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## Isotopic evidence for Late Cretaceous plume–ridge interaction at the Hawaiian hotspot

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When a mantle plume interacts with a mid-ocean ridge, both are noticeably affected. The mid-ocean ridge can display anomalously shallow bathymetry, excess volcanism, thickened crust, asymmetric sea-floor spreading and a plume component in the composition of the ridge basalts<sup>1–4</sup>. The hotspot-related volcanism can be drawn closer to the ridge, and its geochemical composition can also be affected<sup>5–7</sup>. Here we present Sr–Nd–Pb isotopic analyses of samples from the next-to-oldest seamount in the Hawaiian hotspot track, the Detroit seamount at 51° N, which show that, 81 Myr ago, the Hawaiian hotspot produced volcanism with an isotopic signature indistinguishable from mid-ocean ridge basalt. This composition is unprecedented in the known volcanism from the Hawaiian hotspot, but is consistent with the interpretation from plate reconstructions<sup>8</sup> that the hotspot was located close to a mid-ocean ridge about 80 Myr ago. As the rising mantle plume encountered the hot, low-viscosity asthenosphere and hot, thin lithosphere near the spreading centre, it appears to have entrained enough of the isotopically depleted upper mantle to overwhelm the chemical characteristics of the plume itself. The Hawaiian hotspot thus joins the growing list of hotspots that have interacted with a rift early in their history.

For more than 81 Myr (ref. 9), the Hawaiian hotspot has been producing the age-progressive Hawaiian–Emperor chain of volcanic islands and seamounts (Fig. 1). This chain extends for 5,800 km from the present location of volcanic activity on the island of Hawaii and Loihi seamount, to the northernmost Emperor seamount (Meiji) at 53° N. There are over 100 volcanic edifices along the Hawaiian–Emperor chain, and more than 30 of these (in addition to the Hawaiian Islands) have been sampled by dredge and drill<sup>9–11</sup>, making it the most intensively sampled and studied hotspot track on Earth.

Published Sr-isotope ratios of tholeiitic basalts from along the Hawaiian hotspot track show an interesting trend with age (Fig. 2). These ratios remain fairly constant along the Hawaiian Islands and the Hawaiian ridge between Kilauea volcano on Hawaii and Yuryaku seamount (Fig. 1); they then decrease steadily northwards along the Emperor seamounts to Suiko guyot, which was the oldest seamount previously analysed (64.7 Myr; ref. 12). This decrease in the <sup>87</sup>Sr/<sup>86</sup>Sr ratio back in time was attributed to a decrease in distance between the hotspot and the nearest spreading centre<sup>13</sup>. Only the tholeiitic basalts (as opposed to the transitional and alkalic

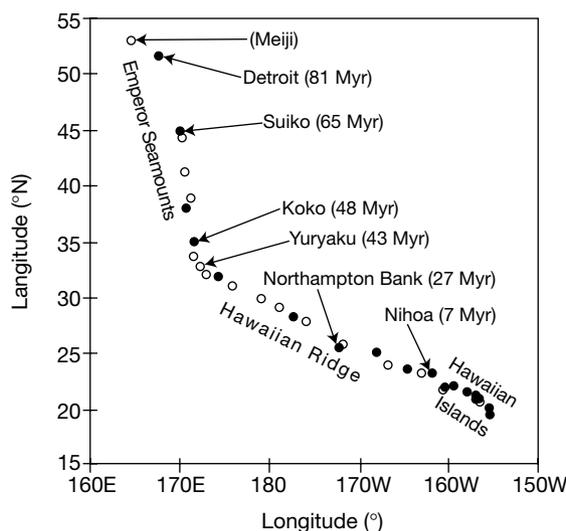
basalts) show this isotopic trend, perhaps because the tholeiites experienced little if any interaction with the lithosphere<sup>14–16</sup>.

Because this trend in <sup>87</sup>Sr/<sup>86</sup>Sr with time was all within the range for the Hawaiian Islands, it could be explained by a sampling bias in the older seamounts. We therefore decided to measure the isotopic ratios of basalts from the two oldest Emperor seamounts (Detroit and Meiji) to determine if the isotopic trend continued further back in time. We also analysed a single sample from Suiko to tie in with the published data. We chose to concentrate our analyses, and our discussion here, on the tholeiitic basalts because only they show the isotopic trend with time.

We measured the Sr, Nd and Pb isotope ratios of basalts from two locations (ODP Sites 883 and 884) on the Detroit seamount platform<sup>9</sup>. The Site 884 basalts have low concentrations of alkalis, TiO<sub>2</sub>, and other incompatible elements at moderate MgO (4.6–8.8 wt%) and Ni (47–94 p.p.m.) contents, and are unambiguously tholeiitic<sup>9</sup>. Basalt recovered at the other drillsite on Detroit (Site 883) are too altered to determine unambiguously if they are tholeiitic (and we wish to limit our study to tholeiites); however, we include them here (Table 1) because they are isotopically similar to the Site 884 basalts (Fig. 3), which bolsters our argument that the Late Cretaceous volcanism was significantly different from the more recent volcanism. The Suiko sample from DSDP Site 192 are tholeiitic basalts<sup>12,17</sup>.

Age-corrected (initial) <sup>87</sup>Sr/<sup>86</sup>Sr, <sup>207</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb values (Table 1) in the Detroit tholeiites (Site 884) are lower than anything previously reported from the Hawaiian–Emperor chain (Fig. 3), while <sup>143</sup>Nd/<sup>144</sup>Nd and <sup>206</sup>Pb/<sup>204</sup>Pb values for these tholeiites overlap the Hawaiian field. Age corrections to these isotopic data are small (Table 1), and even the measured ratios are distinct from the Hawaiian data. The Sr and Pb isotope ratios of the Detroit tholeiites are within the range of Pacific mid-ocean-ridge basalt (MORB), while their <sup>143</sup>Nd/<sup>144</sup>Nd values are lower than modern Pacific MORB, but within the range of Pacific MORB data adjusted to 80 Myr (Fig. 3b). Isotopic data for the Site 883 samples are similar to the Site 884 data, but are not quite as depleted.

The Suiko sample has the highest Sr and Pb isotope ratios measured in this study, and values of <sup>143</sup>Nd/<sup>144</sup>Nd that are similar to those found at Meiji and Detroit (Table 1). Suiko isotopic ratios



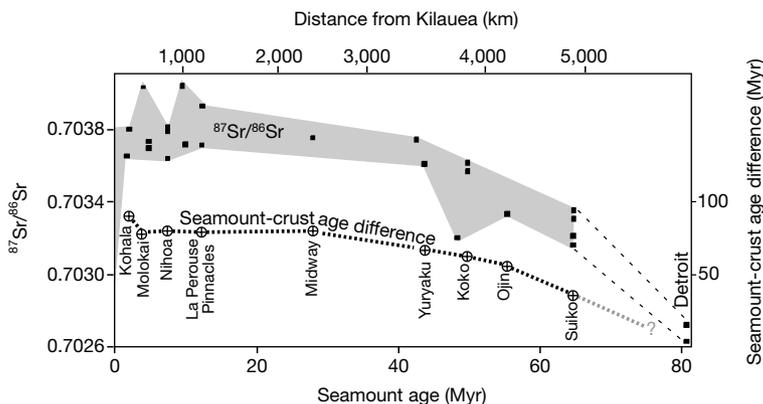
**Figure 1** Location of volcanoes in the Hawaiian–Emperor island/seamount chain that have been dated (the age of Meiji is unknown). Additional locations have been sampled, but not dated. Locations from which tholeiites have been reported are shown by filled circles. Locations where no tholeiites were reported, or the identification of the tholeiites is in doubt, are shown by open circles. Seamounts mentioned in the text are labelled with names and ages. Age data are from a compilation<sup>10</sup>, except for the Detroit seamount age<sup>9</sup>. New isotopic data presented in this study are from Suiko, Detroit and Meiji.

are within the range of published values for tholeiites from the Hawaiian Islands and the Hawaiian ridge (although its  $^{87}\text{Sr}/^{86}\text{Sr}$  is slightly lower than the lowest published  $^{87}\text{Sr}/^{86}\text{Sr}$  for Hawaiian tholeiites that also have Nd and Pb data available; Fig. 3). Our  $^{87}\text{Sr}/^{86}\text{Sr}$  value for the Suiko sample is within analytical error of previous Suiko tholeiite analyses<sup>13,18</sup>. Isotopic ratios of the Meiji sample are generally intermediate between the Detroit and Suiko data (Table 1).

The uniqueness of the Cretaceous Emperor seamount isotopic data compared to the rest of the Hawaiian–Emperor chain is most obvious on the plot of  $^{206}\text{Pb}/^{204}\text{Pb}$  versus  $^{87}\text{Sr}/^{86}\text{Sr}$  (Fig. 3a). The Detroit data overlap the Pacific MORB field, and are distinct from

the Hawaiian Islands/ridge field. The Meiji and Suiko data are displaced from the highly depleted values of the Detroit field, towards the high- $^{206}\text{Pb}/^{204}\text{Pb}$  – low- $^{87}\text{Sr}/^{86}\text{Sr}$  end of the Hawaiian Islands/ridge field. The positive slope of our Meiji–Detroit–Suiko data array on this plot is obviously distinct from the negative slope of the array formed by tholeiites from the 0 to 48 Myr part of the Hawaiian–Emperor chain.

We can quickly rule out several explanations for the unradiogenic nature of the early products of the Hawaiian hotspot. (1) The fact that the Sr and Pb isotope compositions of the Detroit tholeiites are unlike any other known product of the Hawaiian hotspot shows that the trend in Fig. 2, now extended back to 81 Myr, is not due to



**Figure 2** Evolution of the Hawaiian hotspot with time. The shaded field shows the range of published  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of tholeiitic basalts versus seamount age (lower axis) and distance from Kilauea volcano along the Hawaiian–Emperor chain (upper axis). Data points used to define the shaded field are shown by small filled squares, except that the many data points from the Hawaiian Islands have been omitted for clarity. The extension of

the shaded field to include new data presented here for the Detroit seamount is shown by the two dashed lines. The crossed circles connected by the thick dotted line shows the trend in age difference (right axis) between the seamounts and their underlying crust. Age data sources are as in Fig. 1, except that the age difference between the Detroit seamount and the crust is estimated from isochron charts<sup>27</sup> to be <20 Myr.

**Table 1** Isotopic and trace-element ratios of Emperor seamount basalts

	Seamount					
	Suiko	Detroit	Detroit	Detroit	Detroit	Meiji
<b>Location</b>						
Sample	433-31-1	883-1-3	883-2-3	884-6-3	884-10-4	192-6-2
Age (Myr)	64.7	80?	80?	81.2	81.2	85?
Latitude (N)	44°46.6'	51°11.9'	51°11.9'	51°27.0'	51°27.0'	53°0.0'
Longitude (E)	170°1.2'	167°46.1'	167°46.1'	168°20.2'	168°20.2'	164°42.8'
Depth (m)	2237	3208	3219	4720	4751	4069
<b>Measured ratios</b>						
$^{87}\text{Sr}/^{86}\text{Sr}$	0.703297	0.702884	0.702864	0.702768	0.702652	0.703079
$^{143}\text{Nd}/^{144}\text{Nd}$	0.513068	0.513081	0.513100	0.513184	0.513152	0.513072
$^{206}\text{Pb}/^{204}\text{Pb}$	18.609	18.355	18.500	18.175	18.058	18.750
$^{207}\text{Pb}/^{204}\text{Pb}$	15.463	15.443	15.452	15.419	15.410	15.453
$^{208}\text{Pb}/^{204}\text{Pb}$	38.136	37.856	37.809	37.680	37.582	38.036
Rb/Sr	0.015	0.024	0.024	0.012	0.010	0.031
Sm/Nd	0.278	0.285	0.285	0.349	0.350	0.280
Th/Pb	0.833	0.941	0.938	0.321	0.312	0.812
U/Pb	0.300	0.470	0.438	0.143	0.156	0.500
La/Sm	1.99	1.78	1.86	1.06	1.08	1.99
<b>Initial ratios</b>						
$^{87}\text{Sr}/^{86}\text{Sr}$	0.703257	0.702808	0.702788	0.702730	0.702620	0.702972
$^{143}\text{Nd}/^{144}\text{Nd}$	0.512994	0.512987	0.513006	0.513068	0.513035	0.512974
$^{206}\text{Pb}/^{204}\text{Pb}$	18.394	17.938	18.113	18.047	17.918	18.279
$^{207}\text{Pb}/^{204}\text{Pb}$	15.453	15.423	15.434	15.413	15.403	15.431
$^{208}\text{Pb}/^{204}\text{Pb}$	37.945	37.589	37.543	37.588	37.492	37.791

All isotopic ratios were measured at Cornell University using published procedures<sup>25</sup>. All samples were repeatedly leached in hot HCl before digestion, and all isotopic ratios were corrected for fractionation. Sr and Pb isotope ratios are normalized to standards NBS987 (= 0.710248) and NBS981 (= 16.937; 15.493; 36.705), respectively. The average  $^{143}\text{Nd}/^{144}\text{Nd}$  for the La Jolla Nd standard measured at the time of these analyses (via Ames Nd) was 0.511876 ± 14. Two-sigma standard errors based on numerous analyses of standards during the time of the unknown runs are:  $^{87}\text{Sr}/^{86}\text{Sr}$  ± 0.000014,  $^{143}\text{Nd}/^{144}\text{Nd}$  ± 0.000016,  $^{206}\text{Pb}/^{204}\text{Pb}$  ± 0.010,  $^{207}\text{Pb}/^{204}\text{Pb}$  ± 0.011, and  $^{208}\text{Pb}/^{204}\text{Pb}$  ± 0.030. Within-run standard errors are smaller than these external errors. Trace-element concentrations were measured by isotope dilution thermal ionization mass spectrometry (TIMS-ID) of leached samples (Pb, Th and U) and inductively coupled plasma mass spectrometry (ICP-MS), of unleached samples (Rb, Sr, La, Nd and Sm). The ages of the Site 433 and Site 884 samples are known from  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  dating<sup>10,13</sup>. For age correction purposes, the Site 883 samples are assumed to be 1 Myr younger than the Site 884 samples because Site 883 was slightly higher on the seamount, and the Site 883 samples appear to be of transitional composition. The Site 192 sample is estimated to be 85 Myr old for age correction purposes. Errors in the estimated ages of the Site 192 and Site 883 samples of several million years or less are not relevant here.

sampling bias. The Hawaiian Islands are well sampled, and it is unlikely that anything as depleted as the Detroit tholeiites has escaped notice. (2) The low isotopic ratios are not an artefact of the age-correction procedure: age-corrected Sr and Pb isotope ratios for the Site 884 tholeiites are barely different from the measured values. Also, the two Detroit locations (Sites 883 and 884) have similar age-corrected isotopic ratios, despite their different measured ratios and trace-element compositions. (3) The shift in isotopic characteristics of the Hawaiian hotspot from 81 Myr to the present cannot be explained by "plume ageing", that is, radioactive decay within the plume and its source region, as has been proposed for the Kerguelen plume<sup>19</sup>. The trace-element ratios required to account for the differences between the Detroit isotopic data and the Hawaiian Islands/ridge data ( $Rb/Sr = 0.278$ ,  $U/Pb = 0.558-2.11$ ,  $Th/Pb = 2.09$ ) are 4 to 28 times these ratios in the Site 884 tholeiites (and due to partial melting effects, these ratios should be even lower in the source than in the basalts), and 1 to 2 orders of magnitude higher than the maximum ratios likely to be found in plumes<sup>20</sup>.

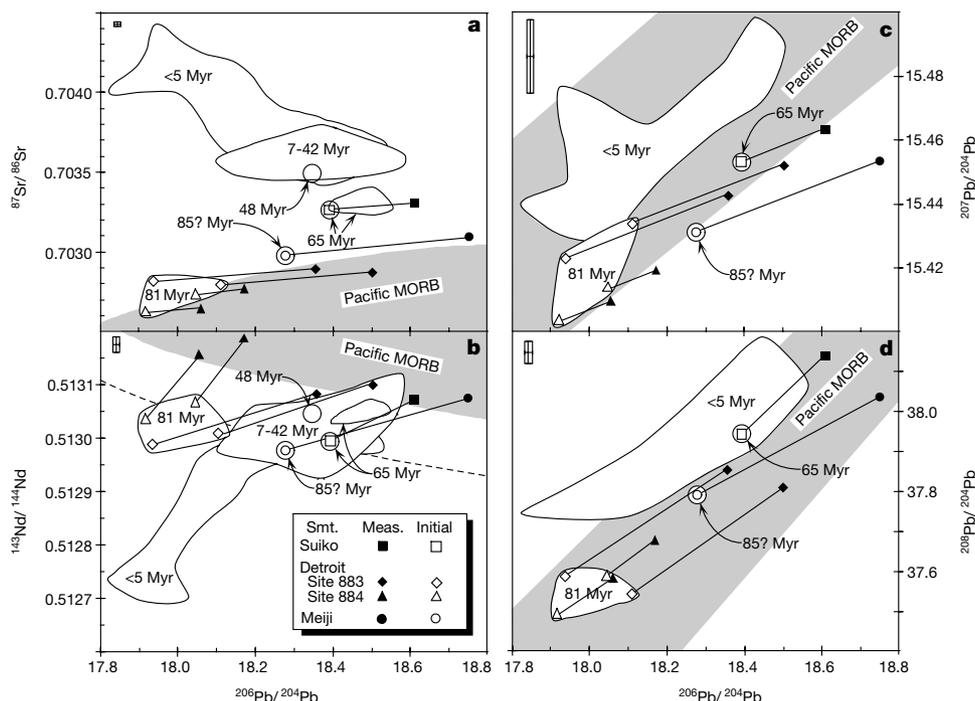
We interpret the displacement of the Cretaceous data points towards MORB values as mixing between a depleted component with the isotopic characteristics of MORB, and a 'Kilauea-like' plume component at the low-<sup>87</sup>Sr/<sup>86</sup>Sr end of the Hawaiian Islands/ridge data set. There is no evidence in the isotopic data from any of the Late Cretaceous samples for the high-<sup>87</sup>Sr/<sup>86</sup>Sr, low-<sup>143</sup>Nd/<sup>144</sup>Nd and low-<sup>206</sup>Pb/<sup>204</sup>Pb ('Koolau-like') component found in the modern plume.

Plate reconstructions place an active spreading centre close to the position of the Hawaiian hotspot at about 80 Myr ago (ref. 8). The rising plume must have entrained more depleted mantle when it came up near the spreading centre. In other locations where a spreading ridge is close to a hotspot (for example, Bouvet, Easter, Galápagos and Iceland) the isotopic characteristics of the hotspot

volcanism extend toward MORB-like values<sup>2,21,22</sup>. A nearby spreading ridge could have created a higher-temperature and lower viscosity and density regime in the upper mantle at ~81 Myr ago, conditions that lead to more entrainment<sup>22</sup>. Hotter upper-mantle temperatures close to the spreading centre also could have allowed greater degrees of melting of the upper mantle<sup>23</sup>, and thus more entrainment. The nearby spreading centre could have caused more vigorous upper-mantle convection and a mechanical stirring effect. Also, younger, hotter lithosphere may be more readily assimilated by the ascending plume melts, thereby imparting a MORB-like isotopic signature to the hotspot basalts. The thick dotted line connecting crossed circles in Fig. 2 shows the changes in the age relationship between the seamounts and their underlying crust. Older lithosphere (at present ~90 Myr at the Hawaiian Islands) is colder, less easily melted, and is thought to play an insignificant role in the genesis of tholeiites on the Hawaiian Islands<sup>14-16</sup>. The thickness (and thus age) of the lithosphere could also determine how much the asthenosphere contributes to hotspot volcanism. If the upwelling mantle plume begins to melt when it reaches a depth of, say, approximately 100 km, and continues to produce melt until it reaches the base of the lithosphere, an area of very young (thin) lithosphere will have a taller melting zone that melts more asthenosphere.

Some workers have suggested that the head of a new mantle plume could entrain large amounts of depleted upper mantle<sup>23-25</sup> (although numerical models suggest that the melting zone of a plume head entrains less rather than more upper mantle<sup>26</sup>), but it is not known if this model can be applied to the Hawaiian plume. This is because the old end of the Hawaiian hotspot track ends at a subduction zone, so the existence of older seamounts or a plume-head-associated flood basalt is speculative.

Finally, a change in the isotopic characteristics of the source region of the plume cannot be ruled out. The region of the mantle



**Figure 3** Isotope ratios for tholeiitic basalts. The figure shows plots of the <sup>206</sup>Pb/<sup>204</sup>Pb ratio against other isotopic ratios; data are for tholeiitic basalts from the Hawaiian Islands (<5 Myr field), the Hawaiian ridge (7–42 Myr field), Koko guyot (48 Myr), and Suiko (65 Myr), Detroit (81 Myr) and Meiji (~85? Myr) seamounts. For Suiko, Detroit and Meiji, filled symbols are measured ratios and open symbols are age-corrected (initial) ratios (Table 1). Only data from tholeiites are shown, with the possible exception of the

Meiji and the Site 883 Detroit samples. Hawaiian Islands and Pacific MORB data are from numerous sources. Fields without data points (7–42 Myr, 48 Myr and 65 Myr) in panels **a** and **b** are also age-corrected<sup>18</sup>. The dashed line in panel **b** is the lower bound of the Pacific MORB field adjusted to 80 Myr ago using a MORB-reservoir Sm/Nd ratio of 0.36 (ref. 28). Age adjustments to the MORB fields in the other panels are negligible. Rectangle in the upper left corner of each panel shows  $\pm 2\sigma$  error box.

where the Hawaiian plume acquires its isotopic signature has probably not been homogeneous and static. Isotopically diverse packages of mantle material may have been randomly swept into this region over (at least) the past 81 Myr. The more depleted composition of the Detroit samples could be explained by a package of relatively depleted mantle having been swept into the plume source region during the Cretaceous. However, this model is untestable, and does not explain why the other two hotspots from which long-term data are available (Kerguelen and Réunion) also appear to have become more enriched with time<sup>19,23–25</sup>. □

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**Filamentous microfossils in a 3,235-million-year-old volcanogenic massive sulphide deposit**

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The record of Archaean microfossils is sparse<sup>1</sup>. Of the few *bona fide* fossil assemblages, most are from shallow-water settings, and they are typically associated with laminated, stromatolitic sedimentary rocks<sup>2–4</sup>. Microfossils from deep-sea hydrothermal systems have not been reported in Precambrian rocks (> 544 million years old), although thermophilic microbes are ubiquitous in modern sea-floor hydrothermal settings<sup>5,6</sup>, and apparently have the most ancient lineages<sup>7,8</sup>. Here, I report the discovery of pyritic filaments, the probable fossil remains of thread-like microorganisms, in a 3,235-million-year-old deep-sea volcanogenic massive sulphide deposit from the Pilbara Craton of Australia. From their mode of occurrence, the micro-organisms were probably thermophilic chemotropic prokaryotes, which inhabited sub-sea-floor hydrothermal environments. They represent the first fossil evidence for microbial life in a Precambrian submarine thermal spring system, and extend the known range of submarine hydrothermal biota by more than 2,700 million years<sup>9</sup>. Such environments may have hosted the first living systems on Earth, consistent with proposals for a thermophilic origin of life<sup>10–13</sup>.

The Pilbara Craton of Australia contains one of the most complete sections of well preserved Archaean volcano-sedimentary rocks and is the site of several previous fossil discoveries<sup>4,14</sup>. From this region, the probable fossil remains of microorganisms were identified in a volcanogenic massive sulphide (VMS) deposit at Sulphur Springs. The deposit is located in low-strain, very low metamorphic grade (prehnite–pumpellyite facies) rocks of the northern Soanesville belt in the eastern Pilbara Craton (Fig. 1a), and displays exceptional textural and structural preservation despite its great age<sup>15</sup>. Mineralization is associated with syn-volcanic faults and lies below a regionally extensive unit of silicified sedimentary and volcanoclastic rocks ('marker chert') which caps a succession of mafic, intermediate and felsic volcanic rocks and syn-volcanic dacitic intrusions (Fig. 1b). These rocks are intruded by the ~3,235-Myr-old co-magmatic Strelley granite, which is synchronous with hydrothermal mineralization<sup>16–18</sup>. The succession was deposited in a deep marine setting, with water depths probably exceeding 1,000 m, as indicated by volcanic textures, sedimentary facies and style of metal precipitation<sup>15,16</sup>.

The deposit (UTM grid 728976594) consists of stratabound lenses of pyrite, sphalerite, chalcocopyrite, tennantite, galena, barite and quartz, and an underlying stringer zone with veins of quartz, pyrite, chalcocopyrite, and minor sphalerite and carbonate. The lower part is characterized by massive, granular, honeycomb and filigree sulphide textures, whereas the upper part contains dendritic, colloform and pellet textures<sup>15</sup>. Bitumen occurs in small quantities throughout the deposit as blocky fragments and films that are intergrown with sulphides, and oil is present as fluid inclusions in hydrothermal barite<sup>19</sup>. The deposit displays an upward and outward zonation from Cu to Zn–Pb, a diagnostic feature of VMS-style mineralization. Galena from the deposit gives a Pb–Pb model age of ~3,260 Myr (ref. 15). The composition of the ore indicates that fluid temperatures probably reached 300 °C (ref. 15), but distal areas were probably much cooler. Much of the mineralization formed by