Atmospheric ^{210}Po anomaly as a precursor of volcano eruptions

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[1] Due to its high volatility, ^{210}Po can be effectively released from active volcanoes prior to their explosions. A continuous record of the fallout rate of ^{210}Po versus that of ^{210}Pb in northern Taiwan reveals that most of the ^{210}Po was derived from pre-eruption gas emission of Philippines'Mayon volcano in the past two years. From this observation we suggest that, if a global network can be properly designed, it may be possible to employ ^{210}Po anomaly as a precursor of volcano eruptions worldwide. *INDEX TERMS:* 0932 Exploration Geophysics: Radioactivity Methods; 0370 Atmospheric Composition And Structure: Volcanic Effects (8409); 4801 Oceanography: Biological And Chemical: Aerosols (0305); 8419 Volcanology: Eruption Monitoring (7280)

1. Introduction

[2] In the atmosphere, ^{210}Po ($T_{1/2}$ = 138 d) is steadily produced from the decay of ^{222}Rn via several short-lived nuclides, followed by ^{210}Pb ($T_{1/2}$ = 22.3 y) and ^{210}Bi ($T_{1/2}$ = 5.01 d). Following its production, ^{210}Po is removed from the atmosphere via its own decay and scavenging by aerosols. However, based on generally accepted residence times of aerosols in the atmosphere (on the order of ~1 week in the troposphere and ~1 year in the stratosphere), global inventory of ^{210}Po in the atmosphere is much higher than that can be produced from its progenitors. Thus, there must exist additional inputs for this nuclide in the atmosphere. To this end, various potential sources have been assessed, including volcanic emission [*Lambert et al.*, 1979, 1982; *Nevissi*, 1984; *Sheng and Kuroda*, 1985; *Nho et al.*, 1996a], coal burning [*Marenco and Fontan*, 1972], air-sea gas exchange [*Turekian et al.*, 1974, 1977; *Bacon and Elzermann*, 1980; *Hussain et al.*, 1995; *Kim et al.*, 2000], dust storms [*Vilenskii*, 1970; *Moore et al.*, 1980; *Carvalho*, 1995; *Nho et al.*, 1996b] and forest fires [*Nho et al.*, 1996b; *Le Cloarec et al.*, 1995]. Among them, only volcanic emission appears to be significant on a global scale [*Lambert et al.*, 1979, 1982].

[3] Due to its volatility, ^{210}Po is highly enriched in volcanic gases in which ^{210}Po/^{210}Pb activity ratios up to ~600 have been reported [*Gauthier et al.*, 2000]. Conversely, the ^{210}Po/^{210}Pb ratios in freshly erupted pyroclastic or lava flows are close to zero indicating effective loss of ^{210}Po to gaseous phases [*Bennett et al.*, 1982; *Gill et al.*, 1985; *Rubin et al.*, 1994]. Although volcanic emissions are highly patchy in space and variable with time, introduction of ^{210}Po from this pathway over time can account for more than one half of the global budget of this nuclide in the atmosphere [*Lambert et al.*, 1979, 1982], exceeding the fairly steady production from its progenitors. Thus, anomalous levels of atmospheric ^{210}Po concentrations or fluxes, especially downwind from volcanoes, are in all likelihood attributable to intensified volcanic emissions.

[4] Here we report fairly detailed time series of over two years, from April 1999 to June 2001, of ^{210}Pb and ^{210}Po fluxes (and hence the ^{210}Po/^{210}Pb activity ratio) at Nankang, a suburban district of Taipei in northern Taiwan. During this period, the Mayon volcano in Philippine experienced many explosive episodes. By correlating our data sets with the timing of Mayon's gas emission and eruption activities in the context of the meteorological conditions during this period, we found that exceptionally high ^{210}Po fluxes at Nankang usually preceded Mayon's eruptions by weeks or months. We hence judge that the unusually high ^{210}Po fluxes are caused by pre-eruption emissions from the Mayon volcano.

1.1. The Mayon Volcano

[5] The Mayon volcano, located 340 km SE of Manila (13°15.4′N, 123°41.1′E) and 1,320 km SSE of Nankang (Figure 1), is the Philippines' most active and volatile volcano. Historically, over 50 eruptions have been recorded since 1616, with a recurrence period averaging ~7 years. However, the frequency of Mayon's activity has been unusually high in recent years. Six years after the major eruption on 2 February 1993, Mayon erupted on 22 June 1999, 5 January 2000, and then almost everyday during 24 February – 7 March 2000 with intense degassing activities. The volcano quieted down in the subsequent nine months but started rumbling and degassing again since January 2001, with ten eruptions already occurred in 2001 alone. So, over twenty eruptions have been recorded during our sampling period of about two years.

2. The Sampling and Analytical Methods

[6] Rainwater (plus dry fallout) was collected at weekly intervals (or shorter when typhoons arrived) using a collector mounted at the rooftop of the Institute of Earth Sciences, Academia Sinica, at Nankang. The collector was fabricated from a PVC cylinder 150 cm in height and 30 cm in diameter, with a conical bottom outfitted with a spigot. Each time after the sample was drained out, 500 ml of 1N HNO_3 and an equal volume of distilled water were used successively to rinse the inner walls of the collector. The washings were combined with the rainwater sample in a beaker and evaporated on a hot plate. After the volume had substantially reduced, the sample was transferred to a smaller beaker to continue the evaporation process. In conjunction with each transfer, the interior of the beaker was acid-cleaned (by refluxing with HNO_3 and $HClO_4$) and the washings were combined with the condensate in the next beaker for further volume reduction. Eventually, the sample was reduced to a nominal size of ~0.5 ml in a 2-ml Teflon beaker, from there the final condensate and beaker washings were transferred to a Kimax tube with a final volume of about 1 ml. Although the work was very tedious, it was meticulously performed. We believe the sample loss is minimal, probably <1%.

[7] The condensed sample was counted using a well-type HPGe detector (EG&G ORTEC GWL-100230) interfaced with a digital γ-ray spectrometer (EG&G ORTEC DSPec™) for the determination of ^{210}Pb and ^{7}Be. Efficiencies of the detector as functions of sample volume and γ-ray energy have been carefully calibrated using ^{152}Eu (SRM-4370C), ^{7}Be (BNL S/M 054911) and Harwell uraninite standard solutions. For 1-ml samples, the absolute count-

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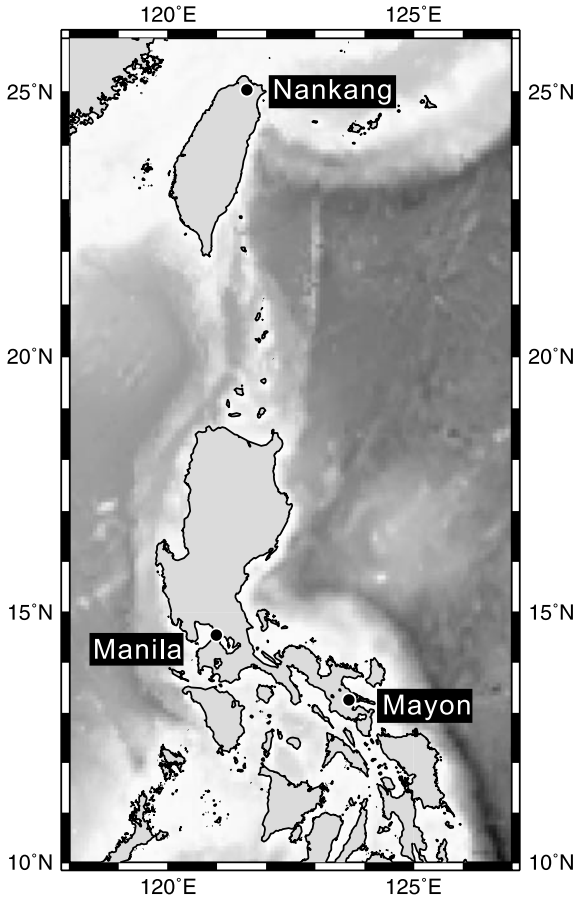

Figure 1. Map showing the locations of Nankang in northern Taiwan and the Mayon volcano in Philippines. Tropospheric transport of Mayon's emissions to Taiwan can be facilitated during the summer monsoon and typhoon seasons when the prevailing winds are toward the north.

ing efficiency is 78% for ^{210}Pb (at 46.52 keV) and 19% for ^{7}Be (at 477.56 keV).

[8] After gamma spectrometry, each sample was spiked with ^{209}Po and processed for alpha spectrometric analysis of ^{210}Po following procedures described elsewhere [*Huh et al.*, 1997; *Huh and Su*, 1999].

[9] The data reported here are decay-corrected back to the time of sample collection. The ^{210}Po data have also been corrected for the ingrowth of ^{210}Po from ^{210}Pb decay during the time span from sample collection to Po plating.

3. Results and Discussion

[10] Figure 2 shows the time series of ^{210}Po, ^{210}Pb and ^{7}Be fluxes at Nankang along with SO_2 emission rate at the Mayon Volcano (http://www.phivolcs.dost.gov.ph). Also indicated on the SO_2 plot are the dates of Mayon's eruption.

[11] A most striking feature in Figure 2 is the high variability of the ^{210}Po flux, which exhibits an overall decrease by more than three orders of magnitude from mid-1999 to late 2000. In contrast, ^{210}Pb and ^{7}Be fluxes do not show such a decreasing trend; they correlate strongly with each other (r = 0.90) but poorly with the ^{210}Po flux (r = 0.19 and 0.31, respectively), suggesting a peculiar source for ^{210}Po during this observation period.

[12] To filter out the effect of wet precipitation as a major factor controlling nuclide fluxes, the ^{210}Po flux is divided by the corresponding ^{210}Pb flux. However, even with this normalization procedure, the ^{210}Po/^{210}Pb ratios still span over three orders of magnitude (Figure 2c) and are usually (especially in the first half of the time series) much higher than values reported for the eastern Pacific [0.03–0.11; *Moore et al.*, 1977] and the northwestern Pacific regions [0.05–0.22; *Yokota and Tsunogai*, 1991].

[13] It is important to point out that, before Mayon's eruption on 22 June 1999, abnormally high ^{210}Po fluxes (with over-equilibrium ^{210}Po/^{210}Pb ratios) had already been measured two months in advance at Nankang. The highest ^{210}Po flux, at 0.101 dpm/cm^2/d, occurred during 19–20 June 1999, 2–3 days before Mayon's eruption on 22 June 1999. Interestingly, higher ^{210}Po fluxes (and hence ^{210}Po/^{210}Pb ratios) could also be noticed 1–2 weeks prior to subsequent eruptions on 5 January and 23 February 2000. Considering the timing of the eruptions mentioned above along with the seasonal changes in the direction of prevailing winds, we couldn't speculate any sources other than Mayon's emission to explain the observed ^{210}Po anomaly. It then follows that what caused the "early arrival" of Mayon-derived ^{210}Po 1,320 km away — even before the volcano erupted at ground zero? We believe it is due to efficient degassing of this highly volatile nuclide followed by long-range transport from the volcanic edifice at an early stage before violent eruptions take place. In fact, although it was not well understood, high levels of ^{210}Po/^{210}Pb ratios in fallout samples prior to major volcanic eruptions have been noted elsewhere before, such as the 1980 eruption of Mount St. Helens [*Nevissi*, 1984] and the 1982 eruption of El Chichón volcano [*Sheng and Kuroda*, 1985].

[14] The tremendous ^{210}Po output from the Mayon volcano prior to the June 1999 eruption must be due to the rather long duration since the previous eruption in 1993. During this period of over six years, the amount of ^{210}Po produced in the magma chamber should be in equilibrium with ^{210}Pb. After the release in 1999 of the accumulated ^{210}Po, it would take some time in a closed system to replenish this volatile nuclide. However, the system was probably not closed, as indicated by incessant gas emission following the June 1999 eruption. Thus, despite the violent eruptions with very high gas emission rates in early 2000, the amount of ^{210}Po introduced in that year was probably insufficient to significantly compensate the decay of the residual ^{210}Po from the eruption in 1999. Alternatively, the low delivery in 2000 of Mayon-derived ^{210}Po at Nankang is probably due to the direction of prevailing winds, which was toward the south during the late winter and early spring when the volcano was active.

[15] An attempt was made to simulate the change with time of the ^{210}Po flux based on the equation:

$$\frac{\partial N_{Po}}{\partial t} = I_{Po} + \lambda_{Pb} N_{Pb} - \lambda_{Po} N_{Po} - \lambda_S N_{Po} \qquad (1)$$

Where N_{Pb} and N_{Po} are respectively the number of ^{210}Pb and ^{210}Po atoms caught in the sampling device in each sampling period; λ_{Pb} and λ_{Po} are the decay constant of ^{210}Pb and ^{210}Po, respectively; I_{po} represents the episodic volcanic input of ^{210}Po; and λ_S is the rate constant of nuclide scavenging by aerosols. Nuclide numbers in the above equation are derived from nuclide fluxes shown in Figure 2 using the equation: F (dpm/cm^2/d) = $\lambda N/A/\Delta t$, where A is the collection area of our device and Δt is each sampling period.

[16] The calculated ^{210}Po fluxes are divided by their corresponding ^{210}Pb fluxes and compared with the observed time series of ^{210}Po/^{210}Pb (Figure 2c). Numerical simulation of the λ_S value suggests a best-fit mean residence time of ~38 days for aerosols carrying Mayon-derived ^{210}Po. Residence times of aerosols in the atmosphere generally increase with altitude [*Reiter*, 1975]. The 38-day residence time falls in the typical range for aerosols in the upper troposphere and near the tropopause [*Krey and Krajewski*, 1970; *Reiter et al.*, 1975].

[17] About 60 of the Earth's 550 historically active volcanoes are in eruption each year [*Simkin et al.*, 1994]. The wax and

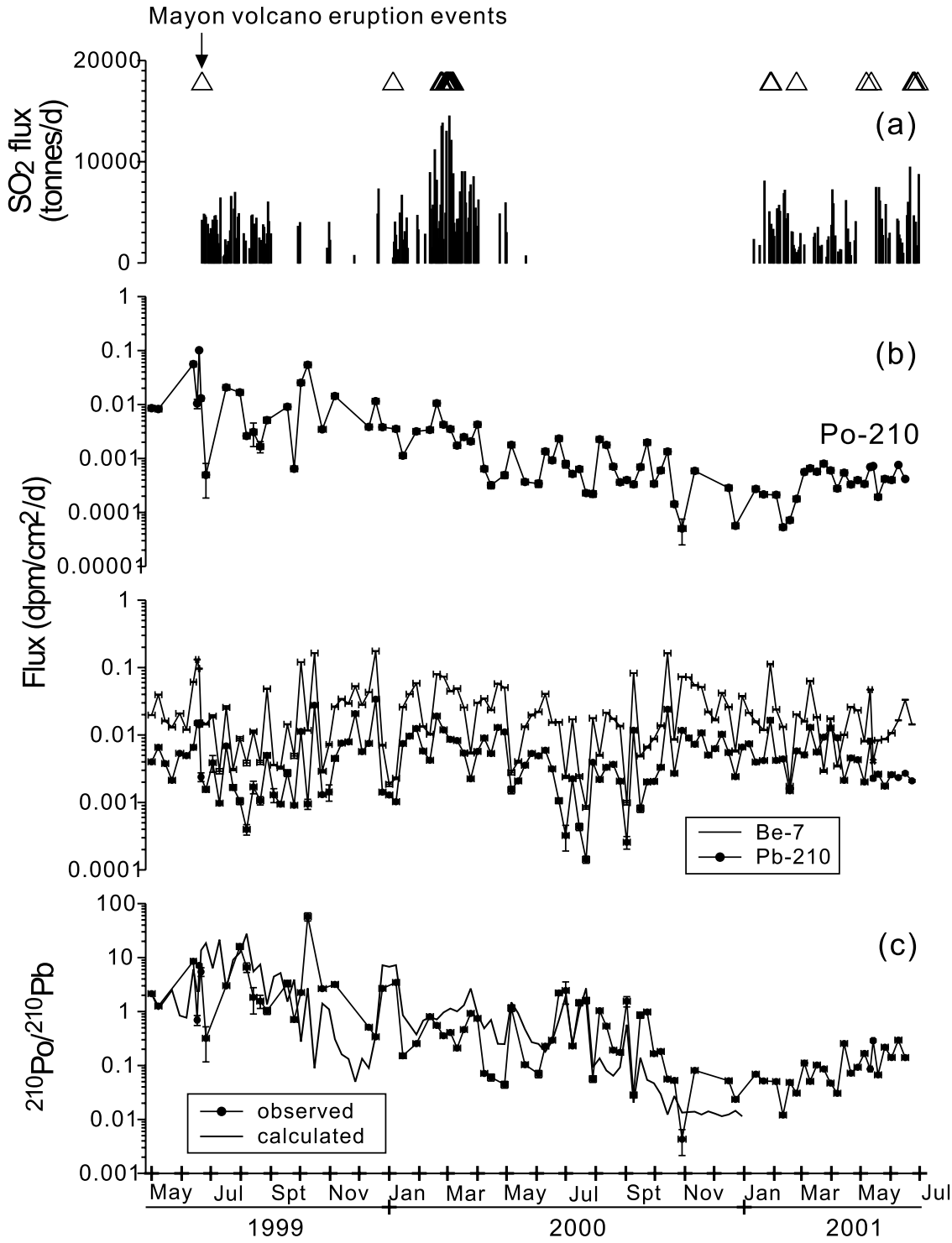

Figure 2. The time series of (a) SO_2 flux, (b) ^{210}Po, ^{7}Be and ^{210}Pb fluxes, and (c) ^{210}Po/^{210}Pb activity ratio. Except the SO_2 flux, which represents Mayon's emission, all other data are obtained at Nankang.

waning of volcanic activities are customarily monitored near volcanoes based on seismic, ground deformation, gas emission and observational monitoring data. Here, by showing the fallout flux in Taiwan of ^{210}Po derived from Mayon's emissions, we suggest that volcanic activities could be monitored remotely downwind. The early emission of ^{210}Po and its enrichment in volcanic gases for long-range transport make this nuclide a highly promising precursor to volcano eruptions around the world.

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