A comparison of measured ¹³⁷Cs and excess ²¹⁰Pb levels in the cultivated brown and cinnamon soils of the Yimeng Mountain area

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Abstract This contribution analyzes the similarities and differences between the measured activities of ¹³⁷Cs and excess ²¹⁰Pb (²¹⁰Pb_{ex}) in the cultivated brown and cinnamon soils of the Yimeng Mountain area, discusses the influence of soil texture on the measurement of ²¹⁰Pb_{ex}, and presents differences between the two types of soils. Fields A and B were selected to represent the fields that contain cultivated brown and cinnamon soils, respectively. From either study field, one site of sectioned core and six bulk cores were collected to measure ¹³⁷Cs levels, ²¹⁰Pb_{ex} levels, and the particle-size composition of soil samples. Three undisturbed soil samples were collected to measure capillary and aeration porosities. The ¹³⁷Cs inventories for the two study fields are very similar. The ¹³⁷Cs is a man-made radionuclide, which means that its measured levels for soils are unaffected by soil texture. In contrast, levels of the naturally occurring ²¹⁰Pb_{ex} in terrestrial surface soils are affected by the emanation of ²²²Rn from the soils. It can be assumed that the coarser the soils, the greater the emanation of ²²²Rn; in addition, the lower the measured ²¹⁰Pb_{ex}, the greater the underestimate of this value. The cultivated brown soils in Field A are coarser than the cultivated cinnamon soils in Field B. As a result, ²²²Rn in Field A will diffuse more easily into the atmosphere than that in Field B. As a consequence, the measured ²¹⁰Pb_{ex} in soils from Field A will diffuse more easily into the atmosphere than that in Field B. As a consequence, the measured ²¹⁰Pb_{ex} in soils from Field A is much lower than the actual value, whereas the value measured for Field B is much closer to the actual value.

Key words ¹³⁷Cs; ²¹⁰Pb_{ex}; brown soil; cinnamon soil; Yimeng Mountain area

1 Introduction

The ¹³⁷Cs and ²¹⁰Pb_{ex} techniques have been used to quantify the rates of erosion and deposition of soils (Walling and Quine, 1990; Walling et al., 1995; Zhang et al., 1990, 2006, 2008; Yan et al., 2002; Golosov, 2003; Yang et al., 2006; Onda et al., 2007; Li et al., 2009, 2012; Gaspar et al., 2013), as well as to reveal the history of sediment deposition in lakes, estuaries, and oceans (DeLaune et al., 1978; He and Walling, 1996; Ritchie et al., 2004; Zhang et al., 2007; Ahn et al., 2010; Olid et al., 2013). The artificial radionuclide ¹³⁷Cs (half-life 30.2 a) has been released into the environment as a result of atmospheric testing of thermo-nuclear weapons. This occurred primarily during the 1950s–1970s, with the maximum rate of ¹³⁷Cs fallout from the atmosphere in 1963. The ²¹⁰Pb (half-life 22.26 a) radionuclide is a natural product of the ²³⁸U decay series derived from the decay of gaseous ²²²Rn (half-life 3.8 d), the daughter of ²²⁶Ra (half-life 1622 a). A fraction of the ²²²Rn atoms produced by the decay of ²²⁶Ra in soils and rocks diffuses upwards into the atmosphere, where it decays through a series of short-life radionuclides to ²¹⁰Pb. Fallout



²¹⁰Pb, like ¹³⁷Cs, is removed from the atmosphere by precipitation or dry deposition, and falls onto the land surface or into lakes and oceans (Peirson & Salmon, 1959; Nevissi, 1985; He and Walling, 1997). Unlike the highly variable rates of ¹³⁷Cs deposition, the natural origin of ²¹⁰Pb_{ex} means that the rates of its deposition have been essentially constant through time (Nozaki et al., 1978). Accordingly, the total ²¹⁰Pb concentrations in surface soils and sediments have shown there are two components: supported ²¹⁰Pb (²¹⁰Pb_{sup}), which is derived from the *in-situ* decay of the parent radionuclide ²²⁶Ra and is in equilibrium with its parent, and unsupported or excess ²¹⁰Pb (²¹⁰Pb_{ex}), which is derived from this atmospheric flux and is not in equilibrium with ²²⁶Ra (Robbins, 1978; Kolker et al., 2009).

Before its application to estimating soil redistribution, the $^{210}Pb_{ex}$ technique was widely used to estimate the ages of lake sediment cores (Koide et al., 1972; Appleby and Oldfield, 1978). The $^{210}Pb_{ex}$ concentrations in the sediments can be determined by subtracting the concentrations of $^{210}Pb_{sup}$ from the measured total ^{210}Pb concentrations (Joshi, 1987), as follows:

$$C_{ex} = C_{tot} - C_{sup} \tag{1}$$

where C_{ex} is the ²¹⁰Pb_{ex} concentration (mBq·g⁻¹), C_{tot} is the total ²¹⁰Pb concentration (mBq·g⁻¹), and C_{sup} is the ²¹⁰Pb_{sup} concentration (mBq·g⁻¹).

Given that sediments are scavenged from the water column and are highly saturated, only a very small fraction of ²²²Rn produced by ²²⁶Ra decay in the sediments will escape into the overlying water column (Appleby and Oldfield, 1992; Du and Walling, 2012). In the sediments, both ²²²Rn and ²¹⁰Pb_{sup} can usually be regarded as being in radioactive equilibrium with the parent radionuclide ²²⁶Ra (Zapata, 2002). Accordingly, experimental determination of the ²¹⁰Pb_{sup} concentrations involves the measurement of *in-situ* levels of ²²⁶Ra, which is measured via its daughters ²¹⁴Pb or ²¹⁴Bi.

However, the case is not true for terrestrial surface soils since some of the ²²²Rn produced by *in-situ* ²²⁶Ra decay is constantly diffused through the soils into the atmosphere. It is this escaping ²²²Rn that decays to ²¹⁰Pb and is the source of ²¹⁰Pb_{ex}. As a consequence, the level of ²¹⁰Pb_{sup} in soils is less than that of ²²⁶Ra, and ²¹⁰Pb_{sup} is never in equilibrium with ²²⁶Ra. Since the ²¹⁰Pb_{sup} activity is calculated from the ²²⁶Ra activity, the levels of ²¹⁰Pb_{sup} measured in the soil samples exceed the actual levels of ²¹⁰Pb_{sup}. Therefore, the measured ²¹⁰Pb_{ex} concentration in the soil samples, as determined using equation (1), will be lower than the actual value. And it can be further assumed that the 222 Rn that emanates from the soils would affect the results of 210 Pb_{ex} measurement.

For different soil types, the proportion of ²²²Rn that emanates from the soils might vary because of differences in physical texture caused by variations in parameters such as particle-size composition, capillary porosity and aeration porosity. Unlike ²¹⁰Pb_{ex}, ¹³⁷Cs is a man-made radionuclide without any natural derivation; accordingly, its measured levels in soils are unaffected by soil texture. Previous studies focused on the emanation of ²²²Rn and determined the emanation coefficients of different types of soils and rocks (Sakoda et al., 2010; Nabil et al., 2011; Künze et al., 2013). Du and Walling (2012) corrected the ²¹⁰Pb_{ex} measurements of sediments on river floodplains by employing the ²²²Rn emanation coefficient. However, few studies on soil erosion have dealt with the influence of ²²²Rn emanation on ²¹⁰Pb_{ex} measurements in soils with different textures.

This study examines the similarities and differences between the measured activities of 137 Cs and 210 Pb_{ex} in the cultivated brown and cinnamon soils of the Yimeng Mountain area, discusses the influences of soil texture on measurements of 210 Pb_{ex}, and presents differences between the two types of soils. The findings are relevant to efforts to use the 210 Pb_{ex} technique to estimate the rates of soil redistribution.

2 Material and methods

2.1 Study area

The Yimeng Mountain area (ca. 17180 km²) is located in the central south of Shandong Province, China (34°22' to 36°13'N, 117°24' to 119°11'E). Elevations range from 150 to 1165 m, with slopes typically within the range of 2°-30°. Geologically, it is characterized by alternating granite (or granite-gneiss) and limestone. The two main soil groups in the area are brown soils developed from granite (or granite-gneiss) weathering crusts and cinnamon soils developed from limestone weathering crusts. The area belongs to the continental monsoon climate area of the warm temperate zone. The mean annual precipitation is ca. 800-900 mm, approximately 70% of which falls between July and September. The mean annual temperature is ca. 13.6°C, and the population density is ca. 630 person km⁻². Two representative study fields, Fields A and B (one on each side of the Benghe River). were selected to represent the zones with brown soils and cinnamon soils, respectively, with the distance of ca.35 km between the two study fields. Both the fields are situated in flat areas at the top of ploughed fields, where erosion over the past 100 years has been negligible.

2.2 Field sampling and laboratory analysis

From either study field, one site of sectioned core and six bulk cores were collected to measure the levels of ¹³⁷Cs and ²¹⁰Pb_{ex}, and the compositions of particle sizes. The undisturbed soil samples were collected by means of metal cylinders (each 100 cm³ capacity) to determine capillary and aeration porosities in triplicate. To collect the two sectioned soil cores (Cores A and B were collected from Fields A and B, respectively), two earth prisms (25 cm×10 cm×44 cm for Core A, and 20 cm×10 cm×48 cm for Core B) were manually carved out by excavating the surrounding earth. A small spade was then used to section the straight earth prisms into 2-cm intervals from the top downwards. The bulk cores were collected randomly in the study fields surrounding the sites of the sectioned cores, using a manual percussion corer equipped with a 5-cm-diameter steel core tube. For each of the three undisturbed sampling sites, four undisturbed soil samples were collected within the depth range of 0-40 cm (one sample for every 10 cm of soil depth).

Samples of bulk cores and sectioned cores were air-dried, disaggregated, let to pass through a 2-mm sieve and weighed. A representative fraction (ca. 150 g) of each sample was then placed into a plastic Marinelli beaker and sealed with polyvinyl chloride tape for 20 days prior to the time of assay. The levels of 210 Pb_{ex} and 137 Cs concentrations in the samples of bulk cores and sectioned cores were measured by gamma spectrometry, using an ORTEC high-resolution, low-background, low-energy, *n*-type, coaxial germanium detector. The samples were put on the detector and counted for ca. 80000 s, providing a precision of approximately $\pm 5\%$ at the 95% level of confidence for the gamma ray measurements. The 137 Cs

concentrations were measured at 662 keV. The total ²¹⁰Pb concentration of the samples was measured at 46.5 keV. The concentrations of ²²⁶Ra were assayed at 351.9 keV by measuring the levels of its short-life daughter ²¹⁴Pb. The absolute grain-size composition of each bulk core and sectioned core sample was determined by means of a laser particle size analyser (Mastersizer, 2000). The capillary and aeration porosities of the undisturbed soil samples were measured using the saturation method (Netto, 1993; Matko, 2004).

3 Results

3.1 ¹³⁷Cs and ²¹⁰Pb_{ex} activities

The experimentally determined ¹³⁷Cs and ²¹⁰Pb_{ex} profiles for Core A are shown in Fig. 1, and those for Core B are shown in Fig. 2. The 137 Cs and 210 Pb_{ex} concentrations in the two sectioned cores are both relatively uniform at a depth of -20 cm, but then decline rapidly with increasing depth. This is consistent with the average plough depth in the area, indicating that the radionuclides are mixed by tillage within the uppermost depth of ~20 cm. For Core A collected from Field A, the measured inventories of ^{137}Cs and $^{210}Pb_{ex}$ are 995 and 10203 Bq m⁻², respectively. The values for Core B collected from Field B are 1001 and 19282 Bq m⁻². respectively. The measured ¹³⁷Cs inventory for Core A is very close to that for Core B. In contrast, the measured ²¹⁰Pbex inventory for Core B is ca. 89% higher than that for Core A. Moreover, the measured ¹³⁷Cs concentrations in the profiles for Cores A and B are very close to each other at their similar depths; by comparison, nearly all of the measured ²¹⁰Pbex concentrations in the profile for Core B are higher than those at similar depths in the profile for Core A.



Fig. 1. ¹³⁷Cs and ²¹⁰Pbex measurement profiles for Core A.



Fig. 2. ¹³⁷Cs and ²¹⁰Pbex measurement profiles for Core B

The measured inventories of ¹³⁷Cs and ²¹⁰Pbex for the six bulk cores are presented in Table 1 (Field A) and Table 2 (Field B). The measured ¹³⁷Cs inventories for the six bulk cores from Fields A and B are very similar. The mean measured inventories of ¹³⁷Cs for the bulk cores from Fields A and B are 998 and 1012 Bq m^{-2} , respectively, which are close to the measured ¹³⁷Cs inventories for the two sites from which sectioned cores were collected. By comparison, the mean measured inventories of 210 Pb_{ex} for the bulk cores from Fields A and B are 10253 and 19104 Bq m⁻², respectively, which are close to the respective ²¹⁰Pb_{ex} measured inventories for the two sites in Fields A and B from which sectioned cores were collected. Clearly, the measured ²¹⁰Pb_{ex} inventory for any bulk core from Field B is higher than that for any bulk core from Field A, and the mean measured inventory of ²¹⁰Pb_{ex} for the six bulk cores from Field B is ca. 86% higher than the mean value for the six bulk cores from Field A.

Table 1Measured inventories of ¹³⁷Cs and ²¹⁰Pbex for the
six bulk cores from Field A (Bq m⁻²)

	A_1	A_2	A ₃	A ₄	A ₅	A_6
¹³⁷ Cs	969	1105	971	956	1051	935
²¹⁰ Pb _{ex}	8555	10673	9254	9896	11836	11306

Note: A1-A6 represent the six bulk cores from Field A.

Table 2Measured inventories of ¹³⁷Cs and ²¹⁰Pbex for the
six bulk cores from Field B (Bq m⁻²)

	\mathbf{B}_1	B_2	B_3	\mathbf{B}_4	B_5	B_6			
¹³⁷ Cs	941	998	1035	953	1016	1127			
²¹⁰ Pb _{ex}	19921	17875	19119	19965	18974	18772			

Note: B_1 - B_6 represent the six bulk cores from Field B.

The results suggest that the measured 137 Cs inventory in Field A is consistent with that in Field B, whereas the measured 210 Pb_{ex} inventory in Field A is ~47% less than that in Field B.

3.2 Grain-size composition

For comparison, the median grain size profiles for Cores A and B are presented together in Fig. 3. The clay (<0.005 mm) content profiles for Cores A and B are presented together in Fig. 4. The median grain sizes for Core A range from 41 to 59 μ m, with a mean value of 47 μ m. In contrast, the median grain sizes for Core B range from 24 to 40 μ m, with a mean value of 31 μ m. The clay contents for Core A range from 8% to 10%, whereas those for Core B range from 13% to 15%. This indicates that the clay contents for Core A are less than those for Core B. Furthermore, whereas the contents of coarse sand (0.5–2.0 mm) for Core A are within the range of -7% to -12%, and the coarse sand contents for Core B are all close to 0.

Whereas the median grain sizes for the six bulk cores from Field A range from 43 to 61 μ m, with a mean value of 52 μ m, and the median grain sizes for the six bulk cores from Field B range from 26 to 41 μ m, with a mean value of 33 μ m. The clay contents for the six bulk cores from Field A range from 7% to 9%, whereas the clay contents for the six bulk cores from Field B range from 10% to 14%. Furthermore, whereas the coarse sand (0.5–2.0 mm) contents for the six bulk cores from Field A range from -9% to -13%, and those for the six bulk cores from Field B are all close to 0. Moreover, during sample preparation, the samples from Field B nearly completely passed

through the 2-mm sieve, except for some withered roots. In contrast, there were always some pieces of gravel in the samples from Field A that could not pass through the sieve.

The results suggest that the cultivated brown soils in Field A are coarser than the cultivated cinnamon soils in Field B. This is consistent with our field observations.



Fig. 3. Median grain size profiles for Cores A and B.



Fig. 4. Clay (<0.005 mm) content profiles for Cores A and B.

The measured capillary porosities for the soils in Field A range from -0.25 to -0.35 cm³ cm⁻³, and in Field B from -0.33 to -0.40 cm³ cm⁻³. The measured aeration porosities for the soils in Field A range from -0.06 to -0.17 cm³ cm⁻³, whereas the values in Field B range from -0.01 to -0.09 cm³ cm⁻³. The capillary and aeration porosities of the soils in Fields A and B are represented by their mean measured values (Fig. 5).



Fig. 5. Capillary and aeration porosities of the cultivated brown sol (Field A) and cinnamon soils (Field B).

As shown in Fig. 5, the capillary porosities of the soils in Field A are lower than those in Field B, whereas the aeration porosities are higher in Field A than in Field B. The moisture (capillary water) capacity of the soils in Field A is lower than that in Field B, whereas the gas permeability of the soils in Field A is greater than that in Field B, which is also consistent with our field observations.

4 Discussion

Theoretically, the activities of ¹³⁷Cs and ²¹⁰Pb_{ex} in soils are associated with either precipitation or dry deposition, but are independent of soil texture. Given the soil erosion in Fields A and B, and differences in the rates of either precipitation or dry deposition between the two fields are negligible, in theory, the measured inventories of both ¹³⁷Cs and ²¹⁰Pb_{ex} in Field A should be consistent with their respective values determined for Field B. However, the present data show that while the measured ¹³⁷Cs inventory for Field A is in good agreement with that for Field B, the measured ²¹⁰Pb_{ex} inventory for Field A is -47% less than that for Field B.

As mentioned in the Introduction, the measured ¹³⁷Cs levels in soils are unaffected by soil texture, given that ¹³⁷Cs is a man-made radionuclide; therefore, the measured ¹³⁷Cs inventories in Fields A and B are very similar. In contrast, for the naturally occurring ²¹⁰Pb_{ex}, the measurement results are affected by the

²²²Rn that emanates from the soils. If only a very little amount of ²²²Rn has diffused into the atmosphere from the soils, the measured ²²²Rn concentrations will be very close to the actual ²¹⁰Pb_{sup} concentrations; conversely, if more ²²²Rn has diffused out, the measured ²²²Rn concentrations will be proportionally greater than the actual ²¹⁰Pb_{sup} concentrations. In other words, according to equation (1), for soils that can retain substantial levels of ²²²Rn, the measured ²¹⁰Pb_{ex} value will be closer to the actual value. In contrast, for soils that can retain limited amounts of ²²²Rn, the measured ²¹⁰Pb_{ex} value will be even lower than the actual value.

In the study area, it can be considered that because the cultivated brown soils in Field A are coarser than the cultivated cinnamon soils in Field B, the aeration porosity or gas permeability should be higher in Field A than in Field B. In contrast, the capillary porosity or the moisture capacity of the soils in Field A is lower than that in Field B. Given that the aeration porosity or the gas permeability of the soils in Field A is higher than that in Field B, the ²²²Rn in Field A will diffuse more readily into the atmosphere than that in Field B. In addition, given that the capillary porosity or the moisture capacity of the soils in Field A is lower than that in Field B, the moisture (capillary water) contents of the soils in Field B will be higher than those in Field A under similar physiographic conditions (e.g. precipitation, solar radiation, temperature, air humidity, and wind speed). Capillary water can restrict ²²²Rn emanation from soils. Consequently, the capillary water in the soils of Field B is anticipated to limit ²²²Rn emanation to lower levels than that in the soils of Field A. So, by comparing the ²¹⁰Pb_{sup} measurements for the two study fields, the measured value of Field A is much higher than the actual value, whereas the measured value of Field B is much closer to the actual value. Consequently, by comparing the ²¹⁰Pb_{ex} measurements for the two study fields, the measured value for Field A is much lower than the actual value, whereas the measured value for Field B is much closer to the actual value. Therefore, the measured ${}^{210}\text{Pb}_{ex}$ inventory in Field A is lower than that in Field B.

Moreover, it is known that soils developed from granites contain higher levels of ²²⁶Ra than those developed from limestones (Yang et al., 2005; Al-Hamarneh & Awadallah, 2009). Consequently, the ²²⁶Ra level is higher in Field A than in Field B. In addition, the proportion of ²²²Rn emanating from Field A is greater than that from Field B, as discussed above. These observations further suggest that, acording to equation (1), the measured ²¹⁰Pb_{ex} level in Field A is less than that in Field B.

It can be assumed that the coarser the soils are,

the lower the measured ²¹⁰Pbex value will be. Accordingly, the measured value will be increasingly less than the actual value. In contrast, the finer the soils, the higher the measured ²¹⁰Pb_{ex} value, and the closer the measured value will be to the actual value. It follows that the empirically determined ²¹⁰Pb_{ex} levels will approach to the actual values as soil moisture contents increase, and that the measured values will be the same as the actual values when the soils or sediments are entirely and constantly covered by the water column. In contrast, the measured ²¹⁰Pb_{ex} value will become increasingly lower than the actual value with decreasing soil moisture content. Moreover, for cultivated soils, the measured ²¹⁰Pbex value of the plough-pan layer is closer to the actual value than that of the plough layer. That is because the soils of the plough layer are annually disturbed by cultivation, which promotes the emanation of ²²²Rn. For undisturbed soils, the measured 210 Pb_{ex} value is closer to the actual value than that of cultivated soils, because more ²²²Rn can be retained in the undisturbed soils than in the cultivated soils. Measurements of ²¹⁰Pb_{ex} approach to the actual value with increasing depth because ²²²Rn is easily retained in the deep-seated soils.

The downward tails in the lower parts of the 137 Cs and 210 Pb_{ex} measurement profiles for Cores A and B might partly reflect the downward movement of the two radionuclides. The 210 Pb_{ex} tails might also reflect the fact that 222 Rn is readily retained in soils at the depth of the tail compared with that at shallower depths, and this may reduce the underestimate of 210 Pb_{ex} measurements.

When the ²¹⁰Pb_{ex} technique is applied to the estimation of soil redistribution, the measurement of the reference inventory would affect the estimation results if differences in soil texture between the study area and the reference site are ignored. For the area analyzed in this study, if the reference site is selected in Field A, the erosion rates of the cinnamon soils will be underestimated, even to such an extent that deposition might be predicted for an eroded site. In contrast, if the reference site is selected in Field B, the erosion rates of the brown soils will be overestimated. In theory, the ²¹⁰Pb_{ex} measurement can be corrected by employing the ²²²Rn emanation coefficient; however, it is difficult to calculate the value of the coefficient because it is influenced by such factors as grain size, capillary porosity, aeration porosity, moisture content, parent material, and soil depth, as discussed above. Therefore, this paper proposes that the reference site needs to be selected in a field with the same soil type as in the study area. In this way, the measurement of ²¹⁰Pbex related to soil texture would not obviously affect the rate estimates of soil redistribution.

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