

Sources of Polonium-210 within the troposphere

By ALAIN MARENCO and JACQUES FONTAN, *Physique des Aérosols et Echanges Atmosphériques, Centre de Physique Atomique et Nucléaire, Université Paul Sabatier, Toulouse*

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ABSTRACT

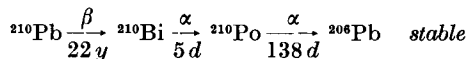
Contemporary technical literature has given consideration to complementary sources of ^{210}Po (fossil, terrestrial, stratospheric, artificial). A discussion of the importance of these various sources, based on short-time measurements of ^{210}Po , of ^{210}Pb , of long-lived nuclides and of products representative of pollution, carried out in S-W France, is given herein. It is concluded that tropospheric ^{210}Po is of natural origin and derived essentially from the decay of the ^{210}Pb present in the atmosphere.

Introduction

Polonium-210, a long-lived daughter of the Uranium-Radium group is formed within the atmosphere by the decay of the Lead-210 produced by the radon diffused from the soil.

The decay-scheme leading to the ^{210}Po within the atmosphere can be summarized as follows:

Rn \rightarrow (short-lived progeny):



It is used as a tracer in atmospheric physics with special application to the determination of the residence-time of the radioactive aerosols present in the air (Burton, 1960; Bhandari, 1963; Jacobi, 1963; Lambert, 1965; Nezami, 1966).

Its origin within the troposphere and the existence of complementary sources of supply have been discussed by a number of authors (Styra, 1960; Stebbins, 1961; Osborne, 1963; Peirson, 1967; Karol, 1969; Mattson, 1970; Martell, 1969, 1970 (1), 1970 (2); Vilenskiy, 1970).

The proportion of ^{210}Po of stratospheric origin, in particular, is not well established.

From simultaneous measurements of the concentrations of various long-lived radioactive elements carried out at a single location, we have studied the probable sources of ^{210}Po in S-W France.

I. The origins of Polonium-210 in the atmosphere

In order to explain the high activity values of ^{210}Po often encountered in the lower atmospheric layers and, in particular, to explain the large differences in the residence-times as calculated from the ratios $^{210}\text{Bi}/^{210}\text{Pb}$ and $^{210}\text{Po}/^{210}\text{Pb}$, a number of authors have discussed the origins of this element. Their hypotheses in no way question the formation of ^{210}Po from natural ^{210}Pb , but they do envisage complementary supply sources within the tropospheric air.

I.1. Fossil origin

Coal, wood, fuels and tetra-ethyl lead comprise "fossil" ^{210}Po in radioactive equilibrium with ^{210}Pb , and combustion of these products puts the two nuclides into suspension in the lower atmospheric layers (Styra, 1960; Peirson, 1967; Martell, 1969, 1970 (1), 1970 (2); Mattson 1970). Given the large radioactive unbalance normally existing in the air between these two elements, such supplementary sources will modify the ratios of their concentrations. Mattson (1970) and Martell (1970) (1) go so far as to predict an eventual enrichment from ^{210}Po -carrying "smokes" in view of the high volatility of the latter.

I.2. Terrestrial origin

Surface deposits of soil and dust also contain ^{210}Po , probably in equilibrium with the ^{210}Pb . Erosion by winds together with the phenomenon

of turbulence can put these substances in suspension within the atmosphere (Karol, 1969; Martell, 1970).

Vilenskiy (1970) discovered, in the superficial ground layers (5 cm), very high (3 to 25) values for the ratio of activities $^{210}\text{Pb}/^{226}\text{Ra}$, due to atmospheric dusts. From his measurements of ^{210}Pb and ^{226}Ra in fallout near Moscow he estimated the contribution of ^{210}Pb of terrestrial origin to be 2–46%. It therefore follows that a large percentage of the atmospheric ^{210}Po , too, is of terrestrial origin.

The extension to the whole atmosphere of the results of fallout determined at ground level is, however, a delicate extrapolation since the larger particles are only to be found in the lower atmospheric layers.

I.3. Stratospheric origin

Such aerosols as can penetrate into the stratosphere (in particular the ^{210}Pb) will remain there on average for from 1 to 2 years, the mean stratospheric residence-time (Martell, 1970); the ^{210}Po may thus approach a state of equilibrium with the ^{210}Pb (activity ratio 0.6 to 1). The stratosphere may serve as a reservoir, and thus as a source of ^{210}Po with respect to the troposphere (Burton, 1960; Lambert, 1965).

Lambert (1965) takes account of this additional source when calculating the residence-times of aerosols within the troposphere on the basis of the concentration ratio Po/Pb . From his estimated stratospheric fraction (varying between 0.05 and 0.50 for an average activity ratio of 0.11), he derived residence-times varying in length of from 33 to 42 days.

I.4. Artificial origin

Latterly, many authors have given consideration to the possibility of the formation of ^{210}Pb in the course of nuclear tests (Stebbins, 1961; Osborne, 1963; Peirson, 1966; Jaworowsky, 1967; Krey, 1967) in particular to explain the high values of this element recorded in the stratosphere at the time of Project Stardust (Krey, 1967; Feely, 1965).

In a manner similar to that for the products of fission, the stratosphere may well act as a reservoir for the ^{210}Pb (of artificial origin) and hence of ^{210}Po once a state of equilibrium is attained.

This hypothesis has been refuted by Feely

(1970) who re-examined the stratospheric samplings provided by Project Stardust which had given anomalous values for the concentrations of ^{210}Pb . The values of from 3 to 4.1×10^{-14} Ci/m³ which had been reported were reduced to 0.5 to 0.8×10^{-14} Ci/m³ and are closely similar to the concentrations generally recorded within the stratosphere.

II. Experimental results and methods of exploitation

Measurements of the concentrations of ^{210}Po and other long-lived radioactive elements in the air were carried out over a period of three years (1965–1968) at a location some 5 km south of Toulouse. Each sampling, taken at 1.5 metres above ground level, continued for an average period of two days, with airflow rates of 600 m³/hour (Marenco, 1967).

The ^{90}Sr , ^7Be , ^{32}P and ^{210}Pb collected by the filter were analysed, at the same time as was the ^{210}Po , in accordance with a method of chemical analysis already presented (Marenco, 1968). The sampling period being of short duration (1 to 3 days), the amount of Polonium formed on the filter from the ^{210}Pb collected, was small. Allowance is made for this when determining the concentration of ^{210}Po in the air.

The chemical recoveries were determined by the use of ^{208}Po as a tracer for the ^{210}Po and standardised carriers for the other nuclides. In the case of stable Lead and Phosphorus, allowance must be made for the considerable amounts of these elements deposited on the filters and which increase the quantities of carriers included in the analysis.

Simultaneous measurements of the concentrations of stable Lead and Phosphorus in the air thus enable the chemical yields to be corrected.

As will be seen, these elements are representative of urban and industrial pollution.

By way of an example Fig. 1 gives a representation of the daily variations in the concentrations of ^{210}Pb , ^{210}Po and ^{90}Sr in the air over a six-month period (Sept. 1967 to Feb. 1968). Fig. 2 presents the monthly variations of activities in the air and in the precipitations of these same elements over the period from Sept 1965 to Sept 1968.

Since the analysis in ^{90}Sr did not commence until Aug. 1966, the monthly values adopted prior to that date were those available from the Ispra station (Italy), as being representative of those for Toulouse; and in fact, very similar activities have been recorded at both stations (Hasl (1) 1966 to 1970), from Sept. 1966 to Dec. 1969.

The daily variations in activities show high concentrations of ^{210}Po , generally associated with high percentages of ^{210}Pb . Generally speaking, both Figures show similarities in the variations of ^{210}Po and ^{210}Pb . On the other hand, these variations do not correspond with those of the ^{90}Sr . In addition, it will be noted that although the monthly variations in the cases of ^{210}Pb and ^{90}Sr , in the air and in precipitations, are closely similar, this is not so in the case of ^{210}Po .

Thus, in the cases of ^{210}Pb and ^{90}Sr , the seasonal effects (the springtime increase in

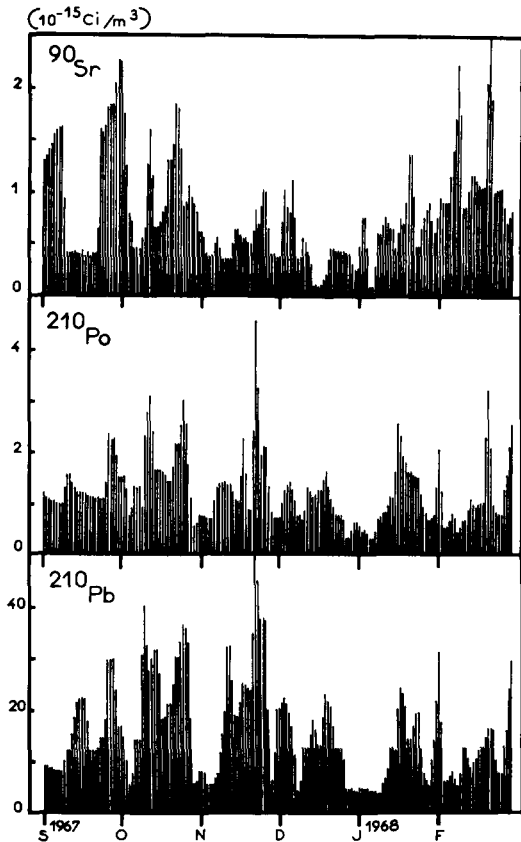


Fig. 1. Daily variations of activities in the air.

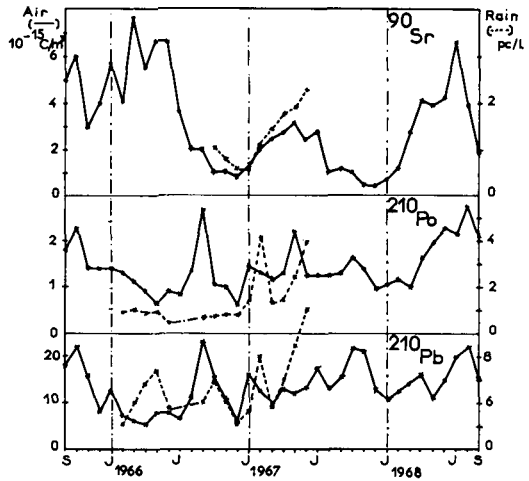


Fig. 2. Monthly variations of activities in air and in precipitation.

^{90}Sr apparent in the air and in precipitation, and the high concentration of ^{210}Pb in low-level air in winter) are of greater importance than the variability in the incidence of precipitation (Lacombe, 1968).

A statistical analysis of the results has been carried out, making use of a computer to determine the correlations existing between the activities and the various parameters. In the case of continuous functions (activities, meteorological parameters with numerical values), the coefficients of linear correlation were calculated (Panofsky, 1968). On the other hand, in the case of the discontinuous functions such as those based on qualitative parameters (the nature of air masses ...) one of the few applicable methods is the determination of cross-correlation curves. The latter are defined for the two variables $X(t)$ and $Y(t)$, dependent on time, in accordance with the relation:

$$P(\theta) = \frac{1}{N} \sum_{t=I}^N X(t) Y(t+\theta)$$

where θ is the time lag between the two variables.

As in the case of the linear correlation, it is possible to normalize the curves with the variance of the two variables. The numerical values of the coefficients obtained in the case of the discontinuous functions are, however, unrepresentative and it is more advantageous

to give consideration to the existence of peaks and to their lags.

The activity results have been reduced to a fixed period of one day, in order to obtain comparable functions, and have been filtered (high-pass filtering) to maintain them within the short-period spectrum band, thus ensuring that the studies of the correlations were unaffected by long-period phenomena. Additionally, the mathematical processing of the results (cutting, filtering) brings about a certain spreading of the variations in activities whose effect is the reduction of the resolution of the peaks and a lowering of the absolute value of the correlation coefficients, which must be taken into account in their interpretation.

III. Discussion of the origins of Polonium-210 in the troposphere

In order to define the origins of ^{210}Po in the troposphere our results have been treated in a statistical manner calculating, in particular, the correlations and cross-correlations existing between two elements, or again those existing between an element and the parameters characterizing the atmosphere.

III.1. Fossil origin

Direct estimation of the importance of such a contribution, based on an overall estimate, would prove to be difficult. It would be necessary to take account of the average content of ^{210}Po in woods and in coals (liable to considerable variation), yields of introduction into the atmosphere, local rates of combustion, etc.,. It would be simpler to seek the effects of such a source directly.

We considered that stable lead would be a good tracer to characterize urban pollution since, although introduced into the atmosphere by the internal combustion engine and the ^{210}Po by domestic and industrial heating, both are produced in the same area—viz. the city of Toulouse.

The production of stable phosphorus in an industrial zone bordering the city was observed in the area of our experiments. Phosphorus may therefore be considered to be representative of the pollution in this region.

The quantity of ^{210}Po , introduced into the

air with the stable lead, is negligible. Effectively, the mean amount of ^{210}Po in the tetraethyl lead as determined in the laboratory was 10^{-16} Ci/ μg of lead, and the concentration of stable lead in the air, as measured at Toulouse, varied between 10^{-2} and $1 \mu\text{g}/\text{m}^3$. The maximum contribution in ^{210}Po is, therefore, 10^{-18} Ci/ m^3 , a value less than one-tenth of the mean concentration of this element in the air.

Studies of the correlations between the concentrations of ^{210}Po and those of stable lead and phosphorus have been made. For purposes of comparison, the same calculations were effected for each of the other radioactive elements. No correlation was observed between the ^{210}Po and the elements characteristic of the pollution. No peak was apparent and the coefficients of linear correlation were very small (0.05 to 0.1). The same applies to the other radioactive elements.

Lastly, a study of the correlations between the activities of the elements and the nature of the air-masses revealed:

(a) For the activities of the elements, a very close correlation with the continental air-masses and a pronounced negative correlation with the maritime air-masses.

(b) For the ratio $^{210}\text{Po}/^{210}\text{Pb}$, no correlation with the continental air-masses but a close correlation with the maritime air masses.

Effectively, the ratio $^{210}\text{Po}/^{210}\text{Pb}$ is higher in maritime air because of the greater concentrations of ^{210}Pb in continental air (due to the exhalation of radon).

Oceanic air-masses being little polluted confirm that the high values of the ratio $^{210}\text{Po}/^{210}\text{Pb}$, as recorded at Toulouse, are not derived from a source of pollution of continental origin.

Thus at Toulouse ^{210}Po of fossil origin can be considered to be negligible.

III.2. Terrestrial origin

A quick evaluation demonstrated that this source of supply should be taken into account. Considering radium concentrations of between 0.1 and 100 pCi/gram of earth (CSNU report—1958) and assuming all daughters of the uranium-radium decay chain to be in radioactive equilibrium, theoretical values for the ^{210}Po concentrations of between 10^{-17} and 10^{-14} Ci/ m^3 can be calculated for high dust concentrations of 100 $\mu\text{g}/\text{m}^3$ in the air.

Table 1. Mean values of ^{226}Ra and ^{210}Pb content for superficial ground layer

Soil	^{226}Ra (pCi/g)	^{210}Pb (pCi/g)	$^{210}\text{Pb}/^{226}\text{Ra}$
Sandy	0.30	0.27	0.90
Loamy	0.34	0.33	0.97

This can no longer be considered to be negligible, particularly if consideration be given to the possibility of the enrichment of the surface layers in ^{210}Pb (and in ^{210}Po in equilibrium).

Measurements of the ^{226}Ra and ^{210}Pb in the superficial ground layer (0–5 cm) were also carried out in southern France. The ratios of the activities $^{210}\text{Pb}/^{226}\text{Ra}$ were found to be less than 1, the activities being virtually in equilibrium (Table 1).

^{210}Po may thus be put into suspension in the air by the dusts swept up from the ground or from plant-life, and the correlations between the variations in concentrations and wind-speeds have been studied.

A definite negative correlation in the case of ^{210}Po and an even more pronounced negative correlation in the case of ^{210}Pb were observed. On the other hand, both ^7Be and ^{32}P showed a slight correlation with this parameter.

Strong winds, in conjunction with a westerly regime at Toulouse are generally associated with disturbances and precipitation. Thus the quantity of dust swept up from the ground is reduced and may well conceal the effect anticipated. On the other hand there exists a prevailing S–E wind (the Autan) which is very dry and can attain windspeeds of 20m/sec at ground level and well capable of putting dusts into suspension in the air.

While good correlations were found between the Autan regime (or its windspeed) and the concentrations of radioactive elements (other than ^{210}Po), poorer correlations were found with ^{210}Po , and none at all for the ratio Po/Pb .

These results support those obtained by Joshi (1968) who, by studying the concentrations of ^{210}Pb and Uranium in the air and in the soils, showed the proportion of ^{210}Pb of terrestrial origin to be less than 2%; it therefore follows that the contribution of ^{210}Po is equally small.

Thus the putting into suspension of terrestrial dusts by the winds has no appreciable in-

fluence as a source of ^{210}Po in the air, at any rate not in the area in which our measurements were made.

III.3. Stratospheric origin

No correlation was to be found between the ^{210}Po and the ^{90}Sr . The ^{90}Sr being representative of products of stratospheric origin, it follows that the high values of ^{210}Po encountered in the lower layers cannot be ascribed to introductions from the stratosphere.

On the other hand, an analysis of the results published by the "Health and Safety Laboratory" (Hasl (2), 1967 to 1970), in respect of the measurements of radioactive elements in altitude, gives a ^{210}Po mean value: $A = 0.6 \times 10^{-14}$ Ci/m³ (altitude: 12 to 18 km; latitude 40° to 60° N) and the evolution in altitude between 1966 and 1968 of the ^{90}Sr concentrations: B (from 2 to 0.15 pCi/m³; mean value of 0.5 pCi/m³).

From the ground level concentrations b of ^{90}Sr in the air or in precipitation at Toulouse we determined the theoretical quantity x of stratospheric ^{210}Po introduced by the ^{90}Sr into the troposphere:

$$x = b \frac{A}{B}$$

The monthly percentage C of the experimental values X of ^{210}Po in the air or in precipitation is:

$$C = \frac{x}{X} 100$$

This percentage is always small, the seasonal variations remaining within the range 0.5% to 6% (Fig. 3).

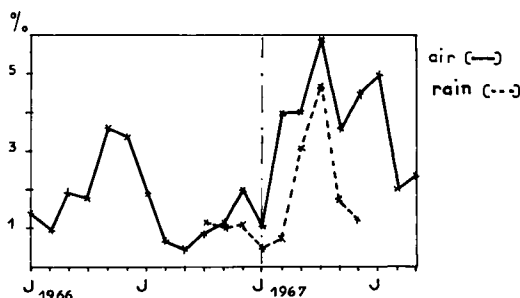


Fig. 3. Monthly percentage of ^{210}Po of stratospheric origin, in air and in precipitations, at Toulouse. (Theoretical values based on ^{90}Sr variations.)

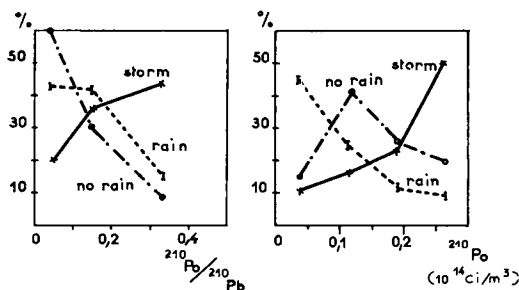


Fig. 4. The statistical distribution of $^{210}\text{Po}/^{210}\text{Pb}$ and of ^{210}Po concentrations, for the three cases: no rain, rain, rainstorm.

Finally, there are a number of methods by which the contribution of ^{210}Po of stratospheric origin, in the evaluation of Polonium, can be determined.

1. *Local method.* (a) By considering the rate of deposition (6.06×10^{-14} Ci/cm² yr) of ^{90}Sr by precipitation at Toulouse, and the mean stratospheric concentrations of ^{90}Sr and ^{210}Po , and ignoring the dry deposits, the introduction of stratospheric ^{210}Po will be found to amount to 0.07×10^4 at/cm² yr. (b) Integration, over a tropospheric column of cross sectional area 1 cm², of the formation of ^{210}Po from a mean ^{210}Pb concentration of 1.5×10^{-14} Ci/m³ (Burton, 1960; Rama, 1961; Karol, 1970) results in a tropospheric production of 1.35×10^4 at/cm² yr. Thus the proportion of ^{210}Po of stratospheric origin amounts to only 5% of the tropospheric production.

2. *Global method.* By consideration of the masses of the tropospheric (4.1×10^{18} kg) and stratospheric (1.1×10^{18} kg) reservoirs (Telegadas, 1968), for a stratospheric residence-time of approximately 12 months (9–13 months) (Roedel, 1967; N'Guyen Ba Cuong, 1968; Krey, 1969), the exchange within the stratospheric mass is of the order of 0.67×10^{18} kg/yr (Chi Trach Hoang, 1969); this corresponds to an introduction of ^{210}Po amounting to 0.26×10^{22} at/yr. The tropospheric production of ^{210}Po originating from ^{210}Pb being some 5.5×10^{22} at/yr, the calculated quantity of ^{210}Po of stratospheric origin amounts to only 4.5%.

Thus all calculations clearly indicate that the introduction of stratospheric ^{210}Po into the troposphere is negligible.

III.4. Storm effects

Simultaneous activity measurements effected at a Pyrenean mountain station (Pic du Midi—altitude 3 000 m) and at a low-level station (Tarbes at the foot of the mountain) have revealed peculiarities in the vertical distribution of ^{210}Po (Croizat, 1969). Higher concentrations of ^{210}Po were recorded at the low-level station during storms.

This, in conjunction with the very special chemical properties of ^{210}Po and its singular behaviour during heavy rainstorms (Lacombe, 1968) led us to investigate the correlations between the ^{210}Po activities and the various types of precipitation. We observed: (a) close negative correlation between the concentrations of all the elements and the daily duration of precipitation (representative of atmospheric washing); (b) nothing of significance in light showers; (c) slight, but unmistakable, correlation for the ^{210}Po only in the course of heavy rainstorms.

A study of the statistical distributions of the Po activities and of the ratio Po/Pb for three distinct conditions (rainstorm, rain, absence of rain) clearly indicates a sharp increase in the percentage of high values in storm conditions (Croizat, 1969) (Fig. 4).

This phenomenon is not easily explained. It may be due to the re-evaporation of the ^{210}Po contained in the precipitation, either in falling through the lower atmospheric layers or on making contact with warmer ground. Moreover, Lacombe (1968) reported greatly increased specific activities for ^{210}Po in precipitation occurring in heavy storms (cumulus, cumulonimbus).

Although this effect is clearly noticeable in individual cases it does not play an important part in the overall evaluation of ^{210}Po owing to the relatively few storms encountered at the latitude in which our experiments were conducted.

III.5. Natural origin

The large differences between the residence-times determined from the ratios of the various activities are insufficient to prove the existence of additional sources of ^{210}Po . Great care must be taken in the interpretation of such residence-times, calculated as they are on the assumption of steady-state conditions and

with no allowances for diffusion (Bhandari, 1970).

Discussions on the various sources of Polonium, suggested in recent technical literature, have shown that such sources have no real importance insofar as our measurements are concerned. On the other hand, we have found very close correlations between ^{210}Po and ^{210}Pb (linear coeff = 0.6), the latter clearly emerging as the main source, and a lesser correlation with ^7Be and ^{32}P (linear coeff = 0.35). This indicates that, like these latter elements, the ^{210}Po depends on the same factors of transport and elimination within the troposphere.

The negative correlation between the concentrations of ^{210}Pb and windspeeds, to which reference was made in paragraph 2, is easily interpreted from considerations of the fraction of ^{210}Pb newly-formed from the radon emanating in the European continental land mass. Assuming constant emission and a westerly, radon-free, low-level airstream reaching the coast, the radon and ^{210}Pb concentrations at a given distance from the coast are proportional, as a first approximation, to the time of contact with the soil, i.e. inversely proportional to the windspeed.

The negative correlation also recorded between the windspeed and the ^{210}Po indicates that it derives in large measure from the ^{210}Pb newly-formed above the European continent.

If consideration be given (Fig. 5) to the variations in the ratio Po/Pb in the air at ground level and in precipitation (representative of air at altitude), it will be noted that the value is always higher in precipitation and that the variations are more pronounced.

By selecting, according to the altitudes of the generative clouds, the precipitations at Toulouse Lacombe (1968) determined the ratio Po/Pb at various altitudes (0.21 at 2 km; 0.24 at 5 km; 0.27 at 9 km. The mean value at ground level was 0.11.

This vertical distribution, integrated over an air-column of 1 cm^2 cross-section, gives a mean tropospheric ratio of 0.22. Burton's (1960) equation then applies and the ratio can be extended to cover the whole of the troposphere:

$$\frac{\text{Po}}{\text{Pb}} = \frac{\lambda_1 \lambda_2}{(\lambda_1 + \Lambda)(\lambda_2 + \Lambda)}$$

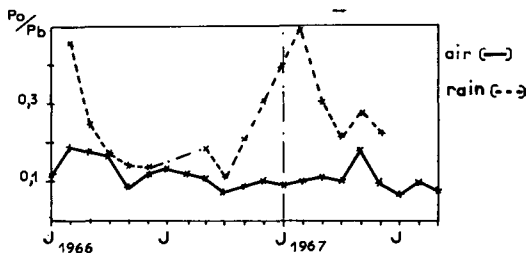


Fig. 5. Monthly variations of the ratio $^{210}\text{Po}/^{210}\text{Pb}$ in the air and in precipitation at Toulouse.

where λ_1, λ_2 = radioactive constants of ^{210}Bi and ^{210}Po

Λ = removal constant

τ = residence-time $\left(\tau = \frac{1}{\Lambda}\right)$

giving τ a value of 65 days.

Conclusions

The results of our experiments have failed to substantiate any of the additional sources put forward in explanation of the high values of ^{210}Po in the lower tropospheric layers.

Fossil or terrestrial sources may sometime make an important contribution (large industrial areas, heavily radioactive soils), but these effects are entirely local.

All our measurements have clearly shown that the fraction of ^{210}Po of stratospheric origin is very small and can generally be neglected.

Although our experiments have shown up a particular effect during rainstorms and have suggested possible re-evaporation of ^{210}Po into the air, such effects are restricted to a very small area and cannot be discerned on a larger scale.

Tropospheric ^{210}Pb remains, therefore, the principal source of ^{210}Po in the troposphere.

Our measurements in air and in precipitation, integrated over the whole troposphere, give a mean value $\text{Po}/\text{Pb} = 0.22$ corresponding to a residence-time of 65 days. This value is high compared to those generally calculated; it corresponds to a global residence-time within the troposphere.

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ИСТОЧНИКИ ПОЛОНИЯ-210 В ТРОПОСФЕРЕ

В современной технической литературе уделяется внимание дополнительным источникам ^{210}Po (древнего, земного, стратосферного и искусственного происхождения). В работе обсуждается важность различных источников, причем дискуссия ведется на основе кратковременных измерений содержания дол-

гоживущих ядер ^{210}Po и ^{210}Pb и продуктов загрязнения, проведенных на юго-западе Франции. Делается вывод, что ^{210}Po в тропосфере имеет естественное происхождение и получается, главным образом, при распаде ^{210}Pb присутствующего в атмосфере.