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This manuscript entitled "Planted mangroves cap toxic oil spill" has been submitted in Marine Pollution Bulletin. Please not that that this manuscript is still under review. Subsequent version of this manuscript may have slightly different content. If accepted, the final version of this manuscript will be available vie the "Peer-reviewed Publication DOI" link on the right-hand side of this webpage. Please feel free to contact any of the authors; we do welcome feedback.

Scientific significance
The majority of the studies on crude oil spills report their negative effect on coastal ecosystems, but none have reported on the role of mangrove restoration in immobilising oil spills (one article in Marine Pollution Bulletin has come close; study by Wells (1999) looked at developing biomonitoring tools to assess toxicity of marine environments (including mangroves and PAHs). Mangroves examined in this study were capable of establishing above an oil spill and immobilising an estimated 39.5 tonnes of toxic hydrocarbons per hectare. The toxicity of sediments within the mangrove rhizosphere (0–30 cm) exceeded safety thresholds by up to 190-fold and by up to 220-fold in sediments below mangrove rhizosphere (30–100 cm).

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Thank you in advance for receiving our manuscript and considering it for review.

Yours sincerely,

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Planted mangroves cap toxic oil spill

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Keywords: total petroleum hydrocarbons, oil spill immobilization, sediment contamination

Abstract

Mangroves are known to provide many ecosystem services, however there is little information on their potential role to cap and immobilise toxic levels of total petroleum hydrocarbons (TPH). Using an Australian case study, we investigated the capacity of planted mangroves (*Avicennia marina*) to immobilise TPH within a small embayment subjected to multiple oil spills throughout the 1980s. Mangroves were planted on the oil rich strata in 1984 to rehabilitate the site, with 2% surviving to form a dense forest. Sediment core examination down to 1 m deep, revealed mangroves have formed a thick (up to 30 cm) organic layer above the TPH spill (39.5 tonnes ha^{-1}) accumulating on average 6.6 mm of sediments a year. TPH levels below this organic layer are extremely toxic (30,441.6 mg kg^{-1}), exceeding safety thresholds up to 220-fold. This research shows that mangroves can play a valuable role in capping highly toxic oil spill material.

Introduction

Oil spills have caused some of the most devastating degradation to coastal ecosystems worldwide (Beyer et al., 2016; Jafarinejad, 2017). Oil has a long residence time affecting almost all levels of the food web (Balthis et al., 2017; Rohal et al., 2020), smothering coastal vegetation, and degrading soil organic matter (Duke, 2016). Restoration of oil-affected ecosystems is costly, and is unfeasible in many situations (National Research Council, 2013; Wallace et al., 2019).

Mangrove ecosystems provide valuable ecosystem services, such as coastal protection, fisheries enhancement, and carbon sequestration (Barbier et al., 2011; Duncan et al., 2016). However,

their ability to accumulate and trap sediments has not been studied in the context of immobilising oil spillages in the marine environment. Based on the documented ability of mangroves to promote recovery of oil-affected marine food webs (Duke, 2016; Ghizelini et al., 2019), mangroves forests might be able to immobilise the petroleum hydrocarbons belowground within their rhizosphere. Many studies have quantified the toxicity of oil pollutants within sediments of coastal vegetation and reported concentrations of major pollutants such as PAHs (polycyclic aromatic hydrocarbons) as function of their ongoing precipitation (Ashok et al., 2019; Santos et al., 2018). However, none of these studies to our knowledge have evaluated the capacity of planted mangroves to lock away a discrete layer of the polluted sediments.

In this study, we assessed the capacity of planted mangroves to cap and immobilise oil-contaminated sediments by their ability to accrue additional sediment on top of the oil layer. Specifically, we investigated: 1) the ability of planted mangroves to cap and isolate a toxic TPH spill within a discrete sediment layer; and 2) the ability of planted mangroves to survive and thrive by growing and accumulating organic matter above toxic TPH sediments.

Method

Study site

The Stony Creek estuary is located in the Melbourne metropolitan area, Australia (Fig. 1). Stony Creek drains an area of 25 km^2 of western Victoria's basalt plains, part of the Maribyrnong catchment (1,408 km^2) to Port Philip Bay. The basalt plains were formed approximately 2–5 million years ago and subsequently weathered to produce the overlying clay soils. The land surrounding Stony Creek is naturally occupied by a She-Oak forest (dominated by *Allocasuarina verticillata*), but was drastically transformed following the European settlement (1850's) through the establishment of quarries, tanneries, and crude oil storage facilities. Following extensive ecosystem degradation of the native grey mangrove (*Avicennia marina)*, restoration efforts focussed on replanting this indigenous species within its natural distribution (Oliver et al., 2002; Sinclair and Boon, 2012; Whitt et al., 2020).

Aerial images and historical records reveal that the first mangroves were planted in 1984 on the western side of the embayment, and they subsequently expanded to the Northern and Eastern sides (Fig. 1; Fig. S1). According to a local newspaper approximately 15,000 grey mangrove seedlings were planted in 1984 (Scott, 1990). Although only 2% of the mangroves survived due to a subsequent oil spill, they spread through the embayment forming a dense forest.

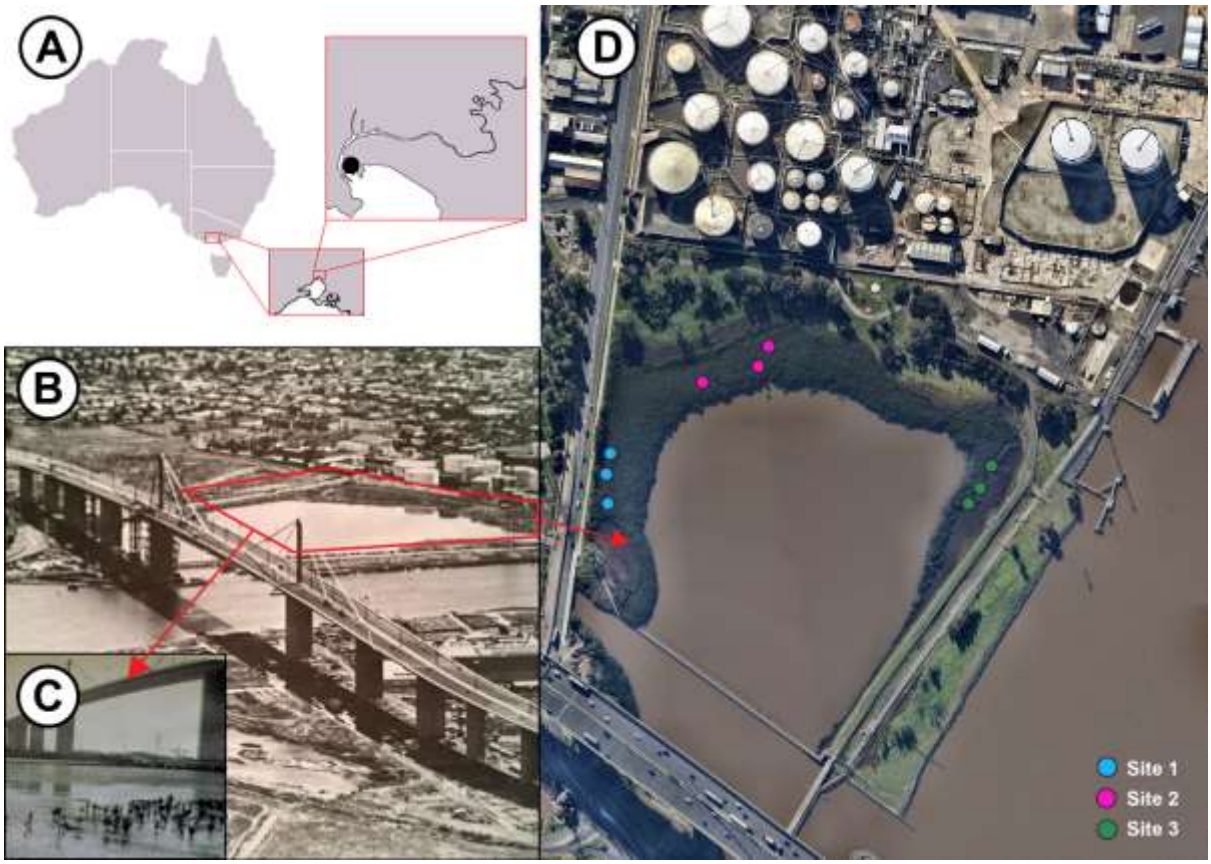

Fig. 1. A) Stony Creek estuary in south-eastern Australia (37°49'30.5"S, 144°53'48.4"E); B) Aerial photograph in the mid-1980s before the establishment of the mangroves; C) Mangrove seedlings (*Avicennia marina*) planted in 1984 (Photo Credit: Port Places); and D) Satellite image taken in 2019 displaying the three sampling sites.

Data Collection

We estimated hydrocarbon content from mangrove sediments at three sampling sites (Fig. 1d). At each sampling site, we collected three soil cores (1 m deep or until bedrock was reached) using a PVC pipe (5 cm internal diameter, 120 cm length) to profile the hydrocarbon content within the

sediment. Nine sediment cores (collected on 03/July/2019) were immediately transported to Deakin University (Burwood campus) and sliced at six depth intervals: 0–10, 10–20, 20–30, 30–50, 50–75 and 75–100 cm. Wet sediment samples were stored at 4℃ and shipped to the Analytical Reference Laboratory (Welshpool, Western Australia) for total petroleum hydrocarbon (TPH) and polyaromatic hydrocarbons (PAH) analyses in accordance with the US EPA 3550C methodology (US EPA, 2007). TPH concentration included five hydrocarbon compound groups (TPH C_{6-9}, TPH C_{10-14}, TPH C_{15-28}, TPH C_{29-36}, TPH $C_{>36}$) and was quantified using gas chromatography / Flame Ionization Detector (Varian CP-3800-4 / Agilent 7890A GC) as described in the Australian national guidelines (National Environment Protection, 1999). Additionally, we quantified 17 PAHs recognised as major pollutants (Tam et al., 2001). The aromatic fractions containing individual PAHs were prepared and analysed following US EPA Method 8270 D (US EPA, 2014), using a Varian 3800 or a Varian 3900 gas chromatograph system, both fitted with Saturn 2000/2100 ion trap detector and using the same column systems.

The dry content of PAHs and TPH and soil organic matter in mangrove sediments was estimated from nine additional cores collected at the three mangrove sites. These additional soil cores were sliced at compatible intervals and dried at 60℃ (until a consistent weight was achieved). The organic matter was estimated by looking at the organic carbon in the sediments above the oil spill prior mangrove arrival (establishment horizon). Mangrove arrival times were clearly delineated within the sediments at 30, 14 and 8 cm depth, which corresponded with mangrove arrival at our sampling sites in 1984 (Site 1), 2002 (Site 2) and 2006 (Site 3), respectively. After grinding the dry soil samples, they were subjected to an effervescence test by addition of 12M HCl to examine the presence of inorganic carbon ($CaCO_3$). One sample contained $CaCO_3$ and was excluded from further analysis. An elemental C:N analyser (EuroEA 3000 Series) was used to quantify soil organic carbon (%) according to the classical Pregl-Dumas method (Bong and Lee, 2007). Data are available from Mendeley Data Repository (Waryszak et al., 2020).

Data analysis

Contamination characteristics

TPH and PAH toxicities were estimated against high sediment quality guideline values (SQGV) issued by Australian and New Zealand Environment and Conservation Council and the Agriculture and Resource Management Council of Australia and New Zealand (ARMCANZ and ANZECC, 2000), as revised by Simpson (2013). The SQGV represent the threshold values associated with adverse biological effects (Warne et al., 2018), which are 280 mg kg^{-1} for TPHs and at 50 mg kg^{-1} for PAHs. Following Simpson & Batley (2016), we assessed the toxicity of PAH concentrations against SQGV by normalising to 1% of total organic carbon (OC). PAH concentrations are normalized to OC (%) as PAHs bind to soil organic carbon, making the PAHs less available to aquatic life, thus lessening their toxicity For example, if the mangrove sediment contained 5 mg kg^{-1} of total PAHs and 0.55% OC, then 1% OC normalised concentration = 5/0.55 = 9.1 mg kg^{-1} of total PAHs (1% OC) within the limit of 0.2—10% (Simpson and Batley, 2016). TPH concentrations in relation to sediment depths was assessed with use of a linear regression on untransformed data. TPH and PAH concentrations detected as below a 0.1 mg kg^{-1} level were converted to 0.05 mg kg^{-1} for computational purposes.

Correlation between PAH and TPH concentrations were assessed with a Pearson's correlation test as indicator if sediment pollution occurred in one or multiple spill events. Additionally, the ratio of fluoranthene:pyrene was assessed as an indicator of PAH source in contaminated sediments i.e., petrogenic versus non-petrogenic (e.g. from tanneries). TPH stocks (tonnes ha^{-1}) were calculated in relation to the oil horizon identified in sediments (below and total). TPH total stock (down to 1 m) and TPH below establishment horizon (capped by organic carbon accumulated above) were compared using regressions.

Capping capability

We assessed the ability of mangroves to cap contaminated sediments by measuring sediment accumulation above the TPH spill and within the detected rhizosphere (top 30 cm). Sediment

accumulation rate (mm year^{-1}) was estimated from ^{210}Pb concentration profiles across the top 30 cm sediments within three mangrove stands (at each site). The fine fraction of the sediment samples (<63 µm) was analysed by alpha spectrometry following method by Sanchez-Cabeza et al. (1998). Sediment accumulation rates were calculated with the Constant Flux: Constant Sedimentation(CF:CS) model (Krishnaswamy et al., 1971). We reported organic matter (%) in the top 30 cm of sediments from organic carbon values (%) acquired in elemental C:N analyser with use of "Van Bemmelen factor" of 1.724 (Allison, 1965).

Results

Contamination characteristics

Mangrove sediment cores at Stony Creek were characterised by a thick rhizome layer extending until 30 cm depth, followed by a distinct anoxic oil-contaminated layer at 30–50 cm, and a mixture of petroleum and clay soil located between the 50-100 cm depth (Fig. 2). The TPH concentrations were significantly above the biologically safe threshold at all depths and sites with exception of five samples below 75 cm (Fig. 2a). The maximum TPH toxicity was detected at 30–50 cm depth and exceeded SQGV thresholds 220-fold (at Site 1, Fig. 1). This TPH value was significantly higher compared to other depth intervals ($t = 4.93$, $P < 0.01$, Table S3). Overall, the concentration of TPH and ranged between 1.6–61,657 mg kg^{-1}. Average estimated TPH stock below 30 cm was 39.5 tonnes ha^{-1} (±12.1 SE) and in the entire sediment profile (down to 1m deep) amounted to 42.7 tonne ha^{-1} (±6.9 SE). The total PAH comprised on average 0.2% (±0.03 SE) of TPH (Table S1, Table S2) and PAH concentrations were strongly correlated with TPH (R^2=0.76, P<0.001). The PAH concentrations did not exceed the SQGV threshold at any of the sampling sites nor depths and ranged between 0.42–35.5 mg kg^{-1}.

Concentrations of TPH and PAH were similar across all sampling depths with an exception of sediments at 30-50 cm depth that contained on average significantly higher concentration of TPH (30,441.6 mg kg $^{-1}$, Table S1, Fig. 2, Fig. S2). The most frequent component of TPH were hydrocarbons

with C-chain size of 15 to 28 carbon molecules accounting for 58% (±2 SE) of all TPH (Fig. S2, Fig. 3). Within total PAH, Pyrene was the most abundant component with an average frequency of 20% (±2 SE, Fig. 3). The average pyrene concentration was 38 times higher than the least abundant PAH type Acenaphthene (Fig. 3). The median fluoranthene:pyrene ratio was 0.06 (range: 0.003–1).

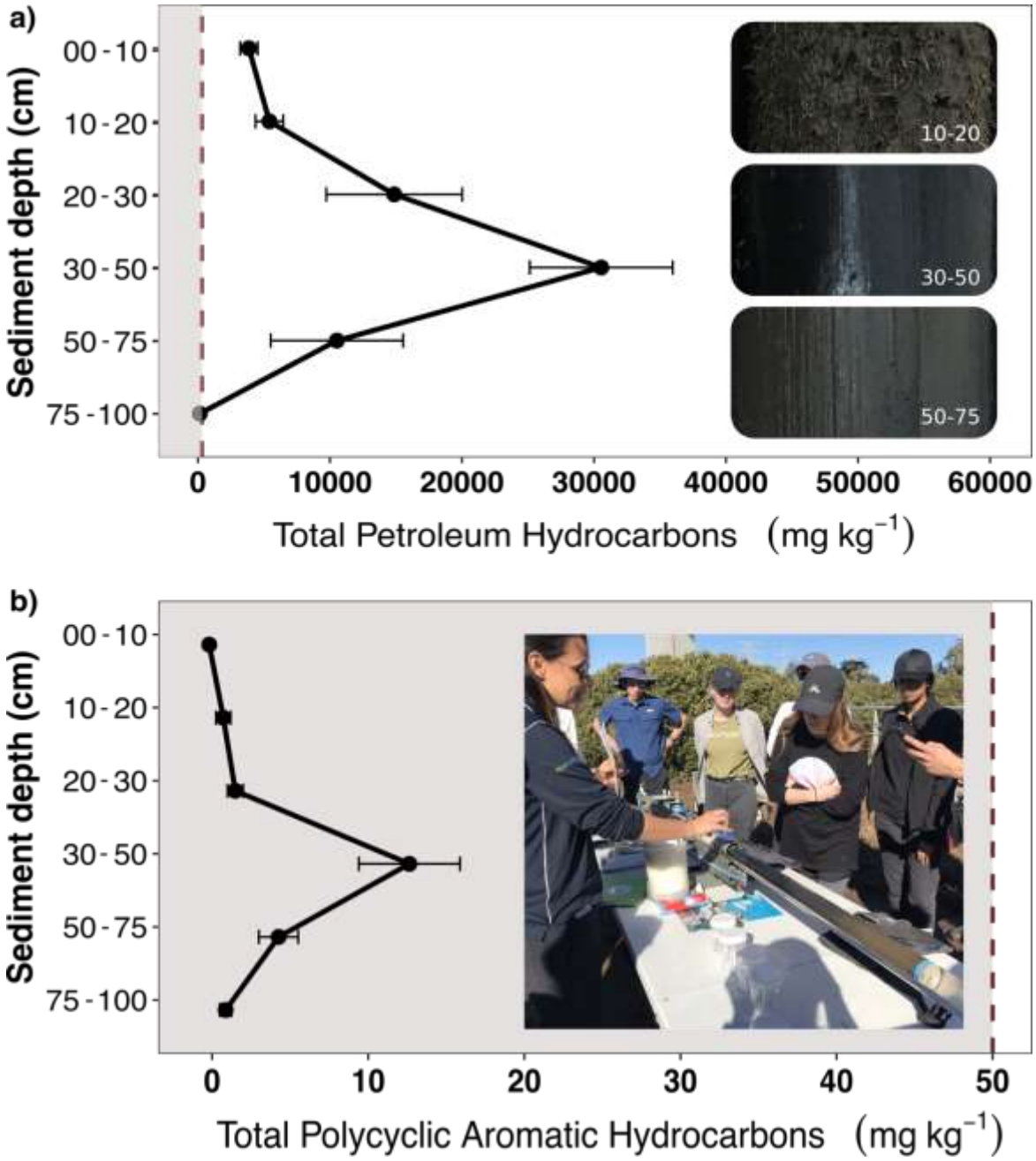

Fig. 2. Concentration of a) TPH and b) PAH (normalized to 1% of organic matter) in a 0-100 cm soil depth profile at the Stony Creek estuary, Australia. Vertical dotted lines indicate SQGV toxicity

level associated with adverse biological effects (280 mg kg^{-1} for of TPHs and at 50 mg kg^{-1} for PAHs). Values denote average ±SE.

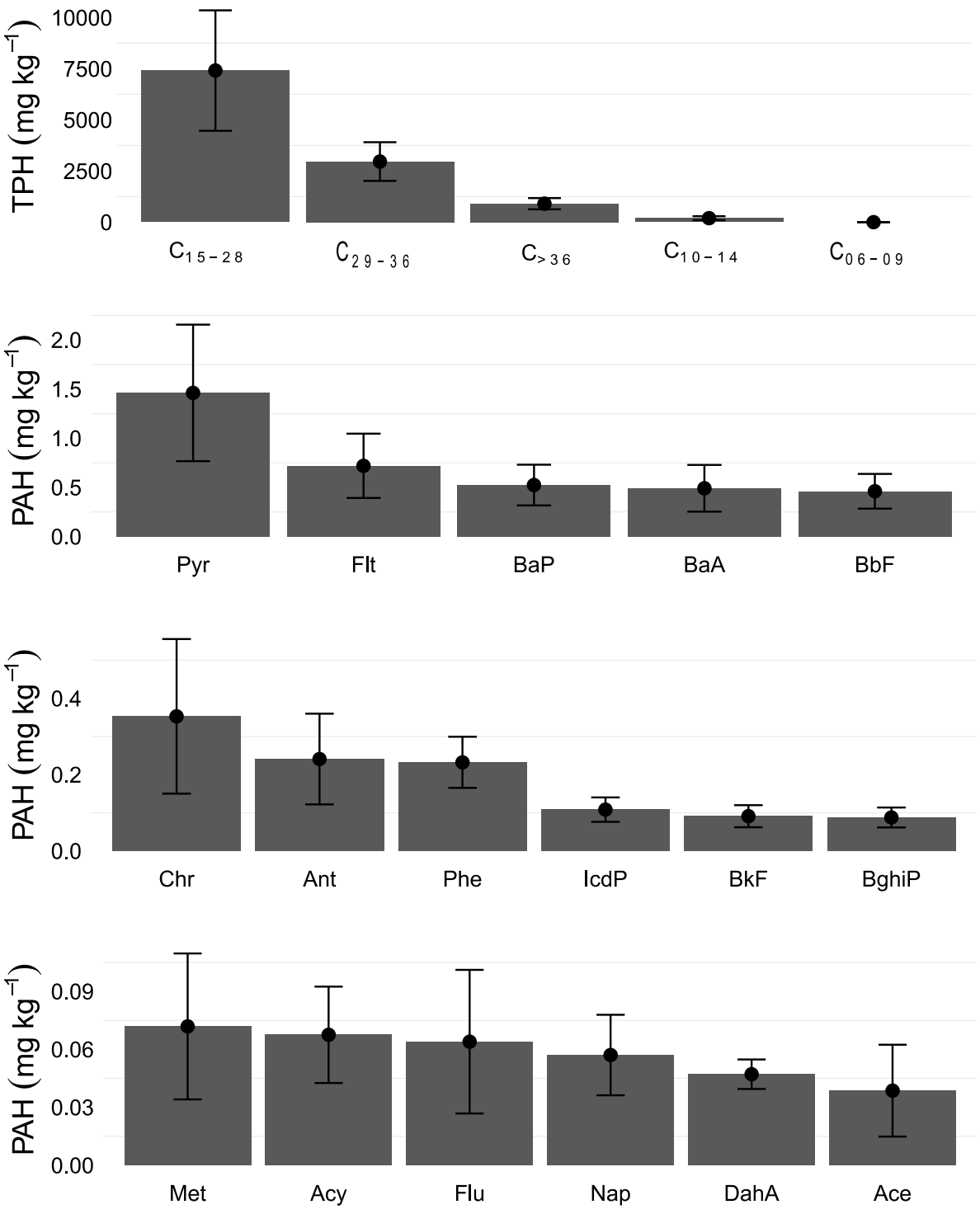

Fig. 3 Concentrations of TPH (TPH C_{6-9}, TPH C_{10-14}, TPH C_{15-28}, TPH C_{29-36}, TPH $C_{>36}$) and PAH: Naphthalene (Nap), 2-Methylnaphthalene (Met), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benz(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (Bkf),

Benzo(a)pyrene (BaP), Indeno(1,2,3-c,d)pyrene (IcdP), Dibenz(a,h)anthracene (DahA), Benzo(ghi)perylene (BghiP), at Stony Creek, Australia. See Table S1 and S2 for detailed information on the relative TPH and PAH content in the sediments (1 m soil profile). Values denote average ±SE. Note different scaling of x-axes.

Capping capability

Rate of sedimentation at planted mangroves sites, as detected in the top 30 cm, was on average 6.6 mm $year^{-1}$ (±0.6 SE) Organic matter content was highest in the top 0–10 cm of sediments (24.2% ±1.5 SE) and gradually decreased with depth: 15% ±0.9 SE at 10–20 cm and 10.6% ±0.8 SE at 20–30 cm.

Discussion and conclusions

This study demonstrates the ability of planted mangroves to cap an oil spill that exceeded SQGV toxicity thresholds by up to 220-fold. We found that mangroves successfully established on top of the oil-contaminated sediments (30–50 cm depth, dating back to 1980's) accumulating relatively high organic matter content within rhizosphere (~16.6% in top 30 cm) and supporting sediment accretion at a rate within the range found in natural mangroves (Ellison, 1999). The high accumulation of sediments was reported in previous studies in relation to mangroves ability to trap marine pollutants (Ashok et al., 2019; Bird, 1986) but has not been reported in relation to their capping service. Here, the characteristics of the site may have facilitated the capacity of mangroves to cap an oil spill. For example, sites that experience naturally high sediment deposition due to the hydrology may allow the oil to settle in such a location. By then planting mangroves at such a site may further help to immobilise and remediate affected sediments. In this study, oil spill was located on impermeable basalt bedrock and likely in-filled with clay soil substrate prior mangrove planting.

Interestingly, the top 30 cm of mangrove rhizosphere also showed high TPH pollution suggesting an upward sorption of oil contaminants (from highly polluted 30–50 cm depth) likely driven by the expanding rhizosphere itself (Patowary et al., 2017). Concentration of TPH pollutants on these

top layers was on average 29-fold above the toxicity threshold (max. 190-fold) and four times above the intervention value (2000 mg kg^{-1}) that indicates immediate soil remediation action (Pinedo et al., 2013).

The high toxicity at these study sites was not driven by any of the analysed PAHs, as their concentrations were below safety thresholds. It is likely that the low PAH concentration is related to the susceptibility of PAH components to physical weathering in intertidal conditions. For example, a study by Ke (2002) suggests PAH concentrations may decrease by 56% within three months of a mangrove oil spill. Given PAHs in this study were still detectable ~35 years after the spill, suggests that PAH concentration was high in the past, enabling us to assess their source presently. For example, the low ratio of two indicator PAH types, fluoranthene:pyrene (<1), suggests strong petrogenic origin of the oil spill (Tam et al., 2001). Similarly, the consistent TPH:PAH ratio distribution in the sediments suggests that the pollutants originated from the same source and within a relatively short period of time, given that weathering processes and oil spill chemical characterisation are a function of time and oxidation levels (Tarr et al., 2016; Wang and Fingas, 2003).

Our results show that mangrove forests may be capable of immobilizing high volumes of toxic petroleum hydrocarbons given their ability to accumulate sediments above the discrete layer of oil-contaminated sediments. However, extreme sea level-related events may pose risk to planted mangroves (Lovelock et al., 2017). To prevent a potential mangrove dieback and the release of hydrocarbons into the coastal environment, it is critical to put in place remediation actions and plans to protect and monitor mangroves (Newton et al., 2020; Ossai et al., 2020). Some potential remediation approaches include the use of hydrocarbon-degrading microbial communities (Daccò et al., 2020; Kumar et al., 2019; Machado et al., 2019), the removal and oxidation of contaminated sediments (Iturbe et al., 2004), the introduction of biochar into affected sediments (Mukome et al., 2020), or phytoremediation involving additional mangrove planting combined with hydrocarbon-degrading microorganisms (Verâne et al., 2020). Climate change, along with the rising sea level, and increasing intensity of storms are already accelerating the degradation and loss of mangrove

ecosystems worldwide (Duke et al., 2017; Zhang et al., 2019). The disturbance or loss of the Stony Creek mangrove forest and the subsequent release of toxic sediments, could have devastating repercussions to coastal fauna and flora.

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Supplementary Information (appendices)

- Table S1 Concentrations of all TPH and PAH types across sampling depths.
- Table S2 Total TPH stock (tonnes ha^{-1}) and TPH stock below the oil spill horizon.
- Table S3. Effect of sediment depths (cm) on TPH and PAH concentrations.
- Fig. S1 Time-stamped satellite imagery of oil spill study area.
- Fig. S2 Concentrations of TPH (Total TPH, TPH C_{6-9}, TPH C_{10-14}, TPH C_{15-28}, TPH C_{29-36}, TPH $C_{>36}$) at six depth intervals (0–10, 10–20,20–30, 30–50,50–75 and 75–100 cm).

- **Table S1 Average concentrations of all TPH (TPH C_{6-9}, TPH C_{10-14}, TPH C_{15-28}, TPH C_{29-36}, TPH $C_{>36}$) and PAH: Naphthalene (Nap), 2-Methylnaphthalene (Met), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benz(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (Bkf), Benzo(a)pyrene (BaP), Indeno(1,2,3-c,d)pyrene (IcdP), Dibenz(a,h)anthracene (DahA), Benzo(ghi)perylene (BghiP). Sediments (1 m soil profile). at Stony Creek, Australia. Total_PAH_norm and Total_TPH denote total concentrations of PAHs and TPHs, respectively. N denotes number of replicates and SE denotes standard error.**

Hydrocarbon type	Sampling depth (cm)	Average concentration (mg kg^{-1})	N	SE	Times above threshold 280 mg kg^{-1}
Ace	00_10	0.02	9.00	0.00	0.00
Ace	10_20	0.02	9.00	0.00	0.00
Ace	20_30	0.01	9.00	0.00	0.00
Ace	30_50	0.11	9.00	0.06	0.00
Ace	50_75	0.03	9.00	0.00	0.00
Ace	75_100	0.04	6.00	0.01	0.00
Acy	00_10	0.02	9.00	0.00	0.00
Acy	10_20	0.03	9.00	0.01	0.00
Acy	20_30	0.03	9.00	0.01	0.00
Acy	30_50	0.17	9.00	0.05	0.00
Acy	50_75	0.11	9.00	0.03	0.00
Acy	75_100	0.04	6.00	0.01	0.00
Ant	00_10	0.03	9.00	0.00	0.00
Ant	10_20	0.05	9.00	0.01	0.00
Ant	20_30	0.10	9.00	0.03	0.00
Ant	30_50	0.90	9.00	0.20	0.00
Ant	50_75	0.26	9.00	0.11	0.00
Ant	75_100	0.05	6.00	0.01	0.00
BaA	00_10	0.06	9.00	0.01	0.00
BaA	10_20	0.18	9.00	0.06	0.00
BaA	20_30	0.26	9.00	0.05	0.00
BaA	30_50	1.78	9.00	0.46	0.01
BaA	50_75	0.36	9.00	0.13	0.00

BaA	75_100	0.21	6.00	0.05	0.00
BaP	00_10	0.09	9.00	0.02	0.00
BaP	10_20	0.23	9.00	0.07	0.00
BaP	20_30	0.29	9.00	0.07	0.00
BaP	30_50	1.45	9.00	0.39	0.01
BaP	50_75	0.79	9.00	0.21	0.00
BaP	75_100	0.19	6.00	0.04	0.00
BbF	00_10	0.13	9.00	0.03	0.00
BbF	10_20	0.30	9.00	0.10	0.00
BbF	20_30	0.31	9.00	0.09	0.00
BbF	30_50	1.12	9.00	0.34	0.00
BbF	50_75	0.61	9.00	0.23	0.00
BbF	75_100	0.22	6.00	0.06	0.00
BghiP	00_10	0.04	9.00	0.00	0.00
BghiP	10_20	0.06	9.00	0.02	0.00
BghiP	20_30	0.05	9.00	0.01	0.00
BghiP	30_50	0.17	9.00	0.05	0.00
BghiP	50_75	0.13	9.00	0.03	0.00
BghiP	75_100	0.07	6.00	0.02	0.00
BkF	00_10	0.04	9.00	0.00	0.00
BkF	10_20	0.06	9.00	0.02	0.00
BkF	20_30	0.06	9.00	0.01	0.00
BkF	30_50	0.18	9.00	0.06	0.00
BkF	50_75	0.13	9.00	0.04	0.00
BkF	75_100	0.07	6.00	0.02	0.00
Chr	00_10	0.05	9.00	0.01	0.00
Chr	10_20	0.13	9.00	0.04	0.00
Chr	20_30	0.15	9.00	0.02	0.00
Chr	30_50	1.37	9.00	0.44	0.00
Chr	50_75	0.22	9.00	0.07	0.00

Chr	75_100	0.12	6.00	0.03	0.00
DahA	00_10	0.04	9.00	0.00	0.00
DahA	10_20	0.03	9.00	0.00	0.00
DahA	20_30	0.03	9.00	0.00	0.00
DahA	30_50	0.06	9.00	0.01	0.00
DahA	50_75	0.06	9.00	0.01	0.00
DahA	75_100	0.07	6.00	0.02	0.00
Flt	00_10	0.06	9.00	0.02	0.00
Flt	10_20	0.23	9.00	0.07	0.00
Flt	20_30	0.31	9.00	0.06	0.00
Flt	30_50	2.46	9.00	0.61	0.01
Flt	50_75	0.82	9.00	0.23	0.00
Flt	75_100	0.29	6.00	0.09	0.00
Flu	00_10	0.02	9.00	0.00	0.00
Flu	10_20	0.02	9.00	0.00	0.00
Flu	20_30	0.02	9.00	0.01	0.00
Flu	30_50	0.23	9.00	0.09	0.00
Flu	50_75	0.04	9.00	0.01	0.00
Flu	75_100	0.04	6.00	0.01	0.00
IcdP	00_10	0.06	9.00	0.01	0.00
IcdP	10_20	0.07	9.00	0.03	0.00
IcdP	20_30	0.06	9.00	0.01	0.00
IcdP	30_50	0.21	9.00	0.06	0.00
IcdP	50_75	0.17	9.00	0.04	0.00
IcdP	75_100	0.07	6.00	0.02	0.00
Met	00_10	0.02	9.00	0.00	0.00
Met	10_20	0.04	9.00	0.01	0.00
Met	20_30	0.04	9.00	0.01	0.00
Met	30_50	0.23	9.00	0.09	0.00
Met	50_75	0.06	9.00	0.02	0.00

Met	75_100	0.04	6.00	0.01	0.00
Nap	00_10	0.02	9.00	0.00	0.00
Nap	10_20	0.03	9.00	0.01	0.00
Nap	20_30	0.03	9.00	0.01	0.00
Nap	30_50	0.15	9.00	0.04	0.00
Nap	50_75	0.06	9.00	0.01	0.00
Nap	75_100	0.04	6.00	0.01	0.00
Phe	00_10	0.05	9.00	0.01	0.00
Phe	10_20	0.15	9.00	0.05	0.00
Phe	20_30	0.15	9.00	0.03	0.00
Phe	30_50	0.48	9.00	0.09	0.00
Phe	50_75	0.35	9.00	0.10	0.00
Phe	75_100	0.20	6.00	0.08	0.00
Pyr	00_10	0.08	9.00	0.02	0.00
Pyr	10_20	0.29	9.00	0.09	0.00
Pyr	20_30	0.92	9.00	0.27	0.00
Pyr	30_50	4.89	9.00	1.26	0.02
Pyr	50_75	1.88	9.00	0.72	0.01
Pyr	75_100	0.33	6.00	0.10	0.00
Total_PAH_norm	00_10	0.84	9.00	0.11	0.00
Total_PAH_norm	10_20	1.91	9.00	0.55	0.01
Total_PAH_norm	20_30	2.81	9.00	0.59	0.01
Total_PAH_norm	30_50	15.96	9.00	3.82	0.06
Total_PAH_norm	50_75	6.08	9.00	1.49	0.02
Total_PAH_norm	75_100	2.08	6.00	0.44	0.01

Total_TPH	00_10	3834.86	9.00	659.96	13.70
Total_TPH	10_20	5385.97	9.00	1045.51	19.24
Total_TPH	20_30	14814.69	9.00	5130.47	52.91
Total_TPH	30_50	30441.67	9.00	5396.28	108.72
Total_TPH	50_75	10481.08	9.00	5004.21	37.43
Total_TPH	75_100	125.18	6.00	63.84	0.45
TPH_06to09	00_10	0.41	9.00	0.02	0.00
TPH_06to09	10_20	0.26	9.00	0.01	0.00
TPH_06to09	20_30	0.23	9.00	0.02	0.00
TPH_06to09	30_50	0.24	9.00	0.02	0.00
TPH_06to09	50_75	0.23	9.00	0.01	0.00
TPH_06to09	75_100	0.23	6.00	0.01	0.00
TPH_10to14	00_10	13.71	9.00	11.04	0.05
TPH_10to14	10_20	19.97	9.00	6.14	0.07
TPH_10to14	20_30	142.32	9.00	50.71	0.51
TPH_10to14	30_50	657.71	9.00	138.25	2.35
TPH_10to14	50_75	275.84	9.00	145.81	0.99
TPH_10to14	75_100	0.77	6.00	0.41	0.00
TPH_15to28	00_10	1666.28	9.00	208.76	5.95

TPH_15to28	10_20	2783.78	9.00	598.80	9.94
TPH_15to28	20_30	8728.04	9.00	3105.63	31.17
TPH_15to28	30_50	21511.70	9.00	4120.69	76.83
TPH_15to28	50_75	7267.28	9.00	3563.20	25.95
TPH_15to28	75_100	88.88	6.00	48.84	0.32
TPH_29to36	00_10	1454.50	9.00	294.51	5.19
TPH_29to36	10_20	1826.15	9.00	350.84	6.52
TPH_29to36	20_30	4426.66	9.00	1480.00	15.81
TPH_29to36	30_50	6683.76	9.00	1050.85	23.87
TPH_29to36	50_75	2384.15	9.00	1080.94	8.51
TPH_29to36	75_100	28.55	6.00	13.21	0.10
TPH_37toUP	00_10	699.96	9.00	202.90	2.50
TPH_37toUP	10_20	755.81	9.00	203.83	2.70
TPH_37toUP	20_30	1517.44	9.00	517.34	5.42
TPH_37toUP	30_50	1588.26	9.00	243.85	5.67
TPH_37toUP	50_75	553.58	9.00	224.21	1.98

TPH_37toUP	75_100	6.76	6.00	6.29	0.02

Table S2 TPH stock (tonnes ha^{-1}) and TPH stock below the oil spill horizon at the three study sites.

Carbon type	Mangrove Arrival Year	Stock (tonnes ha^{-1})	n	SE	Depth (m)
TPH below	1986	41.6	3	6.2	0.3–1
TPH below	2002	35.1	3	15.7	0.14–1
TPH below	2006	41.9	3	14.3	0.08–0.75
TPH total	1986	48.0	3	7.6	0–1
TPH total	2002	37.3	3	16.5	0–1
TPH total	2006	42.7	3	14.6	0–0.75

Table S3. Effect of sediment depths (cm) on TPH (n=51, R2 = 0.45) and PAH content (n=51, R2= 0.58). Bold font indicates significant difference in TPH and PAH content (P < 0.01).

	Total TPH (mg kg^{-1})				Total PAH (mg kg^{-1})			
Depth (cm)	*Estimate*	*SE*	*t*	*p*	*Estimate*	*SE*	*t*	*p*
[0-10 cm] (intercept)	3834.8	3818.4	1.00	0.32	8.2	6.4	1.27	0.21
[10-20 cm]	1551.1	5400	0.29	0.78	5.7	9.1	0.63	0.54
[20-30 cm]	10979.8	5400	2.03	0.05	14.4	9.1	1.58	0.12
[30-50 cm]	26606.8	5400	4.93	**<0.01**	61.7	9.1	6.79	**<0.01**
[50-75 cm]	6646.2	5400	1.23	0.23	21.6	9.1	2.38	0.02
[75-100 cm]	-3709.6	6037.4	-0.61	0.54	-0.7	10.2	-0.06	0.95

Fig. S1 Timestamped satellite imagery of oil spill study area at Stony Creek, Victoria, Australia (as of 17/Jan/2020).

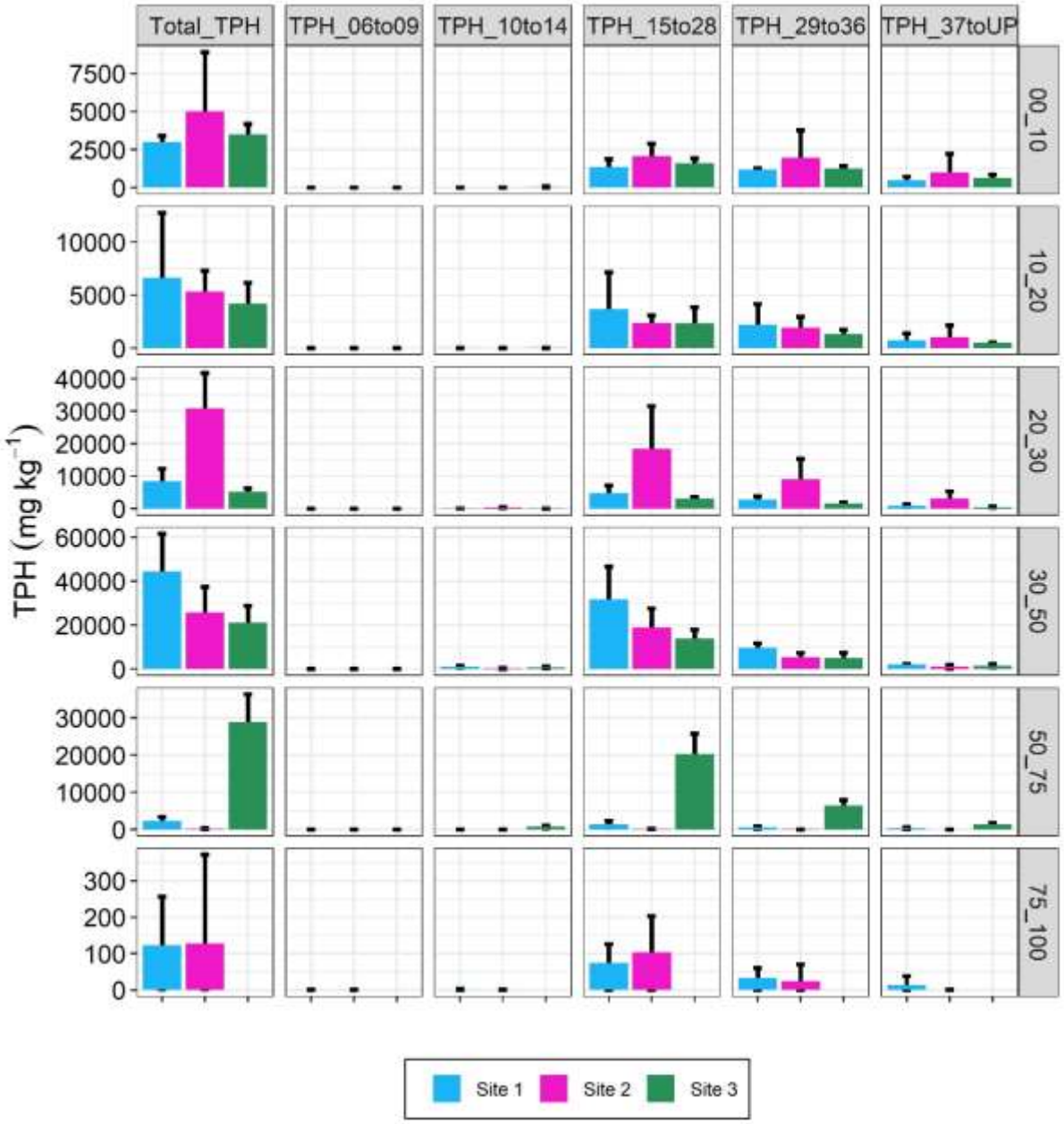

Fig. S2 Concentrations of TPH (Total TPH, TPH C_{6-9}, TPH C_{10-14}, TPH C_{15-28}, TPH C_{29-36}, TPH $C_{>36}$) at six depth intervals (0-10, 10-20,20-30, 30-50,50-75 and 75-100 cm) across three sampling sites, Stony Creek, Australia. Site 3 was sampled till maximum 75 cm depth due to bedrock bottom.