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Rapid heating of carbonaceous matter by igneous intrusions in carbon-rich shale, Isle of Skye, Scotland: an analogue for heating of carbon in impact craters?

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Abstract: The response of organic matter to high-temperature events is important to astrobiology, as it governs the survival of carbon during several processes that may be critical to the origin and spread of life. Impact cratering is a widespread high-temperature process. The behaviour of carbon during impact events is not well understood. But there is the potential to examine other examples of the response of organic matter to high-temperature processes in the terrestrial geological record. In this study, we report on the interaction of Tertiary igneous intrusions (dolerite sills) and carbon-rich Jurassic mudrocks on the Isle of Skye, Scotland. Despite the high temperatures of the igneous intrusion, carbon has been preserved at the dolerite–shale contact and in shale enclaves where partial melting of the shale has occurred. Even though the temperatures achieved by igneous intrusion are much lower than during impact events, it is a valuable analogue, because it represents a rapid introduction of high temperatures to a carbon-rich rock.

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Introduction

Several processes that may be critical to the origin and spread of life, such as the survival of carbon during atmospheric entry of carbon-bearing meteorites and interplanetary dust particles, the impact of carbon-bearing meteorites and meteorite impact into carbon-bearing target rocks, all involve organic matter being subjected to high temperatures. The survival of carbon through these processes thereby has a strong influence over the key issues of panspermia (Melosh 1988; Parsons 1996; Raulin-Cerceau 1998; Coulson 2004) and the impact frustration of life (Maher & Stevenson 1988; Whittet 1997).

The occurrence of carbon in a number of terrestrial impact craters, including the very large Sudbury (French 1968; Avermann 1994; Bunch 1999) and Vredefort (England *et al.* 2002) craters, shows that carbon can survive large-scale impacts. However, the behaviour of carbon during these events is not well understood (French 1998, 2004) and in some cases carbon could be supplied to the crater post-impact (French 1998; Martinez-Ibarra 2003), therefore telling us nothing about survival of organic materials during impact. The greatest uncertainty is over what happens to carbon where the temperature is highest, in impact-generated melts, at crater centres and during atmospheric entry, where melting of rock is likely. Occurrences of carbon in melted impact materials (Chadwick *et al.* 2001; Lindgren & Parnell 2005) require careful interpretation to understand their history.

To help this interpretation, there is the potential to examine other examples of the response of organic matter to high-temperature events in the terrestrial geological record. In this study, we report on the interaction of Tertiary igneous material (dolerite sills) and carbon-rich Jurassic mudrocks on the Isle of Skye, Scotland. This is a valuable analogue for investigation of the effects on carbonaceous material during impact events, because it represents a rapid introduction of high temperatures to a carbon-rich rock.

On the Atlantic seaboard of the United Kingdom, Tertiary intrusion into Jurassic shales is common, where the Tertiary igneous province overlaps with the region of Jurassic marine sedimentation. Intrusion is particularly prevalent in shale horizons because they are mechanically weak, and therefore preferential sites for the emplacement of sills. Thus, sills are commonly encountered in Jurassic shales onshore in Skye (Anderson & Dunham 1966) and in Northern Ireland (Wilson & Manning 1978; McCann 1988), and also in offshore hydrocarbon exploration wells (Ritchie *et al.* 1999). As the shales are the sources of offshore hydrocarbon prospects, there is commercial interest in the potentially destructive consequences of intrusion on organic matter in shales or in

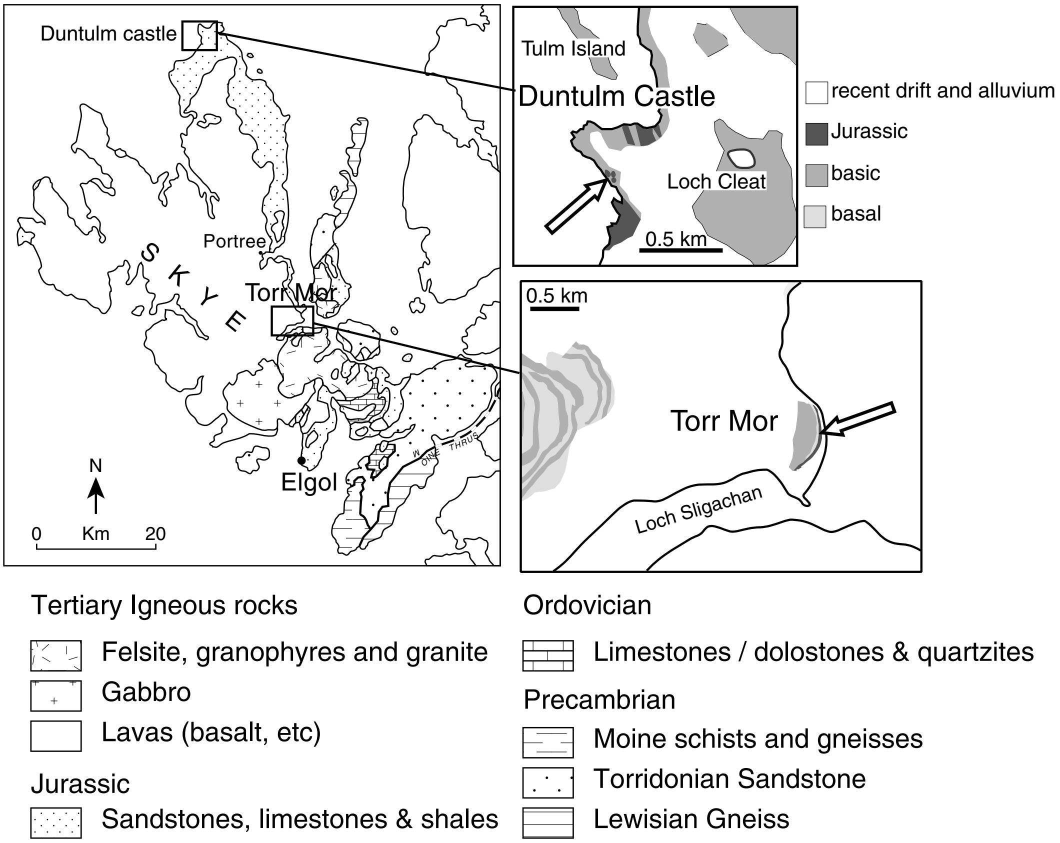

Fig. 1. A simplified geological map of Skye, showing the sample localities and detailed maps of Torr Mor and Duntulm Castle.

oil accumulations along the Atlantic margin (e.g. Price & Whitham 1997; Archer *et al.* 2005; Linnard & Nelson 2005).

Geological setting

The Isle of Skye is situated on the west coast of Scotland. The geology is dominated by Tertiary volcanic plateau basalt lavas and sills (Anderson & Dunham 1966; Fyfe *et al.* 1993). Sedimentary rocks of Mesozoic age outcrop around the margins of the igneous rocks, mainly on the northern and eastern parts of the island. The Mesozoic sedimentary succession includes Jurassic carbon-rich marine mudrocks and shales (Hudson 1983; Fyfe *et al.* 1993). The shales are thermally immature, i.e. they have not been buried deeply enough to generate oil in the normal manner (Kilyeni & Standley 1985).

The contact between the igneous rocks and the carbon-rich shales is exposed at several localities on the island (Fig. 1). The contacts have been the subject of several geochemical studies (Bishop & Abbott 1991, 1993; Katz *et al.* 1998; Farrimond *et al.* 1999; Kemp *et al.* 2005). Examination of material from the sill–shale contact and from enclaves of shale enclosed in the igneous rock allows us to investigate the effect of rapid heating on a carbon-rich rock and its interaction with high-temperature melting.

Specific issues to be addressed are (i) the degree to which carbon survived the process, (ii) whether any mobile carbon phases were formed and (iii) evidence for interaction between carbon and melted rock.

Methods of investigation

Samples were collected from three localities: Torr Mor, Duntulm Castle and Elgol (Fig. 1). Thin sections of the samples were examined in a petrographic microscope and in a scanning electron microscope equipped with an Energy Dispersive X-ray analyser. Total organic carbon (TOC) analyses were carried out with a carbon–sulphur analyser (LECO CS225) after treatment with 10% HCl to remove carbonate (Gross 1971). Raman spectral data were obtained with an inVia Raman microscope, measured with a 514 nm laser and interpreted using parameters specific to carbonaceous materials (Wopenka & Pasteris 1993; Beyssac *et al.* 2003). Samples from Torr Mor were soxhlet extracted with dichloromethane and methanol (93:7) for 48 h and the extracts were analysed in a gas chromatograph mass spectrometer for detection of biological signatures. Particular attention was paid to mass fraction $m/z=217$ and $m/z=218$ (steranes). Steranes are biological markers that can be used as source indicators and as maturity parameters (Peters & Moldowan 1993).

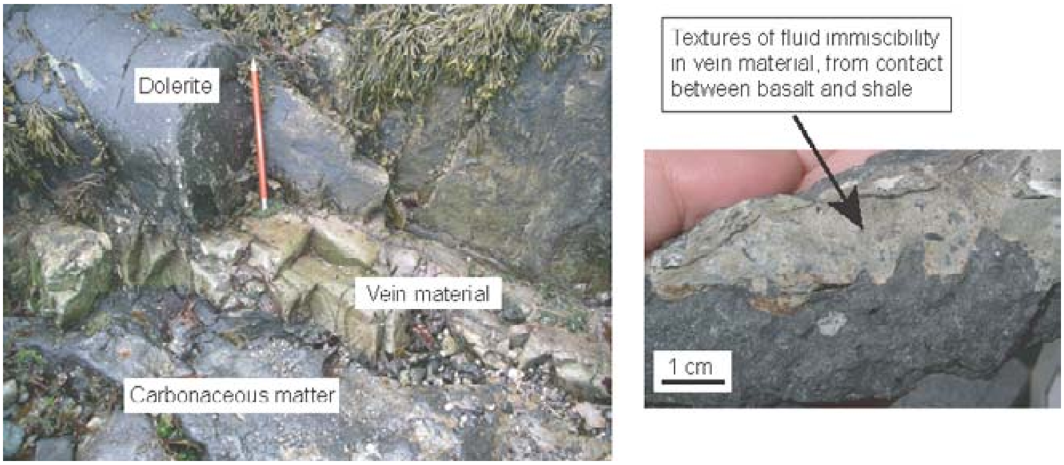

Fig. 2. Contact between dolerite and shale occupied with vein material and carbonaceous matter from Torr Mor. The right-hand image is a close-up of fluid immiscibility between two phases in the vein material: a mineral component (chlorite, calcite and quartz) and carbonaceous matter. The immiscibility textures include sharp boundaries with curved menisci between phases and budding between phases.

Results

Torr Mor

At this locality a 20 cm thick mass of vein-like material occupies the top of the contact between a dolerite sill and an underlying Jurassic shale (Fig. 2). This material consists of two main components, which are greenish and dark grey, respectively. The two components show textures of immiscibility such as sharp boundaries with curved menisci between phases and budding between phases (Philpotts 1979; MacDonald 1993); see Fig. 2. The green component is mainly composed of a fine-grained mixture of chlorite, calcite and quartz. The grey component consists of vesiculated carbonaceous matter. The vesicles are rounded to slightly elongated and sometimes connected by thin stringers (Fig. 3(a)). The vesicles are up to 100 μm in diameter, and filled with chlorite, calcite, quartz or a fine-grained mixture of these minerals. Smaller globules of carbonaceous matter occur inside some of the vesicles and also around the edges of the vesiculated carbonaceous matter (Fig. 3(b)).

Immediately below the first 20 cm, described above (Fig. 2), is an approximately 40 cm thick layer of highly vesiculated carbonaceous matter. This material overlies the laminated shale. The vesicles are filled with the same minerals as above (chlorite, calcite and quartz), but here they are smaller and more irregular. This material also contains chaotically distributed thin (1–2 mm) veinlets of calcite (Fig. 3(c)).

The Raman spectra of the carbonaceous material from the dolerite–shale contact differ from the carbon in the unaltered shale collected at a distance of several metres from the contact. Both spectra give a D1-band at around 1340 cm^{-1} that is indicative of disorder (Beyssac 2003). However, the position of the G-band (with or without merging with a D2-band) at around 1570 cm^{-1} and its peak width of about 30 cm^{-1}, in the carbonaceous material from the contact, characterize it as a more ordered carbon compared to the unaffected shale (Wopenka & Pasteris 1993); see Fig. 4.

Despite carbonaceous matter being heated and mobilized by the igneous intrusion at Torr Mor, biological markers are preserved (Fig. 5). Lower carbon number steranes (e.g. C_{21} and C_{22} pregnanes) are more thermally stable than higher carbon number steranes (C_{27}, C_{28} and C_{29} homologues) (Peters & Moldowan 1993; Saigó 2000). The effect of this can be seen in Fig. 5. The extract of the mobilized carbonaceous matter that was flash heated by the intrusion of the igneous sill contains more pregnane than the Torr Mor shale that has not been heated in this way. Also shown for comparative purposes is a typical North Sea oil. This, in common with the Torr Mor shale, has been heated to oil generating temperatures of probably no more than 150 °C.

Duntulm Castle

The peninsula immediately south of Duntulm Castle is composed of a dolerite sill, within which several large enclaves (of the order of $6 \times 3\ \mathrm{m}^2$) of Jurassic shales are incorporated (Bell & Harris 1986); see Fig. 6(a). Thin veinlets of melt glass are generated in the contact of the shale enclaves and the sill. The veinlets are composed of isotropic glass. They originate at the shale–dolerite boundary and cross-cut the dolerite (Fig. 6(b)). Also along the contact an extreme vesiculation of the dolerite intrusion has taken place (Fig. 6(a)). The vesicles are commonly filled with calcite, chlorite, quartz and carbon (Fig. 6(c)).

Elgol

Another example of a shale enclave contained within a dolerite sill was sampled at Elgol (Morton & Hudson 1995) for comparison with unaltered shale from the same stratigraphic level. However, this shale enclave shows no evidence of partial melting.

Hydrocarbon occurrences in the immediate vicinity of the sill are likely to be derived from the host shale. Some oil showing in adjacent sandstones (Parnell *et al.* 2004) may be the result of regional heating. However, hydrocarbon fluid

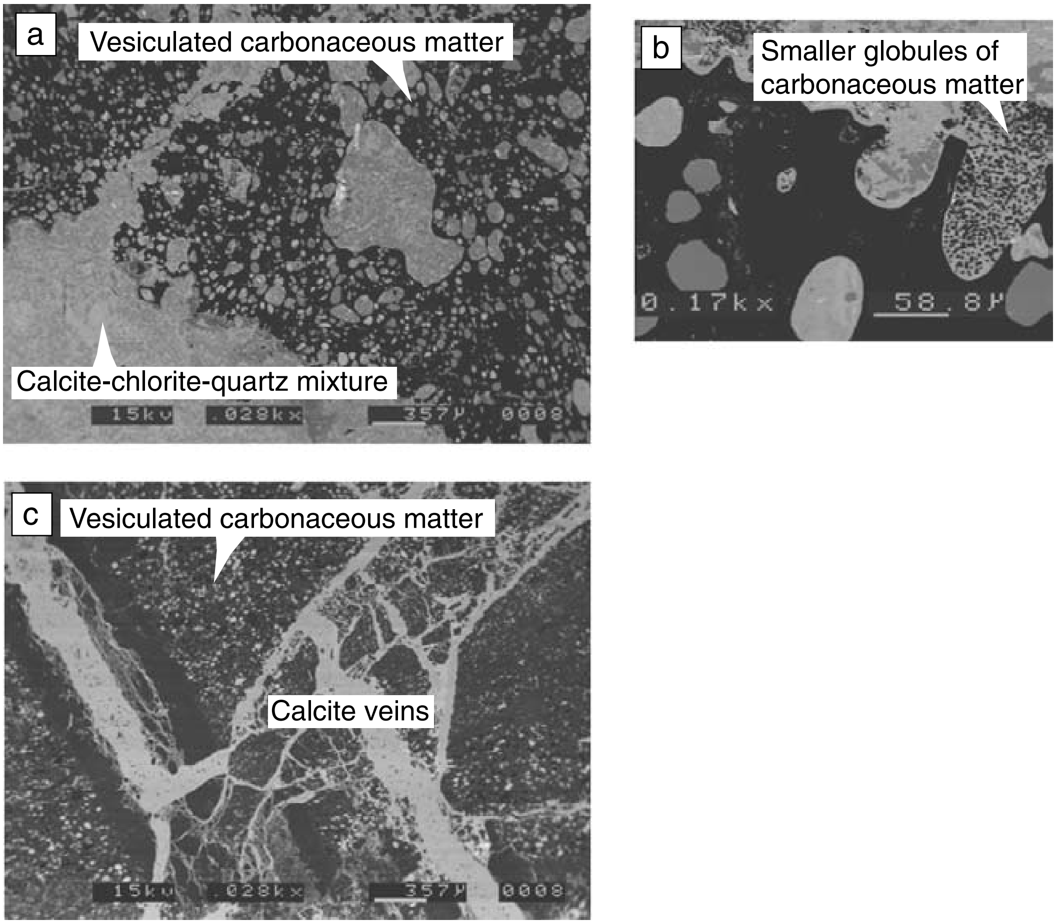

Fig. 3. Scanning electron micrograph of ((a) and (b)) vesiculated carbonaceous material in vein material and (c) vesiculated carbonaceous material below the vein material (all samples from Torr Mor). The vesicles are rounded to slightly elongated and sometimes connected by thin stringers. They are filled with chlorite, calcite, quartz or a fine-grained mixture of these minerals. Smaller globules of carbonaceous matter occur inside some of the vesicles and also around the edges of the vesiculated carbonaceous matter.

inclusions within calcite in veins cutting the sill probably represent volatiles derived from the interaction between the sill and the shale.

TOC analysis

Although the total amount of carbon in the heated shale at the contact with basalt and in shale enclaves is lower than the unheated shale, the results still shows that carbon has been preserved in the shale heated by igneous intrusion. The results also show a high amount of carbon (57%) in the carbonaceous material from Torr Mor (Table 1).

Discussion

Torr Mor

At this locality organic matter was not only preserved but was also mobilized and concentrated at the contact between the dolerite and the shale. The organic carbon content of the carbonaceous material in contact with basalt is 57%, compared to the content of 0.75% in the unaltered shale from this locality. Fluids from the shale were released owing to the effect of heating by the igneous intrusion. These fluids contained a lot of gas, as indicated by the vesicles and veinlets in the carbonaceous material. Localized mobilization of carbonaceous material by heat input from igneous intrusions has also taken place at other localities on Skye, for example at Elgol (Parnell 1992). The Raman data characterize the carbon from the contact with the dolerite as more ordered than the carbon in the shale, which most probably is a result of heating by the intrusion. The immediate contact with the dolerite is a vein-like material composed of two components where one is carbonaceous matter and the other is a fine-grained mixture of chlorite, calcite and quartz. The two components are immiscible, i.e. they are in a fluid state at the same time. The occurrence of smaller globules of carbonaceous matter inside the vesicles and around the edges of the larger areas of carbonaceous matter implies that the globules were disseminated from the larger areas of carbonaceous matter after degassing and when the chlorite/calcite/quartz component was still a fluid. Veins with immiscible hydrothermal fluids, where one phase is carbonaceous material, are similarly reported in Bailey (1959). The carbonaceous matter at Torr Mor is described as graphite by Peach *et al.* (1910), and some of it exhibits a graphitic sheen in the field. However, the Raman data show that it is not fully ordered graphite.

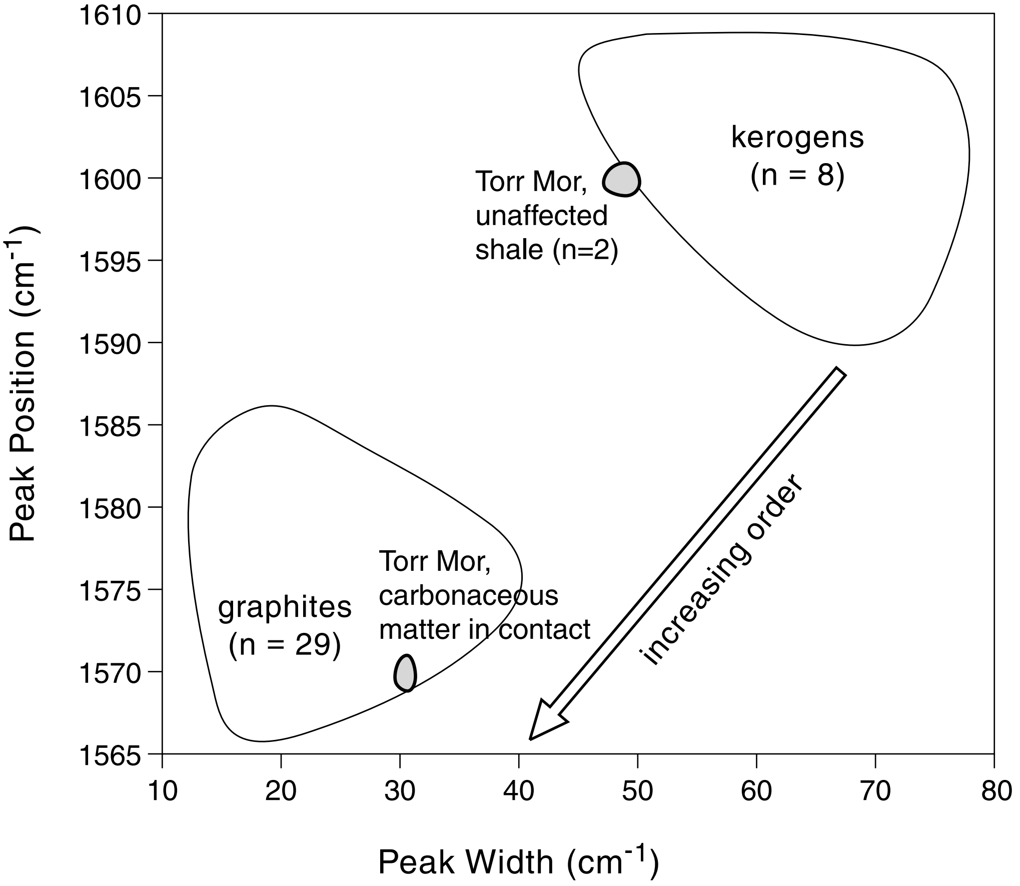

Fig. 4. Cross-plot of peak position and peak width (G-peak with or without merging with a D2-peak) of carbon in unaffected shale and carbonaceous matter in contact with dolerite from Torr Mor. Data of kerogen and graphite from Wopenka & Pasteris (1993).

Duntulm Castle

At this locality enclaves of shale were fully incorporated within the hot igneous melt and veins of melt where generated at the contact between shale and dolerite (Fig. 6(b)). Despite suffering high temperatures, the shale enclave that was analysed in this study has preserved an organic carbon content of 0.43%. The organic carbon content of the Jurassic shales on Skye ranges from less than 0.5% to around 5% (Thrasher 1992), but it is apparent that the carbon content of the enclave is generally lower than at most unheated localities, which suggests that some carbon has been lost. There is evidence for interaction between the shale/carbon and the melt: the basalt is strongly vesiculated at the contact with the shale enclave. The vesicles are filled with calcite, chlorite, quartz and carbon. This indicates that fluids, including carbon, were mobilized from the shale and interacted with the igneous melt.

Related occurrences

Other occurrences of intrusion into Jurassic shales on the Atlantic margin are relevant to this discussion.

(1) At Portrush, Northern Ireland, a Tertiary dolerite sill intruded into Jurassic shale to produce a hornfels (a fine-grained rock baked by contact metamorphism) (Wilson & Manning 1978). This was an important locality in the 18th century debate between Neptunists and Vulcanists over the origin of crystalline igneous rocks. The Neptunists believed that such rocks crystallized from seawater, while the Vulcanists attributed them to volcanic activity. The hard hornfels was initially interpreted as basalt, despite containing abundant ammonites, and was therefore used to support the Neptunist cause. Only after a contact between a sill and metamorphosed shale was recognized (Berger 1816) was the debate resolved. The Neptunist argument had been that fossils could not have survived the heat of molten rock. Over 200 years later, we argue that not only can fossils survive, but organic carbon can also survive the interaction of organic matter with molten rock.

(2) Carbonaceous nodules occur in Tertiary volcanic rocks in a hydrocarbon exploration well in the Erlend Volcanic Complex, offshore of the UK (Kanaris-Sotiriou *et al.* 1993; Kanaris-Sotiriou 1997). The most likely origin for the carbon is from Jurassic shales lower in the succession (Kanaris-Sotiriou *et al.* 1993). Petrographic studies of the carbonaceous matter (Lindgren & Parnell 2006) show that it consists of graphite crystals nested in a matrix of chlorite and quartz. It may have been either mobilized directly from a sedimentary rock or from pre-existing oil derived from such a rock.

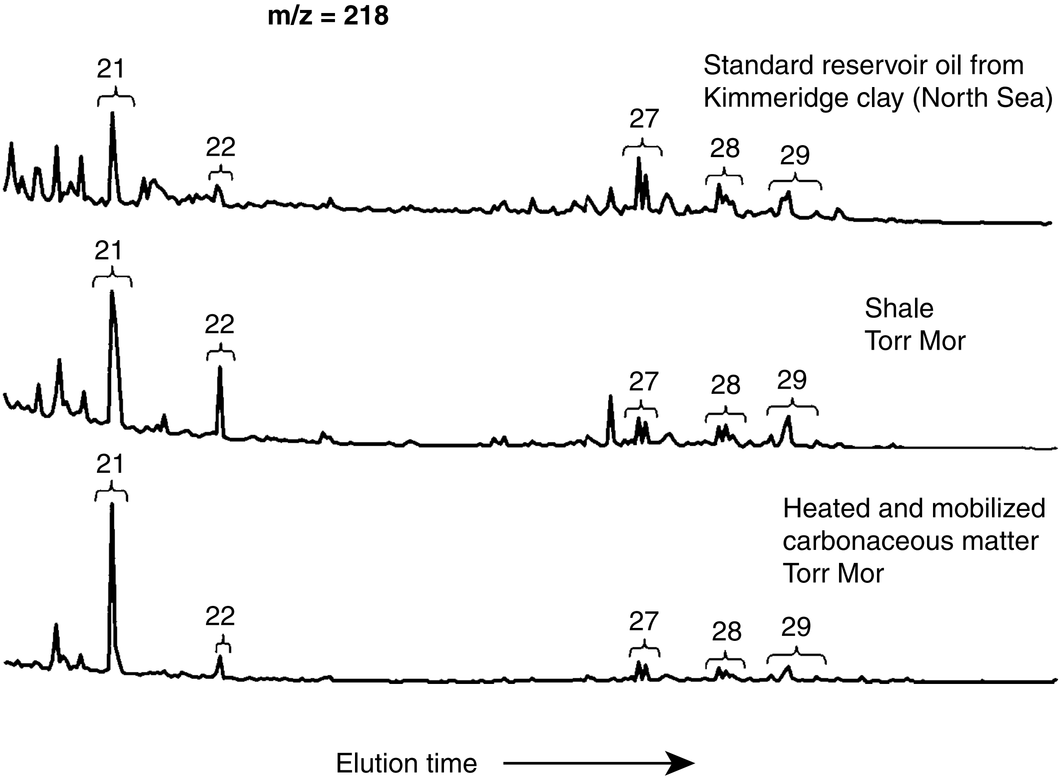

Fig. 5. $m/z = 218$ gas chromatograms of a standard oil derived from North Sea Kimmeridge clay, unheated shale from Torr Mor and carbonaceous matter in the contact zone of the sill and the shale from Torr Mor. The latter sample was heated and mobilized by the igneous intrusion. All three samples show preservation of biological markers: 21, 22 pregnanes and 27, 28, 29 $\alpha\beta\beta$ steranes. The pregnanes/steranes ratio is highest in the mobilized organic matter, showing the highest level of maturity (most heated).

(3) Where a sill intruded Jurassic shale and sandstone at Camas Mòr on the Island of Muck to the south of Skye (Harker 1908), veinlets of melt glass were generated that cross-cut the country rock and the sill. We have not observed carbon in the glass, but the generation of melt veins strengthens the analogy between intrusion emplacement and impact heating, as melt veins are a normal feature of impact craters (Koeberl 2002).

Generation of hydrocarbon through heating by igneous intrusion

The heat of intrusion into an organic-rich sedimentary rock causes thermal alteration of the organic matter. Maturation of the organic matter in the vicinity of the intrusion can lead to the generation of liquid hydrocarbons. There are many cases in the geological record of hydrocarbons at the margins of, and in vesicles of, sills and dykes (Parnell 1988, 2004).

Analogue for heating of carbon in impact craters

Although not as rapid as during instant impact heating, the introduction of heat to the shale by igneous melt occurred relatively quickly. In analogy with impact heating, the target (in this case the shale) was not progressively heated but heated relatively rapidly. The temperatures were not as high as during an impact where temperatures can exceed 3000 °C (French 1998; Osinski *et al.* 2005) in the centre of a crater and in an impact melt. The temperature of a basaltic melt is generally around 1000–1200 °C (Macdonald 1972). Around sills and dykes there is a rapid drop in temperature away from the heat source, as measured, for example, by maturation of organic matter in Jurassic shales around Tertiary intrusions on the Isle of Skye (Bishop & Abbott 1993; Farrimond *et al.* 1999). In impact craters the drop from extreme temperatures is also rapid, and a heating gradient can be measured by thermal maturation of organic matter in a crater from the centre and outward (Parnell *et al.* 2005). Although the temperatures achieved by volcanic lavas and sills are much less than that of a large impact, the effect of heating generated by lavas and sills into carbon-rich targets is to some extent analogous to the effect of impact heating as it involves a rapid introduction of heat and a rapid quenching from high temperatures with no time for equilibration.

The limited database suggests that the intruded shales preserve 10–35 % of the organic carbon content prior to intrusion. For a shale containing Type II kerogen typical of Jurassic shales in North West Europe, strong heating should leave about 30 % of the original carbon resulting from oil generation during normal burial (Cornford 1998), which is a comparable level of preservation. We see no evidence that the organic carbon content is completely burnt out, and there is no obvious mechanism by which this should happen

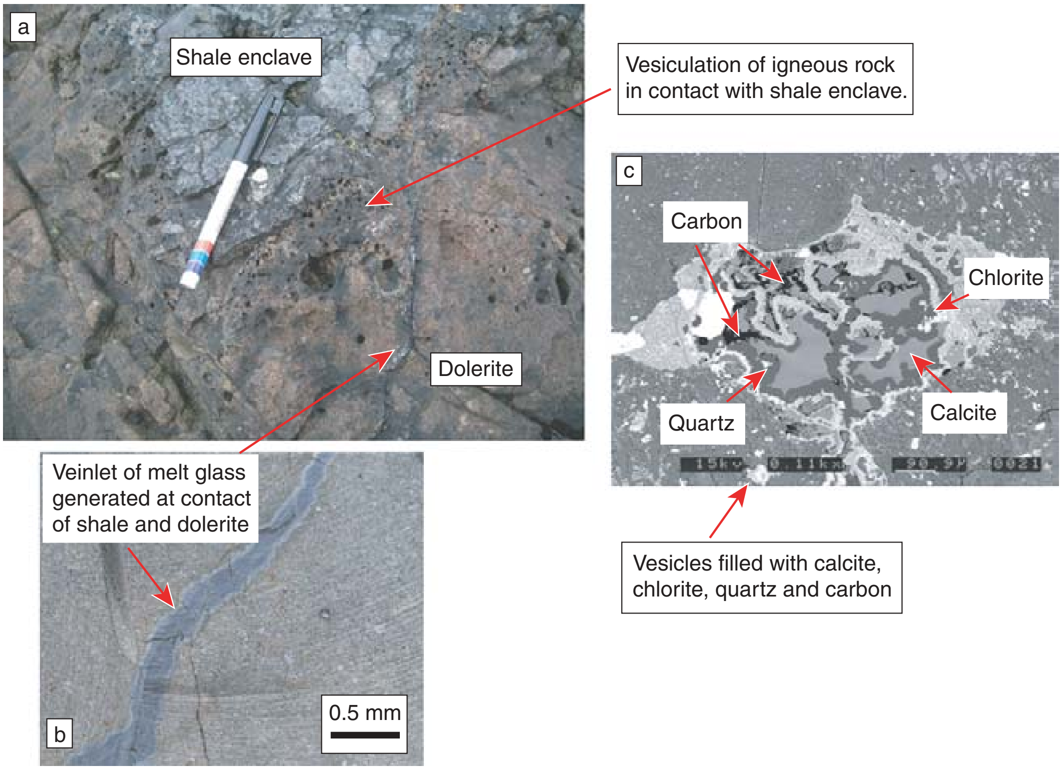

Fig. 6. Duntulm Castle. (a) Shale enclave enclosed in dolerite sill. The dolerite exhibits extreme vesiculation in contact with the shale. (b) Photomicrograph showing a veinlet of melt glass generated at the contact between the sill and the shale enclave. (c) Scanning electron photomicrograph showing a vesicle in basalt filled with calcite, chlorite, quartz and carbon.

Table 1. *Total organic carbon content of carbonaceous material from contact with basalt, unheated shale and shale enclaves*

Locality	Sample	TOC (%)
Torr Mor	Carbonaceous material at contact with basalt	57
	Unheated shale	0.75
Duntulm Castle	Shale enclave	0.43
Elgol	Shale enclave	0.12
	Unheated shale	1.2
Portrush (N. Ireland)	Shale at contact with basalt	0.16

during a subsurface intrusion process. An impact event may be more surficial, but the speed of the event, and limitations on available oxygen, mean that residual organic carbon is also likely to survive this process. In terms of a record for astrobiology, the uncertainty is not over the preservation of organic carbon but over the preservation of molecular markers. Biological markers are preserved in the heated and mobilized carbonaceous matter at Torr Mor, although with a lower abundance compared to oil from the North Sea and the relatively unheated shale from Torr Mor.

Volatile loss from sediments is evident in gas bubbles in the immediately adjacent sill at Duntulm Castle and in the contact zone at Torr Mor. Impact craters in sedimentary target rocks are particularly likely to cause generation of volatiles (Kieffer & Simonds 1980), and on asteroids carbon-rich melts probably devolatilize readily by degassing (Wilson & Keil 1991; Scott *et al.* 1993).

High-temperature processing of organic matter

The response of organic matter to high-temperature events, such as meteorite impacts or emplacement of igneous material, is important to astrobiology as it controls the survival and concentration of carbon during one of the most widespread processes in our Solar System, impact cratering. This is critical to constraining the origin and spread of life in the universe, given that, in order to form and spread 'building blocks' for life, carbon needs to survive volatilization induced by these high temperatures. It may also help to understand the history of carbon in meteorites that have experienced high-temperature processing, such as ureilites (Grady *et al.* 1985; Rubin 1988; Goodrich 1992).

Conclusion

Although the temperatures that carbonaceous matter have experienced during the intrusion of sills into shale are much lower than temperatures achieved at the centre of large impact craters, shallow igneous intrusion in shale is a valuable analogue for investigation of the effect of heating on carbonaceous material during impact events because it represents a rapid introduction of heat, and a subsequent rapid drop in heat, to a carbon-rich rock.

The analogy between impact heating and intrusion emplacement includes:

(i) a rapid introduction of heat;
(ii) heating to temperatures at which melting commences;
(iii) subsequent rapid quenching from high temperatures;
(iv) generation of melt glasses;
(v) injection of fluid veins into the solid rock;
(vi) generation of volatiles that degas from the sedimentary protolith.

In terms of preservation of carbon, the key conclusions are as follows.

(i) Despite high-temperature igneous intrusion, carbonaceous material is preserved both at the dolerite–shale contact at Torr Mor and in shale enclaves within other intrusives.
(ii) Carbonaceous material was mobilized and concentrated at the dolerite–shale contact and in vesicles within the dolerite at the dolerite–shale enclave contact.
(iii) Carbon contained within vesicles of the dolerite shows interactions between carbon and the hot melt where carbonaceous material has survived.

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