ADVANCES IN SEDIMENT FINGERPRINTING

A comparison of geological and statistical approaches to element selection for sediment fingerprinting

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Abstract

Purpose: Elevated sediment loads reduce reservoir capacity and significantly increase the cost of operating water treatment infrastructure making the management of sediment supply to reservoirs of increasing importance. Sediment fingerprinting techniques can be used to model the relative contributions of different sources of sediment accumulating in reservoirs. The goal of this research is to compare geological and statistical approaches to element selection for sediment fingerprinting modelling.

Materials and methods: Time-integrated samplers ($n=45$) were used to obtain source samples from four major subcatchments flowing into the Baroon Pocket Dam in South East Queensland, Australia. The geochemistry of these potential sources were compared to sediment cores ($n=12$) sampled in the reservoir. Elements that provided expected, observed and statistical discrimination between sediment sources were selected for modelling with the geological approach. Two statistical approaches selected elements for modelling with the Kruskal-Wallis H-test and Discriminatory Function Analysis (DFA). In particular, two approaches to the DFA were adopted to investigate the importance of element selection on modelling results. A distribution model determined the relative contributions of difference sources to sediment sampled in the Baroon Pocket Dam.

Results and discussion: Elemental discrimination was expected between one subcatchment (Obi Obi Creek) and the remaining subcatchments (Lexys, Falls and Bridge Creek). Six major elements were expected to provide discrimination. Of these six, only Fe_2O_3 and SiO_2 provided expected, observed and statistical discrimination. Modelling results with this geological approach indicated 36% (+/- 9%) of sediment sampled in the reservoir cores were from mafic-derived sources and 64% (+/- 9%) were from felsic-derived sources. The geological and the first statistical approach differed by only 1% (σ 5%) for 5 out of 6 model groupings with only the Lexys Creek modelling results differing significantly (35%). The statistical model with expanded elemental selection differed from the geological model by an average of 30% for all 6 models.

Conclusions: Elemental selection for sediment fingerprinting therefore has the potential to impact modeling results. Accordingly we believe it is important to incorporate both robust geological and statistical approaches when selecting elements for sediment fingerprinting. For the Baroon Pocket Dam, management should focus on reducing the supply of sediments derived from felsic sources in each of the subcatchments.

Keywords Geochemical fingerprinting • Sediment provenance • Australia • Discriminant Function Analyses

1. Introduction

Elevated sediment loads from accelerated catchment erosion pose a significant challenge to the management of increasingly scarce water resources. Not only does accelerated soil erosion and the concomitant suspended sediment loads threaten aquatic ecosystems (Bunn et al. 2007; Bilotta and Brazier 2008), it significantly increases the costs of maintaining and operating water treatment and transportation infrastructure (Clark 1985; Holmes 1988; Dearmont et al. 1998). It is important to manage sediment sources directly in order to reduce uncertainty in the potential operational funding required to maintain and operate viable water supply networks. Managing sediment supply to waterways requires a detailed understanding of the nature, location and proportional contributions of different sediment sources (Walling 1983; Douglas et al. 2003a; Douglas et al. 2007).

One approach to understanding sediment provenance is to trace the geochemistry of the sediment back to its lithogenic sources (Olley and Caitcheon 2000; Douglas et al. 2003b; Evrard et al. 2011; Navratil et al. 2011; Navratil et al. 2012). This geochemical fingerprinting technique is based on the fact that different rock types often produce soils and sediments with unique geochemical compositions (Klages and Hsieh 1975; Wood 1978; Caitcheon et al. 2006). If these unique geochemical compositions are maintained during generation, transport, and deposition processes, and the geochemistry of sediment sources are indeed distinct, then the spatial origin of transported sediment may be ascertained (Walling et al. 1993; Olley et al. 2001; Douglas et al. 2005). This current research focuses on the analysis and modelling of sediment geochemistry to determine the spatial source of sediment. For reviews of sediment tracing and modelling approaches, see Davis and Fox (2009), Guzmán et al. (2013), Haddadchi et al. (2013), or Koiter et al. (2013b).

Since the early adoption of geochemical fingerprinting techniques (Klages and Hsieh 1975; Wall and Wilding 1975; Wood 1978), research has predominantly focused on the

development of statistical techniques for element selection (Collins et al. 1996; Collins et al. 2012) and refinements in the modelling processes (Walling et al. 1993; Hughes et al. 2009; Collins et al. 2010b). This research has established a solid disciplinary foundation for sediment fingerprinting research. Contrarily, there has not been a comparable level of research on the geologically-based foundation for elemental discrimination that was central to the original development of the geochemical fingerprinting technique.

The fundamental logic for using elemental compositions to discriminate between spatial sources is that different rock types are often formed in different conditions and produce minerals with distinct chemistries. For example, igneous rocks are formed from magma flows either on the Earth's surface (extrusive) or below ground (intrusive) (Tarbuck and Lutgens 2006; Marsh and Kaufman 2012). When the magma cools the ionic component within the magma crystallizes into different minerals with the temperatures and the rate of cooling resulting in rock types with differing mineral compositions (e.g. Bowen's Reaction Series) (Tarbuck and Lutgens 2006; Marshak 2008; Reynolds et al. 2013). Other processes such as weathering and erosion (e.g. sedimentary rocks) over longer temporal scales in conjunction with the differing mineral compositions of rock types results in the unique signatures of soils and sediments traceable with geochemical fingerprinting techniques. Ultimately, this geological foundation for understanding differences between rock types is central to geochemical fingerprinting research.

The goal of this research is to compare geological and statistical approaches to elemental selection for geochemical fingerprinting. The geological approach is based on the work of Koiter et al. (2013a) who applied geological understanding rather than statistical tests to select elements for modelling in the South Tobacco Creek Watershed, Canada. The proposed geological approach selects elements for sediment fingerprinting through an examination of expected, observed and statistical differences between sediment sources. This approach is

then compared to a statistical approach to element selection (Collins and Walling 2002; Wilkinson et al. 2013). These two distinct approaches to elemental selection are investigated in the Baroon Pocket reservoir catchment in the South East Queensland (SEQ) region of Australia.

2. Methods

2.1 Study area

The Baroon Pocket reservoir catchment (74 km²) is subtropical with mean annual temperatures ranging between 24°C and 26°C (BOM 2014). The mean annual rainfall in the catchment is one of the highest in SEQ (1,785mm) with the majority falling during summer (October to February)(BOM 2014). Since the beginning of European settlement, approximately 80% of the native vegetation has been cleared. The region has a population of ~5,000 with the majority located in the urban centre of Maleny.

The Baroon Pocket reservoir catchment is characterized by two distinct areas: A relatively flat plateau in the upper catchment comprised of urban, cattle grazing and intensive dairying, and plantation (e.g. orchards) land uses; and a steep, generally well vegetated, gorge in the lower catchment approaching the reservoir with landslides in areas cleared for cattle grazing that are expected to contribute a significant amount of sediment to the reservoir. The upper catchment is predominantly underlain by Basalt, whereas the lower catchment consists of Basalt, Colluvium, Landsborough Sandstone, and Wappa Ryholite (DME 2008)(Fig 1).

The Baroon Pocket reservoir has an average annual yield of 20,000ML and a storage capacity of 61,000 ML (Dunstan 2007). The catchment of the Baroon Pocket reservoir has the most reliable rainfall out of all water supply catchments in SEQ and also has the highest water supply reservoir elevation in the region. Therefore, this reservoir provides the necessary hydraulic head to supply water throughout the northern section of SEQ's water supply

network. The Baroon Pocket reservoir is receiving high volumes of sediment that are challenging the operational efficacy of this water supply system.

Recent research in the catchment (Kemp et al. 2014) has identified landslides as the primary source of sediment reaching the reservoir. While landslides have been a feature of this landscape for several million years (Willmott, 1983), early scientific studies and the documentary research presented in Kemp et al. (2014) identify European land use as the cause of increased landslide activity over the last 80-90 years and clearly link this increased activity to the extensive clearance of native forest for agriculture that peaked around the end of World War I. Current landslide derived sediment is derived predominately from reactivated or extended historical landslides triggered by a series of wetter years starting in 2010.

The landslide distribution is highest in areas characterized by steep slopes along the incised reaches of Obi Obi and Bridge Creek. Several landslides are directly connected to the drainage network and are hypothesized as supplying a significant volume of suspended sediment to the reservoir. Accordingly, the management objective of this geochemical fingerprinting research is to determine the spatial sources of sediment being deposited in the reservoir.

2.2 Sample collection, processing and analysis

Time-integrated samplers (Phillips et al. 2000) were used to sample sediment and were deployed ~0.5m above the dry-season level (Fig. 1). 45 samplers were deployed between late November and early December in 2012 and retrieved between late July and early August in 2013. Sampling location was determined by pragmatic factors such landowner permission and stream access. Samplers were inspected after each major rainfall event (once in January and twice in February 2013). After the January event, one sampler lost the front end fitting. In mid-February, one missing sampler was reinstalled, and in late February, 3 samplers were

reinstalled. During the retrieval period, 4 additional samplers were missing, likely the result of vandalism. At one of these locations (BP05-Fig. 1) a lag deposit sample was obtained by compiling 20 scrapes of evident deposited materials with a non-metallic trowel over a 100m reach.

Sediment deposited in the Baroon Pocket reservoir was sampled at 14 locations (Fig. 1) with a cable hammer coring device. This corer consisted of a 120 cm PVC 50 mm tube fitted with a one way valve at the top onto which attached a cage containing a sliding weight. The coring device was lowered into the water and supported on the bottom by one cable and another cable attached to the weight was used to raise and lower the weight, effectively hammering the tube into the bed sediments. The sediment core samples were taken downstream of the inputs of each of the major tributaries (e.g. Obi Obi, Bridge, Lexys and Falls Creeks). An additional core-grouping was sampled at the far side of the reservoir (BC04, BC05, and BC09) and is simply referred to as the 'Far' sediment cores. The top 10cm of each sediment core was subsampled for geochemical analysis. In addition, a landslide which was originally mapped by Willmott (1983) and subsequently remobilized in 2008 was opportunistically sampled at two locations during a site inspection with catchment staff. The landslide was sampled similarly to the lag deposit by compositing 20 scrapes of the recently evident landslide subsoils with a non-metallic trowel.

Sediment generation and transportation processes result in fine particle size classes being preferentially eroded and less affected by abrasion and disaggregation during sediment transport (Walling and Woodward 1992; Collins et al. 1997; Dyer and Olley 1999). The resultant fluvial impacts create potential complexities for the direct comparison of the elemental geochemistry of non-mobilized source soils to transported sediment in waterways (Koiter et al. 2013b; Smith and Blake 2014). Accordingly, sediments were exclusively sampled throughout the catchments to facilitate a direct comparison between sediment

184 derived from different source areas, with the exception of the two opportunistic landslide
185 samples. For all samples, the <10µm particle size was isolated with settling columns to
186 further minimize potential differences between source and sediment particle size distributions
187 (Wilkinson et al. 2013). In SEQ, the dominant particle size transported is the <10µm fraction
188 (Douglas et al. 2003b). Accordingly this fraction is appropriate for tracing sediment in the
189 region.

190 Sediment geochemistry was analysed at the Queensland Government Department of
191 Science, Information Technology, Innovation and the Arts (DSITIA) Chemistry Centre with
192 lithium metaborate fusion Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) for the
193 trace element concentrations and Inductively Coupled Plasma-Optical Emission Spectrometry
194 (ICP-OES) for major elements concentrations. Major and trace elements above detection
195 limits are listed in Table 1. Major elements were converted to oxides to compare observed and
196 expected elemental oxide concentrations based on the dominant rock type characteristics.

2.3 Geochemical analysis and modelling

199 A three-step selection process was used to select elements for modelling. First, the
200 upstream lithology of the sediment sampling sites was reviewed to understand whether
201 differences were expected between dominant rock types and, importantly, whether any
202 elements should discriminate between these dominant rock types. Second, scatter plots of all
203 elements were examined to compare the expected to the observed geochemical differences
204 between dominant rock types. Third, observed geochemical differences between sources were
205 tested for significance with the Kruskal-Wallis (KW) H-test (for 3 or more sources) or the
206 Mann-Whitney (MW) U-test (for 2 sources). These non-parametric analyses determine
207 whether elements provide significant discrimination between sources (Collins et al. 1996).
208 Elements with triple source discrimination (expected, observed and statistical) were selected

for modelling. Prior to modelling, the selected elements were examined to ensure that their elemental concentrations plotted within the source range to ensure their conservative behaviour during modelling.

This approach is then compared to a statistical three step procedure (Collins et al. 2010a; Wilkinson et al. 2013) that first tests for conservative behaviour and second applies the KW H-test or MW U-test to determine which elements provide a statistical discrimination between sources. Third, a discriminant function analyses (DFA) selects an optimal group of elements for modelling.

The KW, MW and DFA analyses were performed in R-software (R Development Core Team 2011) with the KLaR package (Roeve et al. 2014). A greedy-wilks test was used for the DFA that allows for the adjustment of the level of the approximate F-test decision (or ‘niveau’) that controls the amount of elements selected into the modelling group. Two niveaux (0.05 & 0.35) were selected, the first to approximate a level of statistical significance (0.05) and the second to expand the sediment source grouping for comparison (0.35). The objective of modelling elements selected with the second niveau (0.35) is to examine the impact of expanded element selection on modelling results.

A distribution mixing model (Lacey and Olley accepted) determined the relative contribution of different sources to in-stream sediment through simultaneously minimising mixing model difference (MMD):

$$MMD = \sum_{i=1}^n \left| \left(C_i - \left(\sum_{s=1}^m P_s S_{si} \right) \right) / C_i \right| \quad (\text{Eq. 1})$$

where n is the number of elements included in the model determined by the three-step selection process; C_i is the sediment core distribution of element property (i); m is the number of sources in the catchment; P_s is the source (s) contribution; and S_{si} is the distribution of element (i) in source (s). P_s was modelled as a truncated normal distribution ($0 \leq x \leq 1$) with a

mixture mean (μ_m) and standard deviation (σ_m). Normal distributions were modelled for each source element (S_{si}) and in-stream sediment element (C_i) (Lacey and Olley accepted). Non-negative constraints were applied to all source and in-stream elements with the absolute sum of all proportional contributions (P_s) equaling 1.

The Optquest algorithm in Oracle's Crystal Ball software (2013) was used to model distributions throughout a Monte Carlo style modelling framework. In Optquest, the μ_m and σ_m for each source's contribution (P_s) distribution were repeatedly varied while minimizing the median MMD when simultaneously solving Eq. 1 2500 times with 2500 random samples (Latin Hypercube – 500 bins) drawn from each in-stream (C_i) and source (S_{si}) distribution. This process of deriving the optimal source contribution mixture (P_s) for all 2500 randomly generated trials was repeated 2500 times. The median proportional contribution (P_s) source from these 2500 additional simulations is reported for each source.

Uncertainty for each source's proportional contribution is calculated by summing the modelled standard deviation of the mixture mean (σ_m) with the median absolute deviation (MAD) of this modelled standard deviation for the additional 2500 model simulations and the MAD of the individual sources median proportional contribution again for the additional 2500 simulations. In summary this uncertainty combines the actual standard deviation modelled for each source contribution with the MAD of this standard deviation and the MAD of the actual source contribution for the additional 2500 simulations.

3. Results

3.1 Expected source discrimination

There are five lithology units in the Baroon Pocket reservoir catchment, three of which are distinct subclasses of igneous rocks including mafic (Basalt), felsic (Wappa Rhyolite), and intermediate rock types (Cedarton Volcanics) (Table 2) (DME 2008). The fourth distinct lithology is a sedimentary rock type (Landsborough Sandstone). Based on its lithographic

composition, including lithofeldspathic labile and quartzose sandstone, this sedimentary rock is likely derived from a felsic igneous rock type. The Amamoor Bed lithology is a mixture of sedimentary and mafic derived materials.

As mineral crystallization is a result of magma chemical composition and cooling histories (Tarbuck and Lutgens 2006; Reynolds et al. 2013), the mafic rock types (Basalt) should have high concentrations of CaO, Fe₂O₃, and MgO and low concentrations of Na₂O, K₂O, SiO₂ (Marshak 2008). Contrarily, the felsic and felsic-derived rock types (Wappa Rhyolite and Landsborough sandstones) should have high concentrations of Na₂O, K₂O, and SiO₂ and low concentrations of CaO, Fe₂O₃, and MgO (Marshak 2008). The Amamoor Beds and the Cedarton Volcanics should consist of an intermediate mixture of the mafic and felsic rock types.

The major element geochemistry of sediment samples are expected to discriminate between mafic (Basalt) and felsic-derived (Landsborough Sandstone and Wappa Rhyolite) rock types and potentially the Amamoor beds and Cedarton Volcanics depending on their degree of intermediacy. There may also be geochemical discrimination for colluvium and alluvium depending on their lithogenic origin, weathering and transport as these potential sediment sources were originally derived from hillslope and riverine erosion processes, respectively

The dominant lithology in the Baroon Pocket catchment is Basalt (73%) (Table 3). Colluvium underlies 10% followed by Landsborough Sandstone (4%), Wappa Rhyolite (3%), Amamoor beds (2%), Cedarton Volcanics (1%), and alluvium (1%). The major streams draining into the Baroon Pocket reservoir are predominantly underlain by Basalt. Walkers Creek has the highest Basalt composition (97%), followed by Obi Obi (88%), Lexys (81%), Falls (74%) and Bridge Creek (59%) (Table 3). As there are only 2 samples from Walkers Creek, which flows directly into Obi Obi Creek, these catchments were merged for the

following analysis and discussion. Colluvium comprises the remaining dominant rock type in Lexys Creek (18%) and Falls Creek (18%). No Landsborough Sandstone was present in Lexys Creek compared to 7% in Falls Creek. In Bridge Creek, colluvium was again the second major dominant rock type (20%), followed by Wappa Rhyolite (11%) and Landsborough Sandstone (8%). The Amamoor Beds (4%) and Cedarton Volcanics (2%) constitute small sections of Obi Obi Creek. Alluvium underlie 3% of Walkers Creek though <1% for the remainder of the catchments. Based on lithology, major element geochemistry should discriminate between sediment sampled in catchments dominated with mafic rock types (Obi Obi Creek) compared to catchments with higher felsic derived rock type compositions (e.g Bridge Creek). Depending on the dominant sources (e.g. mafic or felsic derived), Falls and Lexys sediment may plot between mafic and felsic-derived rock types.

3.2 Observed source discrimination

Scatterplots of the major trace geochemistry confirm discrimination between mafic (Obi Obi Creek) and felsic-derived sediments (Bridge, Falls and Lexys Creek) (Fig. 2). Mafic derived sediments have high Fe_2O_3 and low K_2O and SiO_2 as expected. In contrast, felsic sediment have high K_2O , SiO_2 and low Fe_2O_3 . Bridge, Falls and Lexys Creek all plot similarly indicating non-mafic sediment source origins. The two landslide samples were taken from colluvium (Fig. 1). The increased colluvium area within Bridge, Falls and Lexys Creek in combination with the landslide sample geochemistry indicates the colluvium is most likely derived from elevated felsic-derived sources. The Amamoor Beds and Cedarton Volcanics are likely too small to contribute noticeably to Obi Obi Creek sediment or are comprised of predominantly mafic sources. Accordingly, these two lithologies will be considered mafic sources.

The expected elemental geochemistry resulted in two observed source groupings: mafic derived sediments (Obi Obi Creek) and felsic-derived sediments (Lexys Creek, Bridge Creek, Falls Creek). Although it may be possible to further discriminate between the felsic source group, the elemental overlap and particularly the wide range of Falls Creek and Bridge Creek sediment, makes further felsic source discrimination impractical. Based on the geochemistry, the best approach is to model these two sediment source groupings to determine the spatial origin of the sediment core samples from the Baroon Pocket reservoir. Further, the felsic-derived grouping is not representative of pure felsic material. The three ‘felsic-derived’ catchments (Lexys, Bridge and Falls Creek) are predominantly underlain by the mafic basalts resulting in an intermediate signature for these ‘felsic’ catchments. Pure felsite would have SiO₂ compositions >68% (Marshak 2008). Nonetheless, as these catchments have a greater felsic-derived signature we will refer to this grouping as felsic in the following results and discussion.

There were limited differences between mafic and felsic derived sediments for CaO, MgO, and Na₂O. For Na₂O, this could be related to its non-conservative nature during fluvial transport (Kraushaar et al. 2014). For CaO and MgO we are unaware of the reason for the lack of observed discrimination. There was also observed elemental discrimination between felsic and mafic derived sediments with P₂O₅ and TiO₂. The elevated P₂O₅ could be related to the high concentration of dairy and orchard operations in the predominantly mafic Obi Obi catchment compared the felsic areas lower in the catchment that have more vegetative cover. P₂O₅ also has dissolved phases indicative of potential non-conservative behaviour (Smith and Blake 2014). TiO₂ has low soil mobility and high weathering resistance and, according to Koiter et al., (2013a), the higher values of TiO₂ in the mafic sediments may indicate that they are eroded from a higher position in the soil profile. Conversely, Smith and Blake (2014)

reported poor source discrimination with TiO_2 , particularly between surface and subsurface sources.

Multiple elements from the sediment core samples plot below or above the source sediment (Fig. 2). The sediment core samples have higher Al_2O_3 , and lower CaO , Na_2O , TiO_2 and to a lesser extent – K_2O and MgO . For the geological discriminators, K_2O is affected as 4 core samples plot below the source range excluding this element from modelling. The Baroon Pocket reservoir sediment core samples do plot on a mixing line between the mafic and felsic source samples for Fe_2O_3 and SiO_2 . As these elements together comprise on average 65% (σ 6%) of the total elemental composition of stream sediment samples, they are selected as the elements for modelling.

Fe_2O_3 and SiO_2 significantly discriminate between felsic and mafic sources (Table 1). As there are only two main sources, only one element is required for modelling. Accordingly, Fe_2O_3 and SiO_2 were modelled individually and then simultaneously for all sediment core samples, together as one grouping, as well as for each subcatchment sediment core grouping and the ‘Far’ sediment cores. The Far sediment core grouping, Bridge Creek and Falls Creek all plot clearly within the felsic derived sediment sources (Fig. 3). Both Obi Obi and Lexys Creek plot between the felsic and mafic sources, indicative of a mixture of sources and a higher than expected contribution of felsic sources in sediment cores near the outlet of Obi Obi Creek.

3.3 Statistical source discrimination

As multiple Baroon Pocket reservoir sediment core samples plot outside of the source range, multiple elements were observed not to be conservative. Indeed, without the sampling of the 2 landslides, 29, or almost 75% of the elements would not be conservative. With the inclusion of the landslides, 19 elements or ~50% would not be conservative. High numbers of

non-conservative elements typically indicate a missing source or other factors affecting sediment geochemistry during mobilization, transportation and deposition processes. The effect of the inclusion of the two landslide samples on conservative behaviour suggest that landslides are potentially a significant source that may not have been actively eroding during the in-stream sediment sampling. Of the 20 conservative elements, 14 significantly discriminated between sediment sources. The DFA_{0.05} selected P₂O₅, Zr, SiO₂ and TiO₂ for modelling whereas the DFA_{0.35} included these four elements from the DFA_{0.05} along with Mn, La, Zn and Fe₂O₃. All conservative elements that significantly discriminated between sources were also individually modelled (Table 1) to examine the potential impacts of elemental selection on modelling results.

3.4 Modelling results

For all Baroon Pocket sediment core samples, the Fe₂O₃ only model indicated that 50±13% of all sediment has a mafic origin compared to only 20±7% for the far sediment core grouping (Table 4). The mafic sediment contribution was highest at Lexys Creek (67±9%) with the Fe₂O₃ model, followed by Obi Obi Creek (65±10%) with decreasing contributions for Falls (33±8%) and Bridge (24±7%) Creeks. The average sediment contribution from mafic sources was 13% lower with the SiO₂ only model indicating 25±8% for all sediment cores are derived from mafic sources, 46±8% for Lexys, 45±9% for Obi Obi, 26±7% for Bridge, 23±7% for Falls Creek and 11±6% for the far core grouping. The simultaneous modelling of Fe₂O₃ and SiO₂ provided expectedly intermediate results with average contribution of 36% (σ 19%) for all mafic derived sediments for all catchments (Fig. 4). For the far sediment grouping, only 15±7% of sediment were modelled to derive from mafic origins with this model, compared to 58±9% for Lexys, 54±9% for the Obi Obi, 28±6% for Falls and 23±8% for Bridge Creek. The median modelled source contribution for all sediment core samples with the Fe₂O₃-SiO₂

model ($36\pm 6\%$) was equivalent to the mean of all subcatchment mafic derived sediments ($36\% \pm 19\%$).

The DFA_{0.05} model produced similar results to the Fe₂O₃-SiO₂ model with the exception of Lexys Creek for which only $23\pm 7\%$ of sediment was modelled to derive from mafic origins compared to $58\pm 9\%$ for Fe₂O₃-SiO₂ model. For the remainder of the models, there was a 1% ($\sigma 5\%$) mean difference between the DFA_{0.05} and the Fe₂O₃-SiO₂ models. Conversely, the DFA_{0.35} model results differed considerably from the previous 4 models. The DFA_{0.35} model indicated that on average only 6% ($\sigma 7\%$) of sediments were derived from mafic sources with the highest mafic contributions modelled for the far sediment core grouping ($13\pm 8\%$) and Lexys Creek ($13\pm 8\%$) whereas no mafic contributions were modelled for Obi Obi Creek. Compared to the consistency of the previous four models, the DFA_{0.35} model appears inaccurate.

To demonstrate the underlying complexity with the DFA_{0.35} model, all elements that significantly discriminated between mafic and felsic sources were modelled separately. Quickly it becomes evident that the light rare earth elements (REEs) from the Cerium Group (e.g. La, Pr, Nd, & Sm) perform differently in the modelling process (Fig. 5). In fact, the average modelling results for the 6 sediment core groupings for the light REEs differs from the Fe₂O₃-SiO₂ model by 55% . The light REE models results were predominantly 100% mafic or 100% felsic. The heavy REEs (Yb, Y, & Er) of the Yttrium group performed marginally better with an average difference of 32% for all the 6 different models. Although Zr was selected for the first DFA_{0.05}, it differed from the Fe₂O₃-SiO₂ model by 39% for all 6 models. Zn differed by 35% , and Mn by 26% . The major oxide elements performed best with TiO₂ differing by 22% , P₂O₅ by 10% and both SiO₂ and Fe₂O₃ by 7% . Indeed the DFA_{0.05} achieved a balance between Zr and TiO₂ that likely resulted in the similarity to the Fe₂O₃-SiO₂ model. The DFA_{0.35} model was likely affected by the inclusion of Mn, La and Zn and

could not longer produce comparable results to the other four models. The varying model results for all conservative elements and the DFA_{0.35} model highlights both the underlying complexity involved in modelling sediment sources and the importance of element selection in sediment fingerprinting research.

4. Discussion

The Baroon Pocket reservoir catchment is a complicated tracing environment highlighted by the non-conservative behaviour of multiple elements. There are two main likely factors for this non-conservative behaviour. First, there is either a missing or undersampled source. The impact of the inclusion of two landslide source samples indicates that the missing source is likely these landslides. Second, there was a shift in sediment properties between the riverine and the reservoir environments noted by an increase in Al₂O₃ and a decrease of TiO₂, Na₂O, and to a lesser extent, K₂O. One reason for this transition could be chemical additions (e.g. Al₂(SO₄)₃) to the reservoir to treat algal blooms. These additions may impact sediment elemental geochemistry. The time integrated samplers may have also affected sampled sediment properties as Smith and Owens (2014) reported these devices may sample sediment with potentially different particle size and geochemical compositions than sediment transported in the catchment. Further there could be significant particle size and density sorting effects during transport and deposition in the reservoir. Although we attempt to address potential effects of particle sorting by isolating the <10 µm particle size fraction it is likely that both particle size and density sorting in the reservoir resulted in further separation of the fine and potentially dense mineral components.

Owing to these complications, a geological approach was developed for elemental selection for sediment fingerprinting modelling and compared to statistical approaches. The foundation of the statistical approach to sediment fingerprinting is that the KW/DFA analyses

selects a composite suite of sediment properties that provide the optimal combination of elements for source ascription (Collins et al. 1996; Collins and Walling 2002). The challenge with statistical approaches, as highlighted by Owens et al., (2006), is that they may select elements that are potentially inappropriate for sediment fingerprinting modelling. Further, Smith and Blake (2014) concluded that statistical approaches should not substitute for a detailed understanding of both the chemical and physical foundation of individual tracer property behaviour.

Our research demonstrates that it is important to incorporate both statistical and geological approaches when selecting elements for sediment fingerprinting modelling. For example, in 5 out of the 6 models there was only an average difference of 1% (σ 5%) between the geological ($\text{Fe}_2\text{O}_3\text{-SiO}_2$) and statistical ($\text{DFA}_{0.05}$) modelling approaches (Fig. 4). The similarity between these models provides an increased confidence in results compared to relying on the results from either the statistical or the geological approach. Although this demonstrates the potential of both approaches, one catchment's modelling results differed by 35% which is significant if modelling results are used to inform management interventions. When expanding source selection with the statistical approach ($\text{DFA}_{0.35}$), the source ascription results differed by 25% from the $\text{DFA}_{0.05}$, and 30% from the geological model indicating the potential influence of element selection on modelling results. The utility of using both geological and statistical approaches is that together they provide more certainty to modelling results whilst incorporating the geological foundation for discrimination fundamental to the fingerprinting technique. Future research should investigate statistical and geological approaches to element selection with artificially generated mixtures similarly to Haddadchi et al. (2014) to quantify the influence of elemental selection on the modelling results.

Analyses of in-stream sediment geochemistry in the Baroon Pocket reservoir catchment highlighted two distinct sediment lithogenic groupings: mafic and felsic. For the Baroon

Pocket reservoir, it would be expected that sediment source contributions would relate relatively to dominant rock type area. This would result in 81% of sediment being derived from mafic rock types and only 18% from felsic rock types. When examining all sediment cores with the $\text{Fe}_2\text{O}_3\text{-SiO}_2$ model, only 36% of sediment were derived from mafic and 64% from felsic areas resulting in a felsic contribution-to-area (CTA) ratio of 3.55. This enrichment in sediment derived from felsic origin indicates Landsborough Sandstone, Colluvium or Wappa Rhyolite or combinations of the three contribute more sediment per unit area to the reservoir. Sediments from all catchment groupings had enriched felsic CTA ratios with the highest being for the Obi Obi creek (19), with only 2% of the area being felsic and contributing ~46% of sediments. Lexys (2), Falls (3) and Bridge (2) Creek sediment cores were all enriched in felsic derived sediment.

In-stream sediment ^{137}Cs activity concentrations reveal that sediment are derived from 95% (σ 12%) subsoil sources (Kemp et al. 2014) indicating that channel, gully or landslide erosion is the dominant sediment source from these felsic regions. In addition, the significant difference of including the landslides on element conservative behaviour further suggests the landslides are a potential leading source of sediment in these catchments. More research is required to isolate landslides as the geochemically proven source of sediment to the reservoir and determine which elements can significantly discriminate between colluvium, Wappa Rhyolite and Landsborough Sandstone. It is also important to confirm whether colluvium has a felsic lithology. Accordingly, obtaining at minimum 30 source samples from the landslides and each of major rock type would provide a more detailed understanding of the spatial sources of sediment and their elemental variability in this catchment. Further, a geochemical analyses of sediment core samples near the water supply intake could identify the most problematic sediment sources. Fallout radionuclide or optically stimulated luminescence dating of the sediment cores could also determine source variability over time and provide

rates of sediment accumulation. Indeed, a long-term sampling program is necessary to understand potential inter and intra annual sediment source variations.

5. Conclusions

A geological approach determined the sediment source contributions from two dominant lithologies in the Baroon Pocket reservoir catchment. The results indicate that felsic rock types contribute ~3.5 times more sediment than their unit area to the Baroon Pocket reservoir. As a separate analysis of sediment ^{137}Cs activity concentrations indicated that subsoil sources dominate in this catchment, management should focus on reducing the supply of sediment from felsic channel, gully and landslide sources. The impact of landslide samples on the conservative nature of elements suggests that landslides may be one of the dominant sediment sources in this catchment. A second round of sampling specifically targeting landslides, colluvium and the dominant rock type sources would improve both our geological and our sediment source understanding in the Baroon Pocket reservoir catchment.

The geological and $\text{DFA}_{0.05}$ approaches differed by only 1% (σ 5%) for 5 out of 6 model groupings with only the results for Lexys Creek differing significantly (35%). The $\text{DFA}_{0.35}$ model differed from the geological model by on average 30% for all 6 models. These results demonstrate that element selection may significantly impact sediment fingerprinting results. Accordingly we believe it is important to incorporate both statistical and geological approaches for element selection in sediment fingerprinting modelling.

As the sediment fingerprinting discipline has seen significant advancement with modelling techniques, we believe a similar focus on the geological potential for element selection and source discrimination is needed along with further research examining the impact of element selection with artificial mixtures. Although an underlying premise of the composite fingerprinting technique is that the inclusion of more elements is beneficial to sediment

tracing research (Peart and Walling 1986; Collins and Walling 2002), we believe that it may be as important to understand the utility of a few meaningful elements and their geological foundation for discrimination rather than solely relying on statistical techniques for determining elemental discrimination.

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Tables:

Table 1 All major elements followed by trace elements over detection limits with MW U-test results (p-values and chi-squared) with a * indicating conservative sediment properties with the inclusion of landslides and ^ indicating significant discrimination with the MW U-test

Element	P-value
Al ₂ O ₃	0.269
CaO	0.075
Fe ₂ O ₃ *	0.000^
MgO	0.253
Na ₂ O	0.107
P ₂ O ₅ *	0.000^
SiO ₂ *	0.000^
TiO ₂ *	0.000^
Ba	0.980
Ce	0.000^
Co	0.000^
Cr	0.263
Cu	0.000^
Dy*	0.107
Er*	0.030^
Eu*	0.819
Gd*	0.576
Ho	0.060
La*	0.000^
Lu	0.000^
Mn*	0.000^
Nd*	0.017^
Ni	0.263
Pr*	0.001^
Rb	0.003^
Sc	0.158
Sm*	0.037
Sr	0.684
Tb*	0.751
Th	0.431
Tm*	0.339
U	0.899
V	0.000^
Y*	0.007^
Yb*	0.004^
Zn*	0.000^
Zr*	0.001^

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Table 2 Lithology of dominant rock types in the Baroon Pocket reservoir catchment with geological tracing potential

Unit Name	Dominant Rock Type	Lithographic Description	Age	Expected Elemental Characteristics
Basalt	Basalt	Mainly basalt flows	Tertiary-Quaternary	Mafic - High: Ca, Fe ₂ O ₃ , MgO Low: Na ₂ O, K ₂ O, SiO ₂
Landsborough Sandstone	Sedimentary	Lithofeldspathic labile and quartzose sandstone, siltstone, shale, minor coal, ferruginous oolite marker	Triassic-Jurassic	Felsic - High: Na ₂ O, K ₂ O SiO ₂ ; Low Ca, Fe ₂ O ₃ , MgO
Wappa Rhyolite	Felsites (Lavas, Clastics & High-Level Intrusives)	Rhyolite	Late-Triassic	Felsic - High: Na ₂ O, K ₂ O SiO ₂ ; Low Ca, Fe ₂ O ₃ , MgO
Amamoor beds	Mixed Sedimentary Rocks and Mafites	Mudstone, slate, basic metavolcanics, chert, schist, jasper, greywacke	Late-Devonian-Carboniferous	Intermediate: Ca, Fe ₂ O ₃ , MgO, Na ₂ O, K ₂ O, SiO ₂
Cedarton Volcanics	Mafites (Lavas, Clastics & High-Level Intrusives)	Basaltic to andesitic lava, volcanoclastic arenite	Permian	Intermediate: Ca, Fe ₂ O ₃ , MgO, Na ₂ O, K ₂ O, SiO ₂
Colluvium	Colluvium	Pediment slope wash, clay, scree, soil	Tertiary-Quaternary	Mixture of the above
Alluvium	Alluvium	Clay, silt, sand, gravel; flood plain alluvium	Quaternary	Mixture of the above

Table 3 Baroon Pocket reservoir catchment and major subcatchments dominant rock types

Catchment	Rock Type	Area (km ²)	%
Baroon Pocket reservoir catchment	Basalt	49.9	73
	Colluvium	6.7	10
	Water	4.0	6
	Landsborough Sandstone	2.9	4
	Wappa Rhyolite	2.0	3
	Amamoor beds	1.1	2
	Alluvium	0.8	1
	Cedarton Volcanics	0.7	1
Total		68.2	100
Walkers Creek	Basalt	7.1	97
	Alluvium	0.2	3
	Total	7.3	100
Obi Obi Creek	Basalt	23.9	88
	Amamoor beds	1.1	4
	Colluvium	0.8	3
	Cedarton Volcanics	0.7	2
	Other (inc. water)	0.6	2
	Landsborough Sandstone	0.1	0
	Total	27.1	100
Lexys Creek	Basalt	2.6	81
	Colluvium	0.6	18
	Other (inc. water)	0.1	2
	Total	3.3	100
Falls Creek	Basalt	3.6	74
	Colluvium	0.9	18
	Landsborough Sandstone	0.3	7
	Other (inc. water)	0.1	1
	Total	4.9	100
Bridge Creek	Basalt	9.7	59
	Colluvium	3.2	20
	Wappa Rhyolite	1.8	11
	Landsborough Sandstone	1.4	8
	Other (inc. water)	0.3	2
	Total	16.4	100

Table 4. Model results from the five different modelling approaches including the MMD (mixing model difference), CE (compiled error) and CTA (contribution-to-area) ratios. The CTA was calculated by excluding the area of the reservoir and merging colluvium with felsic rock types. The Amamoor beds and the Cedarton Volcanics were merged with mafic rock types owing to their location within Obi Obi Creek.

Element	Sediment Cores	MMD	Mafic (%)	CE (%)	Felsic (%)	CE (%)	Mafic CTA	Felsic CTA
Fe ₂ O ₃	All Cores	0.145	50	13	50	10	0.62	2.77
	Far Sediment	0.125	20	7	80	8	0.25	4.45
	Lexys Creek	0.129	67	9	33	10	0.82	1.90
	Obi Obi Creek	0.105	65	10	35	10	0.69	14.48
	Falls Creek	0.102	33	8	67	9	0.44	2.75
	Bridge Creek	0.126	24	7	76	7	0.40	1.96
SiO ₂	All Cores	0.073	25	8	75	8	0.32	4.14
	Far Sediment	0.076	11	6	89	6	0.14	4.92
	Lexys Creek	0.072	46	8	54	8	0.57	3.10
	Obi Obi Creek	0.059	46	9	54	9	0.48	22.45
	Falls Creek	0.068	23	7	77	7	0.31	3.15
	Bridge Creek	0.067	26	7	74	7	0.45	1.89
Fe ₂ O ₃ +SiO ₂	All Cores	0.232	36	9	64	9	0.45	3.55
	Far Sediment	0.216	15	7	85	6	0.18	4.73
	Lexy's Creek	0.217	58	9	42	9	0.72	2.41
	Obi Obi Creek	0.184	54	9	46	9	0.57	19.04
	Falls Creek	0.190	28	6	72	6	0.38	2.95
	Bridge Creek	0.206	23	8	77	8	0.40	1.97
DFA _{0.05}	All Cores	0.917	33	8	67	9	0.41	3.70
	Far Sediment	0.882	17	7	83	7	0.21	4.59
	Lexys Creek	1.313	23	7	77	7	0.29	4.36
	Obi Obi Creek	1.249	62	9	38	8	0.66	15.66
	Falls Creek	1.322	29	7	71	8	0.40	2.89
	Bridge Creek	1.339	19	6	81	6	0.33	2.07
DFA _{2,0.35}	All Cores	3.154	7	7	93	8	0.09	5.16
	Far Sediment	2.637	13	7	87	10	0.16	4.84
	Lexys Creek	3.333	13	8	87	10	0.16	4.97
	Obi Obi Creek	2.901	0	7	100	8	0.00	41.68
	Falls Creek	2.962	2	8	98	12	0.03	4.01
	Bridge Creek	3.573	0	7	100	18	0.00	2.56

Figure Captions:

Fig. 1 Map of Baroon Pocket Reservoir catchment, major subcatchments (grey lines) along with time integrated sampler, sediment core (Inset) and landslide sampling locations

Fig. 2 Scatter plots of major element geochemistry for the time-integrated samplers and landslide samples in the Baroon Pocket Reservoir catchment

Fig. 3 Geologically selected discriminators and Baroon Pocket Reservoir sediment cores (BDC)

Fig. 4 Median proportional contribution for mafic derived (Black) and felsic derived (grey) sediments for the five main models (listed on the y-axis). The dashed line represents the modelled mafic contribution from the geological approach ($\text{Fe}_2\text{O}_3 - \text{SiO}_2$). Model uncertainty is listed as the compiled error in Table 4.

Fig. 5 Median proportional contribution modelled for all conservative elements that significantly discriminate between mafic derived (Black) and felsic derived (grey) sediments with the dashed line representing the modelled mafic contribution with the geological approach (SiO_2 and Fe_2O_3). Model uncertainty is listed as the compiled error in a table provided in the supplementary information.

Figure 1
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