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Influence of particle size on geochemical suspended sediment tracing in Australia

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Abstract Suspended sediment tracing is a potentially useful method to identify the sources of suspended sediment in rivers. Sediment tracing relies on the use of representative and conservative tracers. Particle size is known to influence sediment geochemistry and yet its quantitative impact on sediment source discrimination is little known in Australia and worldwide. This paper investigates the influence of particle size on geochemical sediment tracing in southeastern Australia. The effects of particle size on sediment source discrimination are investigated using a multivariate mixing model. The results suggest geochemistry is a useful tracer for metasediment dominated catchments. Element concentrations are enriched in the finer fraction, but the differences of particle size distributions between sources and mixture are not sufficient to result in differences in source discrimination. The study demonstrates the less than 63 μm fraction to be a representative particle size for geochemical suspended sediment tracing in the lithologically-uniform catchments of Australia.

Key words tracing; fingerprinting; sediment tracing; suspended sediment; particle size; geochemistry; metasediment

INTRODUCTION

Numerous studies using sediment tracing (or fingerprinting) to quantify sediment source contribution have been published over the last few decades (e.g. Murray *et al.*, 1993; Olley *et al.*, 1993; Caitcheon, 1998; Wallbrink *et al.*, 1999; Owens *et al.*, 2000; Walling, 2005; Nagle *et al.*, 2007). Despite the increasing popularity of sediment tracing as a research and management tool, insufficient testing of the underlying assumptions have been reported. Sediment tracing requires tracers to be representative and conservative. However, most tracers are likely to be influenced by the particle size analysed. Therefore, it is not feasible to compare tracers with different particle size distributions for sediment source discrimination (Walling & Moorehead, 1989; Collins *et al.*, 2001).

Several approaches have been used to address the impacts of particle size distribution on suspended sediment tracing. Some studies use a narrower particle size fraction for analysis, such as the less than 4 μm (e.g. Wallbrink *et al.*, 2003) or the less than 10 μm fraction (e.g. Douglas *et al.*, 2005). Such approaches may reduce the impacts of particle size distribution on sediment source discrimination, but are unlikely to be representative of the entire suspended sediment. Particle size correction factors, calculated by the specific surface areas of sources and mixture have also been used to adjust the influence of particle size on tracer properties (Collins *et al.*, 1997; Gruszowski *et al.*, 2003). The assumption of such an approach is a linear relationship between particle size and tracer properties. Many tracer properties tend to linearly enrich finer particle size fractions, but exceptions are also observed (Fredericks, 1994; He & Walling, 1996; Singh *et al.*, 1999; Motha *et al.*, 2002). As a result, this approach is only appropriate if the linear relationships between particle size and tracer properties are tested for all the tracers used in the model.

Although it is known that particle size influences tracer properties, and that different approaches can be used to make adjustments, little is known to quantify the variation in sediment source discrimination due to differences in particle size. Most of the particle size adjustment approaches introduce additional laboratory analyses and uncertainties. If the sediment source discrimination is insensitive to particle size at a certain level of variation in particle size distribution, these additional costs can be avoided.

This study presents an investigation of the influence of particle size on geochemical suspended sediment tracing in two coastal catchments of southeastern Australia. Suspended sediment source discrimination is tested at several junctions within the catchments, using different

particle size fractions. The relationships between the geochemistry and particle size fraction, and particle size distributions and catchment characteristics are analysed. The response of sediment source discrimination to particle size is then discussed.

STUDY AREA

The study area is the Moruya-Deua and Tuross River catchments, located on the south coast of New South Wales, Australia (Fig. 1(a)). The Moruya-Deua River drains a catchment of approx. 1500 km^2. The Tuross River catchment is located south of the Moruya-Deua River catchment, and has a total area of approx. 2200 km^2. Both catchments provide important drinking water supplies for nearby towns. The ecological importance of maintaining good water quality in these catchments is also high due to valuable estuarine and adjacent marine habitats.

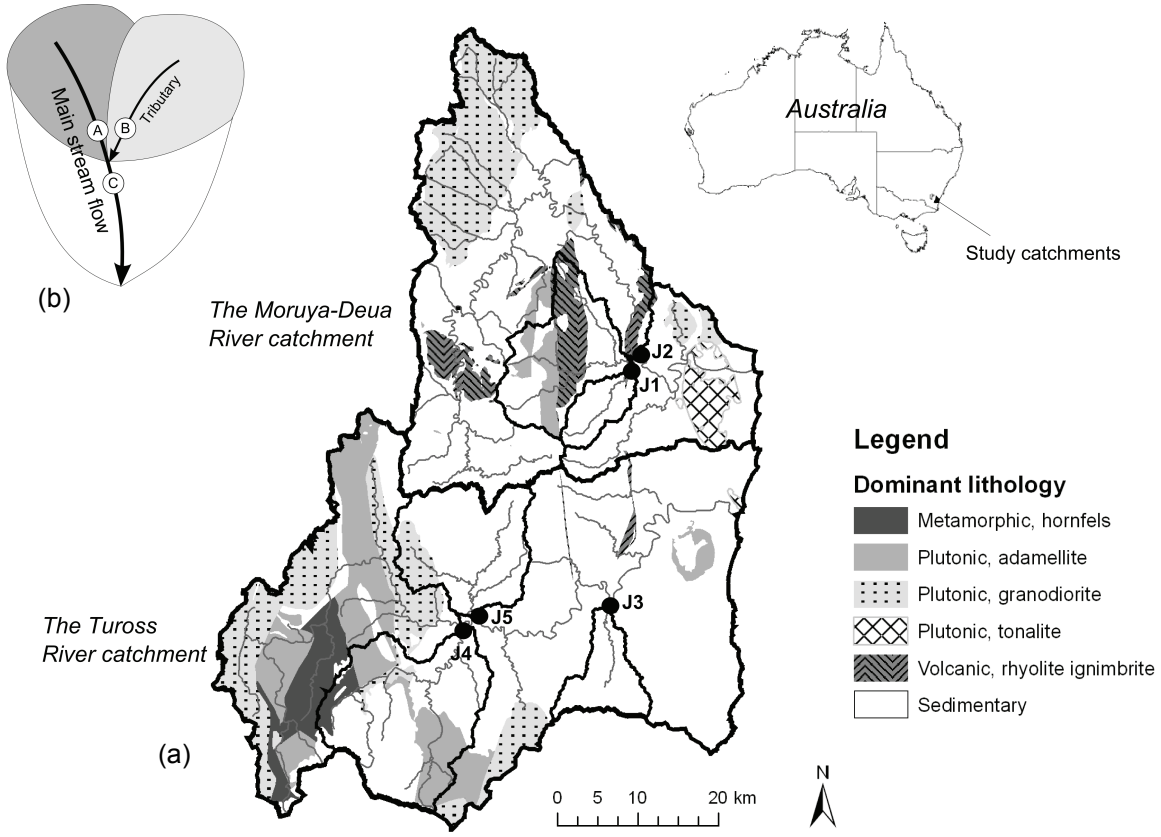

Fig. 1 (a) Catchment location, dominant lithology and the locations of the five sampled junctions J1–5. (b) Diagram showing sampling sites at a hypothetical stream junction. In practice, sites A and B are located approximately 1 km upstream of site C.

Both catchments have a warm temperate coastal climate, with mean annual average rainfall ranging from 1000 mm in the coastal region to 600 mm inland. Over 70% of the catchments are dominated by native eucalypt forests, managed as either production forests or National Parks. The remainder of the catchments are grazing, cropping, and urban areas.

The majority of the Moruya-Deua and Tuross River catchments consist of early Ordovician metasediments, which comprise a turbidite sequence of interbedded sandstones, mudstones, shales, and slates (Lewis *et al.*, 1994). Quartz is the dominant framework grain, with sublitharenites containing mica, plagioclase and K-feldspar. Approximately a quarter of the areas in each catchment are underlain by plutonic rocks dominated by moderately quartz-rich rocks composed

of adamellite, granodiorite and tonalite (Lewis *et al.*, 1994). Basalt is very limited in both catchments, with a scattered and disparate distribution pattern near the coastline.

METHODS OF SUSPENDED SEDIMENT TRACING

Deposited sediment samples were taken from five junctions, with two (J1 and J2) in the Moruya-Deua River catchment and three (J3, J4 and J5) in the Tuross River catchment. These junctions were selected because the subcatchment areas are relatively large and the sample sites were accessible. The locations of the sampled junctions are shown in Fig. 1(a). Each junction contains a complete set of sample sites from two upstream sites (A and B) and one downstream site (C) (Fig. 1(b)). Site A is referred to as the upstream site on the main stream, while Site B is on a tributary. Four to six replicate samples were collected at each site. The samples were taken from sediment deposited in the top 5 mm of the active stream channel. The sediment is usually trapped by vegetation in the stream channel.

In the laboratory, each sample was wet sieved with deionised water to separate the less than 63 μm fraction, then air dried and treated with 30% hydrogen peroxide to remove organic matter. Following acid digestion, the treated samples were analysed with Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES). The elements analysed include major (Al, Ca, Fe, K, Mg, Na and Ti), trace (Ba, Cu, Ga, Mn, V and Zn), and rare earth elements (Ce, Gd, Nd and Pr). Subsamples from each sample site were analysed with X-ray sedimentation by Sedigraph 5100 for particle size distributions. Subsamples from J2 and J3 were separated into three fractions based on Stokes' Law: less than 10 μm, 10 to 40 μm and 40 to 63 μm. These fractions were then analysed with ICP-AES.

The tracers for each junction were selected based on two criteria. Firstly, the median of the tracer concentration at the mixture C is between the median of the two sources A and B. Secondly, the tracer concentrations of sources A and B are significantly different with 95% confidence. The multivariate sediment-mixing model from Collins *et al*. (1998) was simplified to calculate the proportional contributions of suspended sediments from sources A and B in each junction. The simplified multivariate mixing model is expressed below:

$$\text{Minimizing} \sum_{i=1}^{n} \left(\frac{\overline{[C]}_i - [E]_i}{\overline{[C]}_i} \right)^2$$

where $[E]_i = \overline{[A]}_i x + \overline{[B]}_i y$ *c*onstrained by: $x + y = 1$, and $0 \leq x \leq 1$, where $[E]_i$ = estimated concentration of tracer parameter (*i*) in the mixture samples; $\overline{[C]}_i$ = measured median concentration of tracer parameter (*i*) in the mixture samples; $\overline{[A]}_i$ = median concentration of tracer parameter (*i*) in source A samples; $\overline{[B]}_i$ = median concentration of tracer parameter (*i*) in source B samples; x = percentage contribution from source A; y = percentage contribution from source B.

The variation of the estimation is calculated by comparing the estimated concentration in mixture C calculated from the multivariate mixing model ($[E]_i$) and the measured concentrations in the mixture C samples ($\overline{[C]}_i$) (Collins *et al*., 1998).

RESULTS

Particle size distribution of suspended sediment

The suspended sediments from all junctions have high proportions of fine fraction, with an average of 45% of the mass finer than 2 μm (Fig. 2). The sediments from J5 contain the finest particle size distribution, which have 53% of their mass finer than 2 μm, and nearly 80% finer than 10 μm. The coarsest suspended sediments are found from the sources of J3, where an average of

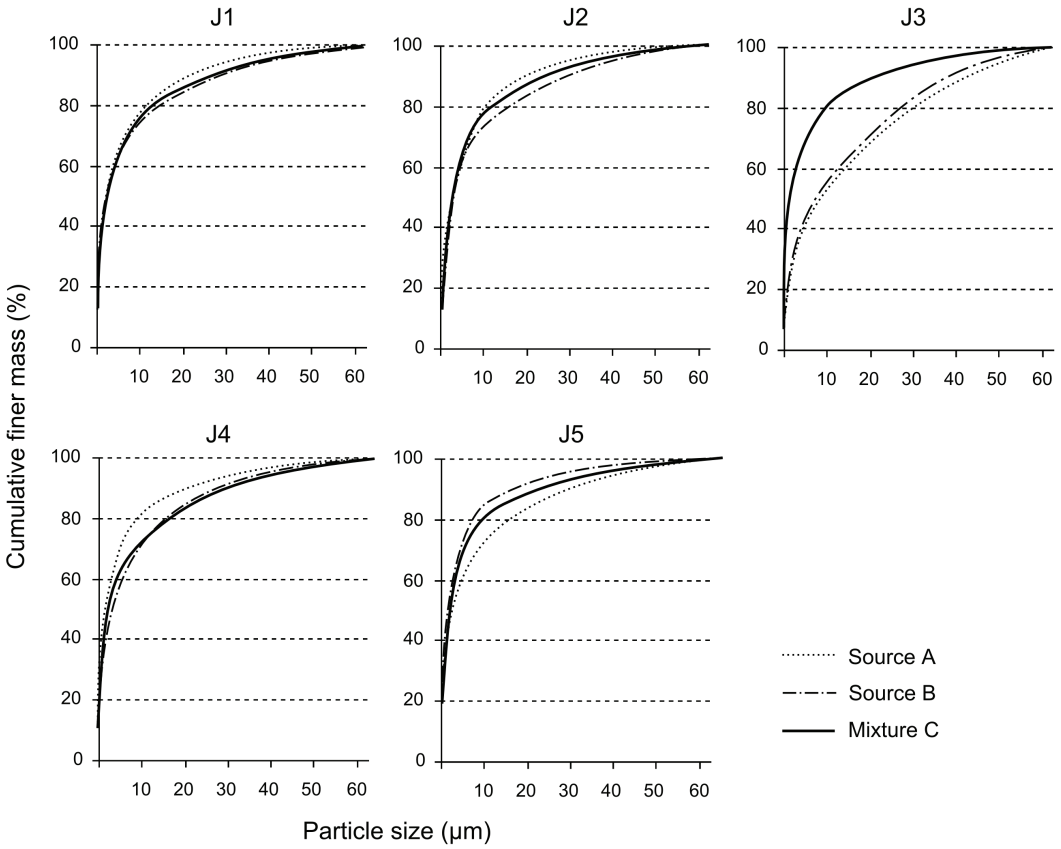

Fig. 2 Average particle size distributions of suspended sediments from sources A and B, and mixture C, of junctions J1–5, expressed by cumulative finer mass percentage.

31% and 55% of the mass is finer than 2 and 10 μm, respectively. The sediments from source A are finer than those from source B in J1, J2 and J4, and coarser than source B in J5. The sediments from J3 have similar particle size distributions for the two sources, but are significantly finer in samples from mixture C.

Particle size distribution and subcatchment characteristics

Assuming the sediments from the sources of each junction are representative of the sediments eroded and transported from the subcatchments upstream, the particle size distributions of sediments may be related to the characteristics of the subcatchments. Such relationships may help interpret possible factors contributing to the variation of the particle size distribution. The particle size distribution of sediments is controlled by erosion and sediment transport. The differences in subcatchment lithology may contribute to selective erosion, and the variation of the catchment size may be an indicator of stream energy which causes preferential transport of sediments.

The relationships between the percentage mass of fine particle size fractions and the percentage area of fine grain lithology are identified. The subcatchment areas of fine grained lithology include the areas dominated by rhyolite ignimbrite, shale, hornfels or aplite. Areas that are dominated by sandstone, adamellite or granodiorite are classified as medium or coarse grain lithologies. The percentage mass of less than 2 and 10 μm fractions for sources of J1–5, as well as the percentage of the subcatchment areas with fine grain lithology are plotted in Fig. 3.

The sediments from 1A, 2A 4A and 5B are finer than from 1B, 2B, 4B and 5A. This is consistent with the higher proportions of fine grain lithologies in subcatchments of 1A, 2A, 4A and 5B. Such a relationship is most obvious from the mass percentage of less than 10 μm fractions. An exception is observed at J3 where the percentage mass of fine sediments from source

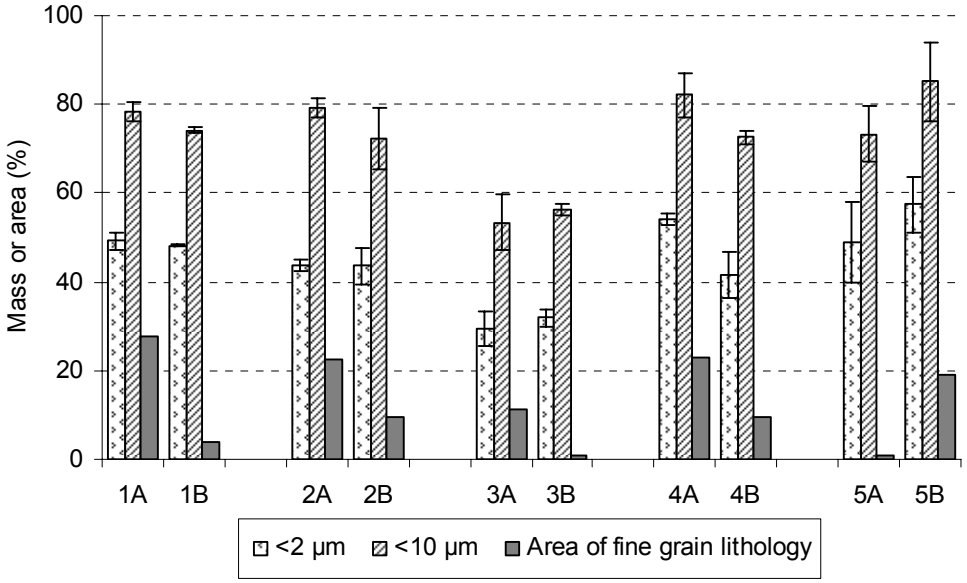

Fig. 3 The relationships between percentage mass and percentage of subcatchment area of fine grain lithology for sources A and B in J1–5.

A does not respond to a higher percentage area of fine grain lithology. When comparing junctions, the percentage mass of sediments is poorly related to the percentage area of fine grain lithology. This is likely because some fine grain areas in sandstone-dominant lithology are omitted from the calculation. Most sandstone-dominant areas consist of an unknown proportion of fine grain minerals such as mudstones, shales and slates. A mineralogical analysis is needed in future research to improve the understanding of the quantitative relationship between particle size distribution and subcatchment lithology.

The relationships between the percentage mass of the fine particle size fractions and the sizes of the subcatchments were investigated. The percentage mass of less than 2, 10 and 40 µm fractions of the suspended sediments are poorly related to the subcatchment areas. This is most likely because the stream energy of the subcatchments is too high to cause the differences in particle size distribution of the less than 63 µm sediments.

Geochemistry and source discrimination

Most element concentrations are significantly different between the less than 10 µm, 10–40 µm and 40–60 µm fractions. The results of Student *t*-tests show that approximately two thirds of the element concentrations are significantly different ($p < 0.05$) when comparing the less than 10 µm and 10–40 µm fractions. These elements include Al, Ba, Cu, Fe, Ga, Gd, K, Mg, Mn, Na and V. When comparing the 10–40 µm with the 40–63 µm fractions, only the concentrations of Ca and Zn are not significantly different. Also, the concentration of Ca cannot be distinguished between the less than 10 µm and the 40–63 µm fractions.

The element concentrations are typically linearly related to the particle size, with higher concentrations observed in finer particle size fractions. Linear models were used to fit the distributions of element concentrations over different particle size fractions at the 95% confidence interval. Among the 17 analysed elements, the concentrations of Al express the best fit to particle size fraction ($R^2 = 0.85$). Other element concentrations that also produce a good fit to the particle size include Mg, Na, K, V and Fe. Most rare earth and trace elements are less sensitive to the particle size distributions of the sediments.

The results of the multivariate mixing model for sediment source discrimination from J2 and J3 are shown in Table 1. No individual tracer from J3 was found satisfying tracer selection criteria. Therefore, the ratios of two elements were used for source discrimination for sediments from J3.

Table 1 Suitable tracers and the proportional contributions of sediments from source A of J2 and J3, using geochemical data for different particle size fractions.

Junction	Particle size (μm)	Suitable tracers	Percentage contribution of source A (%)
J2	<10	Ca, Ga, Mn, Na, Ti, V	46.9 ± 35.4
	10 – 40	Fe, Ga, Na, Ti, V	44.0 ± 9.8
	40 – 63	Ce, Ga, Mn, Na, Nd, Ti, V	44.0 ± 17.6
	< 63	Ca, Ce, Fe, Ga, Mn, Na, Nd, Ti, V	45.2 ± 11.3
J3	<10	27 element ratios	67.5 ± 20.5
	10 – 40	23 element ratios	66.2 ± 19.6
	40 – 63	10 element ratios	69.7 ± 15.2
	< 63	21 element ratios	64.3 ± 12.0

The tracers suitable for sediment source discrimination vary for different particle size fractions. Common tracers for all particle size fractions in J2 include Ga, Na, Ti and V. Elements such as Ga, Mg and Ti are also frequently used in element ratios for J3. Although different tracers are used for the source discrimination in the multivariate mixing model, the results of the proportional contributions of source calculations using geochemistry of different particle size fractions are surprisingly comparable. The proportional contributions of source A in J2 and J3 range from 44% to 47% and from 64% to 70%, respectively, depending on the particle size fractions used in the model. Such differences are minor compared to the variation in the individual results.

DISCUSSION

Sediment source discrimination using different particle size fractions suggests that the geochemical tracing outputs are insensitive to the particle size fractions used for the J2 and J3 sites. The particle size distributions of sediments from J2 and J3 show very similar patterns, with the largest difference lying between source A and mixture C of J3. The differences of particle size percentage mass between source A and mixture C in J3 are 27%, 18% and 9% for the less than 10 μm, 10–40 μm and 40–63 μm fractions, respectively. Such differences in particle size distributions are not sufficiently significant to trigger considerable differences in sediment source discrimination using various particle size fractions. However, given the significant influences of particle size on sediment geochemistry, it is likely that a threshold of particle size differences exists which allows the results of sediment tracing to vary significantly using different particle size fractions.

The insignificant influence of particle size distribution on sediment source discrimination shown in this study suggests that using particle size adjustment approaches without consideration of the threshold of the influence could be misleading. For example, using finer fractions for source discrimination would not have produced significantly different results than using the less than 63 μm fractions. However, the representation of such finer particle size fractions on the suspended sediment as a whole is questionable, particularly when making comparison with other studies that use the less than 63 μm fraction (Rustomji *et al.*, 2008). Besides, collecting sufficient samples to enable analysis of such fine particle size fractions could be difficult, especially from rivers of high stream energy where deposited suspended sediment concentrations are low. Greater uncertainties can also be introduced by additional laboratory processes to extract finer fractions.

The similarity in particle size distributions of various junctions may be caused by the relatively uniform lithology across the catchments. Uniform lithology is often seen as a disadvantage in sediment tracing because of the similarity of many tracer properties between the sources. This study demonstrates the advantages of applying sediment tracing in lithologically uniform areas where the influences of particle size can be minimised, as far as suitable tracers can be identified. In this study, geochemistry is a useful tracer for the Moruya-Deua and Tuross river catchments of Australia where metasediments dominate over 60% of the area. Metasediments are

the typical lithology across the southeast coastal regions of Australia. This study provides an example for sediment tracing in other metasedimentary catchments.

Future research could be focused on the investigation of particle size impacts on sediment source discrimination using different types of tracers and on a wider range of lithologies. These studies may allow for greater differences in particle size distributions between sediment sources and mixture, and thus the particle size threshold could be identified. With better knowledge of the sensitivity of particle size distribution on sediment source discrimination, and of the relationships between the particle size distribution and catchment lithology, cost-effective processes can be designed for sediment tracing.

CONCLUSION

This study investigated the influence of particle size distribution on sediment source discrimination using geochemical tracers. Linear relationships were found between geochemistry of the tracers analysed and the particle size fractions. However, the results of source discrimination using the multivariate mixing model are comparable for the less than 10 μm, 10–40 μm, 40–63 μm and less than 63 μm fractions. Such results suggest that a threshold of differences in particle size distribution between sources and mixture may exist for the particle size to significantly influence source discrimination in sediment tracing. Below this threshold, the less than 63 μm fraction is representative and cost-effective for the source discrimination of suspended sediments. The particle size distributions of the deposited suspended sediments within the same junction were found to be related to the dominant lithology of the subcatchments. Similar studies on a wider range of lithological settings are required to identify the threshold. Nevertheless, this study demonstrates that when undertaking sediment tracing in lithologically uniform areas, geochemical tracers are potentially useful, and the influences of particle size on source discrimination may be minimised, thus using the less than 63 μm fraction may be most efficient.

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