

Australasian asphaltite strandings revisited: their origin and the effects of weathering and biodegradation on their biomarker and isotopic profiles

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Abstract

Reports of bitumen strandings on the coastlines of South Australia, Victoria, Tasmania and Western Australia date from the early 19th Century (Sprigg and Woolley, 1963; Currie et al., 1992; Volkman et al., 1992; McKirdy et al., 1994; Padley, 1995; Edwards et al., 1998 and references therein). The locations of these strandings along Australia's southern margin (Fig. 1), and their greater frequency in southeastern South Australia, western Victoria and southern Tasmania, fuelled early petroleum exploration in the region on the assumption that they were sourced from local submarine seepages (Sprigg, 1986; Volkman et al., 1992; McKirdy et al., 1994). Accounts describe a variety of oily substances that can be assigned to three categories, each with a different origin: oils (crude and refined), waxy bitumens and asphaltites (McKirdy et al., 1986, 1994; Padley, 1995; Edwards et al., 1998).

The focus of the present study involves detailed molecular and isotopic characterisation of the asphaltites (i.e. the Family 4 costal bitumens of McKirdy et al., 1986, 1994). Geochemically quite distinct from the waxy bitumens, these are heavy, sulfur-rich, aromatic-asphaltic crudes (4–18° API; ~4% S; 57–84% asphaltenes) that commonly strand as large jet-black ovoid lumps (up to 670 mm across and 7 kg in weight) at the high water mark on medium to high energy, gently sloping sandy beaches (McKirdy et al., 1994; Padley, 1995). Unlike the waxy bitumens, which have positive buoyancy, the Australasian asphaltites are on average slightly denser than seawater and therefore are likely to have resided mostly within the water column prior to stranding. The fresh strandings have a strong petroliferous odour (Sprigg and Woolley, 1963; Volkman et al., 1992; Padley, 1995; Edwards et al., 1998). Their upper surface is characteristically traversed by shrinkage cracks and, although the interior is pliable when fresh, they become brittle upon storage and exhibit a conchoidal fracture pattern (Fig. 2). Their stable isotopic and molecular compositions (McKirdy et al., 1986, 1994; Currie et al., 1992; Volkman et al., 1992; Dowling et al., 1995; Padley, 1995; Edwards et al., 1998), including their enrichment in metalloporphyrins (Boreham et al., 2001; Totterdell et al., 2008), make them unique among Australasian crude oils. Moreover, historic (>100 years ago) and more recent strandings at sites in Western Australia, South Australia, Victoria and Tasmania and even as far afield as New Zealand and Macquarie Island (Fig. 1) are

of remarkably similar composition, suggesting that they all originated from the same offshore petroleum system (Padley, 1995; Edwards et al., 1998).

The source and age-specific biomarkers of these asphaltites indicate they are derived from a Cretaceous marine shale deposited under anoxic/sulphidic conditions, probably during an oceanic anoxic event (OAE) (McKirdy et al., 1994; Edwards et al., 1998; Boreham et al., 2001). In the Southern Hemisphere the Indian Ocean and contiguous Toolebuc and Blue Whale Seaways were sites of restricted circulation. Accordingly, euxinic marine sediments have been identified in several of the corresponding Australian depocentres, possibly recording the Cenomanian–Turonian OAE2 (Bonarelli Event) and shorter-lived late Albian oceanic anoxic subevent (OAE 1d; Breistroffer Event) (Edwards et al., 1999; Boreham et al., 2001; Struckmeyer et al., 2001; Pancost et al., 2005; Totterdell et al., 2008; Jenkyns, 2010).

The origin of the Australasian asphaltites has long been the subject of debate. No reliable correlation of the asphaltites to any oil produced locally on the southern margin of Australia or globally (using the GeoMark™ database, Summons et al., 2001) has been made. Thus the source of these enigmatic hydrocarbons remains in question.

The close proximity of common stranding sites to the locations of former whaling stations raises the possibility of an anthropogenic origin for the asphaltites. However, the common caulking materials recovered from shipwreck sites differ significantly in composition from the asphaltites (Smart, 1999).

An important clue to the likely origin of the asphaltites is the striking similarity of their biomarker and isotopic signatures to those of the late Albian Toolebuc Formation in the onshore Eromanga Basin (Boreham et al., 2001). Coeval units deposited along the Blue Whale Seaway, which may well host their enigmatic source facies, form part of the post-rift sag phase succession of the Blue Whale Supersequence in the Eyre and Ceduna sub-basins of the Bight Basin and the Otway Supergroup in the Otway Basin (Boreham et al., 2001; Struckmeyer et al., 2001; Bradshaw et al., 2003; Totterdell and Struckmeyer, 2003; Boulton et al., 2005; Totterdell et al., 2008; Blevin and Cathro, 2008; Boreham, 2008). However, is it possible that further clues to their origin might be derived from the asphaltite strandings themselves?

In this study we investigate a suite of asphaltites from four widely separated stranding localities: Limestone Coast (n = 3), Kangaroo Island (n = 4), Eyre Peninsula (n = 2) and Invercargill, New Zealand (n = 2) (Table 1). Sub-samples taken from the external weathered surface and 'fresh' interior of each asphaltite specimen were analysed by gas chromatography-mass spectrometry (GC-MS) and compound-specific isotope analysis

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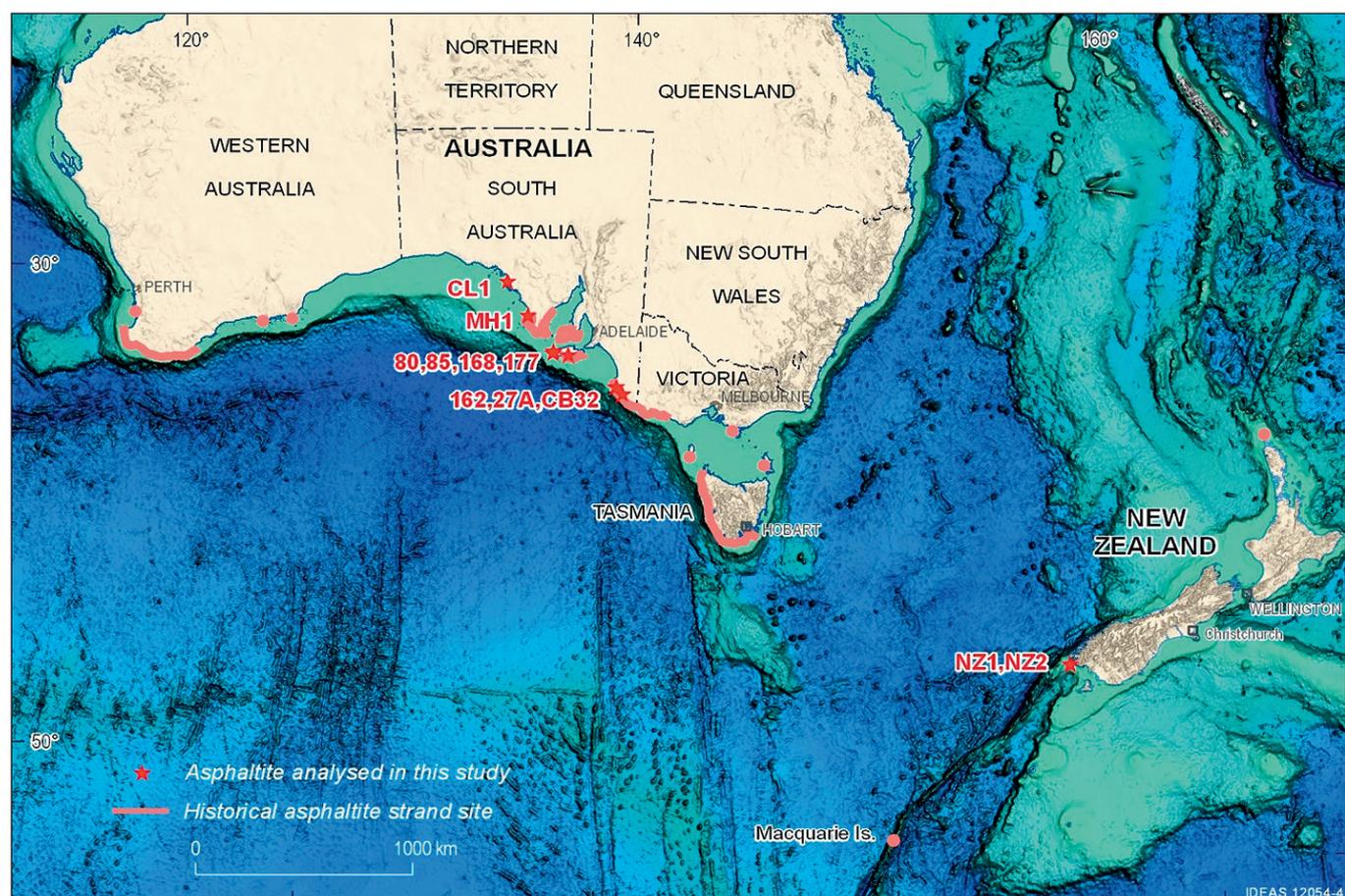


Figure 1. Location of historically documented asphaltite strandings and samples analysed in this study (after Edwards et al., 1998).

(CSIA). Comparison of the resulting data allowed 1) oil-oil correlation; and 2) determination of their degree of weathering which, as a function of residence time in the ocean, may be used to better constrain the location(s) of the parent seafloor seepage.

In investigating the provenance of the asphaltites stranded upon the shores of the southern margin of Australia, and further afield, it seemed appropriate to first reconsider the genesis of such asphaltites. The physical characteristics of the Australasian asphaltites provide additional clues to their origin. Laminations and flow structures observed in some specimens (Fig. 2A, D) are similar to those described in the asphaltic volcanoes of the Gulf of Mexico (Brüning et al., 2010; Schubotz et al., 2011) wherein surface cracking is also observed due to the *in situ* loss of volatiles. In fact, these submarine seeps of viscous bitumen display all the hallmarks of the Australasian asphaltite strandings: devolatilization cracks, large ovoid blocks, benthic bivalve and annelid communities, and the newly described flow characteristics.

Undoubtedly, the asphaltites have lost volatile components subsequent to their escape from the subsurface to the sea floor. This loss is likely to have involved both submarine dissolution, as evidenced by the shrinkage cracks on the upper surface of the stranded asphaltite (Fig. 2D), and subaerial evaporation, which accounts for its loss of plasticity observed once removed from the aqueous environment. The difference in distribution of bulk components between the inner and outer portions of the specimens is relatively small, as shown by the combined loss of saturated and aromatic hydrocarbons (average 5%) and the variation in the

C_{10-19}/C_{30} *n*-alkane ratio (average 13%). Thus, it is probable that most of the light-end loss from the parent crude oil occurred in the subsurface and that the asphaltites were discharged into the ocean as semi-solid bitumen. All the Australasian asphaltites lack 25-norhopanes, implying little in-reservoir bacterial alteration (Volkman et al., 1984). The levels of degradation, classified by Edwards et al. (1998) as being at Level 4 (or PM4) on the biodegradation scale of Peters and Moldowan (1993), which corresponds to moderate degradation on the scales of Wenger et al. (2001) and Peters et al. (2005), are calculated to be Manco 431, 419 [PM4] or ultimate Manco 578 [vector 31043310000] on the recently published scale of Larter et al. (2012), highlighting the fact that the initially generated oil has undergone substantial alteration. However, degradation through bacterial removal and/or intensive water washing had no part in their genesis. The level of biodegradation also suggests that if they emanate from natural submarine seepage it would be of low intensity (Wenger and Isaksen, 2002), an interpretation consistent with the pattern described for the Australian continental shelf (Logan et al., 2010) and ascribed to low recent burial rates. Given its arrival at the sea floor in the form of heavy oil, this modification must have occurred in the subsurface. The most likely mechanism is tar mat formation, due either to deasphalting of the main oil stringer along a flat-lying secondary migration pathway or subsequent invasion of an oil-bearing reservoir by gas from a more mature hydrocarbon kitchen.

Geochemical investigation of the inner ‘fresh’ portion of four previously unanalysed asphaltites from locations in the Bight Basin and New Zealand has shown that these more distant

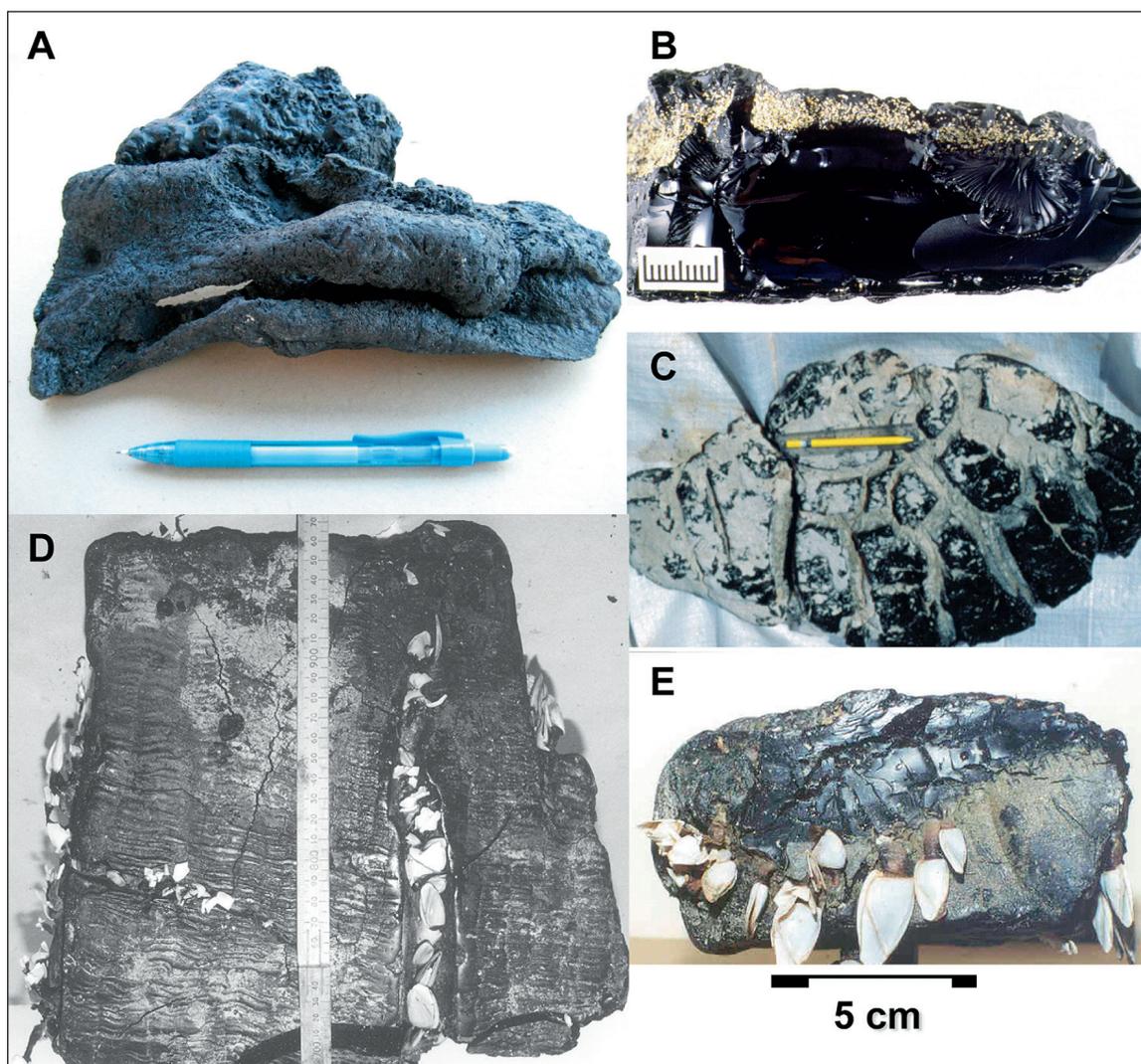


Figure 2. Examples of asphaltite strandings: A) an asphaltite from Port McDonnell (not analysed) with a rolled over edge indicative of viscous flow; B) sample 80, Ravine de Casours, Kangaroo Island, broken open to reveal the conchoidal fracture pattern typical of all asphaltites (scale bar 20 mm); C) sample 177, Bales Bay, Kangaroo Island, a large specimen exhibiting upper surface devolatilization cracks and a characteristic flat ovoid shape (long axis = 75 cm); d) sample NZ1, Invercargill, New Zealand with an unusual internal fabric suggestive of laminar flow, devolatilization cracks and a bivalve colony; e) sample NZ2, Invercargill, New Zealand, also colonised by bivalves. Photographs provided by D. McKirdy (plate A); D. Edwards, nee Padley (plates B and C); and D. Bradley (plates D and E).

Sample	Location	Year	Dimensions L:W:D (mm)	Weight (g)	Degree of Weathering
27A	Pether Rock, Canunda N.P., SA	1990	109:85:60	419	Mild
CB32	Nine Mile Sandhill, Beachport, SA	1990	127:112:49		Moderate
162	German Point, Beachport, SA	1983	328:204:102	2876	Mild
80	Ravine des Casoars, Kangaroo Island, SA	1991	165:100:48	634	Moderate/Heavy
85	Seal Bay, Kangaroo Island SA	1990		764	Moderate/Heavy
168	West Bay, Kangaroo Island SA	1990		1944	Mild/Moderate
177	Bales Bay, Kangaroo Island SA	1991	750:350:40	7000	Mild/Moderate
CL1	Streaky Bay, Eyre Peninsula, SA	2005	138:94:37		Heavy
MH1	S of the Freshmanns, Eyre Peninsula, SA	2005	83:71:26		Mild
NZ1	Invercargill, New Zealand	2002	273:256:87		Moderate
NZ2	Invercargill, New Zealand	2002	116:67:62		Moderate

Table 1. Sample identification, size and weight, location of stranding, year of collection and weathering description.

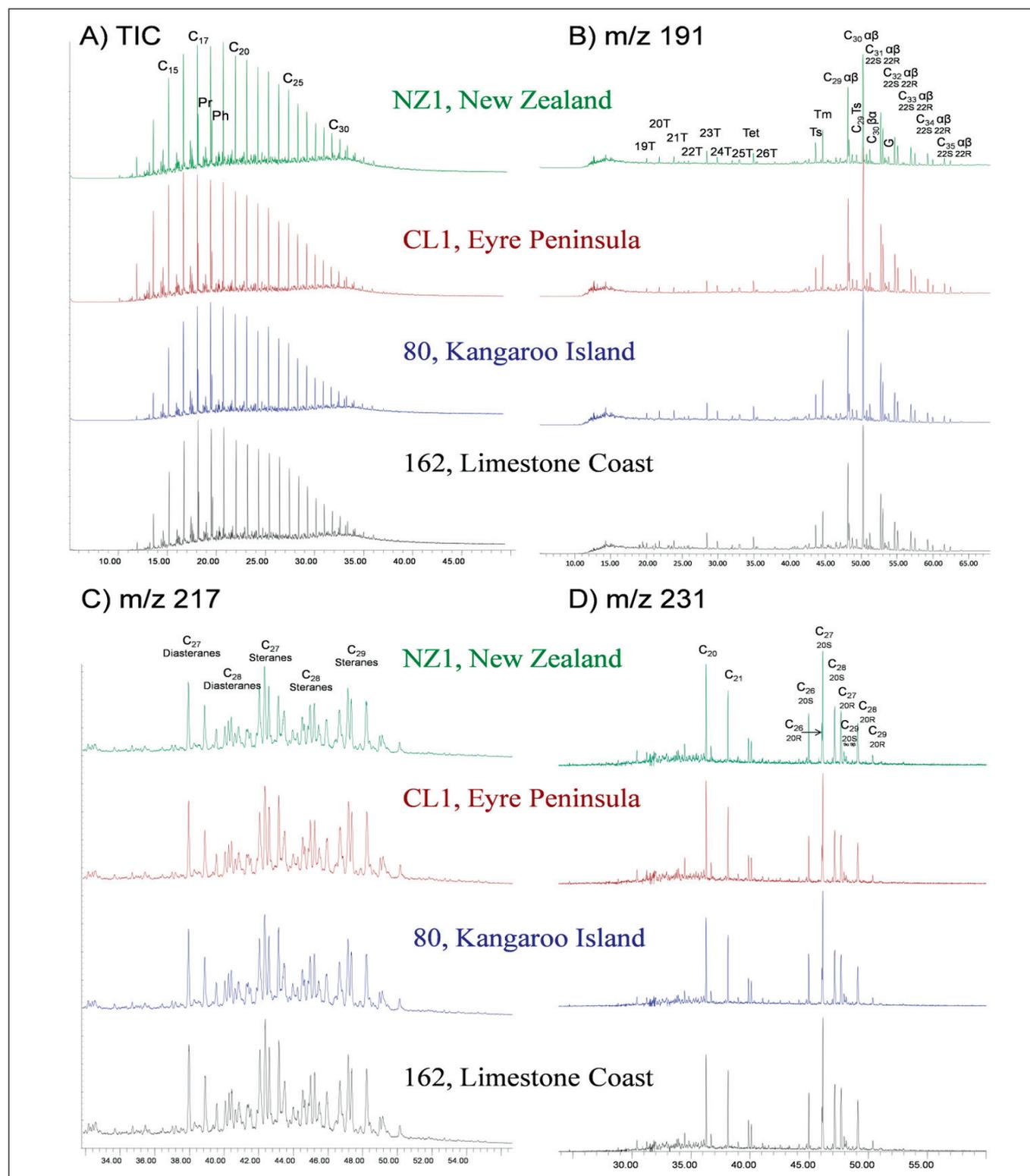


Figure 3. Selected chromatograms of saturated and aromatic hydrocarbons in the interior portion of representative asphaltite specimens from each stranding domain: A) TIC of saturated hydrocarbon fraction; B) *m/z* 191, terpanes; C) *m/z* 217, steranes; and D) *m/z* 231, triaromatic steroids.

Key:

Bulk Composition, normal & acyclic hydrocarbons (fullscan)

C_n = normal alkane C_{carbon number}
 e.g. C₁₅ = pentadecane (C₁₅H₃₂)
 Pr = pristane
 Ph = phytane

Hopanes (m/z 191)

T = tricyclic terpanes note C₂₅ and C₂₆ homologues have a chiral centre at C₂₂ (R & S)
 Tet = C₂₄ tetracyclic terpane
 Ts = C₂₇ 18α(H)-22,29,30-trisnorhopane
 Tm = C₂₇ 17α(H)-22,29,30-trisnorhopane
 C₂₉αβ = C₂₉ 17α(H),21β(H)-hopane

C₂₉ Ts = C₂₉ 30-norhopane
 C₃₀αβ = C₃₀ 17α(H),21β(H)-hopane
 C₃₀βα = C₃₀ 17β(H),21α(H)-hopane
 C_{xx}αβ = C_{xx} 17α(H),21β(H)-homohopane
 (xx = carbon number i.e. 31 through 35)
 G = Gammacerane

Steranes (m/z 217 & 218)

C₂₇ Steranes = C₂₇ 5α(H) steranes (20R + 20S)
 C₂₈ Steranes = C₂₈ 5α(H) steranes (20R + 20S)
 C₂₉ Steranes = C₂₉ 5α(H) steranes (20R + 20S)
 C₂₇ Diasteranes = C₂₇ 13α(H),17α(H) diasteranes (20R + 20S)
 C₂₈ Diasteranes = C₂₈ 13α(H),17α(H) diasteranes (20R + 20S)

Triaromatic Steroids (m/z231)

C₂₀ = C₂₀ Triaromatic Steroid
 C₂₁ = C₂₁ Triaromatic Steroid
 C₂₆ 20R = C₂₆ Triaromatic Steroid 20R
 C₂₆ 20S = C₂₆ Triaromatic Steroid 20S
 C₂₇ 20R = C₂₇ Triaromatic Steroid 20R
 C₂₇ 20S = C₂₇ Triaromatic Steroid 20S
 C₂₈ 20R = C₂₈ Triaromatic Steroid 20R
 C₂₈ 20S = C₂₈ Triaromatic Steroid 20S
 C₂₉ 20R = C₂₉ Triaromatic Steroid 20R
 C₂₉ 20Sα = C₂₉ Triaromatic Steroid 9α,20S
 C₂₉ 20Sβ = C₂₉ Triaromatic Steroid 9β,20S

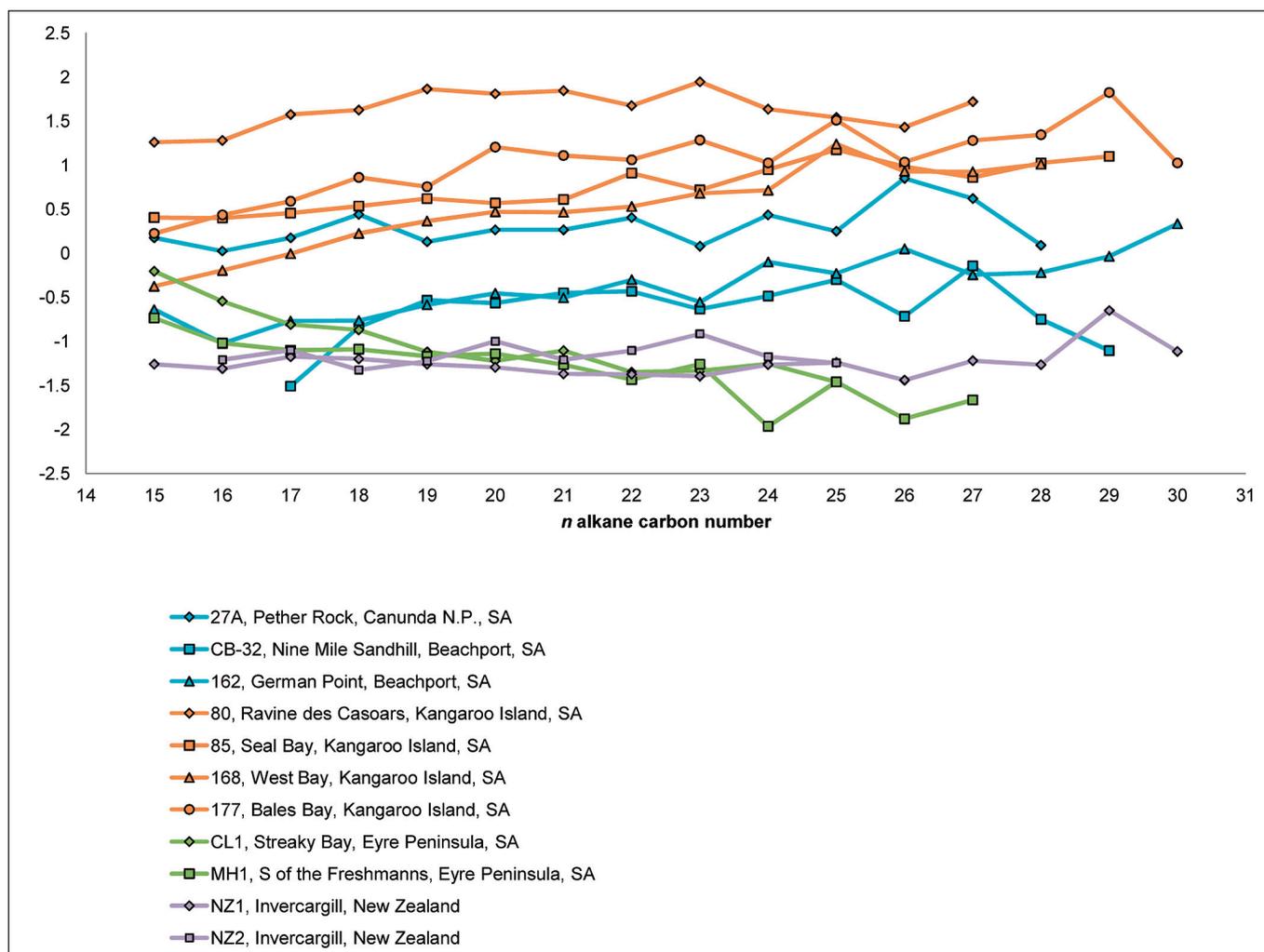


Figure 4. Plots of n-alkane $\delta^{13}\text{C}$ versus carbon number for asphaltite specimens, showing the difference between their inner and outer portions.

strandings may be correlated with a further seven members of the classic ‘Family 4’ asphaltites from common stranding sites on the Limestone Coast and Kangaroo Island. The compositional uniformity of the sample suite, evident across a broad spectrum of parameters, with representative chromatograms from the GC-MS analyses shown in Fig.3, is a clear indication that these asphaltites belong to the same oil family, notwithstanding their disparate stranding localities. Moreover, their bulk composition and biomarker distributions are remarkably similar to those previously reported for other stranded southern Australian asphaltites (Currie et al., 1992; Volkman et al., 1992; McKirdy et al., 1994; Padley, 1995; Edwards et al., 1998; Boreham et al., 2001).

Comparison of inner, fresher portions of each stranding to the outer, weathered surface section revealed subtle but relatively consistent variations in a number of degradation-sensitive components. Molecular fossil and compound-specific $\delta^{13}\text{C}$ signatures (Fig.4) in the saturated hydrocarbons most susceptible to alteration by biodegradation and dissolution suggest that the specimens recovered from the Eyre Peninsula and New Zealand have had longer exposure to weathering in the oceanic realm than those from the Limestone Coast and Kangaroo Island. The corresponding aromatic hydrocarbon distributions of the inner and outer portions differ in a manner consistent with the physical appearance of the strandings.

Within the offshore Bight Basin, the western section of the Ceduna Sub-basin hosts thermally immature organic-rich mudstones with marine biomarker signatures similar (but not identical) to those of the asphaltites (Totterdell et al., 2008), sampled from locations in which shelf-break canyons incise the Cretaceous section. The eastern part of the Ceduna Sub-basin is host to natural seepage and potential source rocks of the required age and thermal maturity (Boult, 2012), where a possible mode of transportation to the inner shelf is available via the upwelling Flinders Current (Middleton and Bye, 2007). However, shelf-break canyons do not appear to expose the Cretaceous section and the seismically interpreted seep features are active on the shelf (Boult, 2012). Migration of a viscous tar mat from a highly specific source rock to the sea floor of the shelf, uncontaminated by the other petroleum systems operating within this sector of the sub-basin, makes this a less likely (though viable) scenario.

The Morum Sub-basin is located within the western offshore Otway Basin, adjacent to the Limestone Coast and southern Kangaroo Island where the least weathered asphaltite strands. This depocentre has what may be the crucial feature, namely a deep shelf-break canyon that cuts deeply into a toe-thrust inversion interpreted to host active source rocks of the appropriate Albian age (Boult et al., 2005). The associated system of steeply dipping faults provides migration pathways along which light hydrocarbons could have continued their movement to reservoirs

higher in the sequence, leaving behind a residual tar mat. This heavy asphaltic bitumen may now be exposed in the floor or walls of the canyon, oozing slowly onto the seafloor to form volcano-like structures. Transport of the asphaltite along the canyon and up onto the shelf can be accounted for by the summer upwelling of the deep oceanic Flinders Current (Middleton and Bye, 2007), where it may be dispersed by the inshore coastal current or accumulate until being washed ashore by winter storm events. Historical distribution patterns support this hypothesis, with the highest concentration of strandings found on the coast closest to the head of the Morum Sub-basin canyons and on Kangaroo Island, which lies westward on the summer route of the Flinders Current. For the remainder of the year the shallow coastal current flows southeastward along the shelf before interacting with the Subantarctic Front of the Antarctic Circumpolar Current system (Rintoul et al., 2001; Middleton and Bye, 2007), thereby providing momentum for the carriage of asphaltite further on towards the Victorian and Tasmanian coasts and beyond.

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