungs, but it is likely from our preliminary measurements of polonium in the bronchial epithelium of lungs of smokers, that these local doses may range from several hundred rem to more than 1000 rem, in the case of an individual smoking two packs a day over a 25-year period. For example, in a 73-year old male who died of cardiac failure, who had smoked "one or more" packs of cigarettes per day for many years, the polonium content in the epithelium of a secondary bifurcation of the right lower lobe was 0.033 pc/cm^2, compared with 0.003 pc/cm^2 in the right main stem bronchus. This man had not smoked after hospital admission for smoke inhalation 10 days prior to death, and he had evidence of bronchial pneumonia at autopsy, so it is probable that equilibrium concentration of polonium in the epithelium at the time he was smoking was substantially higher. Even so, a level of 0.033 pc/cm^2 would give a dose of 165 rem in 25 years. This figure is a minimum value even for this region of epithelium, and higher values could also be present in heavier smokers. Further work will be required to determine these local doses, particularly through use of quantitative radiographs.

With regard to the amount of polonium absorbed into the circulation from tobacco smoke, compared to polonium absorbed from other sources, it is of interest that the urine of nonsmokers contains very little polonium (a mean of 0.011 pc per 24 hours for four subjects). On the other hand, three smokers who smoked an average of two packages of cigarettes a day were found to have an average polonium excretion of 0.065 pc per 24 hours, a nearly sixfold difference. These results suggest that the polonium content of the soft tissues of the body may be significantly elevated in smokers.

A comparison of death rates of lung cancer in smokers may be made to the well-known bronchial cancer incidence in miners exposed to radon daughters (*18*). We calculate the lung-cancer death rate in these miners to be 3 percent per year; their radiation exposure to the bronchial epithelium would be about 20,000 rem in the 17-year induction period (*19*). The radiation dose necessary to account for the lung-cancer death rate in males smoking 40

cigarettes a day or more—about 0.205 percent per year (*20*)—would, on this basis, be about 1300 rem over a 25-year period. A dose of this magnitude from polonium is probable only in localized areas of the bronchial tree, but the causes of lung cancer may not be identical in the two cases (particularly in the Schneeberg miners whose deaths occurred before cigarette smoking was widespread) because of the presence of strong cocarcinogens in cigarette smoke (*21*). Because of the well-known synergistic action of ionizing radiation and cigarette-smoke extracts, or other chemical agents, in experimental cancer production (*22*), the presence of these chemical promoters might lead to cancer from radiation doses at least an order of magnitude less than the figure of 1300 rem. A dose of 100 to 200 rem to the bronchial epithelium may be highly significant, therefore, and even doses at the lower estimate of 36 rem may not be negligible if the dose-response curve for cancer induction is linear for alpha-emitting substances. This general comparison is independent of the relative biological effectiveness chosen for alpha particles.

We support the view that other chemical factors, particularly cocarcinogens, as well as physiological effects, such as alterations of ciliary activity by cigarette smoke, probably play an important part in the genesis of bronchial cancer in smokers. Our present conclusion is that Po^{210} inhaled in cigarette smoke may act as an important initiator in the production of bronchogenic carcinoma.

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23 December 1963

Novel Filter for Biological Materials

Abstract. *Thin plastic sieves with precisely controlled hole size and density can be made by irradiating plastic films with fission fragments and etching out the material traversed by the fragments. These filters may be used for the nondestructive separation of cells of closely similar sizes.*

Studies of radiation-damage tracks in certain solids (*1*) have recently led to a method of drilling fine holes of adjustable size and number in thin sheets of these solids (*2*). The method has now been extended to a number of plastics which are commercially obtainable in sheet form. From plastic film it is now possible to produce fairly large numbers of filters which have important advantages over conventional filters in certain research fields such as cytology and bacteriology.

Briefly, the technique is to bombard a plastic sheet at near normal incidence with fission fragments, which produce continuous trails of radiation-damaged material on the sheet. Fine holes are formed by preferentially dissolving the damage trails in a suitable reagent, which then continues to enlarge the holes at a uniform rate until the desired size is reached. The number of holes is equal to the number of fission particles traversing the plastic film (*2*).

Only heavy, high-energy particles produce continuous trails of chemically altered material. Alpha particles from a Po^{210} source, although not suitable for most plastics, so alter the local structure of cellulose nitrate that holes form (Fig. 1). Lighter particles such as protons, electrons, x-rays, and γ-rays do not produce this effect.

Filters with hole sizes ranging from about 1 μ or less up to about 10 μ and with about 2 percent open area have been made from 0.013 mm (1/2 mil) films of polycarbonate resins and polyester resins. Filters have also been made from a number of experimental plastic films which are not yet commercially available. Extremely uniform holes are formed in polycarbonate film after irradiation and etching (see cover photograph). Figure 2 is a graph of hole diameter plotted against etching time in NaOH solution for an 0.013 mm film. The rate of hole enlargement increases with stirring rate, reagent concentration, and temperature.

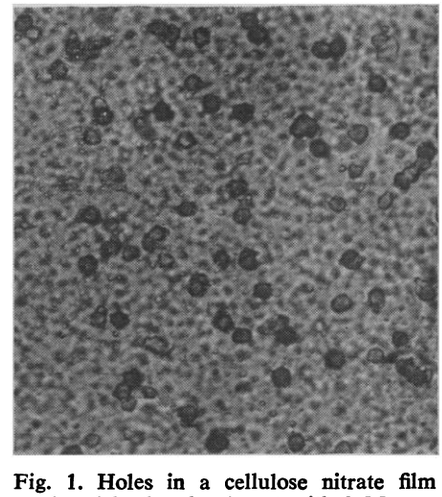

Fig. 1. Holes in a cellulose nitrate film produced by bombardment with 3 Mev α-particles followed by a 3-minute etch in 6*N* NaOH aqueous solution at 75°C.