



Tracing the sources of sediment and associated particulate nitrogen from different land uses in the Johnstone River catchment, Wet Tropics, north-eastern Australia



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ABSTRACT

While the ecosystem of the Great Barrier Reef (GBR), north-eastern Australia, is being threatened by the elevated levels of sediments and nutrients discharged from adjacent coastal river systems, the source of these detrimental pollutants are not well understood. Here we used a combined isotopic ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) and geochemical (Zn, Pt and S) signatures and stable isotope analysis in R (SIAR) mixing model to estimate the contribution of different land uses to the sediment and associated particulate nitrogen delivered to the Johnstone River. Results showed that rainforest was the largest contributor of suspended and bed sediments in the river estuary (both 33.1%), followed by banana (26.7%, 20.4%), sugarcane (21.5%, 21.4%) and grazing (18.7%, 25.1%). However, bananas and sugarcane land uses had the highest contribution to sediments delivered to the coast per unit of area. This will help land managers to prioritise on-ground activities to improve water quality in the GBR lagoon.

1. Introduction

The Great Barrier Reef (GBR) of Queensland, Australia, is the largest protected coral reef ecosystem on Earth and is bordered by a catchment of 423,000 km² (Furnas, 2003). The ecosystem of the GBR is threatened by increasing levels of nutrients and sediment discharged from adjacent coastal river systems (Brodie et al., 2012). Nitrogen enters water in both inorganic and organic forms. The primary inorganic forms of N are ammonia, ammonium, nitrate, and nitrite, while organic N sometimes makes up a significant fraction of soluble and particulate N in natural waters (Inamdar et al., 2015). Particulate N is attached to mineral particles and is associated with sediment losses to water (Inamdar et al., 2015). The understanding of the impact of poor water quality on ecosystem health has been increasingly improved. Elevated delivery of sediment and associated particulate nitrogen (N) to marine environment is considered as the main threats to the water quality in the GBR (Wallace et al., 2016). Increased sediments and nutrients flowing into inshore areas, can cause a range of impacts including higher algal

growth (Bartley et al., 2017). Changes in water quality affect the biodiversity and resilience of reef systems. Higher levels of chlorophyll and lower water clarity indicate higher concentration levels of pollutants, such as suspended sediments and N, which lead to more algae and less coral diversity (Wallace et al., 2015; Bartley et al., 2017). In these conditions, algae take over and reduce the chances for new hard corals to establish and grow (Wallace et al., 2015). The coastal zones, especially areas close to river mouths, are particularly exposed to this runoff. Recent modelling has shown that on average 9398 kt of fine sediment and 48.3 kt of total nitrogen (TN) are discharged to the GBR annually, with estimated 7399 kt and 23.4 kt of these loads, respectively, being considered anthropogenic (McCloskey et al., 2017). In order to improve the health and resilience of the GBR, the Australian and Queensland Governments have implemented the Reef Water Quality Protection Plan designed to reduce loads of fine sediment and nutrients (with a specific focus on N) exported to the GBR through improved management practices and the development of end-of-basin targets for pollutant loads.

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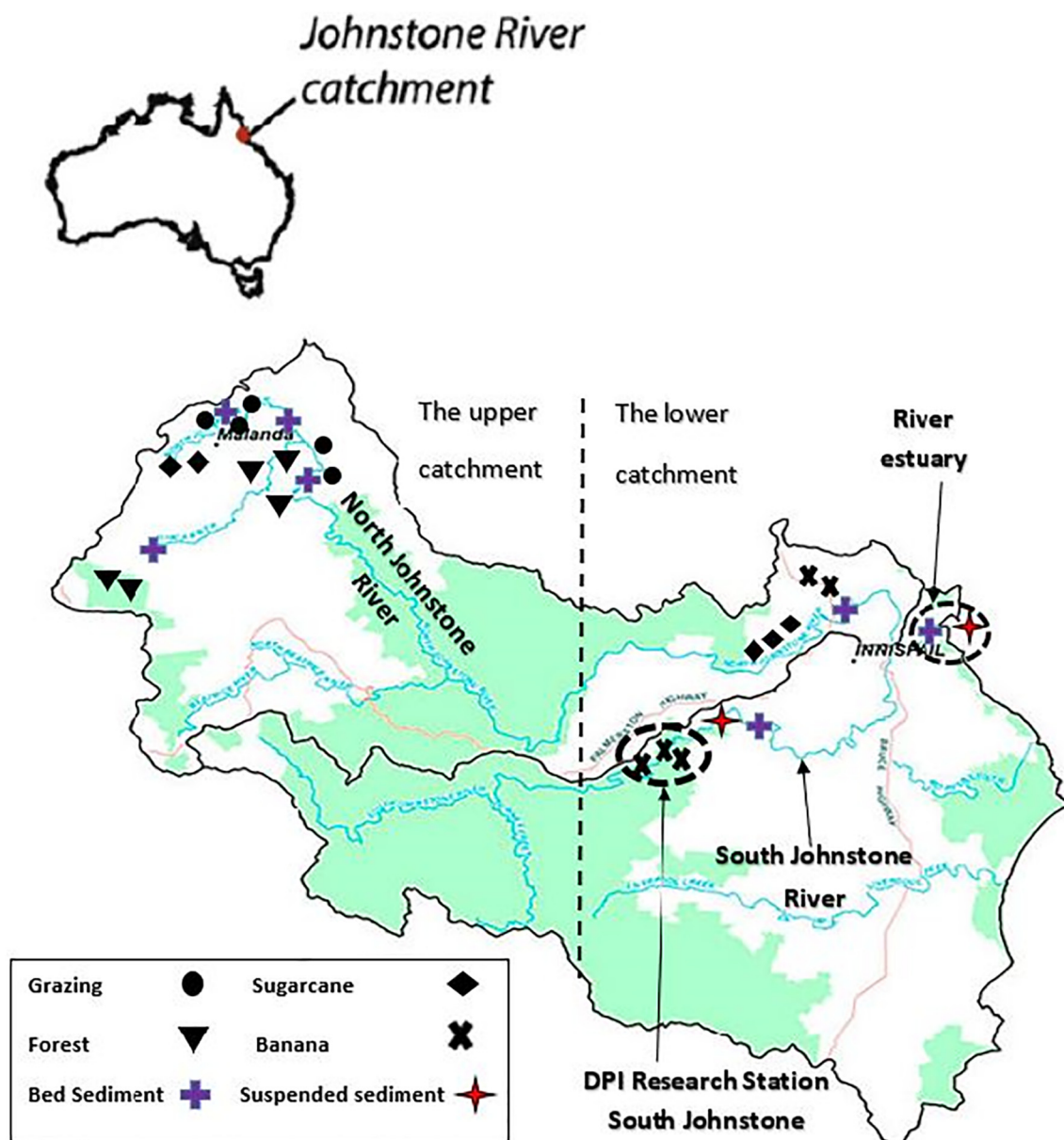


Fig. 1. Location of the study region, soil and sediment sampling sites in the upper and lower Johnstone catchment as well as the Johnstone River estuary, north-east Queensland, Australia (DPI: department of primary industries).

The Wet Tropics region contributes a large proportion of TN loads (31%) exported to the GBR despite comprising only 5.1% of the total GBR catchment area (McCloskey et al., 2017; Hateley et al., 2014a, 2014b). Catchment modelling also suggested that the Wet Tropics region contributed the highest proportion of both dissolved inorganic N (DIN: 42.5%) and particulate N (PN: 31.5%) discharged to the GBR (McCloskey et al., 2017). While the enhanced DIN loading has been well-related to losses from fertilised cropping in the Wet Tropics region (Bainbridge et al., 2009; Mitchell et al., 2009; Hunter and Walton, 2008; Hateley et al., 2014a, 2014b), the relative land use contributions of PN are less certain. Recent modelling suggested that the grazing (including dairy and beef: 32%) and sugarcane (28%) land uses contributed the highest proportions of PN in the Wet Tropics region (Hateley et al., 2014a, 2014b) whereas the available monitoring data place sugarcane and rainforest (natural) as the dominant contributors with grazing a comparatively minor source of PN (Hunter and Walton, 2008). Ascertaining the key land use sources of PN is critical so that management efforts can prioritise investment to reduce loads to the

GBR. While water quality monitoring has occurred in the basins of the Wet Tropics region to determine end-of-river loads (Wallace et al., 2015; Wallace et al., 2016; Garzon-Garcia et al., 2015; Huggins et al., 2017) as well as targeted monitoring to examine specific land use contributions of fine sediments and PN (Hunter and Walton, 2008; Bainbridge et al., 2009), further lines of evidence are required to determine the contributions of sediment and nutrients from different land uses in the Wet Tropics region. In that regard, the Source Catchment Modelling for the region is somewhat constrained by a general lack of empirical data to assist with model validation.

Isotopic signature ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) have been frequently used to discriminate between subsoil and topsoil as the potential sources of sediment and nutrients to the aquatic environment (Garzon-Garcia et al., 2017; Lacey et al., 2016; Mukundan et al., 2010). However, in a catchment with a variety of land uses, the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ signatures alone are not able to discriminate between the land uses under the vegetation types with the same photosynthetic pathways (e.g., grazing and sugarcane as C_4 ; rainforest and bananas as C_3 plants). To tackle this

issue, Bahadori et al. (2019) showed that applying a combination of stable isotopes ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) and geochemical elements (e.g., Zn, Pt and S) within the Stable Isotope Analysis in R (SIAR) mixing model can clearly distinguish the key land uses (rainforest, grazing including dairy and beef, sugarcane and bananas) in the Johnstone River basin of the Wet Tropics region (Bahadori et al., 2019). If this tracing approach can be applied to bed and suspended sediments in the Wet Tropics region, then another line of evidence can be developed to quantify land use contributions of sediment and nutrients in the GBR catchment area.

The key objective of this study is to quantify the contribution of different land use sources (grazing including dairy and beef, sugarcane, rainforest and bananas) to the Johnstone River bed and suspended sediments and associated PN. These sources are the dominant contributors to sediment and nutrients exported during flow events in this area (Hunter and Walton, 2008; Lewis and Brodie, 2011). In this study, the widely used SIAR mixing model is employed to allocate the contribution of each land use source to the mixture of sediments collected from both the river bed and suspended in flow events during 2017 and 2018 based on a novel approach developed to differentiate between sources (Bahadori et al., 2019).

2. Material and methods

2.1. Catchment description and soil sampling

The Johnstone River catchment is located in the Wet Tropics region of NE Queensland, Australia. This catchment covers an area of 2624 km², with relatively dry winters and hot, humid and wet summers (Hunter and Walton, 2008). There are two major arms of the Johnstone River. The North Johnstone River headwaters start in the north-western part of the catchment, while the South Johnstone River starts in the south-western section. Both the northern and southern Johnstone rivers pass through different land uses on their upper and lower sections before merging into a single stream at Innisfail (Fig. 1). Native forest with around 52% is the dominant land use in this catchment followed by grazing pastures (combined beef cattle and dairy) with 20.9%. Sugarcane (14% of area) and banana farming (4.3%) are main intensive agricultural farms in the Johnstone catchment. Urban-related and defence lands also make up 5% of the basin area (other land uses 3.8%) (Bahadori et al., 2019). Within the Wet Tropics region, the summer-autumn months (December to April) are usually associated with intense rainfall, tropical lows/cyclones and the monsoon. The annual average precipitation in the Johnstone catchment varies from 1154 mm in the upper catchment to 3552 mm at Innisfail in the lower catchment (1881–2019) (BOM, 2019).

After an extensive literature review, four key land uses including grazing (beef cattle and dairy), sugarcane, rainforest and bananas were identified as the most likely contributors to sediment and nutrients exported during flow events in this area (Wallace et al., 2015; Hunter and Walton, 2008; Lewis and Brodie, 2011; Hateley et al., 2014a, 2014b). Since these land uses are unequally distributed throughout the catchment, and to collect representative group of samples, the whole Johnstone catchment was divided into three geographical sections consisting of the upper Johnstone, the lower Johnstone and the Johnstone River estuary (Fig. 1). For soil sampling, 20 sampling points (each source was sampled at five locations) were selected throughout the catchment. Soil samples were collected from surface soil (0–10 cm) with an auger after vegetation was removed to ground level. All the soil sampling sites for grazing and rainforest land uses were selected within the upper section of Johnstone catchment. Because this part of the catchment is predominantly covered by grazing pastures, rainforest and small townships including Millaa Millaa and Malanda (population < 3000). Conversely, the lower Johnstone catchment is dominated by sugarcane farms and banana cropping and hosts the townships of Innisfail and South Johnstone (population < 8000). Therefore, banana plantations were sampled exclusively from the lower catchment section

as no banana farm exists on the upper part of the catchment. However, sugarcane sites were selected from both the upper and lower catchment sections, because a minority of sugarcane is also situated within the upper Johnstone catchment. Each land use source was sampled at five locations and at each location, a composite sample of five points was taken, see Bahadori et al. (2019) for more details.

2.2. River bed and suspended sediment collection

For bed sediment sampling, it is expected that collected sediment and associated particulate organic matter along the Johnstone River reasonably reflect the signatures of dominant land uses on each geographical section. All river bed sediment samples were collected along the Johnstone River from different geographical sections including the upper catchment, the lower catchment as well as the river estuary in July 2016. All samples were taken from the top 10 cm using a stainless-steel trowel which was regularly cleaned with milli-Q water to avoid inter-sample contamination. At each location, a composite mixture of 5 sampling points was made to ensure that the collected samples were representative of the river sections.

Suspended sediment samples were collected from the Johnstone River estuary in January 2017 and February 2018, during elevated river flow. One suspended sediment sample was also collected from the South Johnstone River during elevated catchment flows in February 2018, representing a discrete parcel of water/source during the flow hydrograph (Table S1). One of the main analytical challenges in tracing the source of sediments during flood events is collecting enough mass of suspended sediments for biogeochemical analyses. In this study, the SediPump[®] system (Stevens, 2018) was utilised to collect the required amount of suspended sediment during flood events in 2017 and 2018. In this system, a large known volume of water was pumped through a filter cartridge (sediment filter cartridge-wound GW011) using a water pump, and then all the sediment retained in the filter carousel were either recovered by backwashing into a bucket at each site or for the string filter cartridge was cut to release the sediment (Stevens, 2018). The SediPump[®] system allowed us to collect a considerable amount of sediment which enabled us, for the first time, to conduct detailed biogeochemical analysis.

All soil and sediment samples were packed in plastic bags and transported on ice to the laboratory for analysis. In total, 20 soil samples from the four different land use sources, 9 river bed (4 samples from the upper catchment, 2 samples from the lower catchment and 3 samples from the river estuary) and 3 suspended sediment samples (2 samples from estuary collected in 2017 and 2018 plume events, and one suspended sediment sample from the South Johnstone River) were analysed with preparation methods described below.

2.3. Sample preparation and laboratory analyses

In order to ensure consistency and to facilitate the direct comparison of soil and river bed sediments, all the collected samples were air dried, gently crushed using a pestle and mortar and then passed through a 63 μm sieve. Prior to this, the collected samples were passed through sequential 4 mm and 2 mm sieves, and all physically visible organic fragments such as root and leaf litter were removed from samples. Suspended sediment samples were centrifuged upon their arrival at the laboratory to recover as much sediment as possible, and then freeze dried prior to chemical analyses. All the collected soil and sediment samples were directly digested by nitric and perchloric acid (Miller, 1998), and then a total of 21 chemical elements (Na, K, Mg, Ca, Mn, Zn, Al, Cu, Sn, Ni, Co, Cr, Pt, Pb, As, Hg, Fe, Ag, S, P and Au) were measured in the extracted solution using ICP-OES; Perkin Elmer; Optima 8300 (Table S2). For the $\delta^{15}\text{N}$, soil and sediment samples were pelletized in tin capsules with no HCl treatment. For analysis of $\delta^{13}\text{C}$, first inorganic carbonates were removed by 2 ml of 10% hydrochloric acid (HCl), and then all samples were pelletized in silver capsules and

weighed for analysis with a Sercon Hydra 20-22 Europa EA-GSL isotope-ratio mass spectrometer (Garzon-Garcia et al., 2017; Bahadori et al., 2019).

2.4. Statistical analysis and modelling

In this study, the Kruskal–Wallis H-test and stepwise Discriminant Functional Analysis (DFA) were used to select the most discriminative group of signatures ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, Zn, Pt and S) for differentiating the main sources of sediments and particulate N to the Johnstone River (see Bahadori et al. (2019) for more details). The selected group of signatures ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, Zn, Pt and S) were then modelled within Stable Isotope Analysis in R (SIAR) mixing model with omitting concentration dependency and enrichment factor (set to 0) (Parnell et al., 2010). The SIAR model outputs include posterior distributions that represent a true probability density for the mixing contribution of the sources and an overall residual term to provide a quantitative estimation of the contribution of different land use sources to sediments in the Johnstone River (see Supplementary material) (Parnell et al., 2010). In this study, mean percent contributions of sources to sediment, as obtained from model outputs, are reported.

Then, the relative contributions to PN from the different sources were calculated with the SIAR model outputs as follows:

$$\%E \text{ Source } i = \frac{E \text{ Source } i \times \%cont}{\sum_{i=1}^4 E \text{ Source } i \times \%cont} \times 100 \quad (1)$$

with %E source *i* being the contribution of source *i* to PN with *i* varying from 1 to 4 to include all the 4 sources evaluated; E source *i*, the mean PN content of source *i* obtained from elemental analysis of source samples and %cont the mean percent contribution of source *i* to sediment export as obtained from SIAR model outputs. The propagated standard deviation for each source TN contribution was calculated using SGUM (Hall, 2010). In order to find the proportional contribution of different land use sources to the exported sediment and PN to the Johnstone River estuary, the proportion enrichment (PE) factor was calculated as follow:

$$PE = \frac{\%contribution \text{ to the river estuary}}{\%land \text{ use area}} \quad (2)$$

3. Results

3.1. Bed sediments in the upper and the lower Johnstone River

Contributions of different land use sources to the river bed sediment are expected to change along the Johnstone River from the upper to the lower catchment, where banana and sugarcane farms dominantly exists in the lower catchment, while grazing and rainforest land uses cover the main part of the upper Johnstone catchment (Fig. 1).

Fig. 2 shows that rainforest was the largest contributor to the bed sediment in the upper Johnstone River with a mean sediment contribution of 83.4% (SD = 13.2), followed by grazing and sugarcane soils with mean sediment contributions of 9.4% (SD = 10.2) and 7.2% (SD = 7.5), respectively. As there are no banana farms in the upper Johnstone catchment, this source was omitted from the model to avoid any overestimation and miscalculation by the SIAR model for the upper catchment (Fig. 2-A). Rainforest was not only the dominant source of bed sediment but also was the main contributor to the PN (91.7%, SD = 9.6) associated with the bed sediment in the upper Johnstone River (Fig. 2-B). This reflects the higher organic matter content of rainforest soils compared to the other sources (Table 1). Grazing and sugarcane soils only contributed 6.3% (SD = 8.2) and 2.0% (SD = 1.4) of the PN as attached to the bed sediment in the upper catchment, respectively (Fig. 2-B).

Towards the lower section of the Johnstone River, the estimated contribution of land use sources to the river bed sediment and

associated PN changed considerably (Fig. 2-A and B). Banana land use was the largest source to the bed sediment in the lower Johnstone River with a mean sediment contribution of 31.8% (SD = 14.5), followed by sugarcane and rainforest soils with mean sediment contributions of 25.2% (SD = 12.7) and 24.8% (SD = 14.6), respectively. Grazing with 18.2% (SD = 12.6) had the lowest contribution of the four major land uses to the river bed sediments of the lower Johnstone catchment (Fig. 2-A). Banana land use contributed 25.4% (SD = 1.8) to the PN which was the second highest contribution after rainforest land use (43.9%, SD = 1.9) in the lower catchment. Sugarcane had the lowest contribution to PN in the bed sediments of the lower catchment with 11.1% (SD = 5.9) whereas grazing lands accounted for 19.6% (SD = 6.1) (Fig. 2-B).

3.2. Bed and suspended sediment at the Johnstone River estuary

Rainforest was the largest source of bed sediment for the Johnstone River estuary with a mean sediment contribution of 33.1% (SD = 14.5) followed by grazing land use with a mean sediment contribution of 25.1% (SD = 13.9) (Fig. 3-A). The mean proportional contributions of sugarcane and bananas were 21.4% (SD = 13.5) and 20.4% (SD = 13.2), respectively. The contributions from rainforest (33.1%, SD = 19.4) and sugarcane (21.5%, SD = 14.5) to the suspended sediment delivered to the river estuary were similar to the river bed sediments, with rainforest as the dominant source of suspended sediment during the elevated river flows sampled in the wet season (Fig. 3-A). While the contribution of banana farms (26.7%, SD = 15.2) to the suspended sediment were higher compared to the river bed sediment, grazing lands with 18.7% (SD = 13.4) had a lower contribution to the suspended sediments collected during the elevated river flows in the wet season compared to the bed sediment collected from the estuary (Fig. 3-A). The contribution of different land uses to the PN associated with suspended sediment discharged to the GBR lagoon were similar to those attached to the river bed sediment with rainforest being the main source of PN to both suspended (53.5%, SD = 7.3) and bed sediment (52.6%, SD = 11.4). Sugarcane with 8.6% (SD = 1.9) and 8.5% (SD = 0.9) had the lowest contribution to the PN in the suspended and bed sediments, respectively. Grazing lands with 18.4% (SD = 5.2) and 24.3% (SD = 5.5) were the third and second major land use source of PN to the estuary as attached to the suspended and bed sediment, respectively (Fig. 3-B). Banana farms with 19.5% (SD = 3.7) had a higher contribution to PN in the suspended sediment compared to the bed sediment (14.6%, SD = 7.1) at the Johnstone River estuary (Fig. 3-B).

3.3. Bed and suspended sediments collected from the South Johnstone River

The modelling results for the suspended sediment sample collected from the South Johnstone River next to the Department of Primary Industries (DPI) South Johnstone Research Station shows that rainforest was the largest source of suspended sediment for the South Johnstone River with a mean sediment contribution of 96.6% (SD = 1.6). The three other land use sources including grazing (1.0%, SD = 0.8), sugarcane (1.0%, SD = 0.7) and bananas (1.4%, SD = 1.3) had a low cumulative contribution to the 'point in time' suspended sediment sample collected from the South Johnstone River during elevated river flow in the 2018 wet season (Fig. 4-A). However, the contribution of different land use sources to the bed sediment were markedly different from the suspended sediment sample, with bananas being the largest contributor (72.6%, SD = 12.1) followed by rainforest (24.9%, SD = 13.2). Grazing and sugarcane sources with 1.3% (SD = 0.9) and 1.2% (SD = 0.8), respectively, had the lowest contributions to bed sediment of the South Johnstone River (Fig. 4-A). Rainforest with 98.5% (SD = 2.0) was the largest contributor to the PN associated with suspended sediment in the South Johnstone River, while bananas (55.8%, SD = 6.3) and rainforest (42.4%, SD = 5.1) were the main land use sources of PN as attached to the bed sediments in the South

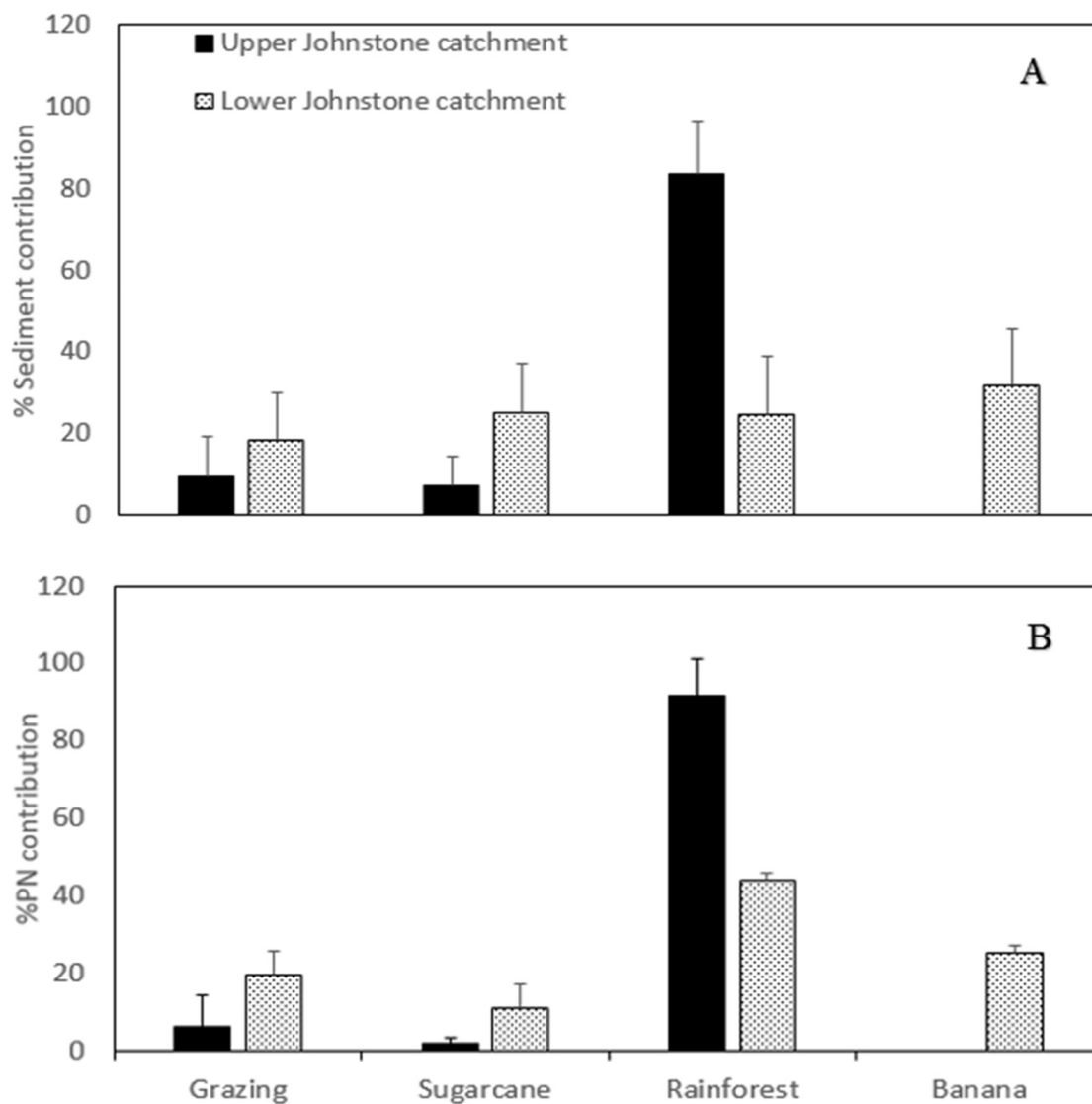


Fig. 2. Mean percent contributions to exported bed sediments (A) and particulate nitrogen (PN) associated with bed sediment (B) to the upper and lower Johnstone River catchments from the land uses of grazing, sugarcane, rainforest and banana (there was no banana land use in the upper Johnstone catchment).

Johnstone River. The cumulative contributions of other land uses to the PN associated with both bed and suspended sediments were < 2% (Fig. 4-B). Details of SIAR modelling output are provided in supplementary documents.

4. Discussion

4.1. Influences of the upper and the lower catchment land uses on river bed and suspended sediments

Recent Source Catchments modelling has revealed that river bank and hillslope erosion are mainly responsible for supplying bedload and

Table 1

Total organic carbon (TOC) and total nitrogen (TN) content of soils collected from different land uses in the Johnstone catchment with standard deviation (STDEV) reported in parentheses, and the proportion enrichment (PE = %contribution to the river estuary/%land use area) for the contribution of different land uses to suspended sediment, bed sediment and associated particulate nitrogen (PN) collected from the Johnstone River estuary.

Land use	Land area (%) total)	%TOC (STDEV)	%TN (STDEV)	Proportion enrichment (PE) (%contribution to the river estuary/%land use area)			
				Bed sediment	PN (associated with bed sediment)	Suspended sediment	PN (associated with suspended sediment)
Grazing	20.9	4.1 (1.1)	0.3 (0.1)	1.2	1.2	0.9	0.9
Sugarcane	14.0	1.9 (0.6)	0.1 (0.0)	1.5	0.6	1.5	0.6
Rainforest	52.0	7.3 (1.6)	0.5 (0.1)	0.6	1.0	0.6	1.0
Banana	4.3	3.3 (1.2)	0.2 (0.1)	4.7	3.4	6.2	4.5

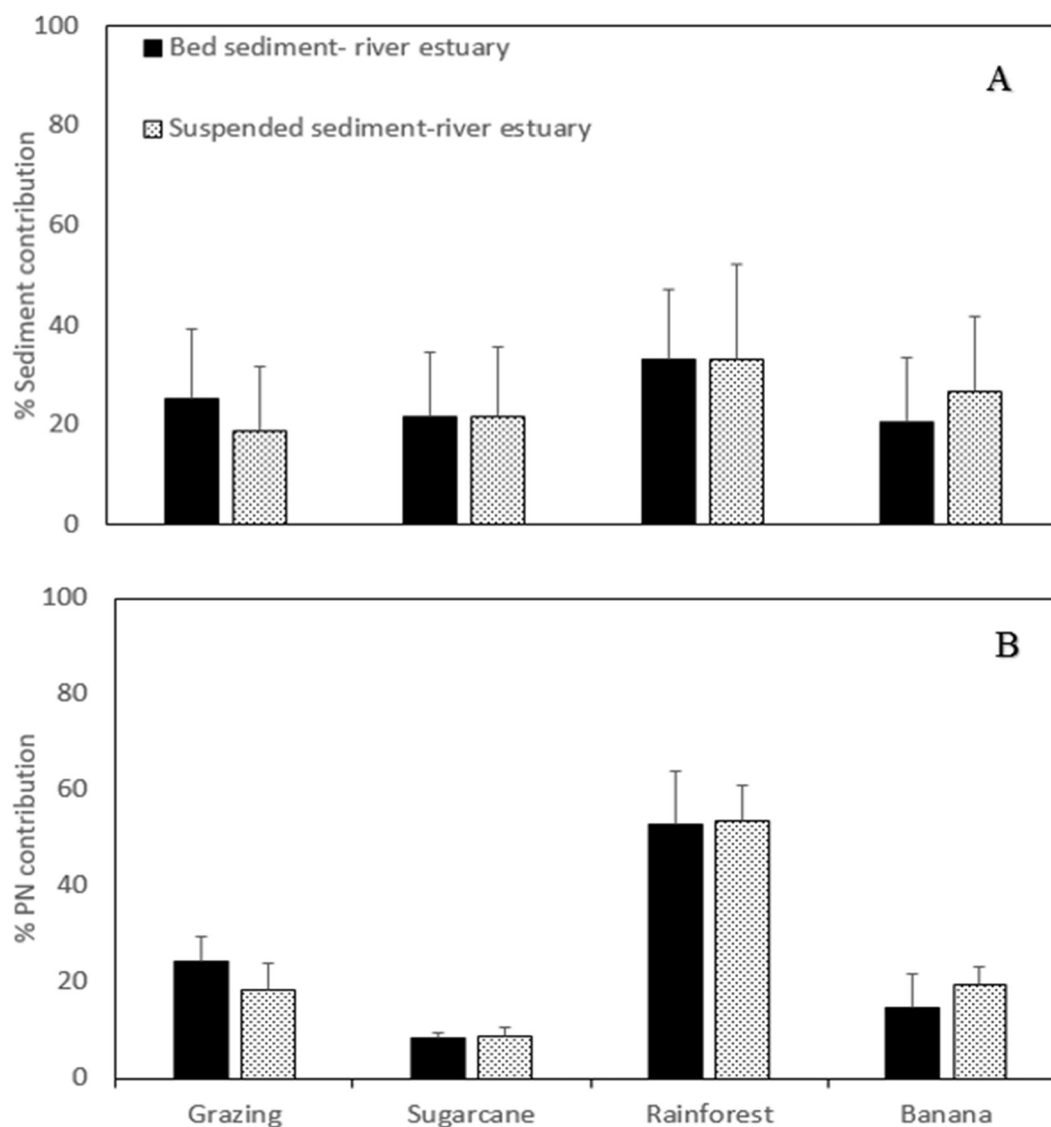


Fig. 3. Mean percent contributions to exported bed and suspended sediments (A) and particulate nitrogen (PN) associated with bed and suspended sediments (B) to the Johnstone River estuary from the land uses of grazing, sugarcane, rainforest and banana.

suspended sediment to the Wet Tropic region including the Johnstone River catchment (McCloskey et al., 2017; Hateley et al., 2014a, 2014b). The contribution of different land uses to the PN and sediments in streams are highly influenced by the proportion of different land uses covering the upstream catchment (Bartley et al., 2017). Hence, the higher contribution of rainforest and grazing sources to bed sediment in the upper part of the Johnstone River is attributed to dominance of these two land uses (Fig. 2-A). In the lower Johnstone catchment, increased areas of sugarcane and banana crops explain their increased contributions to bedload sediment in the lower Johnstone catchment with relatively smaller contributions of sediment and PN from the other sources predominately located on the upper catchment. The relatively low contribution of grazing land use in bed sediment in the upper Johnstone River may be explained by the high levels of grass cover typically observed in beef and dairy pastures throughout the year in the Johnstone catchment (Hunter and Walton, 2008). This is in contrast to the other GBR catchments situated in the Dry Tropics region where the grass cover dramatically reduces during the dry season (Kuhnert et al., 2012).

The contribution of different land uses to the sediment delivered to the estuary is of the greatest interest to guide catchment management programs that aim to reduce sediment and PN delivery to the GBR

lagoon. In this study, results showed that rainforest was the main source of both suspended and bed sediments to the river estuary which is consistent with the result reported by Hunter and Walton (2008) (Fig. 3-A). Moreover, contributions of bananas and sugarcane farms to suspended sediments at 'point in time' samples were higher than their contributions to river bed sediments, while grazing had moderately higher contributions to river bed sediments compared to the suspended load (Fig. 3-A). This could be attributed to the effect of agricultural practices and management systems (tillage, row cropping etc.) on sugarcane and banana farms on generation and transportation of fine particles through sheet and rill erosion (Hateley, 2014b). In addition, banana and sugarcane farms are largely concentrated in the basalt geologies of the catchment area which would produce relatively finer sediment particles which in turn would be preferentially transported through the catchment largely in suspension (McCulloch et al., 2003).

The South Johnstone Research Station is located south-west of Innisfail city, North Queensland. It is situated on Barron River Metamorphic rocks with banana plantations and is surrounded by rainforest land use (Heiner and Smith, 1990) (Fig. 1). This site could be considered as the point that land use changes from the intact world heritage area to agricultural farms. It is clear that the bed sediment collected from the South Johnstone River (next to the DPI Research

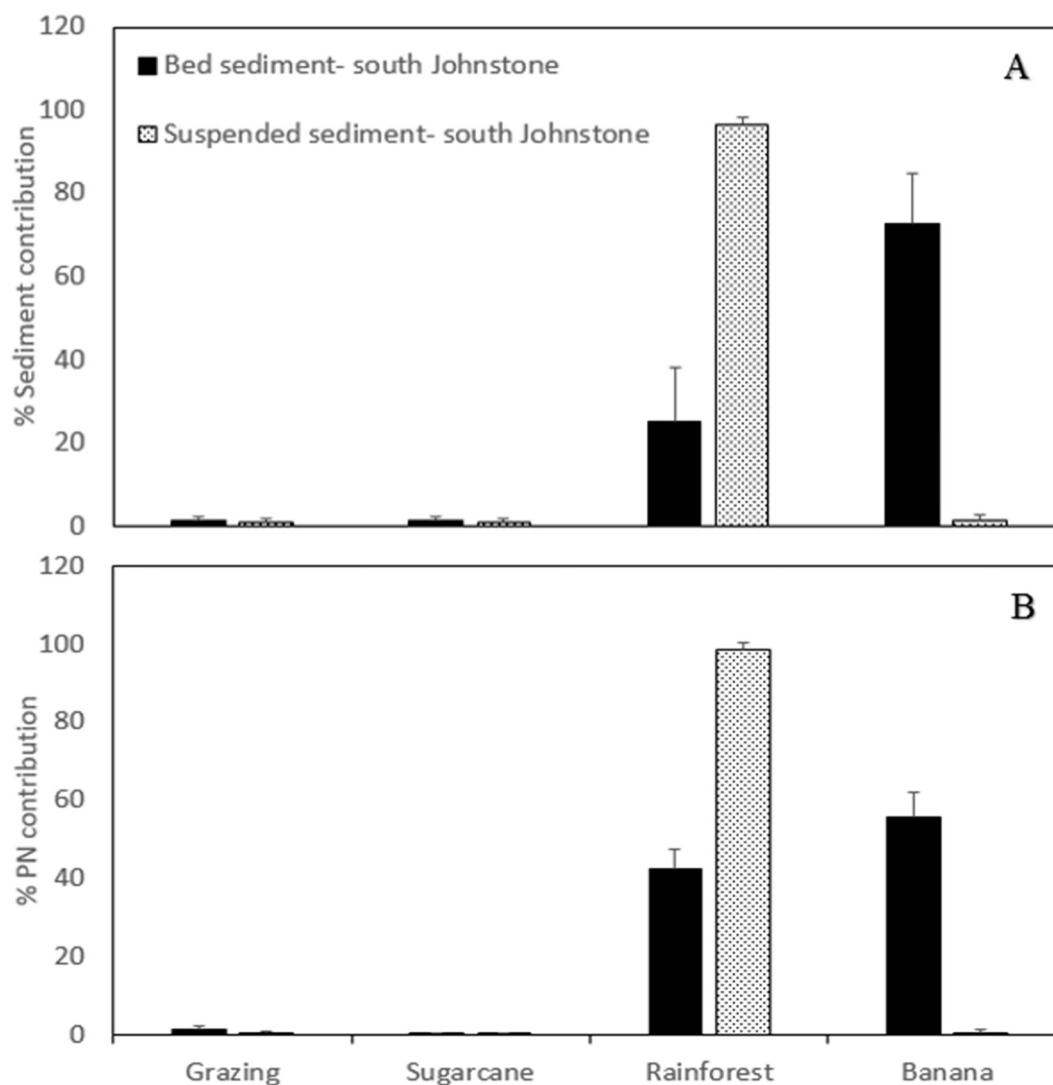


Fig. 4. Mean percent contributions of different land uses (grazing, sugarcane, rainforest and banana) to the suspended and bed sediment (A) and particulate nitrogen (PN) associated with suspended and bed sediments (B) collected from the South Johnstone River next to the department of primary industries (DPI) research station.

Station) has shown the signatures of banana farms, representing runoff from cropping lands combined with a small contribution of sediment and PN from rainforest land use located on the upper catchment (Fig. 4-A and B). However, during the elevated river flow in the wet season, suspended sediment collected at a 'point in time' of flow from the South Johnstone River dominantly carried the signatures of rainforest land use (Fig. 4-A and B). In fact, the suspended sediment in the South Johnstone River was collected over a 2-hour period at a point where water flowing through the river at that time came mostly from the part of catchment dominated by rainforest. Hence, in contrast to the bed-load, suspended sediments in that water would expect to reflect a predominant rainforest signature (Fig. 4-A and B).

4.2. Aggregated effects of land uses on bed sediment, suspended sediment and associated particulate nitrogen export to the Johnstone River estuary

The modelling and monitoring results have already revealed that some land uses in the GBR catchment have much higher flux contributions (*i.e.* $\text{kg}\cdot\text{Ha}^{-1}$) of sediment and particulate nutrients (DERM, 2011b; McCloskey et al., 2017; Hateley et al., 2014a, 2014b; Hunter and Walton, 2008). It has been attributed to the differences in land use and land management practices, as well as the natural features and environmental factors in different areas with similar land uses (DERM,

2011b; Hunter and Walton, 2008). Therefore, it is necessary to find and prioritise the land uses with higher rates of contaminant loads moving off-site, and to adopt the land management practices with the key environmental characteristics in different land uses.

In this study, PN in the Johnstone River estuary are also dominantly sourced from native rainforest (Fig. 3-B). This relatively large contribution of the rainforest source to sediment is due to the larger coverage area of the Johnstone catchment by rainforest (52%). The higher organic matter content in rainforest soils has also led to higher PN associated with sediments originating from rainforest compared to other sources (Table 1). Despite that, the contribution of rainforest is disproportionately small by land use area. It is because this land use covers 52% of the catchment land area.

Results showed that, rainforest and grazing had the lowest proportional enrichment (PE) in the bed and suspended sediment delivered to the Johnstone River estuary. Importantly, once PE was factored, bananas and sugarcane had the highest contribution to both bed and suspended sediment delivered to the coast (Table 1). These results are consistent with results reported by Hunter and Walton (2008) regarding the influence of different land uses on the fluxes of sediment and nutrients from the Johnstone catchment. They highlighted that the relative influence of land uses differ from those estimated on a unit area basis. According to their results, although rainforest had the highest

contribution (41%) to the mean annual fluxes of suspended sediment and nitrogen, this land use had a disproportionately small contribution to the generated suspended sediment per unit of area covered by this land use. By contrast, banana and sugarcane farms had higher contributions per unit of area.

It is well known that ground cover and soil surface condition play a significant role in controlling the rates of run-off and sediment loss (Bartley et al., 2006). The influence of clearing forest in terms of run-off and sediment loss from catchments has been reported in previous studies, suggesting that adequate ground cover, on both hillslopes and riparian zones, needs to be maintained to reduce the potential for gully formation (Silburn et al., 2011; Van Rompaey et al., 2002; Bartley et al., 2017). A global assessment of the significant impact of land use change (particularly clearance of forest land use for cropland expansion) on soil erosion and sediment supply to rivers has also been conducted in previous studies (Borrelli et al., 2017; Van Rompaey et al., 2002). The effect of tree clearing on ecosystem structure and the resultant changes in water and sediment yield have not been well studied in Australia; however, studies in semi-arid rangeland areas in Queensland suggest that converting forest to pasture can increase run-off by ~40% for river catchment scales (Siriwardena et al., 2006; Bartley et al., 2017) (Siriwardena et al., 2006).

The sustainability of farmlands and their impact on downstream water quality are highly related to the crop type, the specific nature of cultivation, the slope/contour of the paddock and also the management systems applied on such areas including N fertiliser regimes (Arnhold et al., 2014). The severity of the erosion on a farmland is controlled by root system and also the extent that vegetation types protect their soil from the impact of runoff and raindrops (Arnhold et al., 2014). The row-cropping system in the banana farms results in a high ratio of exposed ground to raindrops and runoff and can lead to considerable erosion management challenges on this land use (Armour et al., 2013). To address this issue, a plantation design needs to be considered to provide a favourable condition for soil protection and also to ease the access for harvest and farm operations. In cases where a banana farm is located on gentle slopes, it is recommended to use a “grassed inter-row” system. This system provides benefits such as retaining soil cover, building up soil organic matter and allowing nutrient cycling by concentrating litter and trash between the rows (Akehurst et al., 2008; Bagshaw and Lindsay, 2009). In a similar manner, the sugarcane industry has taken effective actions towards minimal tillage systems and also trash blanketing methods in order to reduce soil erosion rate and the delivery of sediment to the rivers (Rayment, 2002; Prove et al., 1995). Despite that, the mean erosion rate from sugarcane land use is 1.2 t/Ha/yr which is the second highest rate after bananas with 1.8 t/Ha/yr (Hateley et al., 2014a, 2014b).

In order to determine the further causes of such high PE in agricultural farms (sugarcane and bananas), it is important to consider the environmental characteristics in parallel with the current land management practices in these land uses (Liu et al., 2018; Chang, 2008). Soils that are susceptible to erosion may not actually erode under an effective management system, while a poor management practice may lead to a high rate of erosion in a soil with a lower inherent erosion potential (DERM, 2011b; Rose, 2017). For instance, it is critical to consider the suitability of lands for intensive agricultural systems, and the way that soil erosion can affect the productivity of agricultural farms, as much as it may impact the water quality downstream (DERM, 2011b; McDowell et al., 2018). Although much of Queensland's horticulture occurs on flat alluvial soils of the narrow coastal plain where erosion risk is relatively low, some agricultural farms are located on slopes and foothills that are vulnerable to water erosion (DERM, 2011b; Carey et al., 2015). A minority of intensive agricultural farms as well as forest and grazing land uses are located on red ferrosols associated with volcanic landscapes (Hunter and Walton, 2008). Basaltic soils such as red ferrosols are well known for having developed structures and physical characteristics that are less susceptible to water erosion

compared to other soil types on the comparable slopes and vegetation types (DERM, 2011a). Although significant rainfall usually occurs throughout the year in the Johnstone catchment, intensive rainfall events associated with low pressure systems and the monsoon dominantly occur during the summer months and they are not evenly distributed over the whole catchment (Hunter and Walton, 2008). The mean annual rainfall of 1154 mm has been reported for the upper Johnstone catchment, while the Innisfail station in the lower Johnstone has recorded an average rainfall of 3552 mm on the coast (1881–2019) (BOM, 2019). Therefore, environmental factors such as spatial rainfall distribution should be taken into account for interpreting the main causes of higher PE in the sugarcane and banana land uses (the lower catchment) compared to the rainforest and grazing sources (the upper catchment).

It is evident from the previous studies that N cycling in interface of freshwater and marine environment is subject to a complex array of regulatory mechanisms involving both physico-chemical and biological factors (Herbert, 1999; Baldock et al., 2004). Particulate organic matter molecules containing nitrogen are available for heterotrophic processes (mineralization), releasing energy and producing NH_4^+ and NO_3^- to the coastal marine environment. Both of the dominant inorganic forms of nitrogen (NH_4^+ and NO_3^-) can be cycled back into organic forms through assimilation and subsequently stored for variable periods of time before N is rereleased via decomposition and mineralization (Scott et al., 2007). Such an anthropogenic nutrient discharge to coastal marine environments is commonly associated with excessive algal growth, eutrophication and ecosystem degradation (Erlor et al., 2020).

4.3. The strengths and limitations of the novel approach of combined isotopic and geochemical signatures and SIAR mixing model

The Australian and Queensland Governments have recently released an update of the Reef 2050 Plan to provide a long-term sustainability framework to protect and manage the GBR until 2050 (Australia, 2018). This comprehensive plan highlights the actions that need to address the key threats to this area, and also to the health and resilience of the GBR in the face of different environmental and anthropogenic pressures. The Reef Water Quality Protection Plan is one of the main themes of this long-term plan which aims to improve agricultural management practices by developing a conceptual understanding of the link between land condition, management practice standards and water quality outcomes (Government, 2018). In this context, identifying the main land use sources of sediment in a catchment is a key requirement for the application of targeted mitigation measures.

Several studies have used isotopic signatures to differentiate between subsoil and topsoil as the potential sources of sediment and particulate nutrients to the rivers (Garzon-Garcia et al., 2017; Lacey et al., 2016; Mukundan et al., 2010). However, these signatures alone are not able to differentiate the land uses covered with the plants that follow the same photosynthetic pathways (grazing and sugarcane as C_4 , and rainforest and bananas as C_3 plants). Bahadori et al. (2019), developed a novel approach of combined isotopic and geochemical properties for discriminating between different land uses as well as estimating the contribution of each land use to sediment and particulate nutrients using the SIAR mixing model. This new tool enabled us to have a discriminative group of signatures for the SIAR mixing model to estimate the contribution of different land uses (grazing, sugarcane, rainforest and bananas) in delivered sediment and PN to the Johnstone River. It provides a useful tool for the GBR authorities to fulfil their catchment management targets in reducing diffuse source pollution and minimising the risk to the GBR from a decline in the quality of water entering the reef from adjacent catchments.

In order to limit particle size effects on the tracers used for fingerprinting, the < 63 μm fraction of soil and sediment was used for measuring isotopic and geochemical signatures in this study. Analysing the < 63 μm fraction is the most common practice in published

fingerprinting studies as the dominant proportion of fluvial suspended sediment loads is represented by this size fraction (Collins et al., 2017). However, some variability in the concentration of different signatures can still exist within the < 63 μm fraction (Hatfield and Maher, 2009; Pulley and Rowntree, 2016). Alternatively, a narrower particle size fraction (< 10 μm) has been suggested in other studies to reduce the impact of particle size variability in the traced fraction (Collins et al., 2017). Although the selection of this narrower particle size has benefited when providing more robust source discrimination (Haddadchi et al., 2015; Lacey and Olley, 2015), the finer fractions are more geochemically active and more susceptible to transformation, with non-conservative behaviour during transport (Collins et al., 2017).

In order to have an accurate estimation of the contribution of different sources in sediment and PN delivered to a river system, it is fundamental to obtain representative samples of suspended sediment. It is well known that a bulk of the suspended sediment transported in Tropical River systems occur within a short time of high rainfall during the wet season. Therefore, there is a need to focus sampling activity on the main periods of suspended sediment transport. In this context, individual instantaneous sediment samples collected at the elevated flow event can be assumed to be representative of sediment transported during a longer period (Phillips et al., 2000). The results of this exploratory study show the potential of a single flood event in representing of sediment originating from a large spatial area of the catchment.

5. Conclusions

This study has provided a quantitative estimation on the contribution of different land uses (grazing, sugarcane, rainforest and bananas) to the sediment and PN discharged to the GBR through the Johnstone River in the Wet Tropics region of north-eastern Australia. Rainforest was the dominant source of sediment and PN to the Johnstone River, while intensive agricultural farms (bananas and sugarcane farms) had the highest rates of sediment and PN delivered to the Johnstone River estuary per unit of area covered by these land uses. These results highlight the required investigations and improvement in the current land-management practices and provide useful information for managers to fulfil their targets to improve the water quality in the GBR by 2050. In particular, this study highlights the importance of adopting the current best management practices in the intensive agricultural farms where the high rates of soil erosion result in several water quality issues downstream in the GBR lagoon, although the type and scale of our sampling did not permit us to identify the key erosion processes that export the sediment from these land uses. Further investigations are also required to examine more vegetative signatures which have the potential to differentiate sources of dissolved organic nutrients to the GBR lagoon.

CRedit authorship contribution statement

Mohammad Bahadori: Conceptualization, Methodology, Software, Writing - original draft, Writing - review & editing, Funding acquisition, Data curation, Validation, Investigation, Formal analysis, Resources, Visualization. **Chengrong Chen:** Supervision, Conceptualization, Methodology, Writing - review & editing, Funding acquisition, Validation. **Stephen Lewis:** Writing - review & editing, Resources. **Mehran Rezaei Rashti:** Writing - review & editing, Resources. **Freeman Cook:** Writing - review & editing, Validation, Software. **Andrew Parnell:** Validation, Software. **Maryam Esfandbod:** Funding acquisition. **Thomas Stevens:** Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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