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

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► **To cite this version:**

John Patrick Laceby, Joe McMahon, O. Evrard, Jon Olley. A comparison of geological and statistical approaches to element selection for sediment fingerprinting. *Journal of Soils and Sediments*, 2015, 15 (10), pp.2117 - 2131. 10.1007/s11368-015-1111-9 . hal-01806082

**HAL Id: hal-01806082**

**<https://hal.science/hal-01806082>**

Submitted on 26 May 2020

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1    ADVANCES IN SEDIMENT FINGERPRINTING

2

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

4    **fingerprinting**

5

6    **John Patrick Laceby • Joe McMahon • Olivier Evrard • Jon Olley**

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## 24 Abstract

1 25 **Purpose:** Elevated sediment loads reduce reservoir capacity and significantly increase the cost of  
2  
3 26 operating water treatment infrastructure making the management of sediment supply to reservoirs of  
4  
5 27 increasing importance. Sediment fingerprinting techniques can be used to model the relative  
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7 28 contributions of different sources of sediment accumulating in reservoirs. The goal of this research is  
8  
9 29 to compare geological and statistical approaches to element selection for sediment fingerprinting  
10 30 modelling.

11 31 **Materials and methods:** Time-integrated samplers ( $n=45$ ) were used to obtain source samples from  
12 32 four major subcatchments flowing into the Baroon Pocket Dam in South East Queensland, Australia.  
13 33 The geochemistry of these potential sources were compared to sediment cores ( $n=12$ ) sampled in the  
14 34 reservoir. Elements that provided expected, observed and statistical discrimination between sediment  
15 35 sources were selected for modelling with the geological approach. Two statistical approaches selected  
16 36 elements for modelling with the Kruskal-Wallis H-test and Discriminatory Function Analysis (DFA).  
17 37 In particular, two approaches to the DFA were adopted to investigate the importance of element  
18 38 selection on modelling results. A distribution model determined the relative contributions of difference  
19 39 sources to sediment sampled in the Baroon Pocket Dam.

20 40 **Results and discussion:** Elemental discrimination was expected between one subcatchment (Obi Obi  
21 41 Creek) and the remaining subcatchments (Lexys, Falls and Bridge Creek). Six major elements were  
22 42 expected to provide discrimination. Of these six, only  $\text{Fe}_2\text{O}_3$  and  $\text{SiO}_2$  provided expected, observed  
23 43 and statistical discrimination. Modelling results with this geological approach indicated 36% (+/- 9%)  
24 44 of sediment sampled in the reservoir cores were from mafic-derived sources and 64% (+/- 9%) were  
25 45 from felsic-derived sources. The geological and the first statistical approach differed by only 1% ( $\sigma$   
26 46 5%) for 5 out of 6 model groupings with only the Lexys Creek modelling results differing  
27 47 significantly (35%). The statistical model with expanded elemental selection differed from the  
28 48 geological model by an average of 30% for all 6 models.

29 49 **Conclusions:** Elemental selection for sediment fingerprinting therefore has the potential to impact  
30 50 modeling results. Accordingly we believe it is important to incorporate both robust geological and  
31 51 statistical approaches when selecting elements for sediment fingerprinting. For the Baroon Pocket  
32 52 Dam, management should focus on reducing the supply of sediments derived from felsic sources in  
33 53 each of the subcatchments.

34 54  
35 55 **Keywords** Geochemical fingerprinting • Sediment provenance • Australia • Discriminant Function  
36 56 Analyses  
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## 60 1. Introduction

61 Elevated sediment loads from accelerated catchment erosion pose a significant challenge to  
62 the management of increasingly scarce water resources. Not only does accelerated soil erosion  
63 and the concomitant suspended sediment loads threaten aquatic ecosystems (Bunn et al. 2007;  
64 Bilotta and Brazier 2008), it significantly increases the costs of maintaining and operating  
65 water treatment and transportation infrastructure (Clark 1985; Holmes 1988; Dearthmont et al.  
66 1998). It is important to manage sediment sources directly in order to reduce uncertainty in  
67 the potential operational funding required to maintain and operate viable water supply  
68 networks. Managing sediment supply to waterways requires a detailed understanding of the  
69 nature, location and proportional contributions of different sediment sources (Walling 1983;  
70 Douglas et al. 2003a; Douglas et al. 2007).

71 One approach to understanding sediment provenance is to trace the geochemistry of the  
72 sediment back to its lithogenic sources (Olley and Caitcheon 2000; Douglas et al. 2003b;  
73 Evrard et al. 2011; Navratil et al. 2011; Navratil et al. 2012). This geochemical fingerprinting  
74 technique is based on the fact that different rock types often produce soils and sediments with  
75 unique geochemical compositions (Klages and Hsieh 1975; Wood 1978; Caitcheon et al.  
76 2006). If these unique geochemical compositions are maintained during generation, transport,  
77 and deposition processes, and the geochemistry of sediment sources are indeed distinct, then  
78 the spatial origin of transported sediment may be ascertained (Walling et al. 1993; Olley et al.  
79 2001; Douglas et al. 2005). This current research focuses on the analysis and modelling of  
80 sediment geochemistry to determine the spatial source of sediment. For reviews of sediment  
81 tracing and modelling approaches, see Davis and Fox (2009), Guzmán et al. (2013),  
82 Haddadchi et al. (2013), or Koiter et al. (2013b).

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

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85 development of statistical techniques for element selection (Collins et al. 1996; Collins et al.  
86 2012) and refinements in the modelling processes (Walling et al. 1993; Hughes et al. 2009;  
87 Collins et al. 2010b). This research has established a solid disciplinary foundation for  
88 sediment fingerprinting research. Contrarily, there has not been a comparable level of  
89 research on the geologically-based foundation for elemental discrimination that was central to  
90 the original development of the geochemical fingerprinting technique.

91 The fundamental logic for using elemental compositions to discriminate between spatial  
92 sources is that different rock types are often formed in different conditions and produce  
93 minerals with distinct chemistries. For example, igneous rocks are formed from magma flows  
94 either on the Earth's surface (extrusive) or below ground (intrusive) (Tarbuck and Lutgens  
95 2006; Marsh and Kaufman 2012). When the magma cools the ionic component within the  
96 magma crystallizes into different minerals with the temperatures and the rate of cooling  
97 resulting in rock types with differing mineral compositions (e.g. Bowen's Reaction Series)  
98 (Tarbuck and Lutgens 2006; Marshak 2008; Reynolds et al. 2013). Other processes such as  
99 weathering and erosion (e.g. sedimentary rocks) over longer temporal scales in conjunction  
100 with the differing mineral compositions of rock types results in the unique signatures of soils  
101 and sediments traceable with geochemical fingerprinting techniques. Ultimately, this  
102 geological foundation for understanding differences between rock types is central to  
103 geochemical fingerprinting research.

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

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

## 114 2. Methods

### 115 2.1 Study area

116 The Baroon Pocket reservoir catchment (74 km<sup>2</sup>) is subtropical with mean annual  
117 temperatures ranging between 24°C and 26°C (BOM 2014). The mean annual rainfall in the  
118 catchment is one of the highest in SEQ (1,785mm) with the majority falling during summer  
119 (October to February)(BOM 2014). Since the beginning of European settlement,  
120 approximately 80% of the native vegetation has been cleared. The region has a population of  
121 ~5,000 with the majority located in the urban centre of Maleny.

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

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

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

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

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

## 151 **2.2 Sample collection, processing and analysis**

152 Time-integrated samplers (Phillips et al. 2000) were used to sample sediment and were  
153 deployed ~0.5m above the dry-season level (Fig. 1). 45 samplers were deployed between late  
154 November and early December in 2012 and retrieved between late July and early August in  
155 2013. Sampling location was determined by pragmatic factors such landowner permission and  
156 stream access. Samplers were inspected after each major rainfall event (once in January and  
157 twice in February 2013). After the January event, one sampler lost the front end fitting. In  
158 mid-February, one missing sampler was reinstalled, and in late February, 3 samplers were

159 reinstalled. During the retrieval period, 4 additional samplers were missing, likely the result of  
1  
2 160 vandalism. At one of these locations (BP05-Fig. 1) a lag deposit sample was obtained by  
3  
4 161 compiling 20 scrapes of evident deposited materials with a non-metallic trowel over a 100m  
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7 162 reach.

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9 163 Sediment deposited in the Baroon Pocket reservoir was sampled at 14 locations (Fig. 1)  
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11 164 with a cable hammer coring device. This corer consisted of a 120 cm PVC 50 mm tube fitted  
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14 165 with a one way valve at the top onto which attached a cage containing a sliding weight. The  
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17 166 coring device was lowered into the water and supported on the bottom by one cable and  
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19 167 another cable attached to the weight was used to raise and lower the weight, effectively  
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21  
22 168 hammering the tube into the bed sediments. The sediment core samples were taken  
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24 169 downstream of the inputs of each of the major tributaries (e.g. Obi Obi, Bridge, Lexys and  
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27 170 Falls Creeks). An additional core-grouping was sampled at the far side of the reservoir (BC04,  
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29 171 BC05, and BC09) and is simply referred to as the 'Far' sediment cores. The top 10cm of each  
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32 172 sediment core was subsampled for geochemical analysis. In addition, a landslide which was  
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34 173 originally mapped by Willmott (1983) and subsequently remobilized in 2008 was  
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36 174 opportunistically sampled at two locations during a site inspection with catchment staff. The  
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39 175 landslide was sampled similarly to the lag deposit by compositing 20 scrapes of the recently  
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41 176 evident landslide subsoils with a non-metallic trowel.

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43 177 Sediment generation and transportation processes result in fine particle size classes being  
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46 178 preferentially eroded and less affected by abrasion and disaggregation during sediment  
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49 179 transport (Walling and Woodward 1992; Collins et al. 1997; Dyer and Olley 1999). The  
50  
51 180 resultant fluvial impacts create potential complexities for the direct comparison of the  
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53 181 elemental geochemistry of non-mobilized source soils to transported sediment in waterways  
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56 182 (Koiter et al. 2013b; Smith and Blake 2014). Accordingly, sediments were exclusively  
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58 183 sampled throughout the catchments to facilitate a direct comparison between sediment  
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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.

197

### 198 **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

209 for modelling. Prior to modelling, the selected elements were examined to ensure that their  
1  
2 210 elemental concentrations plotted within the source range to ensure their conservative  
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5 211 behaviour during modelling.  
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7 212 This approach is then compared to a statistical three step procedure (Collins et al. 2010a;  
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10 213 Wilkinson et al. 2013) that first tests for conservative behaviour and second applies the KW  
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12 214 H-test or MW U-test to determine which elements provide a statistical discrimination between  
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15 215 sources. Third, a discriminant function analyses (DFA) selects an optimal group of elements  
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17 216 for modelling.

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19 217 The KW, MW and DFA analyses were performed in R-software (R Development Core  
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22 218 Team 2011) with the K1aR package (Roever et al. 2014). A greedy-wilks test was used for the  
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24 219 DFA that allows for the adjustment of the level of the approximate F-test decision (or  
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27 220 ‘niveau’) that controls the amount of elements selected into the modelling group. Two  
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29 221 niveaux (0.05 & 0.35) were selected, the first to approximate a level of statistical significance  
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31 222 (0.05) and the second to expand the sediment source grouping for comparison (0.35). The  
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34 223 objective of modelling elements selected with the second niveau (0.35) is to examine the  
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36 224 impact of expanded element selection on modelling results.

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39 225 A distribution mixing model (Lacey and Olley accepted) determined the relative  
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41 226 contribution of different sources to in-stream sediment through simultaneously minimising  
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44 227 mixing model difference (MMD):

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

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51 228 where  $n$  is the number of elements included in the model determined by the three-step  
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54 229 selection process;  $C_i$  is the sediment core distribution of element property ( $i$ );  $m$  is the number  
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56 230 of sources in the catchment;  $P_s$  is the source ( $s$ ) contribution; and  $S_{si}$  is the distribution of  
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59 231 element ( $i$ ) in source ( $s$ ).  $P_s$  was modelled as a truncated normal distribution ( $0 \leq x \leq 1$ ) with a  
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232 mixture mean ( $\mu_m$ ) and standard deviation ( $\sigma_m$ ). Normal distributions were modelled for each  
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2 233 source element ( $S_{si}$ ) and in-stream sediment element ( $C_i$ ) (Lacey and Olley accepted). Non-  
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4 234 negative constraints were applied to all source and in-stream elements with the absolute sum  
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7 235 of all proportional contributions ( $P_s$ ) equaling 1.

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10 236 The Optquest algorithm in Oracle's Crystal Ball software (2013) was used to model  
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12 237 distributions throughout a Monte Carlo style modelling framework. In Optquest, the  $\mu_m$  and  
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14 238  $\sigma_m$  for each source's contribution ( $P_s$ ) distribution were repeatedly varied while minimizing  
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16 239 the median MMD when simultaneously solving Eq. 1 2500 times with 2500 random samples  
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19 240 (Latin Hypercube – 500 bins) drawn from each in-stream ( $C_i$ ) and source ( $S_{si}$ ) distribution.  
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22 241 This process of deriving the optimal source contribution mixture ( $P_s$ ) for all 2500 randomly  
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24 242 generated trials was repeated 2500 times. The median proportional contribution ( $P_s$ ) source  
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26 243 from these 2500 additional simulations is reported for each source.

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29 244 Uncertainty for each source's proportional contribution is calculated by summing the  
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31 245 modelled standard deviation of the mixture mean ( $\sigma_m$ ) with the median absolute deviation  
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33 246 (MAD) of this modelled standard deviation for the additional 2500 model simulations and the  
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36 247 MAD of the individual sources median proportional contribution again for the additional 2500  
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39 248 simulations. In summary this uncertainty combines the actual standard deviation modelled for  
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41 249 each source contribution with the MAD of this standard deviation and the MAD of the actual  
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44 250 source contribution for the additional 2500 simulations.

### 46 251 **3. Results**

#### 48 252 **3.1 Expected source discrimination**

50  
51 253 There are five lithology units in the Baroon Pocket reservoir catchment, three of which are  
52  
53 254 distinct subclasses of igneous rocks including mafic (Basalt), felsic (Wappa Rhyolite), and  
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56 255 intermediate rock types (Cedarton Volcanics) (Table 2) (DME 2008). The fourth distinct  
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58 256 lithology is a sedimentary rock type (Landsborough Sandstone). Based on its lithographic  
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257 composition, including lithofeldspathic labile and quartzose sandstone, this sedimentary rock  
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2 258 is likely derived from a felsic igneous rock type. The Amamoor Bed lithology is a mixture of  
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5 259 sedimentary and mafic derived materials.  
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7 260 As mineral crystallization is a result of magma chemical composition and cooling histories  
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10 261 (Tarbuck and Lutgens 2006; Reynolds et al. 2013), the mafic rock types (Basalt) should have  
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12 262 high concentrations of CaO, Fe<sub>2</sub>O<sub>3</sub>, and MgO and low concentrations of Na<sub>2</sub>O, K<sub>2</sub>O, SiO<sub>2</sub>  
13  
14 263 (Marshak 2008). Contrarily, the felsic and felsic-derived rock types (Wappa Rhyolite and  
15  
16 264 Landsborough sandstones) should have high concentrations of Na<sub>2</sub>O, K<sub>2</sub>O, and SiO<sub>2</sub> and low  
17  
18  
19 265 concentrations of CaO, Fe<sub>2</sub>O<sub>3</sub>, and MgO (Marshak 2008). The Amamoor Beds and the  
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22 266 Cedarton Volcanics should consist of an intermediate mixture of the mafic and felsic rock  
23  
24 267 types.  
25

26 268 The major element geochemistry of sediment samples are expected to discriminate  
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28  
29 269 between mafic (Basalt) and felsic-derived (Landsborough Sandstone and Wappa Rhyolite)  
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31 270 rock types and potentially the Amamoor beds and Cedarton Volcanics depending on their  
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34 271 degree of intermediacy. There may also be geochemical discrimination for colluvium and  
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36 272 alluvium depending on their lithogenic origin, weathering and transport as these potential  
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39 273 sediment sources were originally derived from hillslope and riverine erosion processes,  
40  
41 274 respectively  
42

43 275 The dominant lithology in the Baroon Pocket catchment is Basalt (73%) (Table 3).  
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46 276 Colluvium underlies 10% followed by Landsborough Sandstone (4%), Wappa Rhyolite (3%),  
47  
48  
49 277 Amamoor beds (2%), Cedarton Volcanics (1%), and alluvium (1%). The major streams  
50  
51 278 draining into the Baroon Pocket reservoir are predominantly underlain by Basalt. Walkers  
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53 279 Creek has the highest Basalt composition (97%), followed by Obi Obi (88%), Lexys (81%),  
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56 280 Falls (74%) and Bridge Creek (59%) (Table 3). As there are only 2 samples from Walkers  
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58 281 Creek, which flows directly into Obi Obi Creek, these catchments were merged for the  
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282 following analysis and discussion. Colluvium comprises the remaining dominant rock type in  
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2 283 Lexys Creek (18%) and Falls Creek (18%). No Landsborough Sandstone was present in  
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4 284 Lexys Creek compared to 7% in Falls Creek. In Bridge Creek, colluvium was again the  
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7 285 second major dominant rock type (20%), followed by Wappa Rhyolite (11%) and  
8  
9 286 Landsborough Sandstone (8%). The Amamoor Beds (4%) and Cedarton Volcanics (2%)  
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11 287 constitute small sections of Obi Obi Creek. Alluvium underlie 3% of Walkers Creek though  
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13  
14 288 <1% for the remainder of the catchments. Based on lithology, major element geochemistry  
15  
16 289 should discriminate between sediment sampled in catchments dominated with mafic rock  
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19 290 types (Obi Obi Creek) compared to catchments with higher felsic derived rock type  
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21 291 compositions (e.g Bridge Creek). Depending on the dominant sources (e.g. mafic or felsic  
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23 292 derived), Falls and Lexys sediment may plot between mafic and felsic-derived rock types.  
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26 293

### 29 294 **3.2 Observed source discrimination**

31 295 Scatterplots of the major trace geochemistry confirm discrimination between mafic (Obi  
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33 296 Obi Creek) and felsic-derived sediments (Bridge, Falls and Lexys Creek) (Fig. 2). Mafic  
34  
35 297 derived sediments have high  $\text{Fe}_2\text{O}_3$  and low  $\text{K}_2\text{O}$  and  $\text{SiO}_2$  as expected. In contrast, felsic  
36  
37 298 sediment have high  $\text{K}_2\text{O}$ ,  $\text{SiO}_2$  and low  $\text{Fe}_2\text{O}_3$ . Bridge, Falls and Lexys Creek all plot  
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39 299 similarly indicating non-mafic sediment source origins. The two landslide samples were taken  
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41 300 from colluvium (Fig. 1). The increased colluvium area within Bridge, Falls and Lexys Creek  
42  
43 301 in combination with the landslide sample geochemistry indicates the colluvium is most likely  
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45 302 derived from elevated felsic-derived sources. The Amamoor Beds and Cedarton Volcanics are  
46  
47 303 likely too small to contribute noticeably to Obi Obi Creek sediment or are comprised of  
48  
49 304 predominantly mafic sources. Accordingly, these two lithologies will be considered mafic  
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51 305 sources.  
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306 The expected elemental geochemistry resulted in two observed source groupings: mafic  
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2 307 derived sediments (Obi Obi Creek) and felsic-derived sediments (Lexys Creek, Bridge Creek,  
3  
4 308 Falls Creek). Although it may be possible to further discriminate between the felsic source  
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6  
7 309 group, the elemental overlap and particularly the wide range of Falls Creek and Bridge Creek  
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9  
10 310 sediment, makes further felsic source discrimination impractical. Based on the geochemistry,  
11  
12 311 the best approach is to model these two sediment source groupings to determine the spatial  
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14 312 origin of the sediment core samples from the Baroon Pocket reservoir. Further, the felsic-  
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17 313 derived grouping is not representative of pure felsic material. The three ‘felsic-derived’  
18  
19 314 catchments (Lexys, Bridge and Falls Creek) are predominantly underlain by the mafic basalts  
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21  
22 315 resulting in an intermediate signature for these ‘felsic’ catchments. Pure felsite would have  
23  
24 316  $\text{SiO}_2$  compositions  $>68\%$  (Marshak 2008). Nonetheless, as these catchments have a greater  
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26  
27 317 felsic-derived signature we will refer to this grouping as felsic in the following results and  
28  
29 318 discussion.

31  
32 319 There were limited differences between mafic and felsic derived sediments for CaO, MgO,  
33  
34 320 and  $\text{Na}_2\text{O}$ . For  $\text{Na}_2\text{O}$ , this could be related to its non-conservative nature during fluvial  
35  
36 321 transport (Kraushaar et al. 2014). For CaO and MgO we are unaware of the reason for the lack  
37  
38  
39 322 of observed discrimination. There was also observed elemental discrimination between felsic  
40  
41 323 and mafic derived sediments with  $\text{P}_2\text{O}_5$  and  $\text{TiO}_2$ . The elevated  $\text{P}_2\text{O}_5$  could be related to the  
42  
43  
44 324 high concentration of dairy and orchard operations in the predominantly mafic Obi Obi  
45  
46 325 catchment compared the felsic areas lower in the catchment that have more vegetative cover.  
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48  
49 326  $\text{P}_2\text{O}_5$  also has dissolved phases indicative of potential non-conservative behaviour (Smith and  
50  
51 327 Blake 2014).  $\text{TiO}_2$  has low soil mobility and high weathering resistance and, according to  
52  
53 328 Koiter et al., (2013a), the higher values of  $\text{TiO}_2$  in the mafic sediments may indicate that they  
54  
55  
56 329 are eroded from a higher position in the soil profile. Conversely, Smith and Blake (2014)

1  
2 330 reported poor source discrimination with  $\text{TiO}_2$ , particularly between surface and subsurface  
3 331 sources.

4  
5 332 Multiple elements from the sediment core samples plot below or above the source  
6  
7 333 sediment (Fig. 2). The sediment core samples have higher  $\text{Al}_2\text{O}_3$ , and lower  $\text{CaO}$ ,  $\text{Na}_2\text{O}$ ,  $\text{TiO}_2$   
8  
9 334 and to a lesser extent –  $\text{K}_2\text{O}$  and  $\text{MgO}$ . For the geological discriminators,  $\text{K}_2\text{O}$  is affected as 4  
10  
11 335 core samples plot below the source range excluding this element from modelling. The Baroon  
12  
13 336 Pocket reservoir sediment core samples do plot on a mixing line between the mafic and felsic  
14  
15 337 source samples for  $\text{Fe}_2\text{O}_3$  and  $\text{SiO}_2$ . As these elements together comprise on average 65% ( $\sigma$   
16  
17 338 6%) of the total elemental composition of stream sediment samples, they are selected as the  
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19 339 elements for modelling.

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21  
22 340  $\text{Fe}_2\text{O}_3$  and  $\text{SiO}_2$  significantly discriminate between felsic and mafic sources (Table 1). As  
23  
24 341 there are only two main sources, only one element is required for modelling. Accordingly,  
25  
26 342  $\text{Fe}_2\text{O}_3$  and  $\text{SiO}_2$  were modelled individually and then simultaneously for all sediment core  
27  
28 343 samples, together as one grouping, as well as for each subcatchment sediment core grouping  
29  
30 344 and the ‘Far’ sediment cores. The Far sediment core grouping, Bridge Creek and Falls Creek  
31  
32 345 all plot clearly within the felsic derived sediment sources (Fig. 3). Both Obi Obi and Lexys  
33  
34 346 Creek plot between the felsic and mafic sources, indicative of a mixture of sources and a  
35  
36 347 higher than expected contribution of felsic sources in sediment cores near the outlet of Obi  
37  
38 348 Obi Creek.

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### 42 350 **3.3 Statistical source discrimination**

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44 351 As multiple Baroon Pocket reservoir sediment core samples plot outside of the source  
45  
46 352 range, multiple elements were observed not to be conservative. Indeed, without the sampling  
47  
48 353 of the 2 landslides, 29, or almost 75% of the elements would not be conservative. With the  
49  
50 354 inclusion of the landslides, 19 elements or ~50% would not be conservative. High numbers of  
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355 non-conservative elements typically indicate a missing source or other factors affecting  
1 sediment geochemistry during mobilization, transportation and deposition processes. The  
2 356  
3 effect of the inclusion of the two landslide samples on conservative behaviour suggest that  
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### 27 366 **3.4 Modelling results**

29 367 For all Baroon Pocket sediment core samples, the  $\text{Fe}_2\text{O}_3$  only model indicated that  $50\pm 13\%$   
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31 368  
32 of all sediment has a mafic origin compared to only  $20\pm 7\%$  for the far sediment core grouping  
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380 model ( $36\pm 6\%$ ) was equivalent to the mean of all subcatchment mafic derived sediments  
381 ( $36\% \sigma 19\%$ ).

382 The DFA<sub>0.05</sub> model produced similar results to the Fe<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> model with the exception of  
383 Lexys Creek for which only  $23\pm 7\%$  of sediment was modelled to derive from mafic origins  
384 compared to  $58\pm 9\%$  for Fe<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> model. For the remainder of the models, there was a 1%  
385 ( $\sigma 5\%$ ) mean difference between the DFA<sub>0.05</sub> and the Fe<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> models. Conversely, the  
386 DFA<sub>0.35</sub> model results differed considerably from the previous 4 models. The DFA<sub>0.35</sub> model  
387 indicated that on average only 6% ( $\sigma 7\%$ ) of sediments were derived from mafic sources with  
388 the highest mafic contributions modelled for the far sediment core grouping ( $13\pm 8\%$ ) and  
389 Lexys Creek ( $13\pm 8\%$ ) whereas no mafic contributions were modelled for Obi Obi Creek.  
390 Compared to the consistency of the previous four models, the DFA<sub>0.35</sub> model appears  
391 inaccurate.

392 To demonstrate the underlying complexity with the DFA<sub>0.35</sub> model, all elements that  
393 significantly discriminated between mafic and felsic sources were modelled separately.  
394 Quickly it becomes evident that the light rare earth elements (REEs) from the Cerium Group  
395 (e.g. La, Pr, Nd, & Sm) perform differently in the modelling process (Fig. 5). In fact, the  
396 average modelling results for the 6 sediment core groupings for the light REEs differs from  
397 the Fe<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> model by 55%. The light REE models results were predominantly 100%  
398 mafic or 100% felsic. The heavy REEs (Yb, Y, & Er) of the Yttrium group performed  
399 marginally better with an average difference of 32% for all the 6 different models. Although  
400 Zr was selected for the first DFA<sub>0.05</sub>, it differed from the Fe<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> model by 39% for all 6  
401 models. Zn differed by 35%, and Mn by 26%. The major oxide elements performed best with  
402 TiO<sub>2</sub> differing by 22%, P<sub>2</sub>O<sub>5</sub> by 10% and both SiO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> by 7%. Indeed the DFA<sub>0.05</sub>  
403 achieved a balance between Zr and TiO<sub>2</sub> that likely resulted in the similarity to the Fe<sub>2</sub>O<sub>3</sub>-  
404 SiO<sub>2</sub> model. The DFA<sub>0.35</sub> model was likely affected by the inclusion of Mn, La and Zn and

405 could not longer produce comparable results to the other four models. The varying model  
1  
2 406 results for all conservative elements and the DFA<sub>0.35</sub> model highlights both the underlying  
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4 407 complexity involved in modelling sediment sources and the importance of element selection  
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7 408 in sediment fingerprinting research.  
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#### 11 410 **4. Discussion**

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14 411 The Baroon Pocket reservoir catchment is a complicated tracing environment highlighted  
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17 412 by the non-conservative behaviour of multiple elements. There are two main likely factors for  
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19 413 this non-conservative behaviour. First, there is either a missing or undersampled source. The  
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22 414 impact of the inclusion of two landslide source samples indicates that the missing source is  
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24 415 likely these landslides. Second, there was a shift in sediment properties between the riverine  
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27 416 and the reservoir environments noted by an increase in Al<sub>2</sub>O<sub>3</sub> and a decrease of TiO<sub>2</sub>, Na<sub>2</sub>O,  
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29 417 and to a lesser extent, K<sub>2</sub>O. One reason for this transition could be chemical additions (e.g.  
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31  
32 418 Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) to the reservoir to treat algal blooms. These additions may impact sediment  
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34 419 elemental geochemistry. The time integrated samplers may have also affected sampled  
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36 420 sediment properties as Smith and Owens (2014) reported these devices may sample sediment  
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39 421 with potentially different particle size and geochemical compositions than sediment  
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41 422 transported in the catchment. Further there could be significant particle size and density  
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44 423 sorting effects during transport and deposition in the reservoir. Although we attempt to  
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46 424 address potential effects of particle sorting by isolating the <10 µm particle size fraction it is  
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49 425 likely that both particle size and density sorting in the reservoir resulted in further separation  
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51 426 of the fine and potentially dense mineral components.

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53 427 Owing to these complications, a geological approach was developed for elemental  
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56 428 selection for sediment fingerprinting modelling and compared to statistical approaches. The  
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58 429 foundation of the statistical approach to sediment fingerprinting is that the KW/DFA analyses  
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2 431 selects a composite suite of sediment properties that provide the optimal combination of  
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4 432 elements for source ascription (Collins et al. 1996; Collins and Walling 2002). The challenge  
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6 433 with statistical approaches, as highlighted by Owens et al., (2006), is that they may select  
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8 434 elements that are potentially inappropriate for sediment fingerprinting modelling. Further,  
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10 435 Smith and Blake (2014) concluded that statistical approaches should not substitute for a  
11  
12 436 detailed understanding of both the chemical and physical foundation of individual tracer  
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15 property behaviour.

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17 437 Our research demonstrates that it is important to incorporate both statistical and geological  
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19 438 approaches when selecting elements for sediment fingerprinting modelling. For example, in 5  
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21 439 out of the 6 models there was only an average difference of 1% ( $\sigma$  5%) between the  
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23 440 geological ( $\text{Fe}_2\text{O}_3\text{-SiO}_2$ ) and statistical ( $\text{DFA}_{0.05}$ ) modelling approaches (Fig. 4). The  
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25 441 similarity between these models provides an increased confidence in results compared to  
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27 442 relying on the results from either the statistical or the geological approach. Although this  
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29 443 demonstrates the potential of both approaches, one catchment's modelling results differed by  
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31 444 35% which is significant if modelling results are used to inform management interventions.  
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33 445 When expanding source selection with the statistical approach ( $\text{DFA}_{0.35}$ ), the source ascription  
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35 446 results differed by 25% from the  $\text{DFA}_{0.05}$ , and 30% from the geological model indicating the  
36  
37 447 potential influence of element selection on modelling results. The utility of using both  
38  
39 448 geological and statistical approaches is that together they provide more certainty to modelling  
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41 449 results whilst incorporating the geological foundation for discrimination fundamental to the  
42  
43 450 fingerprinting technique. Future research should investigate statistical and geological  
44  
45 451 approaches to element selection with artificially generated mixtures similarly to Haddadchi et  
46  
47 452 al. (2014) to quantify the influence of elemental selection on the modelling results.

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49 453 Analyses of in-stream sediment geochemistry in the Baroon Pocket reservoir catchment  
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51 454 highlighted two distinct sediment lithogenic groupings: mafic and felsic. For the Baroon  
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455 Pocket reservoir, it would be expected that sediment source contributions would relate  
456 relatively to dominant rock type area. This would result in 81% of sediment being derived  
457 from mafic rock types and only 18% from felsic rock types. When examining all sediment  
458 cores with the  $\text{Fe}_2\text{O}_3\text{-SiO}_2$  model, only 36% of sediment were derived from mafic and 64%  
459 from felsic areas resulting in a felsic contribution-to-area (CTA) ratio of 3.55. This  
460 enrichment in sediment derived from felsic origin indicates Landsborough Sandstone,  
461 Colluvium or Wappa Rhyolite or combinations of the three contribute more sediment per unit  
462 area to the reservoir. Sediments from all catchment groupings had enriched felsic CTA ratios  
463 with the highest being for the Obi Obi creek (19), with only 2% of the area being felsic and  
464 contributing ~46% of sediments. Lexys (2), Falls (3) and Bridge (2) Creek sediment cores  
465 were all enriched in felsic derived sediment.

466 In-stream sediment  $^{137}\text{Cs}$  activity concentrations reveal that sediment are derived from  
467 95% ( $\sigma$  12%) subsoil sources (Kemp et al. 2014) indicating that channel, gully or landslide  
468 erosion is the dominant sediment source from these felsic regions. In addition, the significant  
469 difference of including the landslides on element conservative behaviour further suggests the  
470 landslides are a potential leading source of sediment in these catchments. More research is  
471 required to isolate landslides as the geochemically proven source of sediment to the reservoir  
472 and determine which elements can significantly discriminate between colluvium, Wappa  
473 Rhyolite and Landsborough Sandstone. It is also important to confirm whether colluvium has  
474 a felsic lithology. Accordingly, obtaining at minimum 30 source samples from the landslides  
475 and each of major rock type would provide a more detailed understanding of the spatial  
476 sources of sediment and their elemental variability in this catchment. Further, a geochemical  
477 analyses of sediment core samples near the water supply intake could identify the most  
478 problematic sediment sources. Fallout radionuclide or optically stimulated luminescence  
479 dating of the sediment cores could also determine source variability over time and provide

1 480 rates of sediment accumulation. Indeed, a long-term sampling program is necessary to  
2 481 understand potential inter and intra annual sediment source variations.

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## 6 7 483 **5. Conclusions**

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10 484 A geological approach determined the sediment source contributions from two dominant  
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12 485 lithologies in the Baroon Pocket reservoir catchment. The results indicate that felsic rock  
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14 486 types contribute ~3.5 times more sediment than their unit area to the Baroon Pocket reservoir.  
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17 487 As a separate analysis of sediment  $^{137}\text{Cs}$  activity concentrations indicated that subsoil sources  
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19 488 dominate in this catchment, management should focus on reducing the supply of sediment  
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22 489 from felsic channel, gully and landslide sources. The impact of landslide samples on the  
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24 490 conservative nature of elements suggests that landslides may be one of the dominant sediment  
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27 491 sources in this catchment. A second round of sampling specifically targeting landslides,  
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29 492 colluvium and the dominant rock type sources would improve both our geological and our  
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31 493 sediment source understanding in the Baroon Pocket reservoir catchment.

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34 494 The geological and  $\text{DFA}_{0.05}$  approaches differed by only 1% ( $\sigma$  5%) for 5 out of 6 model  
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36 495 groupings with only the results for Lexys Creek differing significantly (35%). The  $\text{DFA}_{0.35}$   
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38  
39 496 model differed from the geological model by on average 30% for all 6 models. These results  
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41 497 demonstrate that element selection may significantly impact sediment fingerprinting results.  
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43  
44 498 Accordingly we believe it is important to incorporate both statistical and geological  
45  
46 499 approaches for element selection in sediment fingerprinting modelling.

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49 500 As the sediment fingerprinting discipline has seen significant advancement with modelling  
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51 501 techniques, we believe a similar focus on the geological potential for element selection and  
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53 502 source discrimination is needed along with further research examining the impact of element  
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56 503 selection with artificial mixtures. Although an underlying premise of the composite  
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58 504 fingerprinting technique is that the inclusion of more elements is beneficial to sediment  
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1 505 tracing research (Peart and Walling 1986; Collins and Walling 2002), we believe that it may  
2 506 be as important to understand the utility of a few meaningful elements and their geological  
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4 507 foundation for discrimination rather than solely relying on statistical techniques for  
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7 508 determining elemental discrimination.

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24 515 **Acknowledgments** We would formally like to thank Mark Amos of the Lake Baroon  
25  
26 516 Catchment Care Group for assistance contacting landowners; Justine Kemp for field work  
27  
28  
29 517 assistance and advice on the catchment geomorphology; Tanya Ellison, Michael White and  
30  
31 518 Nina Saxton for field work assistance; the Baroon Pocket reservoir Seqwater catchment staff  
32  
33  
34 519 (Tim Odgers, Andrew Smolders, Sean Gibson) for their support and field work assistance;  
35  
36 520 and the landowners who granted us access for this project. This research was funded by  
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39 521 Seqwater.

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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

<b>Element</b>	<b>P-value</b>
Al2O3	0.269
CaO	0.075
Fe2O3*	0.000^
MgO	0.253
Na2O	0.107
P2O5*	0.000^
SiO2*	0.000^
TiO2*	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 <sub>2</sub> O <sub>3</sub> , MgO Low: Na <sub>2</sub> O, K <sub>2</sub> O, SiO <sub>2</sub>
Landsborough Sandstone	Sedimentary	Lithofeldspathic labile and quartzose sandstone, siltstone, shale, minor coal, ferruginous oolite marker	Triassic-Jurassic	Felsic - High: Na <sub>2</sub> O, K <sub>2</sub> O SiO <sub>2</sub> ; Low Ca, Fe <sub>2</sub> O <sub>3</sub> , MgO
Wappa Rhyolite	Felsites (Lavas, Clastics & High-Level Intrusives)	Rhyolite	Late-Triassic	Felsic - High: Na <sub>2</sub> O, K <sub>2</sub> O SiO <sub>2</sub> ; Low Ca, Fe <sub>2</sub> O <sub>3</sub> , MgO
Amamoor beds	Mixed Sedimentary Rocks and Mafites	Mudstone, slate, basic metavolcanics, chert, schist, jasper, greywacke	Late-Devonian-Carboniferous	Intermediate: Ca, Fe <sub>2</sub> O <sub>3</sub> , MgO, Na <sub>2</sub> O, K <sub>2</sub> O, SiO <sub>2</sub>
Cedarton Volcanics	Mafites (Lavas, Clastics & High-Level Intrusives)	Basaltic to andesitic lava, volcanoclastic arenite	Permian	Intermediate: Ca, Fe <sub>2</sub> O <sub>3</sub> , MgO, Na <sub>2</sub> O, K <sub>2</sub> O, SiO <sub>2</sub>
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

<b>Catchment</b>	<b>Rock Type</b>	<b>Area (km<sup>2</sup>)</b>	<b>%</b>
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 <sub>2</sub> O <sub>3</sub>	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 <sub>2</sub>	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 <sub>2</sub> O <sub>3</sub> +SiO <sub>2</sub>	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 <sub>0.05</sub>	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 <sub>20.35</sub>	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

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4 **Figure Captions:**  
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6 **Fig. 1** Map of Baroon Pocket Reservoir catchment, major subcatchments (grey lines) along with time integrated  
7 sampler, sediment core (Inset) and landslide sampling locations  
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9 **Fig. 2** Scatter plots of major element geochemistry for the time-integrated samplers and landslide samples in the  
10 Baroon Pocket Reservoir catchment  
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12 **Fig. 3** Geologically selected discriminators and Baroon Pocket Reservoir sediment cores (BDC)  
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14 **Fig. 4** Median proportional contribution for mafic derived (Black) and felsic derived (grey) sediments for the  
15 five main models (listed on the y-axis). The dashed line represents the modelled mafic contribution from the  
16 geological approach ( $\text{Fe}_2\text{O}_3 - \text{SiO}_2$ ). Model uncertainty is listed as the compiled error in Table 4.  
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19 **Fig. 5** Median proportional contribution modelled for all conservative elements that significantly discriminate  
20 between mafic derived (Black) and felsic derived (grey) sediments with the dashed line representing the  
21 modelled mafic contribution with the geological approach ( $\text{SiO}_2$  and  $\text{Fe}_2\text{O}_3$ ). Model uncertainty is listed as the  
22 compiled error in a table provided in the supplementary information.  
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Figure 1  
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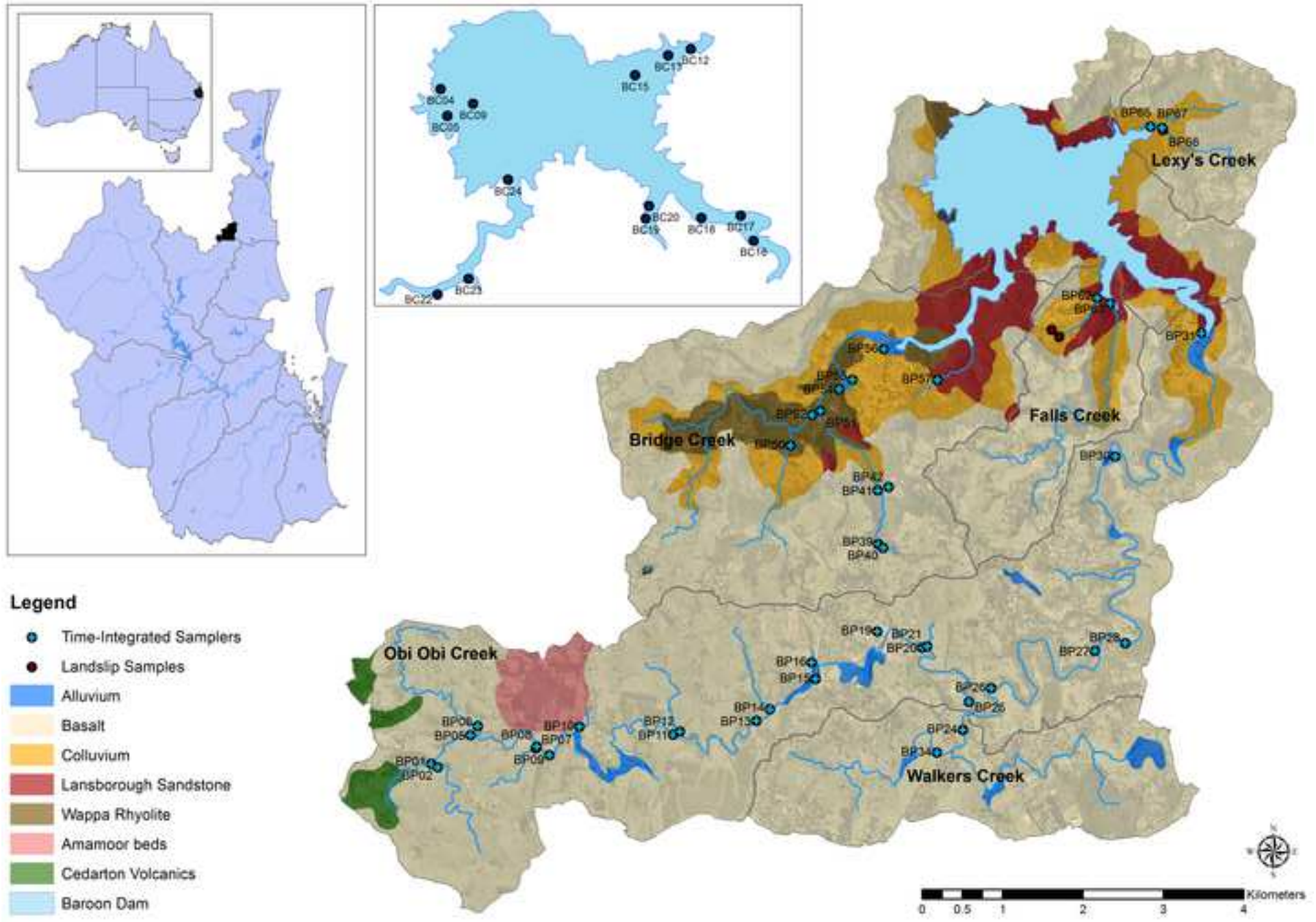


Figure 2  
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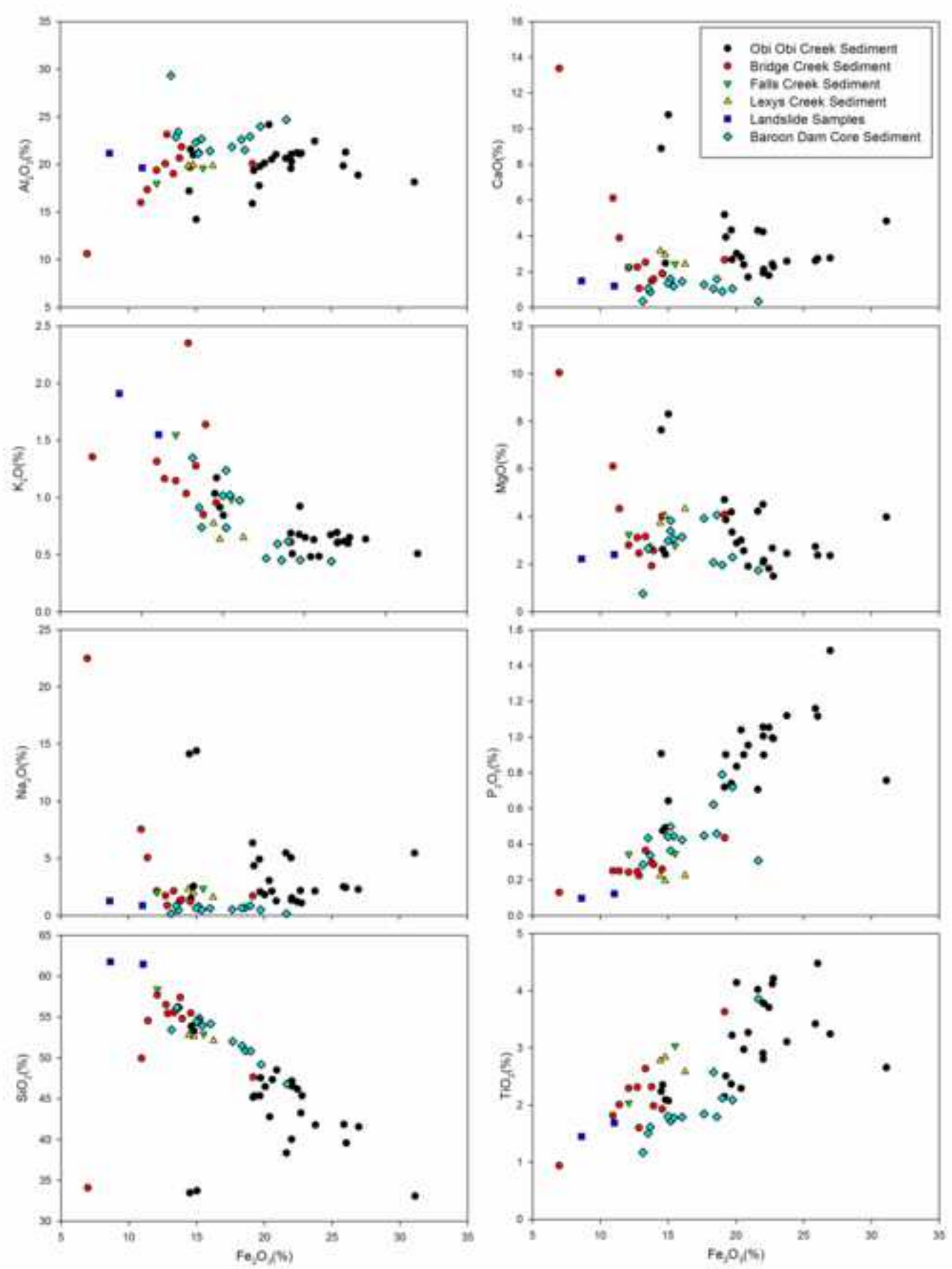




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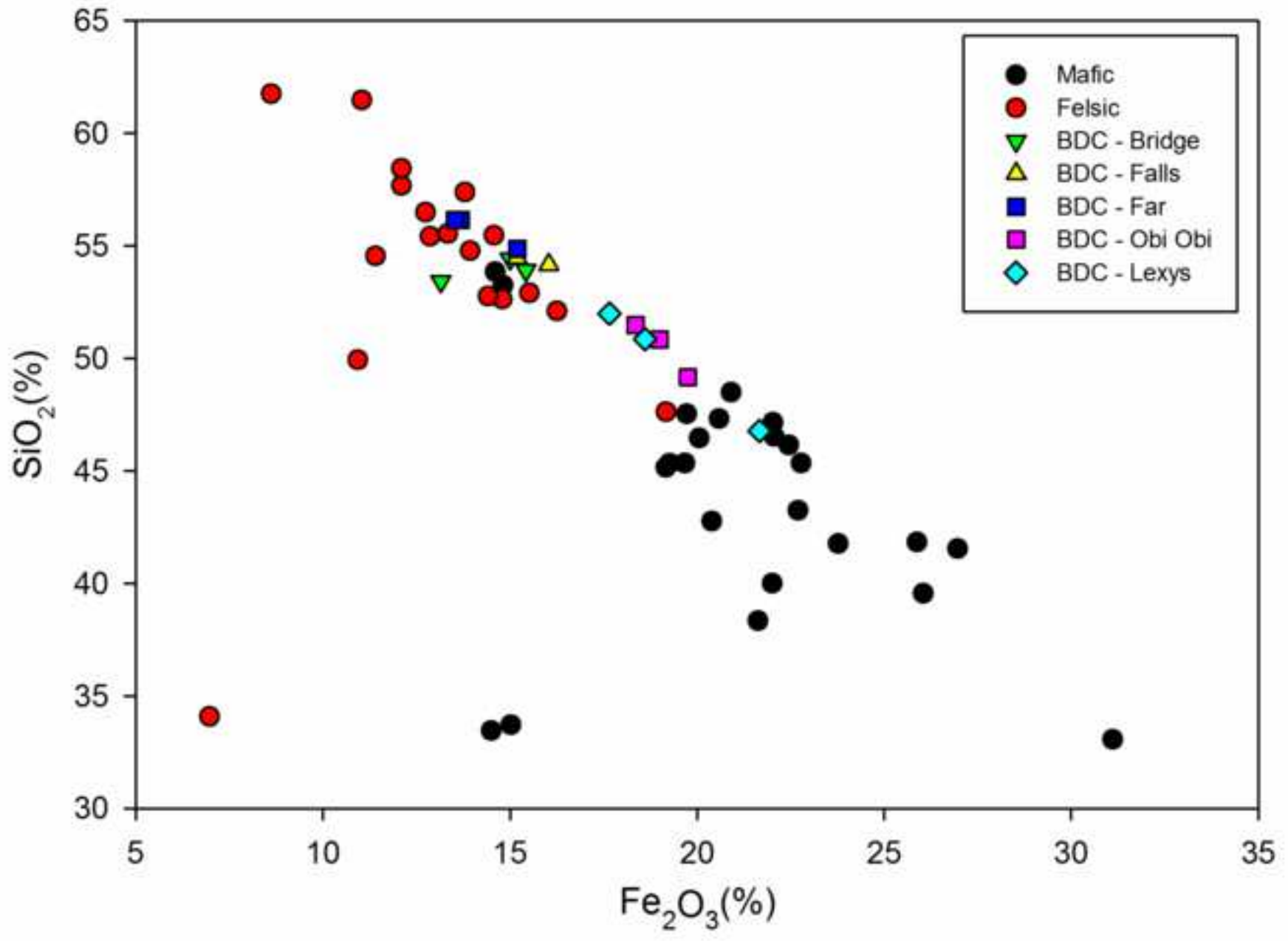


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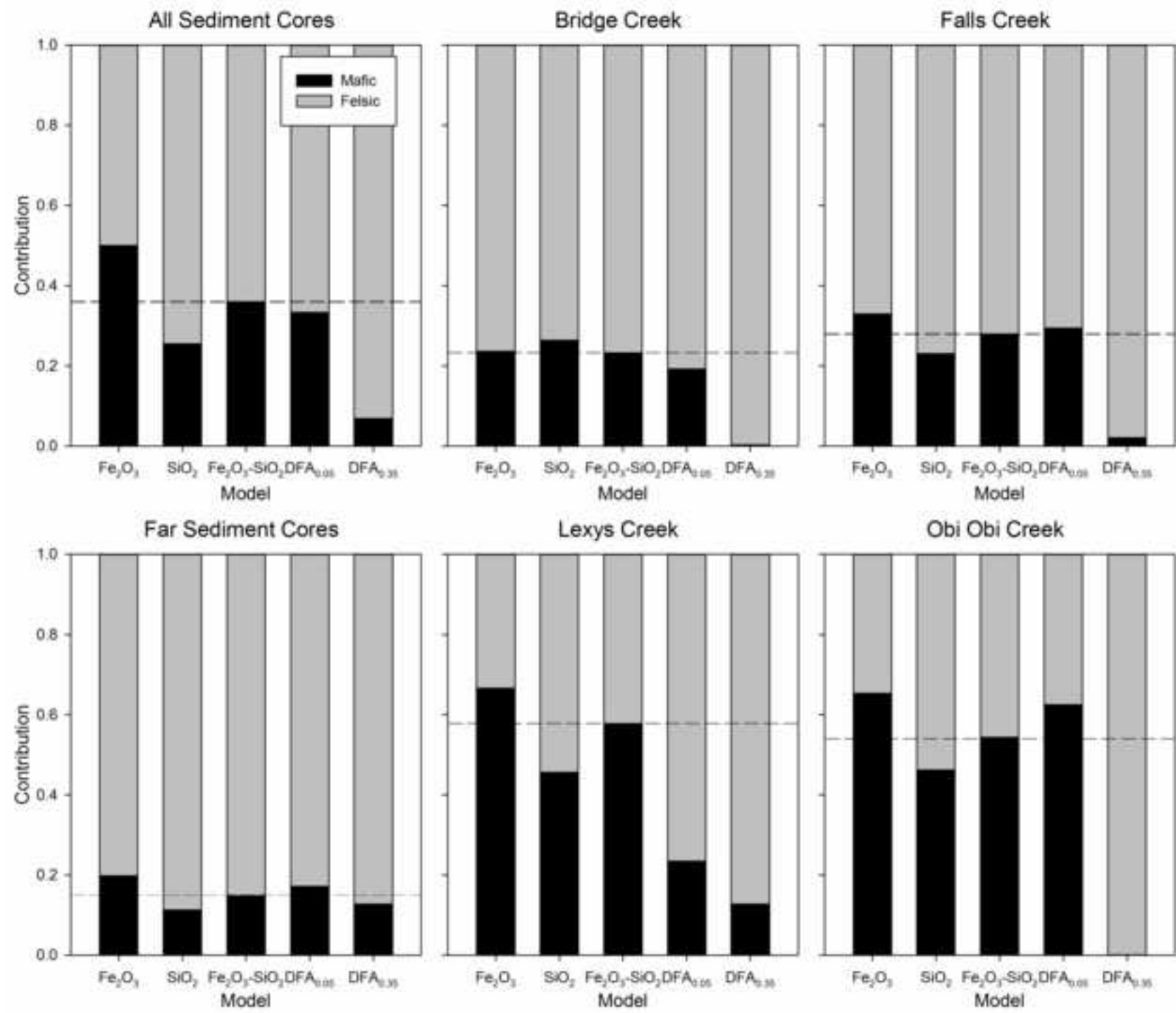


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