Identification of major suspended sediment sources using X-ray florescence analysis: the Oromushi river basin case of study

Kitami Institute of Technology　Student member　Carlos Beitia
Kitami Institute of Technology　Member　Keisuke Nakayama
Kitami Institute of Technology　Non-Member　Naofumi Ohtsu
Kitami Institute of Technology　Non-Member　Misao Yamane
Kitami Institute of Technology　Non-Member　Shintaro Yamasaki
Kitami Institute of Technology　Student Non-member Saadia Bouragba
Kyoto University　Member　Yasuyuki Maruya
Kitami Institute of Technology　Member　Katsuaki Komai

1. INTRODUCTION

The production of suspended sediments (SS) through erosion and transportation mechanisms is essential to diffuse and distribute nutrients and organic matter throughout the alluvial rivers' basin. These substances, which the stream flows transport from the upstream regions to the downstream regions of the catchment, are essential to ensure the integrity of the riverine ecosystems[1)2)3)4)].

However, overland flow resulting from heavy precipitation events increases SS loads affecting the physical and chemical quality of the stream waters, generating negative impacts such as decreases in fishery and crop productivity, increases in water treatment costs and loss of reservoirs storage capacity[5)6)7)8)9)].

Generally, the sediment production occurs unequally within the catchment and thus only some limited areas yield the major part of the sediments. The identification of these areas, considered as main SS sources, is crucial to understand the sediment production and transportation processes.

In this respect, fingerprinting methods have proven to be useful tools to identify the main SS sources within a catchment when an adequate statistical analysis is conducted[10)11)12)]. This type of method endeavors to identify the origin of sediments found at a sink (assumed as the mixture of all sources) within a group of potential sources located in upstream regions as a function of the sediments properties, namely, physical, chemical and biogenic properties.

As for the statistical component, Bayesian statistic offers a stable statistical framework with high applicability to fingerprinting methods, estimating the proportional contributions of different sources to a target area. For instance, the Bayesian model MixSIAR developed by Moore and Semmens (2008) has been successfully applied in pollutant sourcing, determination of carbon sources for soils and calculation of plant water use from soil horizons[13)]. Therefore, in this study we make use of the Bayesian model MixSIAR to estimate the SS yield from 3 large landuse groupings comprising 18 sub-domains within the Oromushi River basin using the geochemical compositions of surface soil and riverbed samples measured by X-ray fluorescence analysis (XRF).

The results obtained from the MixSIAR model were verified by field experiments conducted in the same study area by Ishida et al., 2012[14)]. Moreover, to identify the main SS sources in terms of the major landuses in the catchment, the particle size distribution (PSD) in samples from each sub-domain was measured through laser diffraction analysis and the results were compared with the PSD obtained in the downstream sample.

2. MEHODS

2.1 SITE DESCRIPTION

The Oromushi River is a branch of the Tokoro River with a total length, basin area and mean slope of 9.7 km, 29.3 km^2 and 1/43, respectively. The downstream end is located at 43 43'N and 143 47'E. The river is characterized by high suspended sediment concentrations (e.g. more than 10,000 mg l^{-1}) during flood events and a mean runoff concentration time of about 2 hours. Two predominant landuses can be distinguished within the whole basin: forests and agricultural fields that represent about 80.7 % and 15.8 % of the total coverage respectively.

2.2 FIELD OBSERVATIONS AND LABORATORY PROCEDURES

Firstly, the Oromushi River basin was divided into 18 sub-domains in function of the geographical information system (GIS) data of the catchment such as landuse, elevation, surface soil, and vegetation type. Then, 3 soil samples of approximately 1125 cm^3 were collected from the soil surface layer (i.e. up to 5 cm). For each sub-domain, the 3 samples were combined into one sample in an attempt to represent the spatial heterogeneity of the soil. Additionally, a riverbed material sample of about 4500 cm^3 was collected at the downstream end of the river basin. Because of the low depth and slow flow conditions in this area, this sample was assumed to be the accumulation of the sediments transported from upstream regions.

In order to avoid the contamination of the specimens all sampling tools were cleaned thoroughly between samples stations. Also, double sealed labeled plastic bags were used to store and transport the samples to the laboratory.

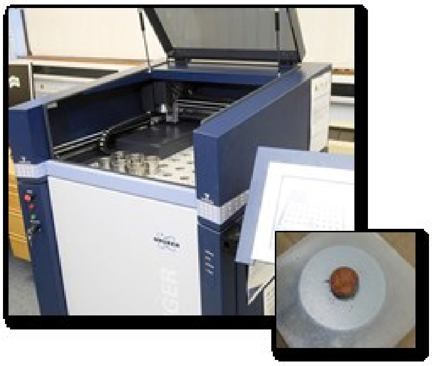

Fig. 1 X-ray fluorescence analyzer and coin-shaped sediment sample

In the laboratory, XRF analysis was performed on samples sieved with distilled water through a 63 μm mesh followed by drying for 24 hours at 105 °C. A 63 μm sieve mesh size was selected in order to focus on fine sediments, which correspond to a grain size between silt and sand. After the sieving, the samples were combusted in a muffle furnace at 750 °C for 1 hour to oxidize compounds and eliminate organic matter. As shown in **Fig. 1**, samples were placed inside 8 mm polyvinyl chloride rings and machine-pressed under 12 tons for 1.5 minutes until a coin-shaped specimen with a smooth surface was obtained. The XRF measurements were conducted using a wavelength dispersive XRF instrument (XRF; S8 Tiger, Bruker AXS) and the concentration of compounds was obtained using the fundamental parameter algorithm installed in the instrument. All the instruments and sample holders used during the analyses were cleaned with ethanol at 99.5 % to avoid inter-sample contamination.

2.3 STATISTICAL ANALYSIS

Principal component analysis (PCA) was applied to identify the most statistically significant compounds within all the compounds detected by the XRF analysis.

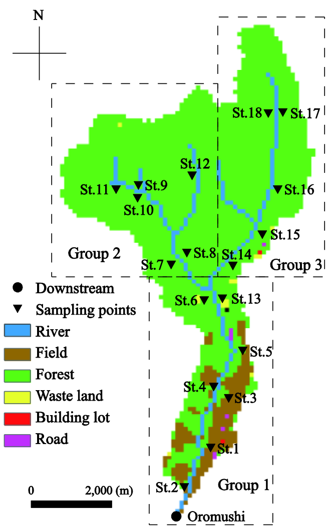

Fig.2 Oromushi river basin land use map, location of the sampling points and groupings with similar area

Based on the PCA results only the most influential compounds were selected estimate the SS contribution from each of the target regions. The outputs of the PCA were used to run the MixSIAR model and the contribution of SS to the downstream end was estimated using the Bayesian equations and parameters of the model. The mean value and standard deviation of the SS contribution were calculated after 10 iterations.

Although it is not possible to include any parameter to consider the effect of SS sources with different areas in the MixSIAR calculations, Ishida et al. (2010) pointed out the importance of considering this effect on the SS yield estimation[15]. Thus, in order to evaluate the SS yielded in different regions of the catchment more accurately the 18 sub-domains were combined into 3 groups with similar areas as shown in **Fig. 2**.

It should be remarked that each group was considered as independent, whereby SS contribution was only to the downstream end and did not influence SS transportation in other areas. Additionally, the SS contributions from the MixSIAR model were verified by field experiments reported by Ishida et al. (2012).

3. RESULTS

3.1 Chemical composition

As expected from natural soil samples, XRF analysis revealed a large number of compounds, from which the PCA identified SiO_2, Al_2O_3, Fe_2O_3, CaO and Na_2O as the main components (See **Fig. 3** and **Table 1**).

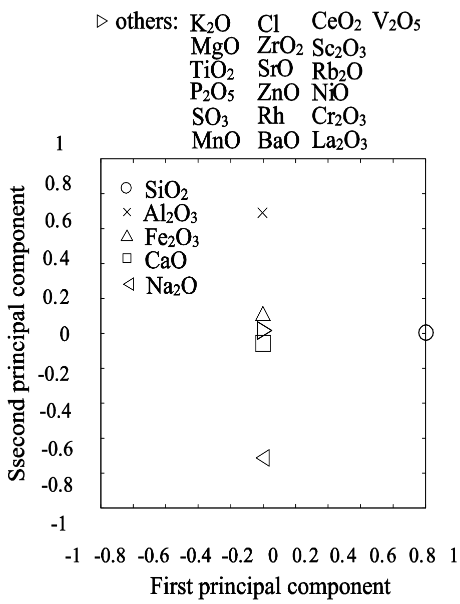

Fig. 3 Principal Component Analysis plot showing the most influential compounds identified by XRF.

The compounds identified were used as SS tracers and the SS contribution from Groups 1, 2 and 3 was estimated. Based on the MixSIAR model the contribution of SS to the downstream end was 38.6 % (±10), 31.1 % (±7.0) and 30.2 % (±11) for

Groups 1 to 3, respectively. Field experiments conducted by Ishida et al. (2012) demonstrated that the contribution for each group was 39.0 %, 34.2 % and 26.8 %, respectively, which is in very good agreement with the contributions obtained from the MixSIAR analysis suggesting that the SS contributions can be accurately estimated by Bayesian methods such as the MixSIAR model (See **Table 2**).

Table 1. Weighted average relative concentration of the most influential compounds for the downstream end and each sub-domain group

Compounds	Downstream (%)	Group 1 (%)	Group 2 (%)	Group 3 (%)
SiO_2	65.2	67.0	61.9	62.2
Al_2O_3	17.7	15.0	15.4	13.5
Fe_2O_3	7.1	6.6	8.0	5.6
CaO	3.0	3.7	5.4	10.7
Na_2O	2.0	2.2	2.4	2.4

3.2 Particle size distribution

The PSD of all sediment samples was measured by laser diffraction analysis. As shown in **Fig. 4,** the results suggest that there is a difference between the predominant grain sizes from the two types of landuses in the catchment (i.e. forest and agricultural fields).

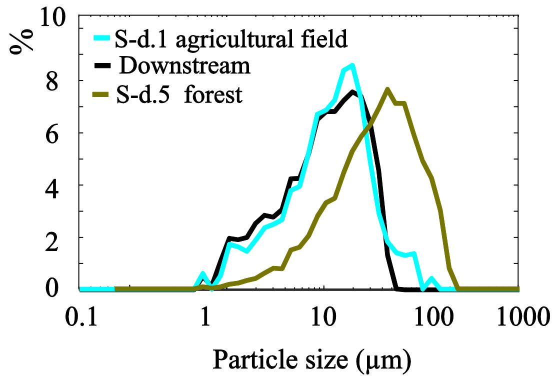

Fig. 4 Particle size distribution for the sediments samples from the downstream end, the forested areas and the agricultural fields.

Sediments from agricultural fields and the downstream region comprise small particles with a predominant diameter of 10 µm, while those collected at forested areas comprise larger particles with a predominant diameter of 60 µm. These results, not only state a difference between regions in function of landuse but also suggest a greater SS contribution from agricultural fields, whose PSD is more similar to the downstream PSD. This suggests a relationship between landuse and the PSD that can be used to differentiate major sediment sources[16)17)18)]. It should be noted that Group 1, with an estimated contribution of 39% of the total SS yield represented 24.07% of the total catchment area from which 34.7% are agricultural fields. PSD analysis results are in good agreement with both the MixSIAR estimations and field experiments based on the chemical composition of the sediments.

A PSD characterize by small particles could be caused by a greater exposure of primary sediment particles and aggregates to weathering forces. This phenomenon is characteristic of agricultural fields and similar areas with poor vegetal cover.

Table 2. Relative average sediment yield estimated by field experiments and the Bayesian model MixSIAR.

	Field experiment estimation (%)	Bayesian model estimation (%)
Group 1	39.0	38.6
Group 2	34.2	31.2
Group 3	26.8	30.2

The high SS yield of Group 1 estimated using both the MixSIAR model and field experiments, along with the similarity of the PSD profiles between the downstream end and the agricultural fields, suggests that in terms of the landuse the agricultural fields are the major source of SS in the Oromushi River basin. Therefore, the application of other analytical techniques, such as X-ray diffraction analysis (XRD) along with the XRF analysis may be useful for making more accurate estimates of the chemical composition of the sediments from both agricultural fields and forests. This will allow more accurate determination of the main sources of SS and clarify the transportation rate from all regions of the catchment to the downstream end.

4. CONCLUSIONS

A fingerprinting method based on the chemical composition of sediment samples was conducted to understand the SS transport from different regions of the Oromushi river basin to the downstream area. XRF was the analytical technique used to assess the chemical composition of the sediments. Among all the compounds identified by XRF analysis the PCA identified: SiO_2, Al_2O_3 and Fe_2O_3 as the main compounds in function of their concentration in the samples.

The MixSIAR model based on Bayesian statistics estimated SS contributions as: 38.6 % (±10), 31.1 % (±7.0) and 30.2 % (±11) for Groups 1, 2 and 3, respectively. These results were confirmed by field experiments that estimated SS contributions of 39.0 %, 34.2 % and 26.8 % for Groups 1, 2 and 3, respectively.

The physical properties of the sediments assessed through the PSD analysis suggest that agricultural fields are likely to be the main source of SS within the catchment which is in good agreement with the field experiments and the MixSIAR model results that identified Group 1 (with 34.7% of agricultural fields) as the main source of SS.

Moreover, these results suggest there is a clear relationship between landuse and the PSD of the surface soil, which indicates the effectiveness of fingerprint methods, especially in a river basin with diverse landuses. Based on the MixSIAR model and the particle size distribution profiles, agricultural fields were revealed to play a greater role in the transportation of SS per unit area than forests.

Therefore, vegetal cover in a river basin may decrease sediment generation and transportation processes by protecting the surface layer of the soil from the action of rainfall kinetic energy and runoff, whilst at the same time preventing landslides and decreasing mudflows into streams. This reinforces the importance of forest conservation and reforestation plans as effective measures to prevent the degradation of water resources and riverine ecosystems.

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