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Modification of the Atmospheric Polonium 210 to Lead 210 Ratio by Volcanic Emissions

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Volcanic gases are very rich in long-lived radon daughters, especially in ^{210}Po . Thus more than 50×10^3 Ci per year of ^{210}Po are injected into the troposphere by a normal volcanic activity, and more than 1×10^3 Ci per year into the stratosphere by volcanic explosions. These fluxes should account for one half of the ^{210}Po content in both of these reservoirs. On the contrary the volcanic sources of ^{222}Rn and ^{210}Pb are generally negligible with regard to the soil production. This new result could explain most of the discrepancies observed by comparing the activity ratios of the different radon daughters, including the existence of $^{210}\text{Po}/^{210}\text{Pb}$ figures superior to one (overequilibria). Stratospheric aerosol residence times calculated on this basis are found to be shorter than the previous figures.

Radon 222 and its decay products have been considerably utilized in atmospheric studies [Jacobi and Andre, 1963; Lambert and Nezami, 1965; Karol, 1970; Turekian et al., 1977]. One reason for this is that the disintegration of the atmospheric gaseous radon is followed, within 1 hour, by a series of long-lived radioactive daughters, quickly fixed on atmospheric aerosol particles:

Assuming that the aerosols are removed from the atmosphere by a first-order process, it is possible to define a scavenging constant λ_s , similar to the radioactive constants λ_{Rn} , λ_{Pb} , etc. Therefore in a closed reservoir the classical equations of the radioactive equilibrium are modified and lead to the following expressions [Burton and Stewart, 1960]:

$$\frac{{}^{210}\text{Pb activity}}{{}^{222}\text{Rn activity}} = \frac{\lambda_{\text{Pb}}}{\lambda_{\text{Pb}} + \lambda_s} \approx \frac{\lambda_{\text{Pb}}}{\lambda_s} \quad (1)$$

since λ_{Pb} is about 10^3 times smaller than λ_s , as shown later.

$$\frac{{}^{210}\text{Po activity}}{{}^{210}\text{Pb activity}} = \frac{\lambda_{\text{Bi}} \lambda_{\text{Po}}}{(\lambda_{\text{Bi}} + \lambda_s)(\lambda_{\text{Po}} + \lambda_s)} \quad (2)$$

It is meaningless to compare directly the concentrations of ^{222}Rn and of its long-lived decay products measured just above the continents, since the vertical gradients due to atmospheric mixing processes are very different. However, far from the radon sources, it is possible to use the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio to obtain a value of λ_s if all of ^{210}Po were directly produced from ^{210}Pb .

Nevertheless, according to Moore et al. [1973], the injection from the stratosphere into the troposphere of lead and polonium 210 approximately in radioactive equilibrium can be significant and results in the overestimating of the aerosol residence time.

Other sources of ^{210}Po such as soil particles and vegetal debris have also been considered by Marengo and Fontan [1972],

who concluded nevertheless that they were of very small importance, and by Poet et al. [1972], who assumed that they could play a significant role.

SUBANTARCTIC ATMOSPHERE

Over the sub-Antarctic ocean the atmospheric concentrations are typically 1.2×10^{-12} Ci/m³ for ^{222}Rn and 0.3×10^{-15} Ci/m³ for ^{210}Pb [Lambert et al., 1966, 1972; Sanak et al., 1978] which gives $\lambda_s \approx 3.7 \times 10^{-1}$ d⁻¹ and a corresponding aerosol half-life of about 1.9 d. This figure of λ_s should give for the $^{210}\text{Po}/^{210}\text{Pb}$ ratio 3.6×10^{-3} instead of an average figure actually observed of about 0.5, i.e., 140 times more.

Lambert et al. [1966] have deduced, from the latitudinal repartition of radioactive aerosols, that in the southern hemisphere ^{210}Pb has essentially a stratospheric origin, even at low altitude. However, this effect should be particularly weak in the subantarctic area, which is known to be, on the opposite, characterized by a very low injection rate of nuclear test stratospheric debris. Moreover, in about 22% of the samples, the activity of ^{210}Po has been found higher than that of ^{210}Pb (overequilibrium).

Consequently, it seems necessary to admit the existence, in the lower atmosphere, of polonium 210 sources independent of the atmospheric radon. Turekian et al. [1974] have observed that the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio, in zooplankton is about 20–30. Consequently, a flux could be expected from the sea surface to the atmosphere, of particles especially enriched in ^{210}Po relative to ^{210}Pb . As mentioned in a previous paper [Lambert et al., 1974], it has been pointed out that this effect was able to account for a limited overequilibrium $^{210}\text{Po}/^{210}\text{Pb}$ in oceanic air masses.

A first approach of this problem could result of a comparison of the ^{210}Po to organic carbon ratios in seawater, in zooplankton and in marine atmospheric particulate matter. On one hand, Nozaki and Tsunogai [1976] and Nozaki et al. [1976] have found about 10 dpm of ^{210}Po per 100 kg of seawater; Bacon et al. [1976] have found very similar results for the dissolved phase in superficial water and about 1 dpm/100 kg in the particulate materials. On the other hand, a particulate organic carbon concentration of 1 to 5×10^{-2} g/100 kg can be inferred from the chlorophyll measures by G. Jacque (private communication, 1978). Thus an activity of about 20–100 dpm/g of carbon could be a good order of magnitude for the seawater particulate phase. Turekian et al. [1974] have also found a ^{210}Po concentration of 2–200 dpm/g of dry zooplank-

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ton in agreement with *Beasley* [1973], who has measured ^{210}Po concentrations comprised between 2 and 20 dpm/g dry in different kinds of zooplankton. With 0.3 g of carbon per gram of zoo plankton, these figures correspond to 6–60 dpm/g of carbon and exceptionally up to 600 dpm/g.

It seems therefore reasonable to admit for ^{210}Po an average concentration of the order of 40 dpm/g, that is, 20 pCi/g of carbon.

In the marine atmosphere, organic carbon concentrations of 0.2–0.8 $\mu\text{g}/\text{m}^3$ have been observed by *Hoffman and Duce* [1977]. *Barger and Garrett* [1970] have reported some figures reaching 6.3 $\mu\text{g}/\text{m}^3$ for a wind velocity of 8.5 m/s, which is the right order of magnitude for subantarctic areas. Consequently, a concentration of 2–8 $\mu\text{g}/\text{m}^3$ seems to be a good evaluation of the range of values in this region.

Finally, an atmospheric ^{210}Po concentration of 0.04 to 0.16 $\times 10^{-15}$ Ci/ m^3 could be attributed to the marine source over the subantarctic ocean where the values measured are generally between 0 and 0.5 $\times 10^{-15}$ Ci/ m^3 .

Figure 1 shows the histograms of the ^{210}Pb aerosol concentrations at Kerguelen Island (49°S) for all data and for different figures of the $^{210}\text{Po}/^{210}\text{Pb}$ ratio. It is clear, by comparing Figures 1b and 1c, that high values of this ratio (>0.5) can only be obtained for low ^{210}Pb concentrations (<1 $\times 10^{-15}$ Ci/ m^3) and therefore could be possibly accounted for by the highest concentrations mentioned above for the marine source of ^{210}Po . However, it can be pointed out in Figure 1d that there is at least one case which is an exception to this interpretation ($^{210}\text{Pb} = 1.4 \times 10^{-15}$ Ci/ m^3 , $^{210}\text{Po}/^{210}\text{Pb} = 1.2$).

ANTARCTIC ATMOSPHERE

Lead 210 concentrations measured at Dumont-Durville (67°S, antarctic coast of Terre Adelie) are shown in Figure 2 together with different figures of the $^{210}\text{Po}/^{210}\text{Pb}$ ratio (original data will be published later as a *Terres Australes et Antarctiques Françaises* report). The concentrations of ^{210}Pb are

clearly higher here than at Kerguelen Island. Moreover, it can be seen (for instance in Figures 2d) that overequilibriums ($^{210}\text{Po}/^{210}\text{Pb} > 1$) are compatible with ^{210}Pb concentrations superior to 1 $\times 10^{-15}$ Ci/ m^3 . In particular from March 1965 to June 1976, 21 cases of ^{210}Po concentrations between 2 and 8 $\times 10^{-15}$ Ci/ m^3 have been recorded, for which the $^{210}\text{Po}/^{210}\text{Pb}$ ratio was between 2.7 and 7.4 in 50% of the cases (reaching even in one case 18.1). According to the preceding considerations, such results cannot be explained by the marine source of ^{210}Po aerosols.

VOLCANIC EMISSIONS OF ^{210}PO

Evidence for the presence of highly enriched volatile elements in volcanic emissions has been published by several authors either during normally active phenomena or during the violent volcanic paroxysms [*Duce et al.*, 1975; *Mroz and Zoller*, 1975; *Lepel et al.*, 1978; *Buat-Menard and Arnold*, 1978].

High concentrations of ^{210}Pb , aerosols (1200×10^{-15} to $22,500 \times 10^{-15}$ Ci/ m^3) have been measured in the plumes of several volcanoes [*Lambert et al.*, 1976, 1979; *Polian and Lambert*, 1979]. As shown in Table 1, these aerosols are very enriched in volatile elements with respect to the radioactive equilibrium: $^{210}\text{Bi}/^{210}\text{Pb}$ was found to be between 3 and 12 and $^{210}\text{Po}/^{210}\text{Pb}$ between 9 and 40. These enrichments, depending on the elements properties, are controlled by several volcanic parameters including the magma temperature. *Vilenskiy* [1977] has confirmed these results by analyzing the $^{210}\text{Po}/^{210}\text{Pb}$ ratio in ashes and condensates of the Tolbachik eruption.

The airflow from the Etna crater has been calculated by *Hauget et al.* [1977] by comparing the SO_2 concentration in the plume and the total SO_2 output and found to be 3×10^{11} m^3/d . This result enables us to calculate for Etna volcano an output of 200 Ci/yr for ^{210}Pb and 2500 Ci/yr for ^{210}Po .

The discharge of ^{210}Po and ^{210}Pb from Mount Erebus has been evaluated by *Polian and Lambert* [1979], by a less precise

Fig. 1. Histogram of the ^{210}Pb atmospheric concentrations observed at Kerguelen Island for different figures of the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio. (a) All data obtained. (b) $^{210}\text{Po}/^{210}\text{Pb} < 0.5$. (c) $0.5 < ^{210}\text{Po}/^{210}\text{Pb} < 1$. (d) $^{210}\text{Po}/^{210}\text{Pb} > 1$.

Fig. 2. Histogram of the ^{210}Pb atmospheric concentrations observed at Terre Adelie for different figures of the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio. (a) All data obtained. (b) $^{210}\text{Po}/^{210}\text{Pb} < 0.5$. (c) $0.5 < ^{210}\text{Po}/^{210}\text{Pb} < 1$. (d) $^{210}\text{Po}/^{210}\text{Pb} > 1$.

method, and found to be 3 Ci/yr for ^{210}Pb and 100 Ci/yr for ^{210}Po .

A direct estimation of the global output of volcanic ^{210}Pb and ^{210}Po is not possible at the present time. However, a comparison between these nuclides and the volcanic SO_2 can give a good order of magnitude of this output estimate.

The only known values of the ratios ^{210}Pb and ^{210}Po to SO_2 are given in Table 2.

It is necessary to point out that the Etna plume is exceptionally rich in sulfur compounds and the Erebus plume exceptionally rich in ^{210}Pb and ^{210}Po relative to sulfur. Consequently, the following ranges seem to be more realistic: ^{210}Pb from 4×10^{-4} to 1×10^{-3} Ci/ton of SO_2 and ^{210}Po from 5×10^{-3} to 4×10^{-2} Ci/ton of SO_2 .

Stoiber and Jepsen [1973] have estimated the total volcanic production of SO_2 to be 1×10^7 tons/yr. The volcanic production of ^{210}Pb is therefore comprised between 4×10^3 and 10×10^3 Ci/yr, i.e., 1.4×10^{23} to 3.5×10^{23} atoms per year and of ^{210}Po between 50×10^3 and 400×10^3 Ci/yr, i.e., 3.2×10^{22} to 25×10^{23} atoms per year.

As a comparison, the mean flux of ^{222}Rn to the atmosphere is about 1 atom/cm² (of continent) per second [Israel, 1951; Servant, 1964; Turekian et al., 1977], i.e., 5.2×10^{25} atoms per year corresponding to a production of a number equivalent of ^{210}Pb atoms, i.e., 1.5×10^6 Ci of ^{210}Pb per year for the whole earth. That represents from 150 to 400 times the volcanic production.

The atmospheric production of ^{210}Po can be estimated by two different ways:

1. A good mean value of the atmospheric activity of ^{210}Pb is about 10×10^{-15} Ci/kg [Sanak et al., 1978] which corresponds to 50×10^3 Ci of ^{210}Pb in the total atmosphere.

The atmospheric production of ^{210}Bi is therefore 5.5×10^{22} atoms/yr.

The residence times of the aerosols marked by this 5-day half-life nuclide being of the order of 5–10 days, at least one quarter of the ^{210}Bi atoms are removed from the atmosphere and the atmospheric production of ^{210}Po is about 4×10^{22} atoms/yr, i.e., 6×10^4 Ci/yr.

2. Let N_{Pb} and N_{Po} be the total number of ^{210}Pb and ^{210}Po

TABLE 1. Volcanic Emissions of Long-Lived Radon Daughters

Site and date	^{210}Pb Concentration, 10^{-15} Ci/m ³	^{210}Pb Output, Ci/yr	Ratio of ^{210}Bi to ^{210}Pb	Ratio of ^{210}Po to ^{210}Pb	Reference
Plume aerosols					
Etna, June 1976 (Sicily)	1,200–3,500	200	5.5	12.5	Lambert et al. [1979]
Etna, June 1978 (Sicily)	1,200–22,500	nd	3	9	Lambert et al. [1979]
Stromboli, Nov. 1978 (Italy)	1,700–2,500	nd	7	40	Lambert et al. [1979]
Erebus, Jan. 1978 (Antarctic)	3,800–12,100	3	10.2	31	Lambert et al. [1979]
Merapi, Aug. 1978 (Indonesia)	600	nd	11	24	Lambert et al. [1979]
Tolbachik, Aug. 1979 (Kamchatka)					
Ashes				17	Vilenskiy [1977]
Condensate				18	Vilenskiy [1977]
Ambient atmosphere	10		~1	0.1	

nd, not determined.

TABLE 2. Annual Output of Volcanic Products

	SO ₂ , tons/yr	²¹⁰ Pb, Ci/yr	²¹⁰ Po, Ci/yr	²¹⁰ Pb/SO ₂ , Ci/ton	²¹⁰ Po/SO ₂ , Ci/ton
Etna	1 × 10 ⁶ *	200†	2500†	2 × 10 ⁻⁴	2.5 × 10 ⁻³
Erebus	1.2 × 10 ³ ‡	3‡	100‡	2.5 × 10 ⁻³	8.3 × 10 ⁻²

*Hauget et al. [1977].

†Lambert et al. [1979].

‡Polian and Lambert [1979].

atoms in the atmosphere, ϕ_{Pb} and ϕ_{Po} the flux per time unit of those atoms to the atmosphere.

Admitting that the residence times of lead and polonium aerosols are the same, a steady state is characterized by

$$\phi_{Pb} = (\lambda_{Pb} + \lambda_S)N_{Pb} \approx \lambda_S N_{Pb}$$

$$\phi_{Po} = (\lambda_{Po} + \lambda_S)N_{Po} \approx \lambda_S N_{Po}$$

or, by using radioactivity units:

$$\frac{(\lambda_{Po}\phi_{Po})}{(\lambda_{Pb}\phi_{Pb})} = \frac{\lambda_{Po}N_{Po}}{\lambda_{Pb}N_{Pb}}$$

which is equal to the activity ratio $^{210}\text{Po}/^{210}\text{Pb}$, i.e., 0.1 in the most important part of the atmosphere [Lambert and Nezani, 1965; Moore et al., 1973].

Therefore the total flux of ^{210}Po to the atmosphere from all kinds of sources should be 1/10 of that of ^{210}Pb , i.e., 1.5×10^5

Ci/yr, or 1×10^{23} atoms per year. This is of the same order of magnitude as the calculated volcanic source.

As a conclusion, we can ascertain that the volcanic emission of ^{210}Po is equal, or superior, to its production in the atmosphere by the ^{210}Pb disintegration. On the other hand, the volcanic ^{210}Pb is generally negligible with respect to the continental production, except perhaps in some very limited areas or during exceptionally strong volcanic events. It is worthwhile to recall that a similar conclusion was drawn by Polian and Lambert [1979] for the Erebus volcano and the antarctic continent.

Therefore it has been possible to connect the observation of overequilibrium $^{210}\text{Po}/^{210}\text{Pb}$ at Dumont-Durville (Terre Adelie) to meteorological conditions in which air masses could go directly from Mount Erebus to this part of the antarctic coast, as shown in Figure 3.

LEAD AND POLONIUM 210 IN THE STRATOSPHERE

The first measurement of lead and polonium 210 in the stratosphere was made by Burton and Stewart [1960] by means of aircraft sampling. Later a large number of measures were made by several authors, but the main series of results have been published by Peirson et al. [1966], Moore et al. [1973], and Krey [1975].

Most of these results give a $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio between 0.6 and 1.0. However, Krey [1975] obtained between 1 and 2 in 6% of the cases (85 among 1300) and in 40% of the high-altitude balloon-borne sampling. The validity of such results has been questioned by their author himself. It is for this reason that since 1976 four experiments have been made by balloon in the lower stratosphere, from Aire-sur-l'Adour (France). The aim of these experiments being to measure the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios, special attention was paid to avoid contamination by tropospheric air during the crossing of the lower altitudes, and before the recovering of the samples. The filters, first put in the sampler, were carefully washed in place by a HNO₃ solution circulation just before the flight, then enclosed between two electric tight shutters, only opened during the sampling. The collected aerosols were recovered by the same operation, always without handling of the filter.

Lead and polonium were separated, as oxides, by using a ^{210}Pb -free lead carrier. The ^{210}Po was deposited on a silver plate and measured by using a very low background α spectrometer. The ^{210}Pb was determined by measuring the ^{210}Po ingrowth during a 4–5 month interval, by using the same α spectrometer. Each experiment included a blank submitted to the same treatment but without air filtration. The results are shown in Table 3.

It may be observed that in the case of flights 3 and 4 the samples show certainly an excess in ^{210}Po relative to equilibrium with ^{210}Pb . This is because of the existence of an independent source for the stratospheric ^{210}Po . According to the

Fig. 3. Meteorological situation on May 4, 1973. $^{210}\text{Po} = 2.28 \cdot 10^{-15}\text{Ci/m}^3$, $^{210}\text{Pb} = 0.84 \cdot 10^{-15}\text{Ci/m}^3$, and $^{210}\text{Po}/^{210}\text{Pb} = 2.72$.

TABLE 3. Stratospheric $^{210}\text{Po}/^{210}\text{Pb}$ Activity Ratios

	Flight 1	Flight 2	Flight 3	Flight 4
Date	Sept. 21, 1976	May 6, 1977	Oct. 26, 1977	April 27, 1978
Altitude, km	10–12	15	17.5	17
^{210}Pb concentration, 10^{-15} Ci/kg	3.3 ± 0.3	7.0 ± 0.7	3.7 ± 0.5	4.4 ± 0.6
Ranges for $^{210}\text{Po}/^{210}\text{Pb}$	0.40/0.67	0.74/1.03	1.35/2.00	1.35/2.02

Blank activities, corresponding to about 20% of that of the sample, are deduced.

preceding considerations the most likely source seems to be an injection of ^{210}Po by the major volcanic explosions.

As previously made, a comparison between lead, polonium, and sulfur compounds should be very useful. In effect, according to Lepel *et al.* [1978] the ^{210}Po enrichment should be particularly high during the initial phase of a volcanic explosion.

Castleman *et al.* [1974] concluded from the study of the stratospheric sulfur isotopic composition that volcanic emissions represent the major source of stratospheric sulfates. The major eruption of Vulcan de Fuego in October 1974 caused an enhancement of 1.6×10^6 tons of SO_4 in the stratosphere 6 months after the eruption [Lazrus *et al.*, 1979]. Cadle [1975] estimated that the SO_2 volcanic emission rate to the stratosphere is 2.8×10^5 tons/yr. This latter figure should correspond to a stratospheric reservoir greater than 2×10^4 tons SO_4 , as estimated by Kriz [1975].

By using the ratios $^{210}\text{Pb}/\text{SO}_2$ and $^{210}\text{Po}/\text{SO}_2$ based on Table 2 the value indicated by Cadle would mean that the ^{210}Pb volcanic injection to the stratosphere could be between 112 and 280 Ci/yr. As a comparison, the average concentration of ^{210}Pb in the stratosphere is about 5×10^{-15} Ci/kg corresponding to 3000 Ci for the whole stratosphere. Therefore as previously pointed out for the troposphere, the volcanic source of ^{210}Pb to the stratosphere is insignificant. On the other hand, the injection of ^{210}Po would be from 1.4×10^3 to 11.2×10^3 Ci/yr, i.e., the same order of magnitude as the flux produced by the disintegration of the stratospheric ^{210}Pb .

Such a polonium source should be visible by analyzing the very large number of stratospheric data published by Krey [1975].

Figure 4 shows the repartition in time and latitude of the cases where a ^{210}Po excess has been observed at altitudes higher than 15 km, together with the main published volcanic explosions. It appears particularly clear that no ^{210}Po excess was obtained during the quiet period from late 1968 to May 1970. But at this time, successive explosions of mounts Hekla (May 1970), Beerenberg (October 1970), and Kliuchevskoi (December 1970) were immediately followed by the reappearance of ^{210}Po excess beginning at the same latitude as these volcanoes.

It can also be pointed out that the higher rate of volcanic explosions in the northern hemisphere than in the southern corresponds to a much more frequent appearance of ^{210}Po excess.

Unfortunately, ^{210}Po was no longer measured by the U.S. program after 1975. However, it can be seen in Table 3 that a systematic difference seems to exist between the experiment relative to 1976 and the results obtained after May 1977 where we found an overequilibrium. This difference has been tentatively attributed to the violent explosion of Mount Niragongo, January 10, 1977.

The results of the flights 3 and 4 would need a stratospheric injection of at least 1×10^3 Ci of ^{210}Po .

It is very difficult to determine whether such an amount of

^{210}Po could have been injected into the stratosphere by this volcanic explosion. In effect, according to Tazieff [1977] the volume of lavas spilled out has been estimated at 20 to $22 \times 10^6 \text{ m}^3$ but after the eruption the increase of the crater volume was measured to be $250 \times 10^6 \text{ m}^3$, i.e., 10 times more. Moreover, we have no information at all on a possible ^{210}Po enrichment of the magmatic gases responsible for the explosion.

It can be questioned whether the changes in nature of stratospheric aerosols observed early in 1977 by Gras [1978] could also be attributed to the Niragongo explosion.

The first consequence of ^{210}Po volcanic injections into the stratosphere is to strongly change the calculation of the aerosol residence time in the stratosphere.

Let N_{Pb} and N_{Po} be the total number of atoms of ^{210}Pb and ^{210}Po , respectively, in the stratosphere, λ_s the scavenging constant, and ϕ_{Po} the flux of volcanic ^{210}Po to the stratosphere.

A steady state is characterized by

$$\lambda_{\text{Pb}} N_{\text{Pb}} + \phi_{\text{Po}} = (\lambda_{\text{Po}} + \lambda_s) N_{\text{Po}}$$

If r is the average ratio $\lambda_{\text{Po}} N_{\text{Po}} / \lambda_{\text{Pb}} N_{\text{Pb}}$, we have

$$\lambda_s = \lambda_{\text{Po}} \left[\frac{1}{r} \left(1 + \frac{\phi_{\text{Po}}}{\lambda_{\text{Pb}} N_{\text{Pb}}} \right) - 1 \right]$$

$\lambda_{\text{Pb}} N_{\text{Pb}}$ is about 3000 Ci, i.e., 6.7×10^{15} dpm; ϕ_{Po} has been evaluated to be at least 1000 Ci/yr, i.e., 1.3×10^{15} atoms/min in the case of a steady state; r is measured to be of the order of 0.8.

We find thus $\lambda_s = 0.5\lambda_{\text{Po}}$ corresponding to a half life of 276 d, which should be a maximum value (instead of 1.5 yr if $\phi_{\text{Po}} = 0$).

Fig. 4. Time and latitudinal repartition of $^{210}\text{Po}/^{210}\text{Pb}$ over-equilibria measured by Krey [1975] at altitudes greater than 15 km. Thin lines indicate $1 < ^{210}\text{Po}/^{210}\text{Pb} < 1.2$, heavy lines indicate $^{210}\text{Po}/^{210}\text{Pb} > 1.2$, stars represent the major volcanic explosion reported, able to give stratospheric injection. Numbers in parentheses indicate sample numbers during the year.

CONCLUSION

Owing to the scavenging processes, the residence time of the aerosols in the troposphere is much too short to allow ^{210}Pb and ^{210}Po to reach radioactive equilibrium with ^{222}Rn . Consequently, the flux from the soil to the whole atmosphere which is 3×10^9 Ci/yr for ^{222}Rn yielding by radioactive decay 1.5×10^6 Ci/yr for ^{210}Pb , and corresponds to a steady state production of 6×10^4 Ci/yr, for ^{210}Po .

This explains why the existence of volcanic sources may be completely negligible on a global scale in the case of ^{222}Rn , very weak for ^{210}Pb , but quite significant for ^{210}Po , especially as this last nuclide is very enriched, relative to ^{210}Pb , in all of the volcanic plumes that we have studied. The volcanic ^{210}Po production of 5×10^4 Ci/yr, indicated in this work, is thus equivalent to the production by the disintegration of the ^{210}Pb present in the atmosphere. Such a value, which represents about 20 times the production by Mount Etna only, accounts for a mean $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio greater than 0.1, and therefore higher than the figure expected from theoretical calculations based on the aerosol residence time as established from other pairs of nuclides.

The existence of this significant volcanic source of ^{210}Po also accounts for the frequent observations of $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios greater than 1, which seem generally impossible to attribute to the other ^{210}Po sources considered: a marine source of ^{210}Po is very likely but should be too weak to be significant, except perhaps when the ^{210}Pb concentration is extremely low.

The high ^{210}Po concentration in the Mount Erebus emissions is probably responsible for the observation of very high $^{210}\text{Po}/^{210}\text{Pb}$ ratios in Terre Adélie.

Volcanic emissions of ^{210}Po reach the stratosphere and there modify significantly the $^{210}\text{Po}/^{210}\text{Pb}$ ratio for several months after the injection. However, the relatively high frequency of overequilibrium observations, only can be explained by these injections from volcanoes.

One of the consequences of these volcanic injections into the stratosphere is to change the calculation of the aerosol half-life in this reservoir. The figures indicated in this work, about 280 days, is shorter than the values initially admitted but in good agreement with the last results published by Moore, Poet, and Martell. However, a systematic study of $^{210}\text{Po}/^{210}\text{Pb}$ ratios in the stratosphere should be continued, possibly in connection with major volcanic eruptions.

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