Lead-210 and polonium-210 in the atmosphere

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ABSTRACT

The concentration of Pb-210 has been measured at several altitudes in the atmosphere
over the United Kingdom during 1958. The gradient into the lower stratosphere was
less steep than observed by BURTON & STEWART (1960) and conformed to the diffusion
models of JACOBI & ANDRE (1963) for the case of short washout times, and MACHTA
(1960).
   The concentrations of Pb-210 and Po-210 in surface air and Pb-210 in rainwater have
been measured continuously since 1961. The ratio of Po-210/Pb-210 exhibits a seasonal
variation during 1962 and 1963: this might be attributed to artificial production of
Pb-210 during the weapon tests of 1961-2. An estimate has been obtained of the global
deposition of Pb-210.

1. Introduction

The usefulness of radioactive isotopes as
meteorological tracers is generally recognised.
Fission products both short- and long-lived
from nuclear weapons have been used to follow
atmospheric movements in the stratosphere and
troposphere: isotopes produced by cosmic rays
have proved similarly useful. In the case of the
radioactive decay products arising from the
exhalation of radon-222 (half-life 3.8 days)
from the earth's surface the behaviour of the
short-lived products (<30 minutes) is charac-
teristic of the boundary layer whilst that of
lead-210 (19.4 years) and polonium-210 (138
days) is affected by atmospheric conditions up
to and beyond the tropopause.

   After exhalation from the ground the trans-
port of radon into the atmosphere is controlled
by turbulent diffusion and limited only by
radioactive decay. The radon decay products
become readily attached to airborne particulate
material and will therefore be subject to remo-
val, by washout and sedimentation, to the
earth's surface.

   The vertical distribution of lead-210 and
polonium-210 have been calculated by assuming
certain conditions of turbulence and the cor-
responding variation with altitude of the diffu-
sion coefficient (see for example JACOBI &
ANDRE, 1963, and MACHTA, 1960. Measure-
ments at various levels in the troposphere and

lower stratosphere have been reported by
BURTON & STEWART (1960), RAMA & HONDA
(1961) and by STEBBINS (1961).

   This paper presents measurements of lead-210
in the troposphere and lower stratosphere that
are more frequent than previously, a continuous
record for several years of the concentrations
of lead-210 and polonium-210 in surface air,
and measurements of lead-210 in rainwater
representative of the global distribution.

2. Method

   The programme of sampling and analysis is
based upon the network and methods used for
fission products from nuclear weapon explo-
sions (CAMBRAY *et al.*, 1964; PEIRSON *et al.*,
1960).

   Airborne dust is sampled continuously for
periods of one week at Chilton (51° N, 01° W),
at 1 metre above ground using cylindrical filters
of esparto grass paper to pass about 1500 kg/day
of air. The sample at an effective altitude of
1.3 km was obtained by aircraft flying over the
eastern Atlantic between latitudes 42° N and
65° N. About 20 samples were obtained each
month, representative of an air mass of more
than 10,000 kg. At higher altitudes, in the
upper troposphere (7.6 km) and lower strato-
sphere (14.3 km) aircraft sorties were less fre-
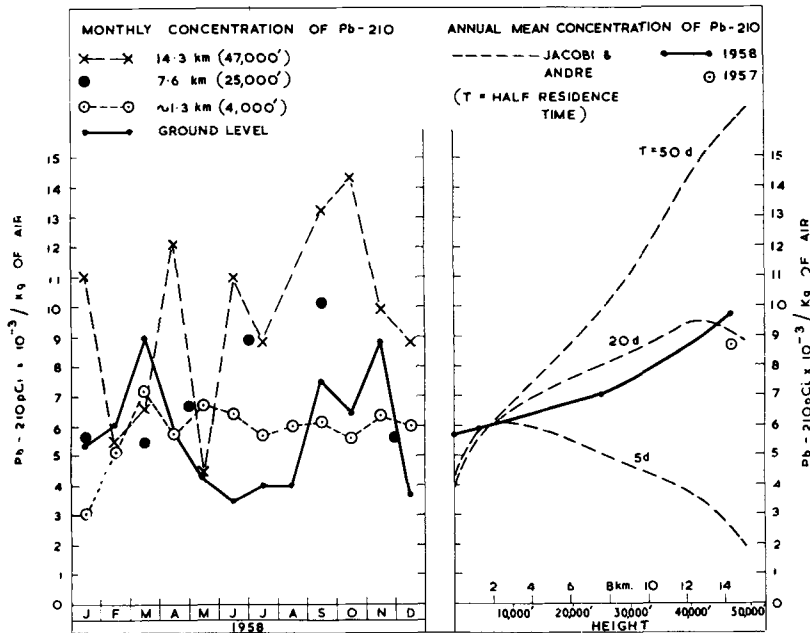quent and of shorter duration: each monthly

FIG. 1. Variation of atmospheric concentration of Pb-210.

aggregate sample was representative of about 500 kg of air.

The sampling efficiency of the esparto filter material is determined regularly for fission product debris by backing esparto filters with high efficiency glass fibre filters at several face velocities (CAMBRAY et al., 1964). It is then assumed that the efficiency for lead-210 and polonium-210 is identical with that for long-lived fission product radioactivity, viz. 82 % at ground level, 90 % at 1.3 km and 100 % at the higher altitudes.

Samples of rainwater have been collected monthly at Milford Haven (51° N, 05° W) in the United Kingdom, and every three months from some of the stations overseas used for fission product sampling. Rainwater is collected using a polythene funnel mounted above a polythene bottle containing a dilute solution in nitric acid of various carriers for the fission product radioactivity. Carriers for lead-210 and polonium-210 are added after collection.

The chemical separation and analysis of lead-210 and polonium-210 have been detailed by SPICER (1965) and WALLACE (1965). A notable feature is the use of polonium-208 (2.9 years) tracer which has overcome earlier difficulties caused by chemical yields that were both low

and variable. This isotope is produced in the cyclotron by the reaction Bi-209 $(p,2n)$ Po-208. The uncertainty in analysis is considered to be equivalent to a relative standard deviation of 20 %.

3. Results and discussion

Atmospheric gradient

The concentration of lead-210 in air during 1958 shows considerable variation, by a factor of two or more in the monthly mean value, at each of the heights of measurement (Fig. 1 and Table 1). The gradient of concentration measured from ground level may be positive or negative although the concentration (per unit mass of air) in the lower stratosphere (14.3 km) is generally larger than at the lower altitudes. In the case of the measurements at ~ 1.3 km over the eastern Atlantic the concentrations refer to oceanic conditions i.e. to the absence of radon emanation from directly below.

Also shown in Fig. 1 is the gradient averaged for all the observations made in 1958. In this case there is a continuous increase with height but more gradually than indicated by the exploratory measurements of BURTON & STEWART (1960) made during 1954-6. The values

lie somewhat above those of RAMA & HONDA (1961): the gradient for 1960 reported by STEBBINS (1961) is negative above about 9 km. However these other results have been obtained from observations over limited periods: it is clear from the monthly record of Fig. 1 that the shape of the curve of concentration versus height may change during the course of a single year. Measurement of samples from the stratosphere not contemporary with results from the troposphere are available for the U.K. during 1957 (Table 1). These and the series of measurements in the stratosphere above the U.S. during 1961–2 (KREY et al., 1965) are consistent with the results for 1958 at 14.3 km.

The possibility of artificial production of lead-210 in nuclear weapon testing should not be ignored. This is considered below in relation to more recent measurements but for the period before 1961 there is no information to decide whether the differences between the reported concentrations of lead-210 could be explained by artificial production.

In a comparison of the results of the present work with theoretical calculation of the variation of concentration of lead-210 with height it is possible to draw only tentative conclusions. This is because the complexities of the physical situation require arbitrary simplification in order to produce a reasonable model. Thus in Fig. 1 the annual mean concentrations for 1958 agree with the curve of JACOBI & ANDRE (1963) that has the following parameters:

(1) a diffusion coefficient in the troposphere of, effectively, 2×10^5 cm²-sec⁻¹,

(2) a much smaller coefficient in the stratosphere,

(3) a half-life of removal by washout of 20 days at all heights up to and including the stratosphere.

The same results are not inconsistent with the model of MACHTA (1960), that is less comprehensive but probably more realistic in the decrease of removal rate with altitude.

Concentration in surface air

The concentrations of lead-210 in surface air measured at Chilton during 1961–5 (Fig. 2 and Table 2) and 1958 (Fig. 1) show a tendency towards minima in the summer months. No attempt has been made to relate this seasonal tendency to local climatic conditions. It is

TABLE 1. *Concentration of Pb-210 in air over the United Kingdom: pCi × 10⁻³/kg.*

	1958				1957
	14.3 km	7.6 km	~1.3 km	Ground level	14.3 km
Jan.	11	5.6	3.1	5.4	
Feb.	5.5	—	5.1	6.0	6.6
Mar.	6.6	5.4	7.2	9.0	6.4
Apr.	12 ⎫	6.7	5.7	5.9	
May	4.4 ⎬		6.7	4.3	
June	11 ⎫	8.9	7.0	3.5	6.4
July	8.8 ⎬		6.3	4.0	7.8
Aug.	11 ⎫		6.0	4.0	
Sept.	13 ⎬	10	6.1	7.5	11.1
Oct.	14 ⎭		5.6	6.4	13.8
Nov.	10 ⎫	5.6	6.4	8.9	
Dec.	8.8 ⎬		6.0	3.6	
Mean	9.7	7.1	5.9	5.7	8.7

reasonable to expect that (a) the exhalation of radon would depend upon the condition of the soil, viz. the temperature and moisture content, and also upon wind velocity and (b) the diffusion of radon upwards would depend upon the atmospheric temperature gradient and vertical air movements (ISRAEL, 1961). It is possible that apparent winter maxima in radon concentration would be related simply to the frequency and intensity of temperature inversions near ground level: GALE & PEAPLE (1958) have demonstrated such a diurnal correlation. Since lead-210 requires a period equivalent to several half-lives of radon to achieve maximum activity, starting from a given quantity of radon, the concentration of lead-210 will not solely reflect local climatic conditions except in a situation of pronounced atmospheric stability. The concentration at a sampling station in the United Kingdom, on the windward side of the European land mass, will be influenced by the transport of air across the Atlantic Ocean from which the exhalation of radon will be zero or very small. Despite the loss of radon by radioactive decay and lack of replenishment during the ocean passage the concentrations of lead-210 at Chilton are comparable with those observed during 1961–2 by PATTERSON & LOCKHART (1964) along the 80th meridian. It follows that the atmospheric residence time of lead-210 is long compared with the time of transit across the Atlantic Ocean.
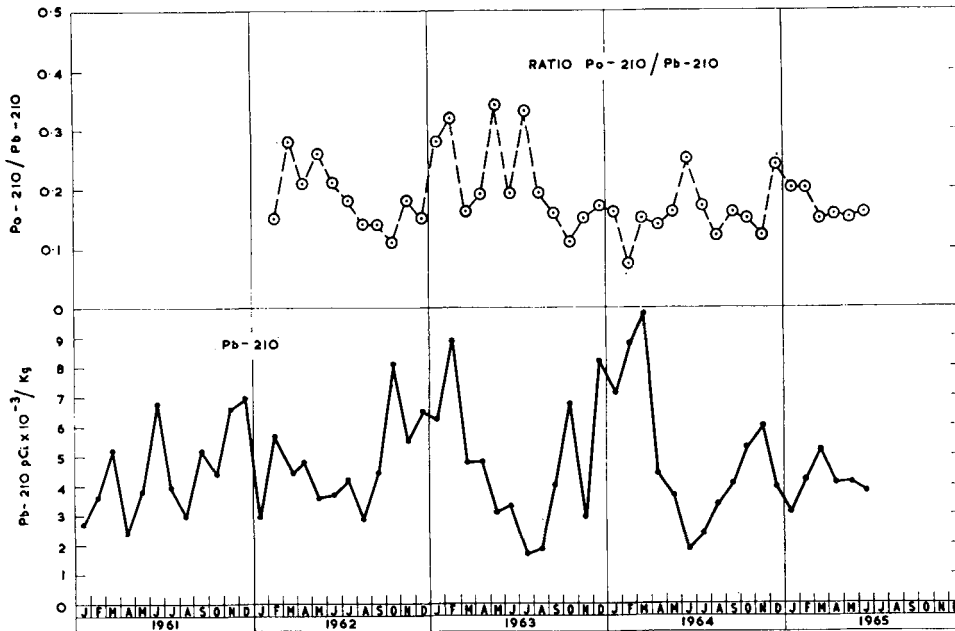
FIG. 2. Concentration of Pb-210 and Po-210 in air near ground level at Chilton.

The curve of the ratio polonium-210/lead-210 in Fig. 2 provides an opportunity to study the age or previous history of the sample. The values of the ratio during the first halves of 1962 and 1963 are noticeably higher than in the second halves whereas in 1964 there is no such apparent seasonal variation. If it is assumed that lead-210 is stored in the stratosphere and is transferred in the spring of each year to the troposphere then a quantity of polonium-210 will be transferred that depends upon the quantity of lead-210 and the storage time. A natural explanation of the phenomenon noted above would be that less lead-210 was transferred, or the storage time was shorter, in the spring of 1964 than in 1962 and 1963. Alternatively it is possible to explain this phenomenon by association with the periods of nuclear weapon testing during the autumns of 1961 and 1962: it is unfortunate that the analyses for polonium-210 made during 1961 without benefit of the tracer technique had to be rejected because of the uncertainty in chemical yield.

The explanation proceeds from the assumption that the normal value near ground level of the polonium-210/lead-210 ratio is observed in Fig. 2 during 1964, 1965 and the second halves of 1962 and 1963. During these periods the average value is approximately 0.15 which compares with 0.13 measured by BURTON & STEWART (1960) in 1955–6 and 0.07 by LEHMANN & SITTKUS (1959) in 1958; on the other hand OSBORNE (1963) observed an excursion to 0.48 in March 1962. It is then postulated that lead-210 was formed artificially in one or more explosions during each of the autumn test series in 1961 and 1962. Any of this material injected into the troposphere would spend several weeks (PEIRSON & CAMBRAY, 1965) reaching the United Kingdom from the Russian test sites during which time growth of polonium-210 would produce a ratio for the artificial material indistinguishable from the normal ratio. However the artificial lead-210 injected into the stratosphere would have been stored from autumn until the following spring: at the time of transfer to the troposphere the artificial ratio would be sufficiently high to enhance the ratio observed in the troposphere after mixing with the natural radioactivity. Thus by assuming reasonable production dates during the periods of testing (HASL, 1964) the maximum reinforcement of the surface air concentration of lead-210 would have accounted for 30 % to 50 % of the amount observed in the early months of 1962 and 1963. It is possible that the artificial lead-210 was formed

TABLE 2. *Concentration of Pb-210 and Po-210 in surface air* $(pCi \times 10^{-3}/kg)$ *and Pb-210 in rain* $(pCi/litre)$.

| | 1961 Surface air (Chilton) | | 1961 Rain (Milford Haven) | | 1962 Surface air (Chilton) | | 1962 Rain (Milford Haven) | | 1963 Surface air (Chilton) | | 1963 Rain (Milford Haven) | |
	Pb-210	Po-210/Pb-210	cm	Pb-210	Pb-210	Po-210/Pb-210	cm	Pb-210	Pb-210	Po-210/Pb-210	cm	Pb-210
Jan.	2.7				2.9		12.08	1.6	6.2	0.28	2.26	2.7
Feb.	3.5				5.6	0.15	2.28	3.2	8.9	0.32	5.51	2.1
Mar.	5.1				4.4	0.28	9.59	1.7	4.8	0.16	12.87	1.6
Apr.	2.4				4.8	0.21	5.35	2.2	4.8	0.19	10.88	1.6
May	3.8				3.5	0.26	6.59	2.1	3.0	0.34	4.78	4.8
June	6.7				3.7	0.21	3.39	3.3	3.3	0.19	9.56	2.5
July	3.9				4.2	0.18	4.48	3.1	1.7	0.33	5.14	3.4
Aug.	2.9				2.8	0.14	11.40	2.5	1.8	0.19	8.00	1.9
Sept.	5.1		8.04	2.1	4.4	0.14	13.73	2.2	4.0	0.16	9.36	2.4
Oct.	4.3		18.04	2.1	8.0	0.11	3.28	2.2	6.7	0.11	7.26	4.1
Nov.	6.6		9.11	2.6	5.5	0.18	8.68	1.5	2.9	0.15	20.93	1.9
Dec.	7.0		9.57	2.6	6.5	0.15	8.75	1.7	8.2	0.17	3.45	2.4

| | 1964 | | | | 1965 | | | |
	Pb-210	Po-210/Pb-210	cm	Pb-210	Pb-210	Po-210/Pb-210	cm	Pb-210
Jan.	7.1	0.16	2.63	3.9	3.1	0.20	11.48	2.5
Feb.	8.8	0.07	4.86	5.0	4.2	0.20	0.62	5.6
Mar.	9.8	0.15	11.68	7.0	5.1	0.15	7.97	1.3
Apr.	4.4	0.14	5.60	3.5	4.0	0.16	4.80	1.7
May	3.7	0.16	6.59	4.3	4.1	0.15	5.33	3.4
June	1.8	0.25	5.33	2.6	3.8	0.16	6.91	3.4
July	2.3	0.17	7.31	1.9				
Aug.	3.3	0.12	4.92	2.3				
Sept.	4.0	0.16	3.05	3.7				
Oct.	5.2	0.15	8.97	2.0				
Nov.	6.0	0.12	6.33	3.2				
Dec.	3.9	0.24	12.44	0.71				

TABLE 3. *Concentration of Pb-210 in rain: pCi/litre.*

| Station | Lat. | Long. | 1961 | | | | 1962 | | | | 1963 | | | | 1964 | | | |
			1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	4
Tromsö	70°N	19°E							1.5	0.9								
Bodö	67°N	19°E			2.8		2.0	2.0			3.8	3.0	1.4	4.6	2.2			
Milford Haven	51½°N	05°W				2.4	1.8	2.4	2.5	1.7	1.9	2.5	2.5	2.5	6.1	3.5	2.4	
Linz	48½°N	14½°E					2.5	3.2	3.4		3.8	3.0	3.0					
Gosausee	48°N	13½°E	2.8	2.9														
Ottawa	45½°N	76°W		5.6	5.1	5.6	5.2	4.1	3.7		9.7	5.9	4.1	3.4		4.1		
Nassau	25°N	77½°W									3.1	1.8	2.7	2.2	1.4	2.1	2.0	
Lagos	6½°N	03½°E			4.8		4.5											
Singapore	01°N	104°E		1.3		1.1	1.6		1.6		1.3	2.1	1.3		1.0	1.6	1.8	
Melbourne	38°S	145°E									3.2	2.0	3.6	4.0				
Ohakea	40°S	175°E			0.6		0.5	0.5	0.54	0.80								
Stanley	51½°S	58°W									0.85	1.3	1.2	2.0	0.9	1.2		
Argentine Is.	65°S	64°W														0.2		

by the nuclear reaction lead-208 $(2n, \gamma)$ lead-210, and that a fraction of a megacurie was formed in each year. There is some support for the suggestion of artificial production of lead-210 in 1962, but not in 1961, in that the concentration in the lower stratosphere increased suddenly in December 1962 (KREY et al., 1965). Other radioactive isotopes were produced in nuclear explosions during the period 1961–2 by neutron activation, e.g. antimony-124, and, as in the case of lead-210, possibly by second-order neutron capture, manganese-54 and antimony-125 (KAURANEN, 1964; CAMBRAY et al., 1964, 1965).

The normal value of the ratio in surface air of 0.15 may be used to make an upper limit calculation of the tropospheric residence time. Neglecting turbulent diffusion the activity ratio of polonium-210/lead-210 is given by $[\lambda E/(\lambda F + \lambda)]$ $[\lambda E/(\lambda F + \lambda)]$, where λ is the washout removal factor assumed exponential and λE and λF are radioactive decay constants for bismuth-210 and polonium-210. For a ratio of 0.15 the mean residence time is 40 days.

Concentration in rain

The monthly concentrations of lead-210 in rain at Milford Haven are listed in Table 2 for the period 1961–5. The rain concentration shows less tendency to summer minima than the air concentration curve of Fig. 2. This may not be a fair comparison in view of the different locations of Milford Haven (coastal) and Chilton (inland). Nevertheless a "washout factor" may be extracted from these data by calculating the ratio of concentrations for equal masses of rain and air. The annual mean values of this ratio for the period 1962–4 lie in the range 600 to 800. This is similar to the range of values calculated for caesium-137 in nuclear weapon debris (PEIRSON & CAMBRAY, 1965).

Values of the ratio polonium-210/lead-210 in rainwater at Milford Haven are available but have not been presented. These values are more erratic than those measured in surface air at Chilton. This behaviour, subject to further investigation, is provisionally attributed to factors dependent upon the chemical state of polonium-210 during the washout process and during storage in the collecting bottle.

Although it was not possible to measure the concentration of lead-210 in rain regularly at all overseas stations, the results listed in Table 3 give some impression of the global distribution. This distribution is similar to that reported by PATTERSON & LOCKHART (1964) for concentrations in air: the measured rain concentrations are roughly one-half those derived from the air concentrations, by using the washout factor determined above. As a general conclusion the northern hemisphere is somewhat richer in lead-210 than the southern hemisphere. This is in agreement with lower radon concentrations in the southern hemisphere due to the smaller area of land and to slow mixing across the equator (JUNGE, 1963). The concentrations at Ottawa and Melbourne appear higher than the averages for the respective hemispheres: this is probably due to their continental positions relative to prevailing winds. Integration of the concentrations of Table 3 weighted by the distribution of rainfall with latitude (MÖLLER 1951) indicates that the annual global deposition (by rain) of lead-210 is of the order of one megacurie.

Acknowledgement

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СВИНЕЦ-210 И ПОЛОДИИ-210 В АТМОСФЕРЕ

Концентрация Pв-210 в атмосфере была измерена на различных высотах над Объединенным Королевством в течении 1958 г. Градиент концентрации в нижней стратосфере был меньше, чем градиент наблюдавшийся BURSTON & STEWART (1960) и соответствовавший диффузионной модели JACOBI & ANDRE (1963) для случая малых времен размыва, а также меньше, чем градиент определенный MACHTA (1960).

Концентрации Pв-210 и Po-210 в приземном воздухе и концентрация Pв-210 в дожде измеряется непрерывно с 1 1 г. В отношение Po-210/Pв-210 наблюдались сезонные изменения в течение 1962–1963 годов. Это может быть связано с искусственным образованием Pв-210 во время ядерных испытаний 1961–1962 годов. Была получена оценка для мирового отложения Pв-210.