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Factors controlling temporal and spatial variations of atmospheric deposition of ^{7}Be and ^{210}Pb in northern Taiwan

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[1] Fluxes of ^{7}Be and ^{210}Pb monitored at two contrasting sites in northern Taiwan were studied along with meteorological data to elucidate factors controlling their temporal and spatial variations. The 9 year time series of both nuclides at Nankang in the Taipei Basin show dependence on wet precipitation and follow an annual cycle regulated by typhoons, monsoons, and mei-yü. Superimposed on the annual cycle are interannual variation caused by El Niño–Southern Oscillation and intraseasonal oscillations due to the passage or invasion of fronts, cold surges, dust storms, and the Pacific high-pressure system. The observed ^{7}Be/^{210}Pb ratios are indicative of the source regions of the fallout nuclides, with higher ratios from high-altitude rain and lower ratios from dry fallout and low-altitude rain. The 2 year time series at Yangminshan shows that ^{7}Be and ^{210}Pb fluxes at the mountainous site are in phase with those at Nankang. However, nuclide fluxes at Yangminshan are 4–5 times those at Nankang. After combining results from this and other ancillary studies at Yangminshan, we suggest that (1) in applying fallout nuclides to study the Earth's surface processes, the assumption of constant flux should be made judicially according to the timescales involved, and that (2) the application of fallout nuclides to study soil erosion in hilly areas must consider not only control by wet and dry precipitation but also by direct contact and interaction of clouds with soils enhancing nuclide fluxes.

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1. Introduction

[2] Be-7 ($T_{1/2}$ = 53.3 days) and ^{210}Pb ($T_{1/2}$ = 22.3 years) are two fallout radionuclides which have been extensively used as tracers to study the Earth's surface processes. Be-7 is produced by spallation of oxygen and nitrogen nuclei by cosmic rays in the stratosphere and upper troposphere [*Lal et al.*, 1958]. The atmospheric production of ^{7}Be has no relationship with season and longitude, but its tropospheric fluxes vary with altitude, latitude, and season as the stratospheric/tropospheric exchange occurs in spring [*Lal and Peters*, 1967; *Hötzl et al.*, 1991; *O'Brien et al.*, 1991; *Koch and Mann*, 1996]. Following its production, ^{7}Be is rapidly attached to aerosol particles [*Junge*, 1963], with its atmospheric distribution governed by the interaction and movements of air masses and its removal from the atmosphere controlled by aerosol scavenging and precipitation to the Earth's hydrosphere and pedosphere [*Feely et al.*, 1988].

[3] Pb-210 in the atmosphere comes primarily from the decay of ^{222}Rn emanated from continental soils. Oceanic areas contribute merely ~1% of the global ^{222}Rn flux into the atmosphere [*Samuelsson et al.*, 1986; *Nazaroff*, 1992]. Thus atmospheric ^{210}Pb concentration decreases with altitude and is to a large extent regulated by land-sea distribution. As for the transport in and removal from the atmosphere, both ^{210}Pb and ^{7}Be are essentially controlled by the same factors and processes. Thus owing to their contrasting source terms, different half-lives, but same transport and removal mechanisms, these two nuclides are very powerful tools, especially when used in tandem, for studying atmospheric dynamics and scavenging of aerosols and associated materials.

[4] Following the deposition of ^{7}Be and ^{210}Pb onto the Earth's surface, both nuclides are strongly attached to soil and sediment particles. If the depositional fluxes of ^{7}Be and ^{210}Pb in a catchment basin or landscape unit are monitored, then from the distribution and inventories of these two nuclides in top soils or surficial sediments it is possible to study erosion, transportation and deposition of soils and sediments from episodic to multidecadal timescales. The issue of soil conservation is important for Taiwan, an island experiencing the highest erosion rate in the world [*Li*, 1976; *Dadson et al.*, 2003].

[5] In Taiwan, located between the Asian continent and Pacific Ocean, one is in a unique position to study the effects of complex atmospheric dynamics and meteorological phenomena on the deposition of ^{7}Be and ^{210}Pb. The climate system of Taiwan is regulated by the annual cycle of

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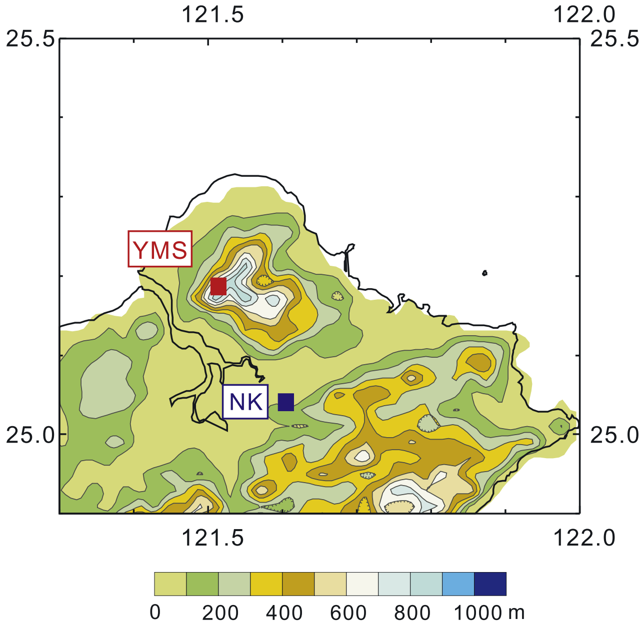

Figure 1. Geographic location of Nankang (NK) and Yangminshan (YMS) in northern Taiwan.

East Asian monsoon, mei-yü, typhoons, and dust storms, and by quasi-biennial oscillation and El Niño–Southern Oscillation (ENSO) on multiyear timescales [*Su et al.*, 2003]. On shorter timescales (<1 month), Taiwan's weather conditions are influenced by the passage of cold fronts in the fall and winter months and the waxing and waning of the Pacific high year-round [*Lee and Liu*, 2004]. In order to apply ^{7}Be and ^{210}Pb as tracers to study the Earth's surficial processes more rigorously, it is mandatory to understand factors controlling their atmospheric fluxes first. To this end, a time series data with adequate time length and resolution are required. We have been monitoring the fluxes of fallout radionculides in northern Taiwan since the late 1996 and previously reported some interim results (until mid-2001) [*Su and Huh*, 2002; *Su et al.*, 2003]. Since then, the time series data have been expanded substantially with more details emerging. A more thorough and updated overview of our observations is warranted.

[6] The study sites, Nankang (NK) and Yangminshan (YMS), are both within the Greater Taipei metropolitan area in northern Taiwan. NK (25°1′26″N, 121°34′48″N, 15 m above sea level (asl)) is in the Taipei Basin, whereas YMS (25°9′54″N, 121°33′55″N, 730 m asl) is situated in the Tatun volcano group in the north of the Taipei Basin (Figure 1). Although NK and YMS are in the same mesoscale meteorological domain, they have very different weather and climatic conditions due to differences in topography. A comparison of the time series data between these two sites may shed light on orographic and related effects, thus providing the necessary conditions for improved soil erosion studies in Taiwan [*Huh and Su*, 2004].

2. Sampling and Analytical Methods

[7] Rainwater sampling started since 31 October 1996 at NK where samples were collected at monthly to biweekly intervals at the initial stage and then, for the majority of the 9 year period, we felt compelled to increase the sampling frequency from weekly to daily in order to better distinguish signals from intense rainfall events. Sampling at the YMS site started on 26 August 2003 and was synchronized with that at NK so that the YMS time series can be studied vis-à-vis the NK time series.

[8] The rainwater collector was fabricated from a PVC cylinder 150 cm in height and 30 cm in diameter, with a conical bottom outfitted with a spigot. The sample collection and processing procedures have been described elsewhere [*Su and Huh*, 2002; *Su et al.*, 2003]. Briefly, sizes of rainwater samples were reduced (via evaporation) to within 2 mL and transferred to Kimax® tubes for gamma counting. Two well-type HPGe detectors (EG&G ORTEC GWL-100230), each interfaced to a stand-alone digital γ-ray spectrometer (EG&G ORTEC DSPec Plus®), were used for this work. Efficiencies of the detectors as a function of γ-ray energy and sample size have been carefully calibrated using certified liquid standards containing ^{7}Be (BNL S/M 054911) and mixed nuclides (IAEA-133B). Depending on sample sizes (within 2 mL) and detectors used, the absolute counting efficiencies fall within 64–79% for ^{210}Pb (at 46.52 keV) and 19–20% for ^{7}Be (at 477.56 keV).

3. Results and Discussion

[9] Space limitation does not allow the massive data sets to be tabulated here, but they can be accessed electronically as auxiliary material[1]. The data are also stored at the Data Management Center of the Institute of Earth Sciences, Academia Sinica and can be located at http://dmc.earth.sinica.edu.tw/Contributor/Huh/Huh_et_al2006. To facilitate the following discussion, the occurrences of typhoons, dust storms and notable weather conditions are annotated on the archived database. Plotted in Figure 2 are the time series of ^{7}Be and ^{210}Pb fluxes and rainfall intensities at NK during 1996–2005. Also shown in Figure 2 are the $^{7}Be/^{210}Pb$ ratios and daily mean concentrations of PM_{10} (suspended particulate materials less than 10 μm in diameter); the latter data are obtained from Taiwan EPA's air quality data monitored at Sijhih, ~3 km in the east of NK. In what follows, we shall focus on the 9 year time series from NK, followed by a comparison between NK and YMS for the latter part of the time series (from 2003 to 2005).

[10] With a total of 427 data points (and much more for the PM_{10} data) in the NK time series, temporal variations are depicted at a fairly high resolution. The correspondence between the peaks of nuclide fluxes and episodes of heavy rain is distinct. However, the correlation between rainfall and time-averaged daily fluxes for the overall data sets is not strong (Figure 3), suggesting large temporal variability in the source regions, hence characteristics, of air masses and the complexity of atmospheric dynamics and scavenging processes (to be discussed later).

[11] The correlation of rainfall with ^{7}Be (r^2 = 0.35) is significantly better than with ^{210}Pb (r^2 = 0.14), suggesting that wet precipitation carries a heavier weight in removing ^{7}Be from the atmosphere. This is consistent with previous

[1]Auxiliary materials are available at ftp://ftp.agu.org/apend/jd/2006jd007180.

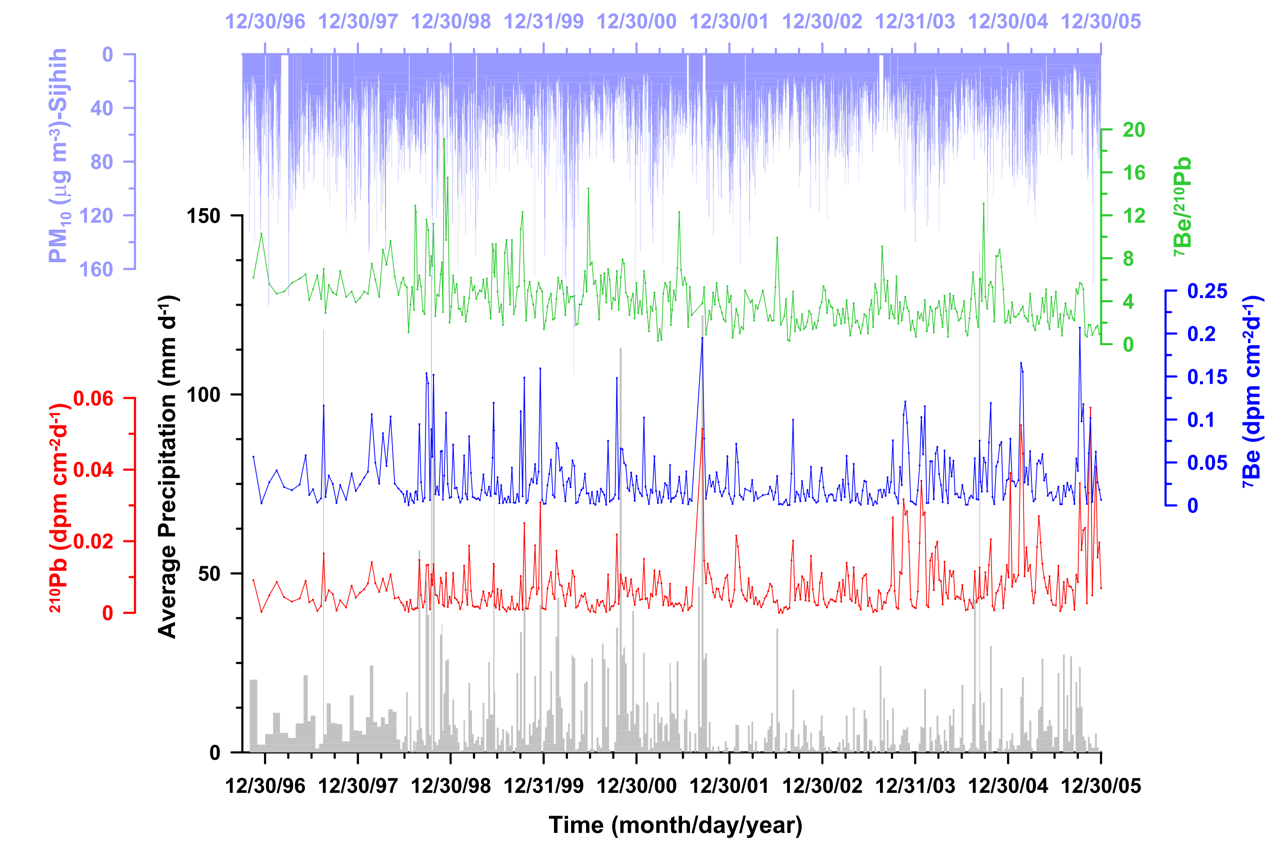

Figure 2. Time series of the fluxes of ^{7}Be (third row) and ^{210}Pb (fourth row) and daily rainfall (bottom row) during 1996–2005 at NK. Also plotted for comparison are the daily mean PM_{10} concentration (top row) and the ^{7}Be/^{210}Pb ratio (second row).

studies and supports the notion that dry deposition as a scavenging process is more important for ^{210}Pb than for ^{7}Be [*Olsen et al.*, 1985; *Schuler et al.*, 1991; *Caillet et al.*, 2001; *McNeary and Baskaran*, 2003]. In contrast to the mediocre correlation between the fluxes of individual nuclides and rainfall, the correlation between ^{7}Be and ^{210}Pb fluxes is much better (r^2 = 0.62; Figure 4). This is also a rather common observation [*Baskaran et al.*, 1993; *Kim et al.*, 2000; *Caillet et al.*, 2001; *McNeary and Baskaran*, 2003] and suggests that deposition of both nuclides are largely governed by the same processes despite their different source functions and somewhat different behaviors.

[12] Since it has long been recognized that wet precipitation is mainly responsible for removing ^{7}Be and ^{210}Pb from the atmosphere [*Small*, 1959; *Todd et al.*, 1989; *Baskaran*, 1995; *Caillet et al.*, 2001; *McNeary and Baskaran*, 2003; *Su et al.*, 2003; *Su and Huh*, 2006], it is conducive to interpret the time series data in the context of rainfall climatology in northern Taiwan. Shown in Figure 5 is an averaged annual cycle of monthly precipitation at Nankang during 1996–2005, which is consistent with rainfall characteristics in northern Taiwan synthesized from Taiwan Central Weather Bureau's data for the period from 1961 to 1998 [*Chen and Chen*, 2003]. Thus despite very large interannual variations in the NK time series, the combined data set may still be representative of climatological mean phenomena on decadal or even longer timescales in northern Taiwan.

3.1. Factors and Processes Controlling the Characteristics of ^{7}Be and ^{210}Pb Deposition

[13] Be-7 and ^{210}Pb fluxes and the ^{7}Be/^{210}Pb ratio in northern Taiwan are regulated by a number of factors and processes, some of them are discussed below in the context of major climatological and weather phenomena over an annual cycle.

3.1.1. Typhoons

[14] Located near the western Pacific warm pool and intersected by the tropic of Cancer, Taiwan is frequently

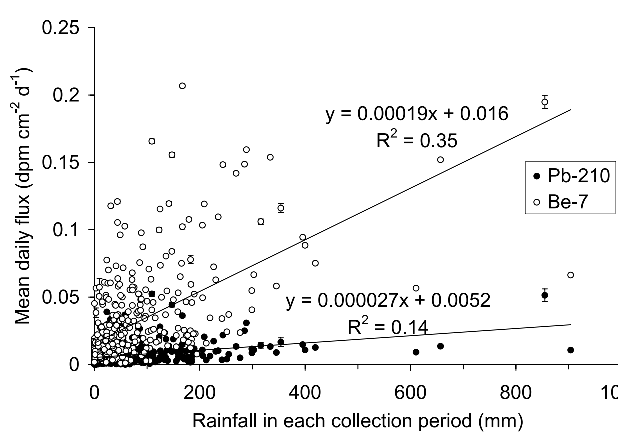

Figure 3. Correlation between the fluxes of individual nuclides (^{7}Be and ^{210}Pb) with rainfall at NK.

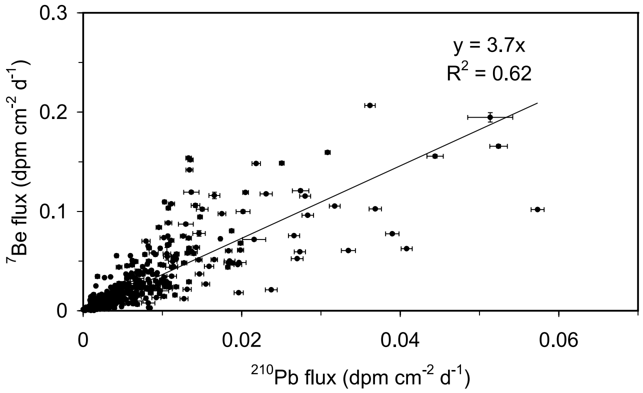

Figure 4. Correlation between the fluxes of ^{7}Be and ^{210}Pb at NK.

affected by typhoons formed in the northwestern Pacific. In the summer and fall seasons (primarily during July–September) of 1997–2005, northern Taiwan was directly hit or indirectly influenced by a total of 53 typhoons. In Figure 2, approximately 50% of the sampling episodes with mean rainfall intensity exceeding 15 mm/d were related to typhoons whereas all those above 50 mm/d were caused by typhoons. Overall, precipitation associated with typhoons accounts for 31% of the total rainfall in the 9 year time series.

[15] While typhoons often induce heavy rain, they also contribute significantly to the deposition of ^{7}Be and ^{210}Pb. Taking the 9 year time series as a whole, 16% of the total ^{7}Be flux and 12% of the total ^{210}Pb flux are derived from typhoons. These percentages are proportionally lower than the contribution of typhoons to total rainfall. Possible causes for this discrepancy include the dilution effect by larger amounts of rainwater and lower concentrations of ^{7}Be and ^{210}Pb in low-latitude maritime air masses in which typhoons are formed.

[16] During typhoons, strong convection in the troposphere promotes downward transport of ^{7}Be-enriched upper air and upward transport of ^{210}Pb-enriched lower air, thus resulting in an increase of the ^{7}Be/^{210}Pb ratio in the lower troposphere [*Su et al.*, 2003]. The ^{7}Be/^{210}Pb activity ratios from typhoon-derived rain reflect the source region, path, and maximum cloud height of typhoons. Higher ratios are typical of high-altitude maritime air masses usually associated with more intense typhoons, while lower ratios may result from typhoons modified by continental air masses on their trajectories.

3.1.2. Monsoons

[17] Located between the world's largest continent and the largest ocean, Taiwan is strongly affected by the East Asian Monsoons. During the northeast monsoon season following the typhoon season, frequent passages of cold fronts and cold surges in northern Taiwan combined with orographic effects render the autumn the wettest season during the annual cycle (Figure 5). It is important to note that precipitation during the northeast monsoon season contributes to ~60% of the annual rainfall and accounts for ~56% of the annual fluxes of ^{7}Be and ^{210}Pb, with ^{7}Be/^{210}Pb ratios similar to annual means. Thus if normalized against the amount of rainfall, monsoon rain is ~2 times more effective than typhoon rain in removing fallout nuclides from the atmosphere. The difference is most likely related to rainfall characteristics. Precipitation associated with typhoons in the warm season is of strong convective type, as mentioned earlier. Under typhoon conditions, below-cloud washout could be an important process removing ^{7}Be and ^{210}Pb. In contrast, with a more stable stratification in the cold season, most of the cold fronts, cold surges and northeast monsoon flow bring in more stable precipitation in northern Taiwan, sometimes with overcast conditions lasting for longer periods of time. The contrasting scavenging efficiency between monsoon rain and typhoon rain may very well reflect the different effect of rainout versus washout processes [*Su and Huh*, 2006].

3.1.3. Mei-yü

[18] Each year from mid-May to mid-June, Taiwan is influenced by the so-called mei-yü. In Chinese words, mei-yü means "plum rain," which happens in the time of the year when southwest monsoon flow begins to prevail and plum begins to ripe in southern China. During this period of approximately one month, mei-yü fronts frequently bring in heavy rainfall over the Taiwan area, causing a small rainfall peak superimposed on a decreasing trend from late winter to early summer [*Chen and Chen*, 2003]. The mei-yü rain contributes to about 8% of the annual rainfall and ~7% of the annual fluxes of ^{7}Be and ^{210}Pb at Nankang during 1996–2005. The ^{7}Be/^{210}Pb ratio in this period (4.27) is not significantly different from the overall mean (4.20).

3.1.4. Dust Storms

[19] Between the late season of the northeast monsoon and the onset of mei-yü, there is a drier period when Taiwan may be affected by dust storms originating in arid and semiarid regions in northern China, Mongolia, and central Asia. During this time of the year, as part of the winter Asian monsoon system, the Siberian high dominates the weather pattern of East Asia, transporting dusts to Taiwan via northeastern airflow at the southeastern part of the high-pressure system. As PM_{10} levels in the air rises above 100 μg m^{-3} in northern Taiwan, they are often attributed to long-range Asian dust transport [*Chou et al.*, 2004]. In recent years, there has been a clear trend of increased frequency and intensity of dust storms. The number of times Taiwan was affected by dust storms from China increased from 1–3 times per year during 1994–1999 to 5–11 times per year during 2000–2002 [*Lee and Liu*, 2004].

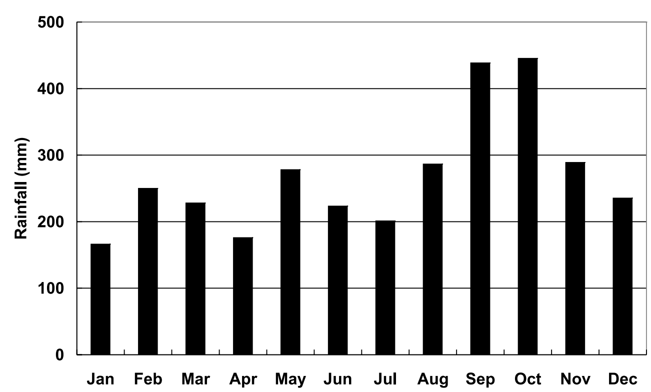

Figure 5. Annual cycle of monthly rainfall at NK averaged from 1996 to 2005.

[20] Figure 2 shows that, in periods of dust storm incursion during the spring of 2001 through 2005, the $^{7}Be/^{210}Pb$ ratios are abnormally low, generally around one or even lower. It is due in large part to contribution from resuspended dust particles. While ^{210}Pb in dust particles retains the memory of ~32 years (i.e., the mean life of ^{210}Pb), ^{7}Be retains only for 77 days and hence, the resuspension of "aged" particles will result in low $^{7}Be/^{210}Pb$ ratios in the fallout. Besides the deposition of dust particles, ^{7}Be and ^{210}Pb are also removed from the ambient air by dry scavenging and precipitation. As mentioned earlier, although dry precipitation is less effective compared with wet precipitation in scavenging aerosols and airborne particles, it removes higher proportions of ^{210}Pb relative to ^{7}Be. Thus preferential removal of ^{210}Pb from ^{210}Pb-laden continental air masses by dry precipitation would also result in low $^{7}Be/^{210}Pb$ ratios in the fallout.

3.1.5. High-Frequency Oscillations

[21] Superimposed on the annual cycle of climatological phenomena are transient disturbances due to rapid changes of weather conditions such as the invasion of the Pacific high in the winter. In northern Taiwan, there is about one frontal passage per week in winter and spring. Between two frontal passages, high-pressure fair weather usually prevails, which is characterized by weak surface winds and a clear diurnal cycle in surface temperature [*Lin et al.*, 2004]. Under these drier and warmer weather conditions, not only are the fluxes of ^{7}Be and ^{210}Pb reduced sharply, the $^{7}Be/^{210}Pb$ ratios are exceedingly low due to stable atmospheric condition and the dominance of dry fallout whereby ^{210}Pb is preferentially removed over ^{7}Be.

3.1.6. Interannual Variability and Unusual Events

[22] Besides the seasonality and intraseasonal oscillations embedded in the annual cycle, interannual variabilities are also revealed in the Nankang time series. This is not unexpected considering that two ENSO cycles happened during 1996–2005, including the strongest El Niño of the 20th century during 1997–1998 [*Su et al.*, 2003]. It may have some bearing on the striking imbalance in water supply Taiwan experienced in these years, with devastating floods during 1998–2001 followed by severe droughts during 2002–2003.

[23] As part of the climatic change, annual rainfall varied by a factor of 3.8 (from 1431 mm in 2003 to 5420 mm in 2000) in the time series. During the same period, annual deposition of ^{7}Be and ^{210}Pb differed by a factor of ~3 between years of high and low fluxes. It is important to point out that the correlation between the fluxes of fallout nuclides and rainfall gradually deteriorates as the timescale increases (from weekly to yearly). This probably suggests the influence of other yet unknown factors or processes operating at lower frequencies. A case in point is the unusually high ^{7}Be and ^{210}Pb fluxes measured in 2005, which is a year of moderate rainfall. Could it be that enhanced stratospheric-to-troposphere exchange occurred in 2005? Evidence for this speculation remains to be sought.

3.2. A Comparison of the Time Series Between Nankang and Yangminshan

[24] Although NK and YMS are only ~20 km apart, their altitudes differ by more than 700 m. Furthermore, the YMS site is at the windward side of the Chi-Shin Mountain overlooking a valley opening in the northeast direction (Figure 1). Such a terrain makes the YMS site frequently shrouded in cloud mists due to the passage of fronts, especially during the northeast monsoon season. Thus a comparison of the time series between these two localities allows us to assess the effects of topography and clouds on the deposition of ^{7}Be and ^{210}Pb in northern Taiwan.

[25] Figure 6 shows that, during the period from September 2003 to October 2005, the time series of ^{7}Be and ^{210}Pb fluxes and $^{7}Be/^{210}Pb$ ratio at NK are clearly in phase with those at YMS, reflecting that these two sites are in the same meteorological regime. However, the amount of rainfall at YMS is about twice of that at NK during this period. Meanwhile, cumulative fluxes of ^{7}Be and ^{210}Pb at YMS are ~5 and ~4 times, respectively, of those at NK. Thus concentrations of ^{7}Be and ^{210}Pb in YMS rainwater are higher than those in NK rainwater by a factor of about two or more, suggesting more effective scavenging of fallout nuclides at YMS.

[26] In addition to this work, we have also made other ancillary measurements at YMS, with a goal to better understand aerosol transport and scavenging processes. These experiments entailed (1) measuring ^{7}Be and ^{210}Pb in cloud water and comparing their respective differences in cloud water and rainwater [*Su and Huh*, 2006] and (2) measuring the distribution of natural and anthropogenic fallout nuclides in soils at YMS and comparing observed inventories in soils with inventories expected from overhead precipitation [*Huh and Su*, 2004]. It's found that ^{7}Be and ^{210}Pb concentrations are higher in cloud water than in concurrent rainwater samples. Thus the higher fluxes of ^{7}Be and ^{210}Pb measured at YMS can be attributed to the dominance of in-cloud scavenging (rainout) in an environment often immersed in clouds. In comparison, below-cloud scavenging (washout) carries more weight at NK where thermal convection in the Taipei Basin often causes localized, abrupt afternoon rainshowers. This type of rain, like typhoon rain, can also dilute ^{7}Be and ^{210}Pb concentrations in wet precipitation.

[27] As for the inventories of fallout nuclides in soils of YMS, it is found that the observed values are much higher than expected from overhead precipitation alone [*Huh and Su*, 2004]. Thus by integrating our rainwater, cloud water and soil studies, we have assembled data to support our previous conjecture that low-lying clouds play an important role in transporting fallout nuclides (as well as pollutants) directly into soils in mountainous areas.

4. Summary and Conclusions

[28] Atmospheric fluxes of ^{7}Be and ^{210}Pb have been monitored at NK and YMS for more than 9 years and 2 years, respectively, in order to study the factors/processes governing spatial and temporal variations of their deposition. Our main findings are summarized below.

[29] 1. As observed in numerous previous studies, depositional fluxes of ^{7}Be and ^{210}Pb correlate well with each other, indicating that, despite their different source functions, these two nuclides are removed from the atmosphere by the same mechanisms. Although both ^{7}Be are ^{210}Pb are scavenged mainly by wet precipitation, the correlation of

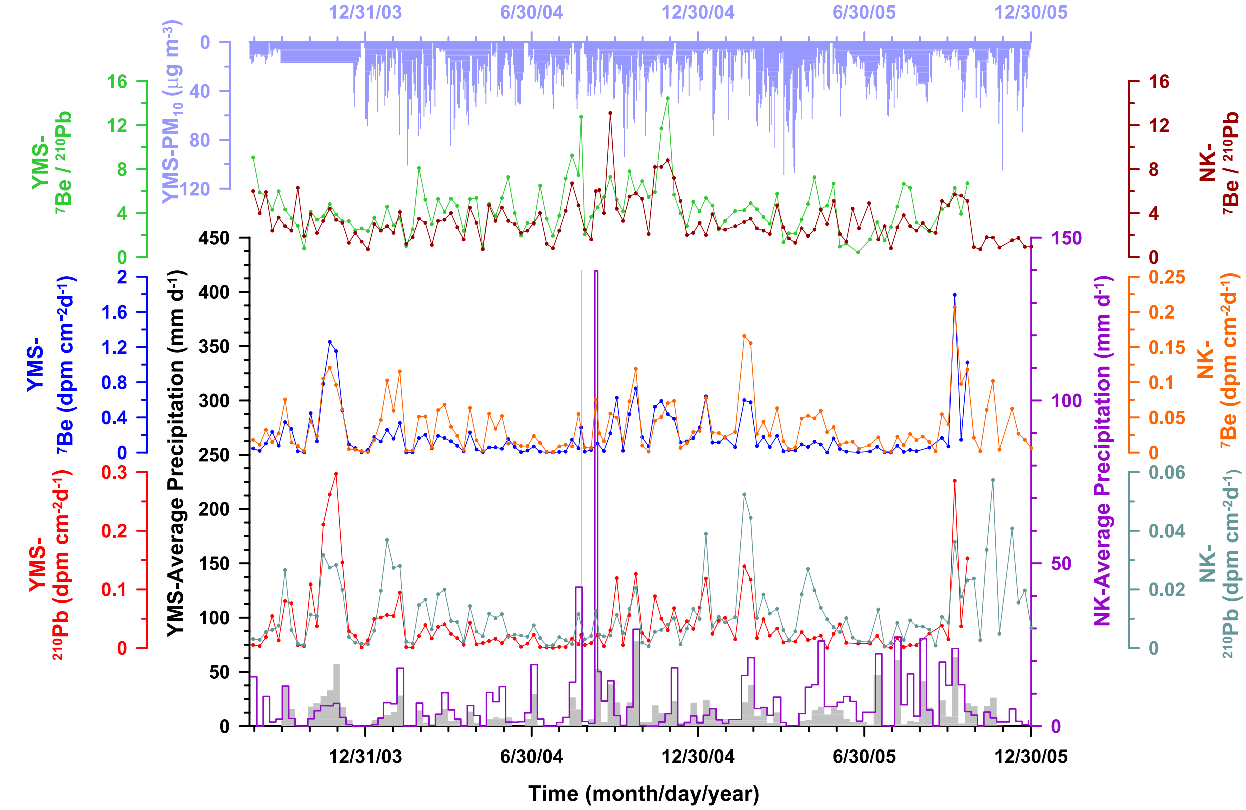

Figure 6. Times series of mean daily rainfall, fluxes of ^{7}Be and ^{210}Pb, and ^{7}Be/^{210}Pb and PM_{10} at YMS during 2003–2005 plotted together with the same data at NK for comparison. Note the scale difference between the two data sets.

^{7}Be with rainfall is better than that of ^{210}Pb, suggesting that ^{210}Pb has a larger fraction of dry to total deposition.

[30] 2. Much of the wet precipitation at NK is caused by monsoonal, typhoon, and mei-yü rains. Together, they contribute to ~80% of the ^{7}Be flux and ~75% of the ^{210}Pb flux, with dry fallout and other types of wet precipitation accounting for the remainder of the fluxes.

[31] 3. Fluxes of ^{7}Be and ^{210}Pb in northern Taiwan show seasonality and follow the annual rainfall cycle regulated by typhoons, monsoons and mei-yü. Embedded in the annual cycle are intraseasonal oscillations due to the passages/incursions of fronts, cold surges, dust storms, and the Pacific high. Over longer timescales, the effect of multiyear oscillation, such as ENSO, is also seen, which can cause large interannual variations in rainfall and nuclide fluxes. The results suggest that when ^{7}Be and ^{210}Pb are used as chronometers or tracers in environmental applications, the assumption of constant flux should be made with caution.

[32] 4. The ^{7}Be/^{210}Pb activity ratio could be taken as a diagnostic indicator of the source regions and altitude of air masses. For instance, rainwater samples derived from high-altitude and maritime air masses are characterized by higher ratios. On the other hand, wet precipitation from low altitude and dry precipitation are expected to carry lower ratios. However, this ratio should be used judicially; it may become indistinguishable if an air mass is substantially modified by other air masses.

[33] 5. A comparison of the time series between NK and YMS points to the importance of the orographic effect at mountainous areas. Although NK and YMS are only 20 km apart, mean annual rainfall at YMS is twice of that at NK and measured fluxes of ^{7}Be and ^{210}Pb at YMS are 4–5 times of those at NK. Furthermore, soil inventories of ^{7}Be and ^{210}Pb at YMS are even higher than what can be expected from atmospheric fluxes. Integrating the results from this and other ancillary studies, we conclude that clouds play an important role in transporting fallout nuclides directly into soils in mountainous areas. This finding has important implications to the application of fallout nuclides to soil erosion studies in Taiwan.

[34] **Acknowledgments.** We would like to thank J.-Y. Cheng, J.-Y. Chu, and C.-T. Hsieh for assistance in the laboratory and Charles C. K. Chou of the Research Center for Environmental Change, Academia Sinica, for providing the PM_{10} data. This work is supported by the National Science Council and the Atomic Energy Commission of the Republic of China under grant NSC 94-NU-7-001-002.

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