Tellus (1990), *42B*, 76–82

Atmospheric transport of trace elements toward Antarctica

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(Manuscript received 29 November 1988; in final form 5 June 1989)

ABSTRACT

Since 1960, at the French station of Dumont d'Urville (Terre Adelie coast), during more or less long periods, we have monitored Rn-222 and its decay product Pb-210, fission products due to atmospheric nuclear tests, and the cosmonuclides Be-7 and P-32. Almost all these species show seasonal variations, whose general features are maximum concentrations during the local summer (December–January), and minima in the middle of winter (June–July). A long-term change in the Pb-210 concentrations was also observed at Dumont d'Urville, for all seasons, with a more-or-less continuous decrease since 1969 to 1986, from 2.5 to 1.5 dpm per 1000 m^3. Such variations seem to be quite usual, because an ice core sampled at the South Pole also displays important fluctuations of the Pb-210 concentration in the fresh snow between 1888 and 1974, from 1 to 3 dpm/kg, particularly from 1920 to 1954. The latitudinal profiles of both Rn-222 and Pb-210 showing minima between 40° and 60° South at sea level, the long-range transport of these nuclides from mid-latitudes toward Antarctica should occur through the higher rather through the lower layers of the troposphere. Therefore, the Pb-210 long-term changes observed can be ascribed to global changes of the general circulation in the Southern hemisphere. The concentrations of Pb-210 at Dumont d'Urville, and the ozone vertical column in October at Halley Bay, show common features, more particularly a significant decrease in the 1980s, meaning that changes in the atmospheric circulation occurred in the Antarctic area from the middle of the 1970s, which can partly account for the ozone decrease beside variations of trace species concentrations.

1. Introduction

The Antarctic continent is probably the only large area on Earth that acts exclusively as a sink for energy, water vapor, and gaseous and particulate trace materials: all these are transported to the Antarctic continent from sources, in regions of lower latitudes. During their long range transport toward and over Antarctica, the chemically active trace species are subjected to their usual removal processes. Moreover, some peculiar processes occur in the very cold Antarctic atmosphere, involving heterogeneous reactions not observed in other terrestrial areas, which have been considered as possible explanations for the so-called ozone hole.

Natural and man-made radioactive tracers are very useful for studying these transport processes, because they have well-known sources and, for Rn-222, are evenly spread and almost continuous. Moreover, they do not participate in atmospheric chemical reactions. Rare gases are removed from the atmosphere by their own radioactive decay, and aerosols also by scavenging.

Rn-222 is outgassed from the surface of continents, except Antarctica which is almost totally ice covered. The outgassing rate depends on soil characteristics such as U/Ra content, porosity, moisture content and on local meteorological conditions (Tanner, 1980). However, it does not change significantly with time, and a rather constant mean annual value of the Rn-222 atmospheric concentration is observed at continental monitoring stations (Lambert et al., 1982).

Pb-210 aerosols are produced by the radioactive decay of atmospheric Rn-222. Pb-210

* CFR Contribution no. 1628.

atoms are trapped by submicron aerosols (Sanak et al., 1981). With the Pb-210 half-life being about 20 years, the main sink of this nuclide is aerosol scavenging, with a global tropospheric mean residence time of about 1 week. However, a Pb-210 reservoir of longer residence time also exists in the stratosphere. A general budget of atmospheric Rn-222 and Pb-210 was made by Lambert et al (1982).

Since 1960, at the French station of Dumont d'Urville, on the Terre Adelie coast (Fig. 1), during more-or-less long periods, beside Rn-222 and its decay product Pb-210, we have monitored fission products due to atmospheric nuclear tests, and the cosmonuclides Be-7 and P-32. The techniques utilized have been described in preceding papers: Lambert (1963) for fission products; Sanak et al. (1985), for cosmonuclides; Lambert et al. (1970) for Rn-222; Lambert et al. (1966, 1975) for Pb-210.

Almost all these species show important seasonal variations, whose general features are maximum concentrations during the local summer (December–January), and minima in the middle of winter (June–July). It is worthy of note that the major errors of measurement are due to stochastic variations in radioactive countings. In this paper, we only use mean monthly, seasonal and annual values, calculated from at least 30 measurements, and sometimes thousands. The statistical errors are therefore significantly reduced to less than a few %, and generally still less. They can be totally neglected relative to the variations under study. By contrast, non-stochastic errors are mainly due to inaccurate calibrations, particularly of the radioactive counter efficiency: these errors obviously do not compensate by averaging, and they forbid a precise study of the long term evolution of most of the species, but Pb-210.

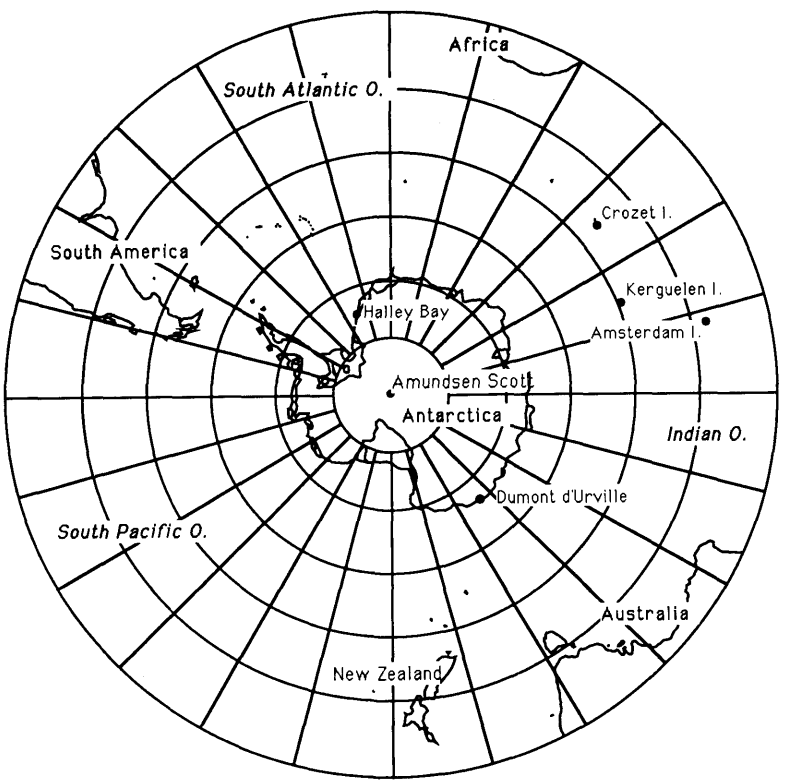

Fig. 1. Antarctic map.

2. Seasonal variations of Rn-222 and Pb-210

Rn-222 atmospheric concentrations in subantarctic and antarctic areas are controlled by their long-range transport from mid latitudes, which varies with the season. This nuclide has been monitored from 1965 to the present at Kerguelen island, and from 1967 to 1981 at Dumont d'Urville, at the rate of 12 times per day. Fig. 2 shows its seasonal variations at Dumont d'Urville, each figure being calculated from about 5000 values. During the same time, Pb-210 has also been monitored at Dumont d'Urville on a daily basis. Its seasonal variations are shown in the same figure, where each point represents about 400 values. The similarity of the two curves is particularly remarkable, suggesting that the residence times of the two nuclides are extremely close to each other over the subantarctic ocean, namely 5.5 days, which is the Rn-222 radioactive mean life, instead of 1 week for the whole troposphere.

This rather short life suggests that the transport of these nuclides from their remote continental sources is rapid. However, Sanak (1983) and Polian et al. (1986) pointed out that in summer, at sea level, the latitudinal profiles of both Rn-222 and Pb-210 show minima between 40° and 60° South. This observation applies particularly to Pb-210, as shown in Fig. 3, in which we have gathered the results obtained by several authors for subantarctic traverses along different meridians, as well as the US network at 80° West, operating from 1974 to 1976. It is worthy to note that the figure relative to Dumont d'Urville is the mean value of the 4 summer months (November to February), from 1960 to 1980, i.e., of about 1200 values.

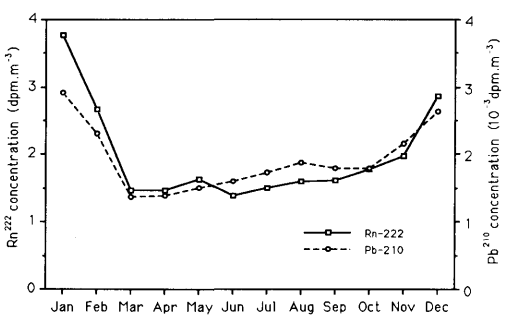

Fig. 2. Seasonal variations of Rn-222 and Pb-210 at Dumont d'Urville (Terre Adelie) for the period 1967–1981. Each point has been calculated from about 5000 values of Rn-222, and 400 values of Pb-210.

Consequently, during the summer, the long-range transport of these nuclides from mid-latitudes toward Antarctica cannot occur through the lower layers of the troposphere. On the other hand, a stratospheric circulation is ruled out by the long time involved. For all these reasons, Polian (1984) concluded that there is a transport of Rn-222 and Pb-210 through the higher layers of the troposphere.

3. Seasonal variations of fission products and cosmonuclides

The preceding hypothesis is supported by the study of the atmospheric concentrations of fission products at Dumont d'Urville, which show a clear maximum in summer, especially in February (Fig. 4). During the periods *free of nuclear tests,* the fission products collected at ground level have been injected into the higher troposphere from their stratospheric reservoir. Lambert et al. (1965, 1966) showed that the geographical distribution of the fission products was then similar to that of Rn-222 and Pb-210, with a minimum at around 50° South.

The very important role played by transport through the high troposphere in the observed seasonal variations is emphasized by analysis of the short-lived cosmonuclides Be-7 (53.4 days) and P-32 (14.3 days). These nuclides are mainly produced in the stratosphere (Lal and Peters, 1962) at a very constant ratio Be-7/P-32 = 100. It may be observed in Fig. 5 that the longer-lived of these nuclides has an almost constant mean monthly value, whereas the shorter-lived one, whose concentration at ground level strongly depends on its recent injection from the stratosphere, shows the same already-mentioned seasonal variations.

Therefore, the similarity of the seasonal variations at Dumont d'Urville of Rn-222, Pb-210, fission products and short-lived cosmonuclides can be simply ascribed to the well-known fact that, at spring time (around October),

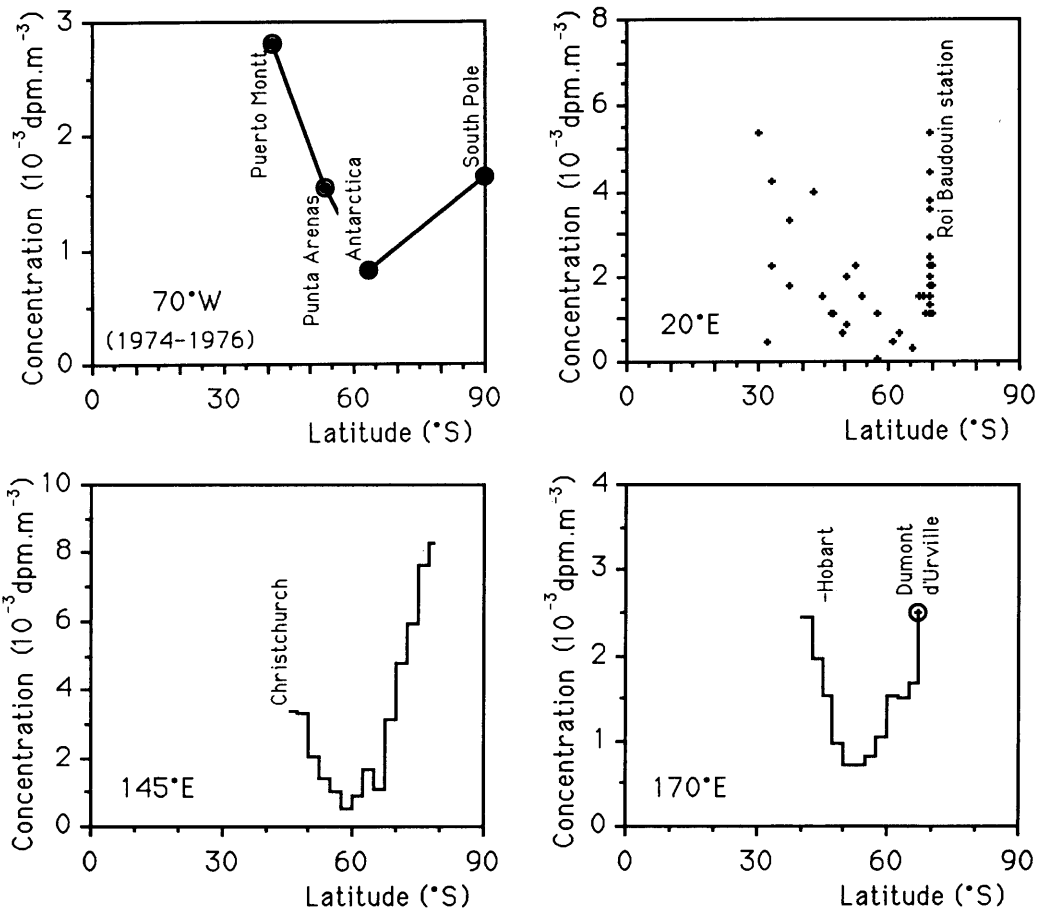

Fig. 3. Latitudinal profiles of Pb-210 in Subantarctic areas along the meridians 70°W, 20°E, 145°E and 170°E. Large circles represent the average of a large number of measurements.

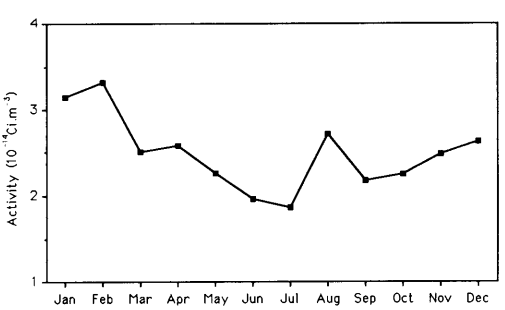

Fig. 4. Seasonal variations at Dumont d'Urville of fission products from stratospheric origin, i.e., during periods without atmospheric nuclear tests.

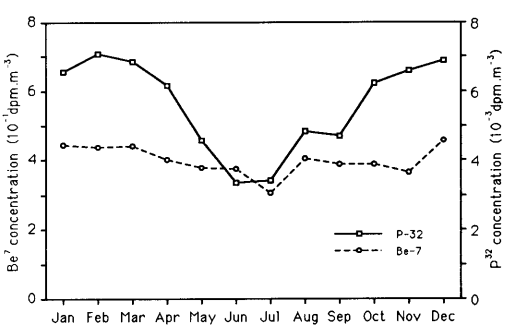

Fig. 5. Seasonal variations of Be-7 and P-32 at Dumont d'Urville for the period 1977–1981. Each point represents 100 to 150 values.

when the winter polar vortex vanishes, the whole Antarctic atmosphere is overturned by air advections from the north.

4. Long-term variations of Pb-210

Our Be-7 and P-32 monitoring at Dumont d'Urville has been too short to enable a long term trend study. A similar study would be unsound for atmospheric fission products whose variations are primarily due to the nuclear test history.

As above mentioned, Rn-222 has been monitored from 1965 to the present at Kerguelen island, and from 1967 to 1982 at Dumont d'Urville. However the interannual calibration of the measurement devices is not very accurate. Nevertheless, it may be seen in Fig. 6 that, at these stations, the Rn-222 mean annual values are almost constant, within $\pm 25\%$, which is about the same as the long-term reproducibility. Consequently, these observations cannot be interpreted in terms of atmospheric circulation changes.

Pb-210 has also been monitored at Dumont d'Urville since 1960, on a daily basis. In fact, the aerosols are only collected at this station, by filtration, and the filters are subsequently measured in the laboratory after their return to France. The Pb-210 activity is determined through that of its granddaughter Po-210, which is an alpha emitter with a 138 day half-life. For this reason, and because the Po-210 activity is typically 10 times less than that of Pb-210 in the troposphere, a two-year delay is necessary to let Po-210 reach its radioactive equilibrium with Pb-210 on the filter. Therefore, the measurements relative to 1987 are not already available.

Owing to the utilized technique, the efficiency of the measurements was continuously controlled by regularly remeasuring several filters. No shift was detected in this efficiency (Lambert et al. 1988). Consequently, long term variations of the order of a few % can be considered as significant.

A record of the mean annual concentrations is shown in Fig. 7, where each point represents the mean value of about 365 daily measurements, together with the values relative to December–February (seasonal maximum) and May–August (seasonal minimum), where each point is the mean value of about 100 measurements. Except in 1964 and 1967, these curves show the same general trend, characterized by a more-or-less continuous decrease since 1969, from 2.5 to 1.5 dpm per $1000m^3$ for the mean annual value. Therefore, the long-term change in the Pb-210 concentrations at Dumont d'Urville, observed for all seasons, should be due to a global change of the general circulation in the Southern hemisphere.

Such variations seem to have been quite usual in the past. In effect, an analysis of an ice core, sampled at South Pole, also displayed important fluctuations of the Pb-210 concentration in the fresh snow between 1888 and 1974, from 1 to 3 dpm/kg (Sanak and Lambert, 1977). A large change of the Pb-210 concentration was observed in the Antarctic snow, particularly from 1920 to 1954: it could therefore be possible that similar

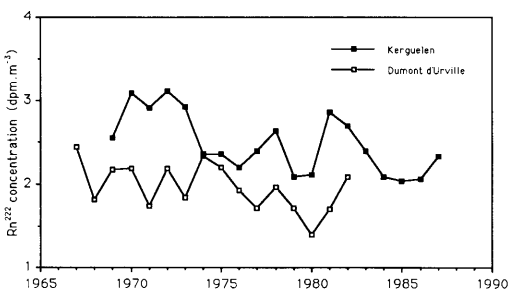

Fig. 6. Long-term monitoring of Rn-222 at Kerguelen island and Dumont d'Urville. Each point represents about 4000 measurements; however, the interannual calibration is not better than $\pm 25\%$.

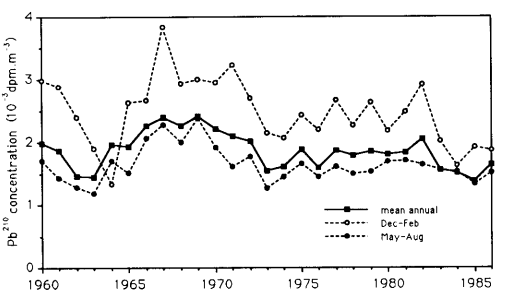

Fig. 7. Long-term monitoring of Pb-210 at Dumont d'Urville: mean values annual, December to February, and May to August calculated from daily measurements.

atmospheric circulation changes already occurred in the past, even though they were not detected through classical meteorological measurements. They cannot be explained for the moment.

5. Comparison of Pb-210 and other trace species

It is interesting to examine to what extent the preceding changes have affected other natural and/or man made species, whether chemically inactive like Pb-210 aerosols, or active such as ozone. This gas is likely (beside Pb-210) the only chemical species for which a long-term record is available in the Antarctic atmosphere.

Farman et al. (1985), and Stolarski et al. (1986), showed a remarkable downward trend of the ozone vertical column during the springtime over Antarctica. It is generally accepted that ozone variations are controlled by photochemical processes, and by the atmospheric circulation that is driven by a particularly stable winter polar vortex. Most models of this phenomenon have focused on chemical interactions of ozone with natural and/or anthropogenic trace species (Solomon et al., 1986; McElroy et al., 1986a, b; Crutzen and Arnold, 1986; Molina et al., 1987), although dynamical processes have also been considered (Tung et al., 1986).

However, the atmospheric concentrations of Pb-210 do not depend on chemical processes, and their record in Antarctica, since 1960, also shows a clear decrease since 1979. This trend seems to be rather similar to that of ozone. The concentrations of Pb-210 at Dumont d'Urville, relative to October, are reported in Fig. 8, together with the ozone vertical column measured during the same months at Halley Bay (Farman et al., 1985), which is also situated on the Antarctic coast, but at a longitude almost diametrically opposed to Dumont d'Urville. We plot in the same figure the ozone measurements deduced from the Nimbus-7 satellite observations (Stolarski, 1988). All these curves show common features, more particularly a significant decrease in the 1980s, but the correlation coefficients are small: -0.66 (confidence level 99.2%) for 1960–1975; $+0.71$ (confidence level 97.8%) for 1977–1986. All correlations are considerably weakened when data relative to 1976 are also utilized, the correlation coefficients falling to -0.49 and $+0.41$, respectively.

To conclude, it seems that a drastic change occurred during the period 1975–1977 in the mean meridional transport of trace materials from mid-latitudes to the Antarctic atmosphere. A similar observation can be made from the Be-7 record in aerosols at the South Pole and Punta Arenas (Feely et al., 1988), where the year 1976 seems to be unusual, more particularly in October. This change is also visible in the chemical analyses of Antarctic ice cores by Legrand and Kirchner (1988). All these observations mean that, beside variations of trace species concentrations, which can account for a local as well as general ozone decrease (Bowman, 1988; Reinsel et al., 1988), changes in the atmospheric circulation also occurred in the Antarctic area, from the middle of the 1970s, i.e., before the ozone decrease became important.

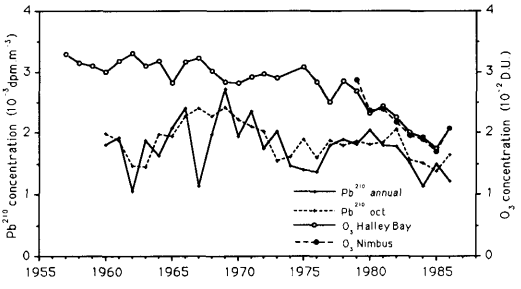

Fig. 8. Comparison of ozone and Pb-210 in Antarctica.

6. Acknowledgements

This research was supported by the French CNRS, CEA, the administration of Terres Australes et Antarctiques Françaises, and the Expéditions Polaires Françaises. The authors thank all scientists and technicians for aerosol sampling in Antarctica, and J. C. Duplessy for fruitful discussions.

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