

Article

Differentiating Potential Suspended Sediment Sources Using Radionuclide Tracers and Soil Organic Matter Analysis in a Headwater Catchment in Chuncheon, South Korea

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Abstract: Identifying potential sources of suspended sediment (SS) in headwater catchments is crucial for water quality management. To differentiate these potential SS sources, we investigated the distribution of two fallout radionuclides (FRNs), ¹³⁷Cs and ²¹⁰Pb_{ex}, using gamma spectrometry along with soil organic matter (SOM) analysis in a headwater catchment with five potential SS sources: cultivated land, non-harvested forest floor, eroded hillslope, harvested forest floor, and stream bank. The ¹³⁷Cs and ²¹⁰Pb_{ex} concentrations and the SOM content were considerably higher in the harvested forest floor materials than in the other four potential SS source materials. FRN concentrations revealed distinct properties according to the type of potential SS sources. Specifically, the combination of FRNs (with the effect of SOM content removed) associated with the mineral fraction and SOM showed distinguishable differences among the potential SS sources, except for no difference between cultivated land and eroded hillslope. Therefore, SOM and FRNs, or their combination, can be effective indices to differentiate or trace potential SS sources on various land use/land cover types within a catchment. Further field tests will allow the tracing techniques that bind FRNs with SOM to contribute to understanding SS transport from non-point sources within a catchment.

Keywords: suspended sediment; radionuclide; soil organic matter; surface soil; tracing technique; non-point source



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1. Introduction

Sediment transport in headwater catchments primarily involves suspended loads consisting of clay, silt, and sand particles suspended in water owing to turbulence, in contrast to bedload, which involves rolling, sliding, and bouncing of larger particles along the streambed [1,2]. For most streams, suspended sediment (SS) consists of fine sand, silt-sized, and clay-sized particles with diameters ≤ 0.2 mm [2]. The suspended load comprises a major portion of the total sediment load in most water systems [3] and significantly influences physical and biological processes within catchments from upstream to downstream [2]. Consequently, changes in land use leading to increased SS

loads or concentrations [4,5] have long been a dominant concern in water quality and aquatic [6,7] management of catchments. Several studies have explained that accelerated SS load input into downstream water can impair drinking water quality by exceeding turbidity guidelines [8,9], cause heavy siltation of fish spawning habitats [10,11], and reduce channel capacity, affecting flood risk [12] and inundating offshore corals [13]. Thus, understanding the links between catchment characteristics, including land use and SS dynamics, is essential for effective water quality management at the catchment scale.

63.1% of the total land area in South Korea is covered by steep-slope forests [14], and headwater streams (i.e., first- to third-order streams) account for 88.9% of the total river channel length nationwide [15]. Gomi et al. [2] reported that the SS dynamics in headwater streams play a significant role in water quality management. Despite the importance of freshwater supply, highland agriculture, which may have induced increases in SS, has concentrated in headwater streams [16,17]. Additionally, the amount of SS originating from non-point sources in highland agriculture or mountainous areas is discharged during the East Asian summer monsoon, which accounts for 60–70% of the annual precipitation in South Korea [18,19]. For these reasons, South Korea has been classified as a nation with very high water erosion vulnerability [20,21]. Moreover, intensive highland agriculture on steep slopes can cause severe soil erosion, resulting in downstream water quality impairment [22–24]. However, water resource management policies for turbidity reduction are concentrated downstream of the catchments [25]. To protect and manage water quality, we must provide information on SS originating in non-point sources within a catchment.

Information on SS sources is essential for interpreting the SS dynamics and constructing a sediment budget in a catchment [26,27]. Walling [28] noted that identifying SS sources is a key parameter for designing effective sediment management and control strategies. According to several studies [2,29,30], SS in streams is derived from sources both external (e.g., bank erosion, mass movement, roads and trails, and surface erosion on slopes) and internal (e.g., storage within the channel system from perennial and ephemeral reaches) to the stream channel. Gomi et al. [2] and Collins et al. [30] also explained that source types and spatial distributions differ according to catchment dynamics (i.e., man-made and/or natural disturbance, land use change) in a given catchment.

Techniques using fallout radionuclides (FRNs) such as caesium-137 (^{137}Cs), lead-210 (^{210}Pb), and beryllium-7 (^7Be) have recently been used to identify SS sources within catchments in many regions [31–34]. In particular, ^{137}Cs and excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) have been more widely used for identifying SS sources [31,35,36] as ^7Be has a very short half-life ($t_{0.5} = 53$ days) compared with ^{137}Cs ($t_{0.5} = 30$ years) and $^{210}\text{Pb}_{\text{ex}}$ ($t_{0.5} = 22$ years) [37]. FRNs can allocate the amount of SS contributed by each source via physical or biogeochemical properties [38,39]. Several researchers have explained that this method can determine soil and sediment redistribution dynamics within agricultural [40,41] and forested [31,36,42] catchments. Moreover, FRNs have been used to complement conventional methods for assessing SS movement and tracing SS sources within catchments [31,32,35]. The FRNs of SS sources can vary among catchments depending on a number of spatial characteristics, such as land use and management practices, geology of the parent material, and geomorphologic history [43,44]. Owing to these characteristics, research on reliable tools for evaluating dominant SS sources has led to the development of nuclear techniques based on FRNs [45].

Understanding the distribution and properties of FRNs is necessary for the field application of the tracing technique using FRNs. However, several domestic studies have been conducted in only a few geographical areas [36,46,47], and hence, FRN-based tracing techniques have been poorly understood in South Korea. The objectives of this study were (1) to analyze the distribution of FRNs (^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$) concentrations along with soil organic matter (SOM) content in potential SS sources across various land use/land cover types within a small headwater catchment and (2) to differentiate these potential SS sources based on differences in FRN concentration and SOM content.

2. Materials and Methods

2.1. Study Area

The investigated catchment (0.15 km²) is located in the Chuncheon-si of mid-eastern Korea (37°55' N, 127°47' E) (Figure 1). Chuncheon-si, situated in the upper region of both the capital Seoul and the Han River, holds important multiregional water sources [16], including the Lake Soyang, Gongjicheon, and Uiam dam watersheds. Certain areas within Chuncheon-si contribute a significant amount of SS to the Lake Soyang, Gongjicheon, and Uiam dam watersheds (Figure 2a,b) [16,48,49]. The continuous expansion of agricultural activities in upstream areas of the Lake Soyang watershed has led to sediment and nutrient export from highland agricultural fields, causing serious water quality problems for Lake Soyang [50]. Hence, the management of headwater catchment, including various non-point sources, has been recognized as an important regional task to protect water quality and the aquatic ecosystem of a lowland river [50].

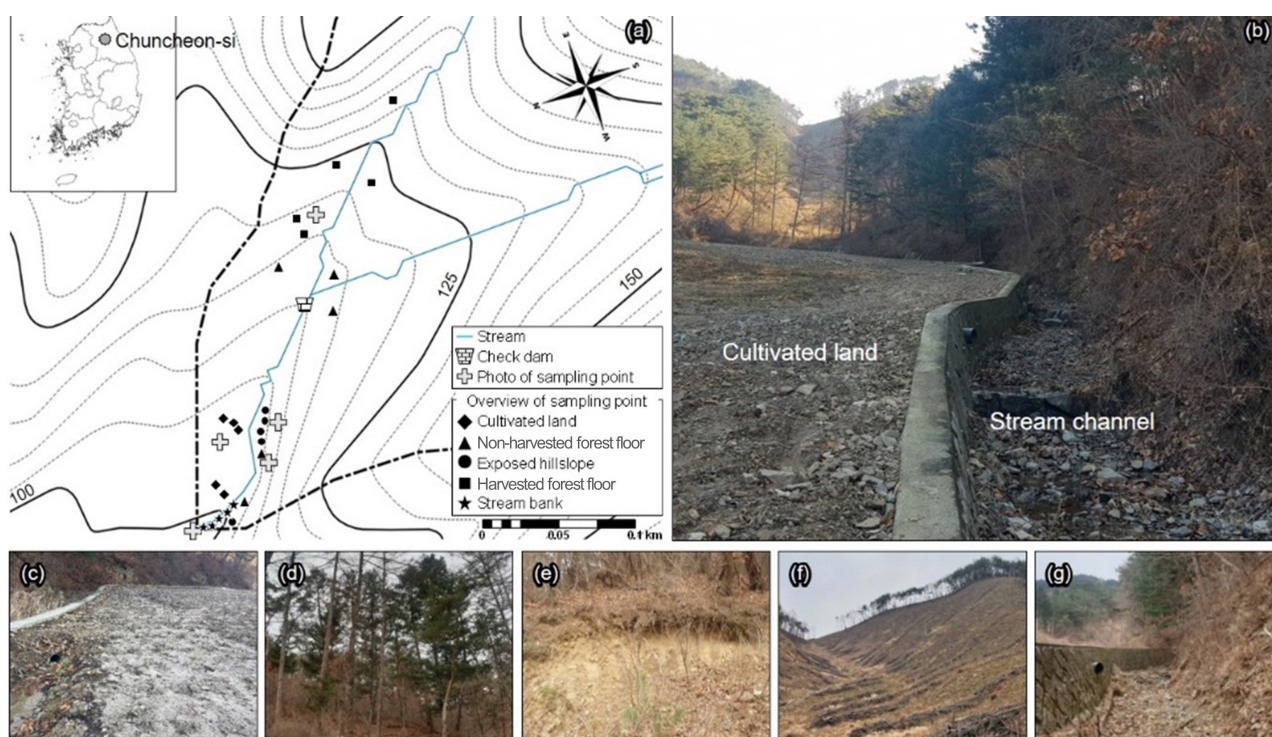


Figure 1. Location and field photos of the study site. (a) Location map with sampling points and (b) upstream view from the outlet. Potential sources of suspended sediment (SS) at this study site: (c) cultivated land, (d) non-harvested forest floor, (e) exposed hillslope, (f) harvested forest floor, and (g) stream bank. Photos in (c–g) also show overview of sampling points for potential sources of SS.

The Chuncheon climate station (Automated Synoptic Observing System, ASOS) recorded an average annual precipitation of 1367.3 mm with a standard deviation of 330.0 mm from 2001 to 2020, with 63.1% concentrated during the East Asian summer monsoon. The mean annual temperature is 11.6 °C with a standard deviation of 0.5 °C. The catchment ranges from 100.0 to 147.5 m above sea level, and the underlying geology consists of garnet–biotite gneiss. The stream channels are 0.3 km long and 3.2 m wide and have a 0.15 m/m slope (Figure 1a,b). The catchment is occupied by forests (64.7%), harvested areas (34.3%), and cultivated land (1.0%) (Figure 1c–g). The partial harvesting was conducted in 2017 (Figure 1f). Of the forest area, 68.7% is covered by coniferous trees (*Pinus densiflora*, *Pinus koraiensis*, *Pinus parviflora*, *Pinus pumila*, *Pinus strobus*, *Larix gmelinii*, and *Larix kaempferi*), 4.1% by deciduous broad-leaved trees (*Quercus variabilis*), and 27.2% by mixed forest.

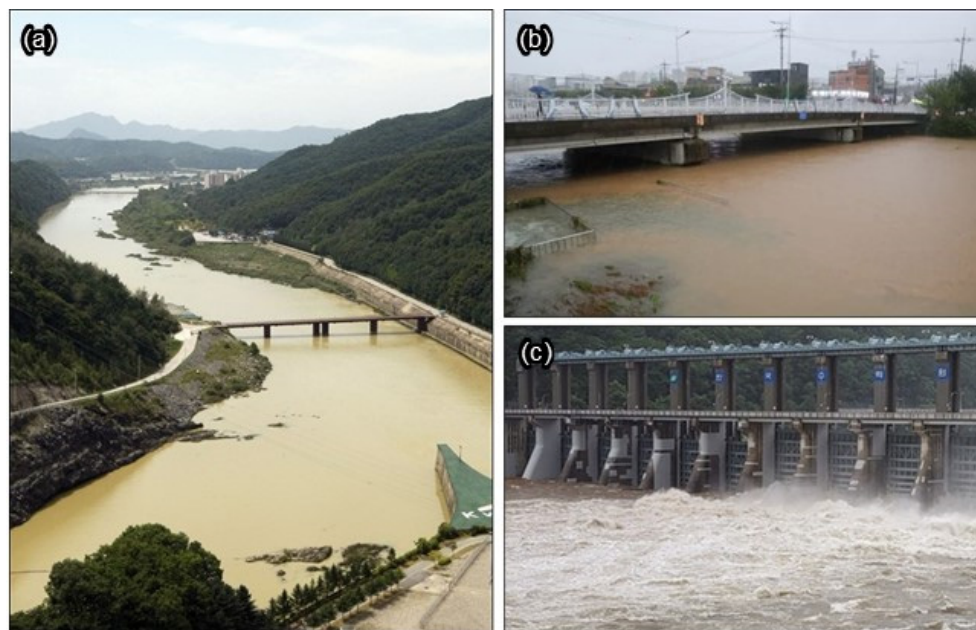


Figure 2. Views of turbid water occurrence due to suspended sediment transport during East Asian summer monsoon in (a) Lake Soyang [51], (b) Gongjicheon [52], and (c) Uiam dam [53] watersheds in Chuncheon-si, South Korea.

2.2. Soil Sampling and Pretreatment

Soil sampling was conducted to obtain representative samples from each of five potential SS sources (i.e., five samples for each SS source type) in the catchment. Using a soil core sampler (stainless steel; 5 cm diameter; 5 cm length), the surface material (<5 cm depth) was sampled from the cultivated land ($n = 5$), non-harvested forest floor ($n = 5$), eroded hillslope ($n = 5$), harvested forest floor ($n = 5$), and stream bank ($n = 5$) over two days on 26 November and 8 December 2020. The depth of nuclide penetration in cultivated soil can be assumed to be equal to the depth of plowing (for example, 20 cm) [54]. In other words, the tillage of cultivated land is assumed to mix soil uniformly to a certain depth, resulting in a uniform distribution of radioactive nuclides throughout the soil. The soil in the cultivated land of the study site is tilled once a year, such as with rotary tillage to a depth of 0–12 cm or deep rotary tillage to a depth of 0–25 cm. However, the sampling was conducted before tilling, assuming that most of the radioactivity is concentrated near the surface during plant growth [55], so the sampling depth of the cultivated soil was set to the surface.

The collected soil samples were oven-dried at 105 °C for 24 h, weighed, disaggregated, and passed through a 2 mm open sieve by referring to the procedures of Mizugaki et al. [31] and Anh et al. [56]. Each soil sample (<2 mm) was placed in a polycon container (5 cm diameter; 2.5 cm length) to determine the FRNs (i.e., ^{137}Cs , and $^{210}\text{Pb}_{\text{ex}}$) in this study.

2.3. Radiometric Analysis for FRNs

The ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ contents were analyzed using gamma spectrometry. The $^{210}\text{Pb}_{\text{ex}}$ activities were determined from the difference between the total and supported ^{210}Pb activities as follows [57,58]:

$$^{210}\text{Pb}_{\text{ex}} = ^{210}\text{Pb}_{\text{total}} - ^{214}\text{Pb} \quad (1)$$

where $^{210}\text{Pb}_{\text{ex}}$ represents unsupported (excess) lead-210 ($^{210}\text{Pb}_{\text{ex}}$), which is determined from the difference between the total ($^{210}\text{Pb}_{\text{total}}$) and supported ^{210}Pb activities. Assuming equilibrium between the ^{226}Ra and ^{222}Rn in the soil samples, the supported ^{210}Pb activity can be calculated from the activity of its short-lived daughter product ^{214}Pb (half-life of 27 min), which is derived from ^{222}Rn in the ^{238}U decay series [31,57,58].

For FRN analysis, the gamma activity of each sediment sample was measured for 40,000 s. Gamma-ray emissions at 661.6 keV (^{137}Cs), 46.5 keV (^{210}Pb), and 351.9 keV (^{214}Pb) energies were measured using a Broad Energy High-purity Germanium detector system (BEGe) (BE3825, Canberra, GA, USA), coupled to a preamplifier (iPA-SL10, Canberra, GA, USA) (PSC822, Canberra-Eurisys, GA, USA), and a multichannel analyzer (DSA-LX, Canberra, GA, USA). Energy and efficiency calibrations of the detector were performed using standard and background samples.

It is known that FRNs can be adsorbed by both mineral and organic fractions of surface soil [31,59,60]. In this context, FRNs may be associated with potential source materials depending on SOM. Therefore, we calculated FRN activity associated with the mineral fraction (C_m) to remove the effect of organic matter on ^{137}Cs or $^{210}\text{Pb}_{\text{ex}}$, according to Mizugaki et al. [31]:

$$C_m = \frac{C_{\text{total}} - C_{\text{om}}\text{SOM}}{1 - \text{SOM}} \quad (2)$$

where C_{total} represents the total FRN activity concentration associated with the material, and C_{om} is the FRN activity concentration associated with the organic fraction, following the procedure of Mizugaki et al. [31]. SOM is the organic matter of the potential SS source material in cultivated land, non-harvested forest floor, eroded hillslope, harvested forest floor, and stream bank. The C_{om} values for FRN activity were not detectable and were assumed to be zero for all source materials as in previous studies [31,36].

2.4. SOM Analysis

Dried soil samples (<2 mm) were analyzed for soil organic carbon (SOC) using an elemental analyzer (Vario MAX CN; Elementar Analysensysteme, Langenselbold, Germany). Subsequently, SOM was calculated by multiplying the percentage of SOC by 1.724 [61].

2.5. Statistical Analysis

All data were analyzed using a non-parametric equivalent of one-way analysis of variance (ANOVA) (Kruskal–Wallis test) for comparisons, with significance values adjusted by multiple tests for pairwise post hoc analysis. For some of the variables, the correlation between two variables was analyzed using the Spearman coefficient (r). All statistical analyses were performed using Statistical Package for Social Sciences (SPSS) version 24 (IBM SPSS Statistics, IBM Corporation, New York, NY, USA).

3. Results and Discussion

3.1. Characteristics of FRNs and SOM for Potential SS Sources

Table 1 illustrates the SOM values and FRN characteristics for each sample from potential SS sources (cultivated land, non-harvested forest floor, eroded hillslope, harvested forest floor, and stream bank). SOM (%) ranged from 0.69 to 1.61 for cultivated land, from 2.89 to 8.62 for non-harvested floor, from 0.48 to 2.98 for eroded hillslope, from 5.96 to 11.09 for harvested forest floor, and from 2.75 to 5.05 for stream bank. In terms of FRN concentration, ^{137}Cs (Bq/kg) ranged from 1.8 to 5.8 for cultivated land, from 8.6 to 44.0 for non-harvested floor, from 0.7 to 3.6 for eroded hillslope, from 16.8 to 89.3 for harvested forest floor, and from 1.5 to 22.3 for stream bank. $^{210}\text{Pb}_{\text{ex}}$ (Bq/kg) ranged from 168.4 to 221.2 for cultivated land, from 222.9 to 741.4 for non-harvested floor, from 58.8 to 165.0 for eroded hillslope, from 659.9 to 1464.3 for harvested forest floor, and from 154.3 to 591.4 for stream bank.

Among the potential SS sources, the mean value of SOM was highest in the harvested forest floor and lowest in cultivated land (Figure 3). The mean value of FRN concentration was highest in the harvested forest floor and lowest in the eroded hillslope (Figures 3 and 4a). In particular, the difference between harvested forest floor and cultivated land or eroded hillslope for SOM (Figure 3a) and the difference between harvested forest floor or non-harvested forest floor and eroded hillslopes were significant for FRNs (Figure 3b,c) according to the statistical analysis ($p < 0.05$).

Table 1. Summary of sampled soil and FRN characteristics in potential SS sources in this study.

	No.	SOC (%)	SOM (%)	^{137}Cs (Bq/kg)	$^{210}\text{Pb}_{\text{ex}}$ (Bq/kg)
C_l	1	0.64	1.11	2.6	221.2
	2	0.40	0.69	2.7	168.4
	3	0.48	0.84	3.7	207.3
	4	0.74	1.28	1.8	219.3
	5	0.93	1.61	5.8	189.8
N_f	1	5.00	8.62	8.6	494.5
	2	4.15	7.16	42.0	741.4
	3	2.81	4.84	44.0	593.3
	4	1.67	2.89	17.3	390.0
	5	3.04	5.24	12.6	222.9
E_h	1	1.73	2.98	3.6	91.0
	2	0.64	1.10	1.4	156.8
	3	0.28	0.48	2.1	165.0
	4	0.34	0.58	0.7	58.8
	5	0.49	0.85	0.9	109.3
H_f	1	5.30	9.14	42.2	1183.1
	2	3.46	5.96	28.4	659.9
	3	6.43	11.09	53.9	1464.3
	4	3.82	6.58	16.8	724.3
	5	6.07	10.47	89.3	1385.1
S_b	1	1.59	2.75	6.0	167.5
	2	2.47	4.26	9.7	283.6
	3	2.16	3.72	1.5	154.3
	4	1.82	3.14	5.6	408.3
	5	2.93	5.05	22.3	591.4

Note: C_l : cultivated land, N_f : non-harvested forest floor, E_h : eroded hillslope, H_f : harvested forest floor, S_b : stream bank, No.: number of samples, SOC: soil organic carbon.

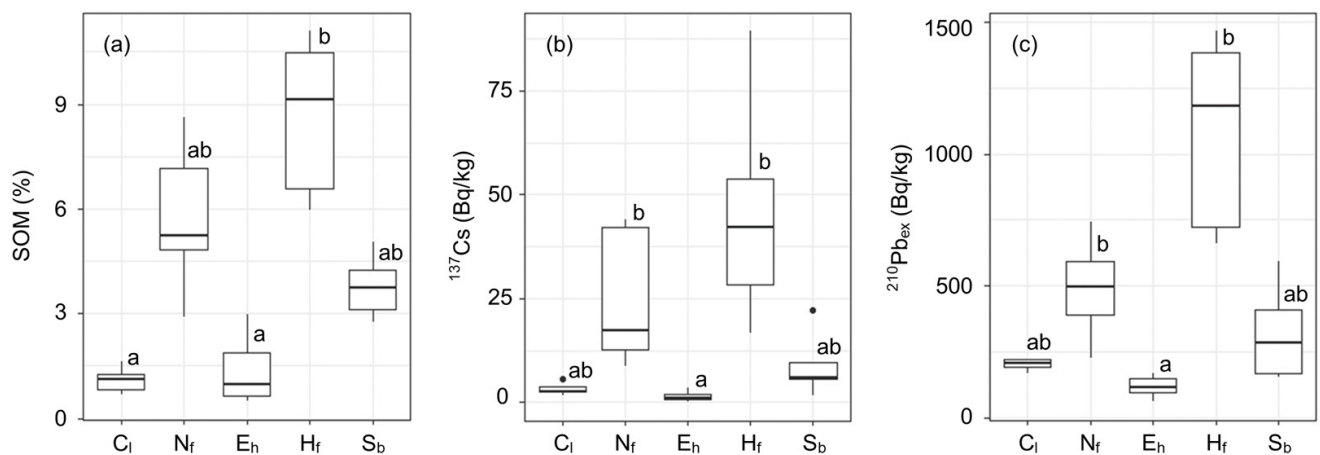


Figure 3. Comparison of differences in (a) SOM, (b) ^{137}Cs , and (c) $^{210}\text{Pb}_{\text{ex}}$ concentrations among the potential SS sources. C_l : cultivated land, N_f : non-harvested forest floor, E_h : eroded hillslope, H_f : harvested forest floor, S_b : stream bank. The upper and lower hinges correspond to the first and third quartiles (the 25th and 75th percentiles). The whiskers extend from the hinges to the highest and lowest values that are within 1.5 (the interquartile range). Values beyond the end of the whiskers are outliers and plotted as points. Different letters indicate statistical significance (Bonferroni's post hoc tests after one-way ANOVA and Mann–Whitney U).

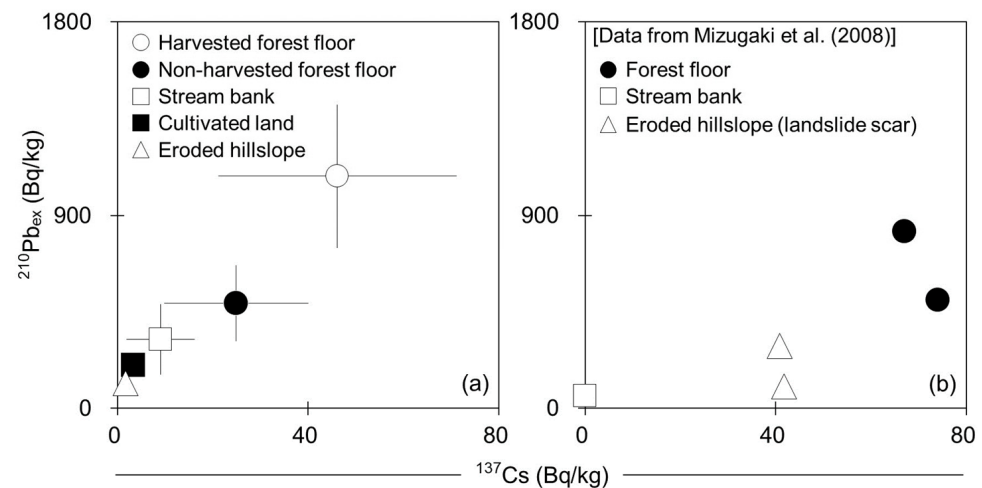


Figure 4. FRN concentrations associated with the potential SS source materials in (a) this study and (b) the previous study [31]. The error bar in (a) indicates the standard deviation. (b) was plotted using the approximate median values calculated by the authors.

The results of this study (Figure 4a) were similar to those of Mizaugaki et al. [31], who found that ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activities were much higher in forest floors than in other potential SS sources, including stream banks and landslide scars (Figure 4b). Figure 5 presents a comparison of the ^{137}Cs values analyzed in this study with the results of other domestic studies. Although there are no reference data on the $^{210}\text{Pb}_{\text{ex}}$ concentration in other domestic studies [36,46], it is expected to exhibit a similar pattern to the ^{137}Cs concentrations of potential SS sources. Harvested and non-harvested forest floors exhibited high concentrations, whereas eroded hillslopes, forest roads, and channel banks exhibited low concentrations owing to subsurface material exposure on the surface areas, as observed in the Woldong catchment [36] and Yeongsan River [46] (Figure 5).

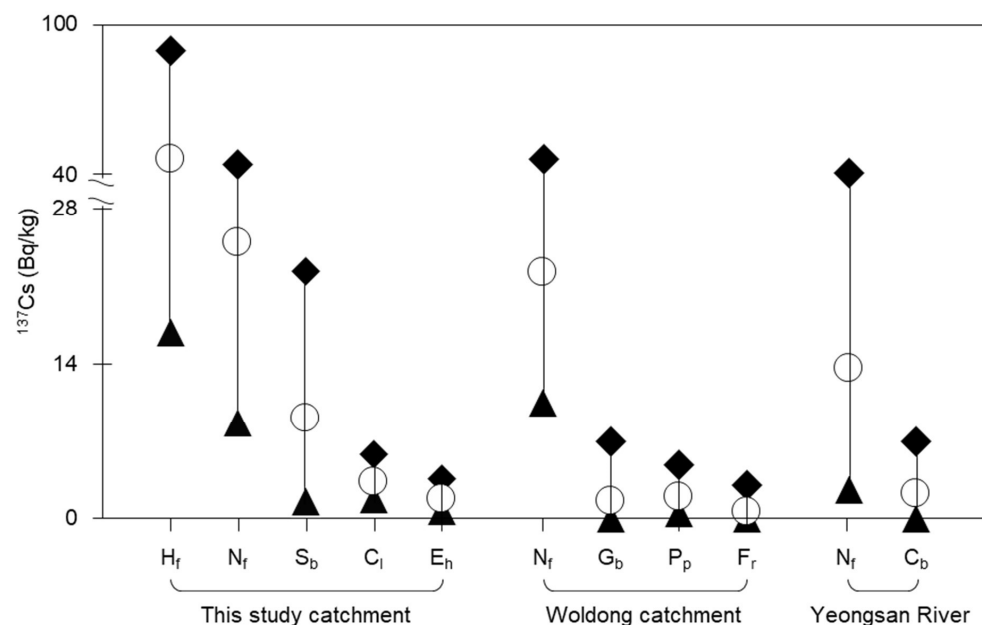


Figure 5. Maximum ($\blacklozenge$), minimum ($\blacktriangle$), and mean ($\circ$) values of ^{137}Cs concentration in the potential SS source materials for difference between this study catchment and both Woldong catchment [36] and Yeongsan River [46]. The sources are H_f (harvested forest floor), N_f (non-harvested forest floor), S_b (stream bank), C_l (cultivated land), E_h (eroded hillslope), G_b (gully bank), P_p (paddy plain), F_r (forest road), and C_b (channel bank).

The FRN concentrations can vary depending on the degree of surface soil disturbance. Kato et al. [62] reported that 80% of deposited FRNs were found in the upper 2 cm of the soil layer. He and Walling [59] reported that if erosion occurs, sediment is transported from surfaces containing higher levels of FRNs. Generally, it has been reported that undisturbed surface soil on the forest floor showed significantly high values of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activities [31,63], but with high variability due to spatial variations in fallout, throughfall, and soil conditions [31,64,65], and erosion–deposition affected by surface runoff [66]. In particular, forest clear-cutting can significantly contribute to the high inputs of FRNs to surface soils due to the decomposition of logging residue (e.g., needles and lichens) [67,68] and increases in continuous FRN fallout caused by decreases in interception loss by the forest canopy [69,70].

Subsurface material exposed to the surface may exhibit significantly low FRN concentrations. Wallbrink and Murray [71] and Mizugaki et al. [31] indicated that the low activities of FRN concentrations observed in the materials of SS sources suggest the loss of surface materials by erosion. These studies support the low FRN concentrations observed in the materials of eroded hillslopes and stream banks in this study (Figures 3 and 4). Here, the reason for higher FRN concentrations in the stream bank than in eroded hillslope can be explained by the difference in vegetation presence (Figure 1e,g). In the study site, the stream bank was partially covered by vegetation and shrubs, which can trap sediment transported from adjacent hillslopes by surface erosion [31]. Therefore, this suggests that the presence of vegetation can influence the redistribution of FRN concentration. Low levels of FRN concentration in cultivated land materials are due to human disturbances such as soil reconditioning [36] and severe erosion of disturbed soil [72].

Additionally, differences in SOM among the potential SS sources can be explained by the reasons mentioned above for FRN concentration since FRNs are adsorbed by soil minerals and organic matter [31,59,60]. For the potential SS source materials in this study (Table 1), SOM also demonstrated a significant positive correlation with ^{137}Cs ($r = 0.84$, $p < 0.001$) and $^{210}\text{Pb}_{\text{ex}}$ ($r = 0.84$, $p < 0.001$), indicating their adsorption.

3.2. Differentiating the Potential SS Sources by Binding FRNs and SOM

As mentioned above, the results of this study show that the FRN concentration was related to SOM content (Table 1, Figure 3), similar to previous studies' findings [35,41]. These indicate that FRN concentrations may be largely dependent on SOM content.

Consequently, after removing the effect of SOM content on FRN concentration, the results for the combination of SOM and ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ associated with C_m are shown in Figure 6. The mean concentration of ^{137}Cs associated with C_m exhibited the highest value in the harvested forest floor (51.0 ± 28.3 Bq/kg) and the lowest value in the eroded hillslope (1.8 ± 1.1 Bq/kg) (Figure 6a). The mean concentration of $^{210}\text{Pb}_{\text{ex}}$ associated with C_m exhibited the highest value in the harvested forest floor (1194.6 ± 389.6 Bq/kg) and lowest value in the eroded hillslope (117.5 ± 40.1 Bq/kg) (Figure 6b). The combination of both FRNs associated with C_m and SOM showed distinguishable differences among the potential SS sources, except for a similar value between cultivated land and eroded hillslope.

Based on the results of this study, we suggest that the combination of FRNs associated with C_m and SOM can be effectively used to differentiate or trace potential SS sources on various land use/land cover types within catchments.

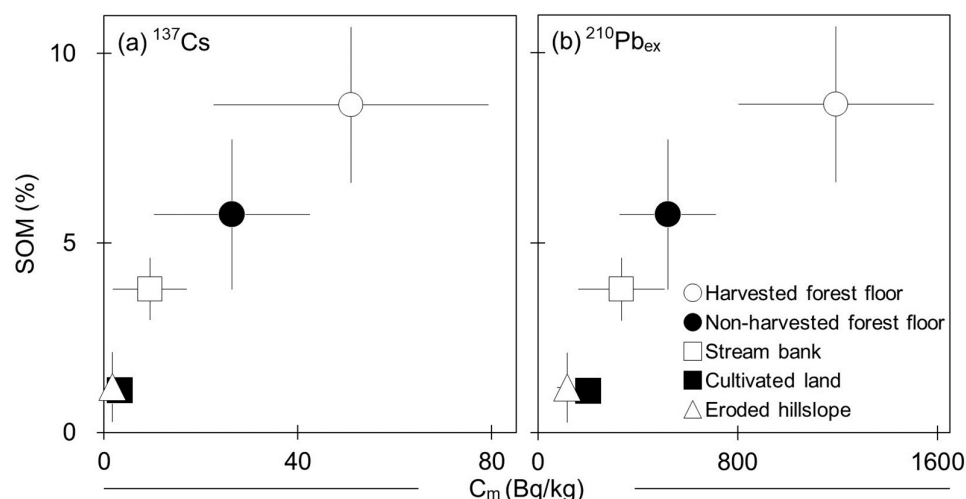


Figure 6. Relationship between fallout radionuclides activity ((a) ^{137}Cs and (b) $^{210}\text{Pb}_{\text{ex}}$) associated with the mineral fraction (C_m) and SOM. The error bar indicates the standard deviation.

4. Summary and Conclusions

This study analyzed two FRNs, ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ concentrations, and SOM content to differentiate potential SS sources within a headwater catchment in Chuncheon, South Korea. Among the five potential SS sources, the harvested forest floor materials exhibited the highest mean values of SOM content and FRN concentrations, whereas the mean value of SOM content and FRNs concentrations was lowest in the cultivated land and eroded slope materials, respectively. However, there were no statistically significant differences in FRN concentrations and SOM content between some of the potential SS sources.

FRN concentrations revealed distinct properties according to the type of potential SS sources. Furthermore, the combination of the FRNs (with the effects of SOM content removed) associated with C_m and SOM also showed distinguishable differences among the potential SS sources, except for the difference between cultivated land and eroded hillslope.

These results suggest that the potential SS sources on various land use/land cover types within a headwater catchment can be differentiated using SOM and FRNs or their combination. In the future, the combination of FRNs and SOM as source tracers should be tested via field monitoring to assess the results of this study and further understand SS dynamics in the catchment.

Author Contributions: Conceptualization, S.N. and S.K.; methodology, S.N., M.K. and S.K.; analysis and investigation, S.N., K.K., M.K. and S.G.; resources and data curation, S.N., K.K., S.J. and J.L.; writing—original draft preparation, S.N.; writing—review and editing, K.K., J.K.K. and S.K.; supervision, J.K.K. and S.K. All authors have read and agreed to the published version of the manuscript.

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