Factors controlling atmospheric fluxes of ^{7}Be and ^{210}Pb in northern Taiwan

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[1] Fluxes of ^{7}Be and ^{210}Pb were continuously monitored at two sites in northern Taiwan during 1996–2001. The time series of both nuclides show seasonality and dependence on precipitation, annual monsoon cycle and episodic events (typhoons and Asian dust storms). Besides, anomalously high ^{7}Be and ^{210}Pb fluxes occurred in 1998, which are most likely caused by enhanced atmospheric convection in the north Pacific resulting from significantly increased SST in the western Pacific warm pool. *INDEX TERMS:* 0932 Exploration Geophysics: Radioactivity methods; 1040 Geochemistry: Isotopic composition/chemistry; 1854 Hydrology: Precipitation (3354); 1610 Global Change: Atmosphere (0315, 0325). **Citation:** Su, C.-C., C.-A. Hu, and F.-J. Lin, Factors controlling atmospheric fluxes of ^{7}Be and ^{210}Pb in northern Taiwan, *Geophys. Res. Lett.*, *30*(19), 2018, doi:10.1029/2003GL018221, 2003.

1. Introduction

[2] This study involves the application of two fallout nuclides: ^{7}Be and ^{210}Pb. ^{7}Be (half-life 53.3 days) is a cosmogenic radionuclide produced by spallation of oxygen and nitrogen in the upper troposphere and lower stratosphere by cosmic rays. The production of ^{7}Be has negligible dependence on season or longitude, but varies with altitude, latitude and the ∼11-year solar cycle [*Koch et al.*, 1996]. Activities of ^{7}Be in the atmosphere increase sharply with altitude and have been reported to range from 0.06 to 1.2 dpm m^{-3} in the troposphere and 10 to 35 dpm m^{-3} in the stratosphere [*Kaste et al.*, 2002]. The amount of ^{7}Be that reaches the surface of the earth is controlled by factors or processes including cosmic-ray intensity, wet and dry deposition, vertical transport in the troposphere and horizontal transport from the subtropics and mid-latitudes into the tropics and polar regions [*Feely et al.*, 1989; *Talpos and Cuculeanu*, 1997]. The atmospheric flux of ^{7}Be observed worldwide in previous studies ranged from ∼6 to ∼40 dpm cm^{-2} yr^{-1} [e.g., *Turekian et al.*, 1983; *Olsen et al.*, 1985; *Schuler et al.*, 1991].

[3] ^{210}Pb (half-life 22.3 years) is a member of the ^{238}U decay-series nuclides. The primary source of ^{210}Pb in the atmosphere is ^{222}Rn emanation from continental soils. The global ^{222}Rn flux into the atmosphere is estimated to be about 8 to 11 dpm cm^{-2} d^{-1} from continental areas but merely ∼0.1 dpm cm^{-2} d^{-1} from oceanic areas [*Samuelsson et al.*,

1986; *Nazaroff*, 1992]. Therefore, spatial variation of atmospheric ^{210}Pb concentration or flux is to a large extent controlled by land-sea distribution. *Beks et al.* [1998] estimates that, downward fluxes of ^{210}Pb are 1.3–2 dpm cm^{-2} yr^{-1} in continental areas but only ∼0.02 dpm cm^{-2} yr^{-1} in oceanic areas, with a global mean of about 0.4–0.6 dpm cm^{-2} yr^{-1}.

[4] Owing to their contrasting source terms, ^{7}Be and ^{210}Pb have been widely used as tracers for studying atmospheric transport and exchange, aerosol removal processes, and global circulation models [*Balkanski et al.*, 1993; *Baskaran et al.*, 1993; *Baskaran*, 1995; *Rehfeld and Heimann*, 1995; *Koch et al.*, 1996; *Tokieda et al.*, 1996]. Although such studies are fairly common, the data are still lacking in key areas such as Taiwan and its vicinity. It is important to fill the data gaps to establish a globally comprehensive database.

[5] Located between the world's largest continent and the largest ocean and intersected by the tropic of Cancer, Taiwan is well suited for studying complex and important atmospheric dynamics. The weather system in Taiwan is regulated by northeast monsoons in the winter and southwest monsoons in the summer. Superimposed on the annual cycle of monsoons are Mei-yü (plum rain) and dust storms derived from the Asian continent and typhoons coming from the tropical Pacific. Using ^{7}Be and ^{210}Pb as tracers, we attempted to examine the inner workings of the atmospheric system and various meteorological factors and processes in controlling the sources, transport and removal of ^{7}Be and ^{210}Pb from the atmosphere in northern Taiwan and in the vicinity of the southern East China Sea.

2. The Sampling and Analytical Methods

[6] The sampling sites, Nankang and Peng-Chia Yü, represent two rather different settings (Figure 1). Nankang (NK) is a suburban district of Taipei in northern Taiwan and Peng-Chia Yü is a remote islet (area = 1.14 km^{2}) ∼55 km offshore to the north. Rainwater samples were collected at weekly intervals at NK but monthly intervals at PCY. 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 and evaporated on a hot plate. During the evaporation and sample transfer processes, the sizes of the beakers were also reduced step-by-step. Whenever a
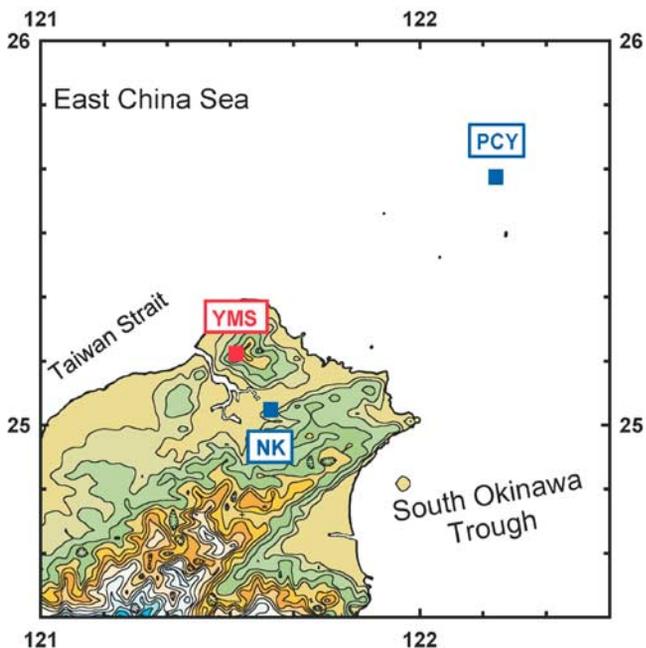
Figure 1. The position of Nankang (NK), Peng-Chia Yü (PCY) and Yang-Ming Shan (YMS) in northern Taiwan.

beaker was emptied, it was acid-washed (by refluxing with HNO_3 and $HClO_4$) and the washings were combined with the condensate in the next, smaller beaker for further evaporation. Eventually, the sample was reduced to a nominal size of $\sim$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.

[7] Gamma counting was performed 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 ^{7}Be (BNL S/M 054911) and Harwell uraninite standard solutions. For 1-ml samples, the absolute counting efficiency is 78% for ^{210}Pb (at 46.52 keV) and 19% for ^{7}Be (at 477.56 keV). The data reported here have been decay-corrected to the mid-point of sample collection.

3. Results and Discussion

[8] Figure 2 shows the time series of ^{7}Be and ^{210}Pb fluxes along with the amounts of precipitation at PCY (Figure 2b) and NK (Figure 2c). Although the sampling resolution at NK is one week (to be presented later), the data are integrated to monthly intervals in Figure 2c to facilitate inter-site comparison. Also shown in Figure 2 for the discussion to follow are the time series of Niño3 + 4 SSTA (Climate Prediction Center/NOAA; ftp://ftpprd.ncep.noaa.gov/pub/cpc/wd52dg/data/indices/sstoi.indices), 1st EOF of Pacific Warm Pool's SST (Climate Diagnostics Center/NOAA-CIRES; http://www.cdc.noaa.gov/Correlation/pacwarm.data), and PM$_{10}$ concentration at Yang-Ming Shan (data from Taiwan EPA), another suburban site near Taipei.

[9] Fluxes of ^{7}Be and ^{210}Pb show strong seasonality, with peaks in the winter and low values in the summer. There are

also inter-annual and inter-site variations. For the entire multi-year time series, the mean fluxes of ^{7}Be and ^{210}Pb are 6.7 and 1.1 dpm cm^{-2} yr^{-1}, respectively, at PCY and are substantially higher at NK (11.0 and 1.9 dpm cm^{-2} yr^{-1} for ^{7}Be and ^{210}Pb, respectively). There exists a strong correlation between long-term nuclide fluxes and average rainfall at these two sites. More details of the time series are discussed below.

[10] The Asian continent constitutes the dominant source of ^{210}Pb in the western Pacific. When the Mongolia High moves southward in the winter, the northeast monsoon prevails and transports ^{210}Pb-enriched air masses towards Taiwan. Conversely, the southwest monsoon in the summer introduces into Taiwan ^{210}Pb-depleted air masses from the South China Sea and western Pacific high-pressure systems. Therefore, the switch of monsoon can largely explain the annual cycle of the ^{210}Pb flux. As for ^{7}Be, its production does not have any direct relationship with land-sea distribution. The introduction of ^{7}Be is controlled mainly by stratosphere-troposphere exchange and vertical mixing within the troposphere [*Husain et al.*, 1977; *Viezee and Singh*, 1980; *Feely et al.*, 1989; *Baskaran*, 1995]. The transport of ^{7}Be from the stratosphere downward is enhanced during tropopause folding which in subtropic areas usually takes place during late winter and early spring. During this period the height of
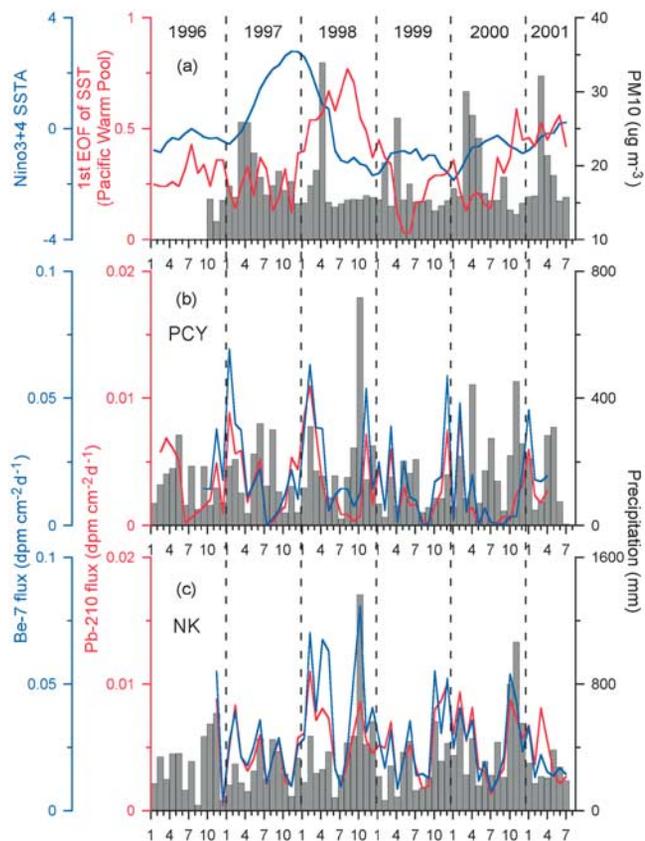
Figure 2. Time series of (a) Niño3 + 4 SSTA, 1st EOF of Pacific Warm Pool's SST, and PM$_{10}$ concentration at Yang-Ming Shan, suburban Taipei, (b) ^{7}Be and ^{210}Pb fluxes along with monthly precipitation at PCY, and (c) ^{7}Be and ^{210}Pb fluxes along with monthly precipitation at NK.
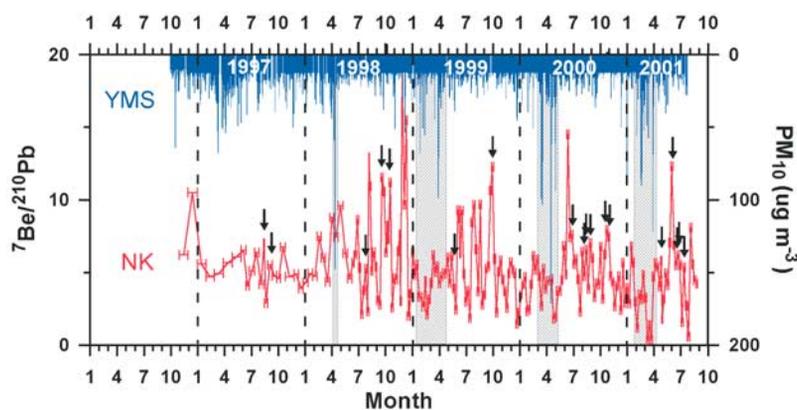
Figure 3. Correlation between the flux ratio of ^{7}Be/^{210}Pb at Nankang (bottom, scale at the left), timing of typhoons (indicated by arrows) and PM$_{10}$ concentration measured at Yang-Ming Shan, Taipei (top, scale at the right). Note the correspondence between: (1) typhoons and ^{7}Be/^{210}Pb peaks, and (2) PM$_{10}$ peaks and low ^{7}Be/^{210}Pb ratios.

tropopause over northern Taiwan reaches its annual low (down to ~6 km, compared with ~16 km in the summer and fall).

[11] Regardless of the distinct difference in source terms of ^{7}Be and ^{210}Pb, their fluxes (as shown in Figures 2b and 2c) correlate well with each other (r = 0.89, n = 113, P < 0.001) and fluctuate with wet precipitation. In fact, this is also a common phenomenon elsewhere and suggests that both nuclides are removed from the atmosphere mainly by rainfall [*Baskaran et al.*, 1993; *Caillet et al.*, 2001].

[12] In addition to factors and processes mentioned above, other episodic events can also modify the time series of ^{7}Be and ^{210}Pb fluxes. Most noteworthy are the effects of typhoons and Asian dust storms. During the period covered in this study, 25 typhoons stroke Taiwan (Figure 3), with the hardest hit years being 1998 (5 hits), 2000 (7 hits) and 2001 (5 hits). Typhoons cause strong convection and mixing in the troposphere, promoting downward transport of ^{7}Be-enriched upper air and upward transport of ^{210}Pb-enriched lower air. Consequently, the ^{7}Be/^{210}Pb ratio in the lower troposphere increased substantially during a typhoon. This can be clearly seen in Figure 3.

[13] Dust storms can affect the relative flux of ^{7}Be and ^{210}Pb in a different way. The primary source regions of Asian dust storms are the Taklimakan desert, the Gobi desert and the Loess Plateau in China [*Zhang et al.*, 1993; *Xuan and Sokolik*, 2002]. Every spring, dust storms formed in conjunction with the expansion of high-pressure systems over these regions can reach Korea, Japan, Taiwan and Hong Kong [*Fan et al.*, 1996; *Chung et al.*, 2001; *Yang*, 2002]. The invasion of dust storms into Taiwan each year is clearly reflected in the PM$_{10}$ data from Taiwan-EPA's air quality monitoring station at Yang-Ming Shan, which usually happens around March and April (Figure 3). Unlike typhoons, which cause high ^{7}Be/^{210}Pb ratios in precipitation, dust storms usually result in low ^{7}Be/^{210}Pb ratios due to their association with ^{210}Pb-laden continental air masses.

[14] Finally, in addition to intra-annual variabilities discussed above, we wish to examine if there are any inter-annual changes in the 5-year time series. Possible mechanisms for long-term (>1 year) changes include solar activity cycle (for ^{7}Be only), El Niño/Southern Oscillation (ENSO), and quasi-biennial oscillation [*Lal and Peters*,

1967; *Reiter et al.*, 1983; *Koch et al.*, 1996; *Gerasopoulos et al.*, 2003]. Although our time series is not adequate to address the effect of solar activity on ^{7}Be, it covers the period (i.e., 1997–1998) when the most intense ENSO in the 20th century occurred. This gives us an opportunity to evaluate the effect of ENSO on the flux of fallout nuclides, although it is sometimes difficult to differentiate
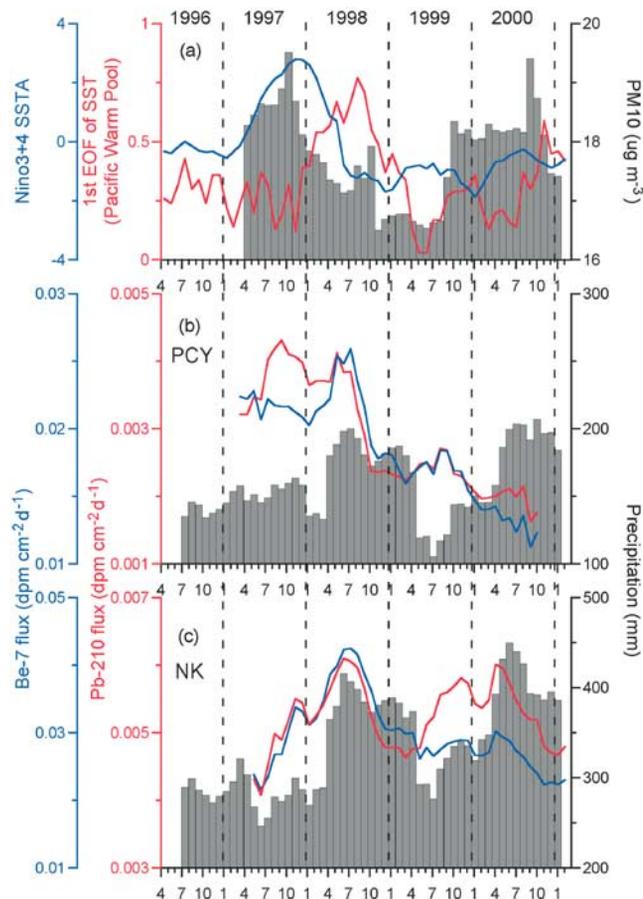
Figure 4. 13-month moving average derived for the attributes in Figure 2 to smooth out short-term fluctuations and highlight inter-annual oscillations.

between ENSO and quasi-biennial oscillation [*Koch and Mann*, 1996; *Baldwin et al.*, 2001].

[15] To facilitate the delineation of long-term trend, we plotted the data to show the 13-month moving average (Figure 4) so as to smooth out short-term periodicities or noises. Figure 4a shows that the 1997–1998 ENSO is depicted by pronounced sea surface temperature anomalies, namely, the maximum in late 1997 of Niño3 + 4 SSTA, followed by that of 1st EOF of Pacific Warm Pool's SST in mid-1998, with a 9-month time lag. Concurrent with the peak in 1998 of 1st EOF of Pacific Warm Pool's SST is the peak in the ^{7}Be plot for both PCY and NK. It should be noted that during the 1997–1998 ENSO, SST around Taiwan increased similarly and was in phase with the 1st EOF of Pacific Warm Pool's SST curve [*Chen*, 2002]. This large increase in SST caused large-scale warming and enhanced mixing in the troposphere [*Hoerling et al.*, 2001], and thus more frequent typhoons and higher amounts of precipitation. As discussed earlier, typhoons are responsible for the introduction of relatively higher ^{7}Be flux and higher ^{7}Be/^{210}Pb ratio.

[16] It should also be noted in Figure 4 that, there were two instances when the 13-month moving average of ^{7}Be and ^{210}Pb showed large disparities, with unusually high ^{210}Pb fluxes and the trend of its moving average mimicking that of PM$_{10}$ concentration. The reason that the ^{210}Pb anomaly was not observed at NK during 1997–1998, nor at PCY during 1999–2000, was probably because of insufficient amounts of precipitation or height of cloud cover for effective scavenging of ^{210}Pb-laden airborne particles.

[17] In conclusion, Taiwan is situated in a sensitive location where the interplay of various meteorological factors and processes could be deciphered using ^{7}Be and ^{210}Pb as aerosol tracers. In this study, we have shown that the time series of ^{7}Be and ^{210}Pb fluxes (and their relative magnitudes) at two contrasting sites can be interpreted by atmospheric transport and precipitation associated with monsoons, typhoons and dust storms for intra-annual variability and by that associated with ENSO (or SST anomaly) for long-term oscillations. If used in combination with other tracers of natural or anthropogenic origin, these fallout nuclides could find more practical applications such as the study of volcanic eruptions [*Su and Huh*, 2002], air pollution and global change issues.

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