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Dating climatic change in hot deserts using desert varnish on meteorite finds

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Abstract

A thin coating of desert varnish occurs on Forrest 009 and Nurina 004, both equilibrated ordinary chondrite (L6) finds from the Nullarbor Plain, Australia. This finely laminated deposit is chemically and petrographically comparable to the varnish found on terrestrial rocks. Forrest 009, which has a terrestrial age of 5.9 kyr, has a 100-130 µm thick coating of desert varnish that has a laterally consistent chemical micro-stratigraphy comprising a narrow Ba- and Mn-poor lower region, a thick Ba- and Mn-rich central area and a narrow outer zone almost devoid of both cations. The interior of the meteorite contains Fe-oxide and oxyhydroxide veins that have formed by chemical weathering of metals and sulphides. As these veins do not cross-cut the varnish, it must have accreted rapidly relative to the weathering rate of the meteorite. The \leq 70 µm thick varnish on Nurina 004, which has a terrestrial age of 33.4 kyr, lacks a consistent chemical microstratigraphy, but it is cross-cut by Fe-oxide and oxyhydroxide veins, some of which have supplied Fe to the varnish. This implies that the chemical weathering rate of Nurina 004's interior was slow in comparison to the accretion rate of the varnish. The petrography and chemical composition of varnish on Forrest 009 indicates that this meteorite may have resided in a relatively humid environment for most of its 5.9 kyr terrestrial history and that the Nullarbor recently became more arid. This conclusion supports results from an analysis of Fe-bearing weathering products in the interior of the meteorite by Mössbauer spectroscopy, which also indicate that Forrest 009 experienced an early period of rapid weathering under relatively humid conditions. The petrography of varnish on Nurina 004 shows that the interior of the meteorite weathered relatively slowly, probably because it fell during an arid time, which is again in agreement with previous Mössbauer spectroscopy results. Results from both meteorites are in agreement with palaeoclimate data derived from a number of other proxies. The implications of this work are that the large number of meteorites that have been collected from several hot deserts of the world may be a powerful source of information on climate change over the last 30-35 kyr.

Keywords: Desert varnish; ordinary chondrite; meteorite; weathering; Nullarbor.

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1. Introduction.

Determining the nature and chronology of climate change during the Quaternary is currently an extremely important area of research that incorporates many disciplines. Earth Scientists have concentrated on identifying finely layered sedimentary sequences, deposited in terrestrial or marine environments, that record past climates by chemical, isotopic, mineralogical or palaeontological means. One finely laminated deposit that has attracted considerable recent interest is desert varnish (see review by [1]). This is a rock coating that is predominantly composed of clay minerals cemented by Mn-Fe oxyhydroxides [2, 3]. Desert varnish accretes very slowly (<1 - 40 μ m/kyr) and rarely exceeds 200 μ m in thickness regardless of the duration of subaerial exposure [4]. The presence of anthropogenic pollutants in recently formed varnish indicates that most constituents are atmospherically-derived [5, 6] and precipitation plays a major role [1]. Bacteria are also probably important in fixing the Mn and Fe [2, 3]. Despite the variability of accretion rates, even between samples from the same locality [4], the chemical microstratigraphy of varnish may be correlated on a regional scale. Variations in Ba and Mn concentrations, which produce colour banding in ultrathin sections examined by transmitted light, are believed to reflect fluctuations in humidity of the environment and so provide a proxy for climate change [1, 2, 7]. Despite a considerable body of research that supports the utility of varnish as a climate proxy, detailed studies of varnish from a number of sites within a limited geographical area have shown that the chemical microstratigraphy may vary considerably [8]. A detailed discussion of this issue is beyond the scope of the current study and here we adopt the most recent model [1]. In order for data extracted from varnish to be of any practical use in analysing climate change, it is necessary to study samples derived from rock surfaces whose duration of subaerial exposure is precisely known. This necessity of using dated surfaces has significantly limited the application of desert varnish in climate research. Meteorites of known terrestrial ages provide dated surfaces that may be useful in these studies.

Meteorites recovered from hot deserts are another potential source of palaeoclimate data. It has been demonstrated by [9, 10] that a climate signal can be extracted from some terrestrially weathered meteorites (equilibrated ordinary chondrites) by analysing two properties: (i) the ratios of different valence states of iron in weathering products and (ii) the ratios of paramagnetic and magnetically-ordered Fe^{3+} . The ratio of Fe^{0} and Fe^{2+} to Fe^{3+} can be measured using Mössbauer spectroscopy. When equilibrated ordinary chondrites fall to Earth all iron is present as Fe⁰ and Fe^{2+} but it is progressively oxidised to Fe^{3+} during chemical weathering. The weathering of hot desert meteorites takes place in two distinct stages [9-11]. Prior to terrestrial weathering the equilibrated ordinary chondrites have a high intergranular porosity (average of 15% [10]). Weathering agents (atmospheric gas & water) readily gain access to the interior of the meteorites via these pores. Fe,Ni metal and troilite is rapidly converted to Fe-oxide/oxyhydroxides, which occlude the intergranular pores and coat, or 'armour', mineral grains in a process termed 'passivation'. Following passivation, weathering rates slow dramatically [11]. Meteorites that have fallen in hot deserts during relatively humid periods (inferred from other palaeoclimate indicators such as former lake levels and periods of speleothem formation) had been more heavily weathered during the pre-passivation phase than those that have fallen during more arid times [9, 10]. In addition to quantifying Fe^{3+} concentrations, Mössbauer spectroscopy can also be used to measure the abundance of paramagnetic Fe^{3+} and magnetically-ordered Fe^{3+} . Paramagnetic Fe^{3+} is associated with the minerals akagenéite, goethite or lepidocrocite whereas the magneticallyordered Fe³⁺ is associated with maghemite and magnetite. These data are expressed as a ratio of paramagnetic Fe³⁺/total Fe³⁺ where total Fe³⁺ is paramagnetic Fe³⁺ plus magnetically-ordered Fe^{3+} [9]. These authors suggest that weathering during relatively humid periods will produce a lower value of paramagnetic Fe^{3+} /total Fe^{3+} and this initial mineralogy is preserved through subsequent periods of weathering.

This paper describes results of a petrographic and chemical study of varnish that coats two meteorites recovered from the Nullarbor desert, Australia. The Nullarbor is an excellent location

for this study because the surface has been stable for the last ~30 kyr and during this time the region has experienced a number of humid-arid cycles that have been studied using proxies including speleothems and former lake levels. Results of this work show that the varnish and its parent meteorite are both valuable sources of palaeoclimatic data and that analysing varnish on meteorites has a number of advantages over using varnished terrestrial rocks. In addition, we show that the varnish may provide information on the terrestrial history of the meteorite, for example the relative timing of periods of burial and exhumation.

2. Materials and methods

The two meteorites discussed here, Forrest 009 and Nurina 004, were both recovered from the Nullarbor Plain, Australia. Five pieces of Forrest 009 (L6), totalling 1 kg, were recovered in 1979 (latitude S: 30° 09', longitude E: 128° 05') [12]. One piece of Nurina 004 (L6) weighing 28.3 g was recovered in 1986 (latitude S: 30° 47', longitude E: 126° 27') [13]. These two meteorites were selected for study for a number of reasons. Firstly, the terrestrial ages of these meteorites have been determined and weathering products in their interiors have also been studied in detail [9]. Secondly, both meteorites have a well preserved sequence of desert varnish. Lastly, the degree of weathering of the interior of Forrest 009 is comparable to that of Nurina 004, although the two meteorites have very different terrestrial ages (Fig. 1). Work by [9] suggests that this similarity in the degree of weathering is because Forrest 009 fell during a relatively humid period whereas Nurina 004 fell when the Nullarbor was much more arid. Thus, these two meteorites provide an excellent test of the utility of desert varnish for describing climate change.

Double-polished thin sections, prepared to standard petrographic thickness (~30 μ m), were made of Forrest 009 and Nurina 004. Backscattered electron (BSE) images were acquired using a Cambridge Instruments S360 SEM operated at 20 kV. X-ray maps of the varnish and underlying meteorite were acquired using an Oxford Instruments ISIS system attached to the SEM. Quantitative chemical analyses of the varnish on Forrest 009 were obtained using a Cameca SX50 wavelength-dispersive electron probe operated at 15kV with a 10 nA beam current. The spot was defocused to 10 μ m in diameter to limit volatile loss from beam-sensitive constituents and to provide averaged analyses of the micrometre-scale laminations.

3. Chemical and mineralogical composition of meteorite interiors

Both meteorites are L6 equilibrated ordinary chondrites. The average mineralogy of L class ordinary chondrites is 44.8% olivine, 24.2% orthopyroxene, 5.0% clinopyroxene, 10.3% plagioclase 8.4% Fe,Ni metal, 5.8% troilite [14]. Forrest 009 has a ¹⁴C terrestrial age of 5.9 ± 1.3 kyr (B.P) and is relatively highly weathered (oxidation of 38.4%), whereas Nurina 004 has a ¹⁴C age of 33,400 \pm 2300 kyr (B.P) but has undergone a comparable degree weathering (oxidation of 40.4%; visual weathering grade "C") [10, 13] (Fig. 1). The values of paramagnetic Fe³⁺/total Fe³⁺ for Forrest 009 and Nurina 004 are 0.54 and 0.70 respectively [9] (Fig. 1). More data on the distribution of total Fe between various primary and secondary minerals and valence states are listed in [9].

Almost all of the original Fe,Ni metal in Forrest 009 has been replaced by Feoxide/oxyhydroxides, although considerable volumes of troilite remain. Ramifying veins of Feoxide/oxyhydroxides also cross-cut the interior of the meteorite (Fig. 2a, b, c). Olivine and pyroxene grains adjacent to the Fe-oxide/oxyhydroxide pseudomorphs and veins have been partially dissolved to leave skeletal relicts (Fig. 2d). The interior of Nurina 004 shows a comparable degree of terrestrial weathering to Forrest 009 and again contains pseudomorphs and veins of Fe-oxide/oxyhydroxides (Fig. 3). Significantly, some veins are partially occluded by an iron sulphide (?pyrite or marcasite), demonstrating that hydrous Fe-oxides and oxyhydroxides are not the only weathering products of troilite. There is again evidence for dissolution of silicates to producing skeletal relicts (Fig. 4a). Outer parts of the meteorite contain 'pockets' up to 0.5 mm deep that are filled with rounded grains ~40-3 μ m in diameter and cemented by Feoxide/oxyhydroxides and desert varnish (Fig. 3). The rounded grains include desert varnish (Fig. 3b) and quartz (Fig. 3c).

4. Structure and composition of the desert varnish

Light microscope and BSE images show that the varnish on outer surfaces of Forrest 009 is 100-130 µm thick (Fig. 2b, c) and is thickest in local depressions, or 'microbasins', in the outer surfaces of the meteorite (Fig. 2b). Another surface of Forrest 009, probably that which rested on the ground, is encrusted with a layer of mixed carbonate and quartz. As the varnish directly overlies olivine, pyroxene, Fe,Ni metal and troilite grains, it has clearly formed directly on the interior of the meteorite rather than on the fusion crust. Significantly, none of the Feoxide/oxyhydroxide veins that pervade the interior of the stone can be traced into the varnish and they terminate abruptly at its base (Fig. 2b, c). A few quartz grains occur at the interface of the outer meteorite surface and the base of the varnish, but quartz is absent from within the varnish itself. Some of the outer layers of the varnish have been exfoliated by displacive growth of Cacarbonate within fractures (Fig. 5). These fractures are oriented both parallel and perpendicular to layering in the varnish. Those fractures that cut through lower levels of the varnish appear to have been partially 'healed', although contain Ca-carbonate in the upper levels (Fig. 5). Electron probe data show that the varnish is dominated by Al, Si, Mn and Fe (Table 1). Correlation coefficients between all of the elements analysed are also listed in the table. These data demonstrate two related elemental groups: Si-Al-Mg-K and Ba-Mn. Data for Fe are less clear, but this element is most strongly associated with the Si-Al-Mg-K group. Through most of its thickness the varnish is opaque in transmitted light apart from the outermost part, which is dark orange. BSE images show that the varnish is laminated on the micrometre scale and the outermost part, which is orange in transmitted light, has a lower mean atomic number (Fig. 5). Where thickest, within local microbasins (Fig. 2c), the varnish has a narrow and discontinuous lower zone that is relatively poor in Ba and Mn, a wide middle zone that is Ba- and Mn-rich and a narrow uppermost zone that is almost devoid of both cations (Fig. 5).

The varnish on Nurina 004 is \leq 70 µm thick (Figs 3, 4) and has a complex relationship to the parent meteorite. The interface between the outer surface of the meteorite and the varnish can be abrupt (Fig. 3a), but veins of Fe-oxide/oxyhydroxide may also penetrate from the interior of the meteorite into the varnish (Fig. 4b, c). In some places these veins are abruptly truncated along an internal 'unconformity' (Fig. 4d), whereas elsewhere the vein connects with a Fe-rich layer within the varnish (Fig. 4b, c). The chemical stratigraphy of the varnish, as revealed by BSE images, X-ray maps and linescans, is complex and highly variable so that a consistent pattern could not be recognised. Most of the varnish is Ba- and Mn-rich although small Ba- and Mn-poor areas occur in the lower parts of microbasins.

5. Discussion

Previous work on varnished meteorites and varnish from Australia: A number of varnished meteorite finds have been previously described, including the Namibian find Gobabeb [15] and meteorites collected from the Gold Basin strewn field (Mojave Desert) by [16, 17]. Considerable work has been undertaken on varnish in arid regions of Australia by Dragovich, predominantly in western New South Wales [18-22]. This author has noted the ubiquity of varnished rock surfaces in arid Australia. The varnish ranges from 50-150 µm in thickness and commonly shows a layering of Mn-rich and Mn-poor zones when studied in cross-section [19].

Accumulation of varnish on the Nullarbor meteorites: The presence of varnish on both meteorites studied and the absence of layers of coarse detrital material within the varnish supports the assertion of [10] that the Nullarbor meteorites have been subaerially exposed for most of their terrestrial history. As [10] also note, most of the Nullarbor finds are too small to have formed a crater on impact, which could also have lead to burial. The absence of a fusion crust beneath the

varnish of both finds suggests that it has either weathered away or spalled off soon after impact. If the crust had persisted for tens or hundreds of years prior to being eroded from the outer surface of the meteorite some of the earliest layers of desert varnish may have been lost. This possibility is difficult to evaluate with the information currently available.

The small rounded grains of quartz and desert varnish within 'pockets' in the outermost parts of Nurina 004 are interpreted to be wind-blown grains that accumulated within depressions in the surface of the meteorite. Some early varnish occurs between the detrital grains and has probably cemented them (Fig. 3b). The presence of detrital grains of desert varnish within these pockets demonstrates that it was being actively eroded from rock surfaces elsewhere shortly after Nurina 004 fell. Although the varnish on Forrest 009 shows no evidence of abrasion, indicating that it may have been accreting continuously since the fall of the meteorite, 'unconformities', presumably produced by localised erosion, occur within varnish on Nurina 004. Significantly, the Fe-oxide/oxyhydroxide veins that are very common in the interior of Forrest 009 do not cross-cut the varnish and so must predate it. By contrast, Fe-oxide/oxyhydroxide veins within Nurina 004 commonly penetrate into the varnish, showing that the interior of the meteorite was still undergoing weathering after the varnish had started to form. The potential palaeoclimatic significance of these observation is discussed later. The fact that some of the Feoxide/oxyhydroxide veins within Nurina 004 connect with Fe-rich layers within the varnish (Fig. 4b, c), indicates that some components of the varnish have been derived from within the parent rock.

The varnish on Forrest 009 and Nurina 004 is chemically comparable to that found on terrestrial rocks from hot deserts in the USA [8] and arid Australia [20]. From electron probe analyses of varnish on terrestrial volcanic rocks [8] likewise identified the presence of a silicate phase (containing Si, Al, K and Mg), a Mn-bearing phase and a Fe-bearing phase. The <70 μ m and 100-130 μ m thickness of the Nurina 004 and Forrest 009 varnishes gives accretion rates of \leq 2-22 μ m/kyr respectively. This is in good agreement with the 3-15 μ m/kyr mean growth rates of varnish on young (<30 kyr) substrates in the USA [4].

Palaeoclimate data from meteorite weathering products: Forrest 009 and Nurina 004 are among of a suite of nine L and LL class ordinary chondrites from the Nullarbor whose terrestrial ages and degrees of oxidation were quantified by [9, 10] (Fig. 1). They found that when the degree of oxidation of these finds was compared with their terrestrial age (2.2-33.4 kyr), some meteorites showed greater rates of weathering than others. The three finds with terrestrial ages of 5.9-7.6 kyr, which include Forrest 009, stand out as having been relatively highly weathered (Fig. 1) and also have low values of paramagnetic Fe³⁺/total Fe³⁺ (0.47-0.57) (Fig. 1). These data have been interpreted to indicate that the three meteorites fell during a humid period [9]. By contrast some of the older finds, including Nullarbor 012 and Nurina 004, have relatively low degrees of weathering for their age and higher values of paramagnetic Fe³⁺/total Fe³⁺ (Fig. 1), suggesting that they fell during a more arid time [9].

Data from a number of sources confirms that the climate in the Nullarbor has changed over the last 30 kyr, although it must be emphasised that throughout this time the climate has been essentially arid (Alex Bevan, personal communication 2002). Data from lake levels indicates that the Nullarbor was relatively humid from 25-20 kyr then became more arid, with a minimum level at 15 kyr and returned to humid conditions with another peak in lake levels at ~7 kyr (BP) [23-25]. In addition, botanical evidence suggests that effective precipitation in the Nullarbor increased from ~180 to 250 mm from 10-8 to 5-4 kyr (Alex Bevan, personal communication 2002) [26]. A period of halite speleothem growth at 2.5 \pm 1.2 kyr has also been identified by [27]. Humid periods identified by these independent climate proxies are indicated on Figure 1 (shaded areas) and show a good correspondence with the humid periods identified by rapid rates of weathering and low paramagnetic Fe³⁺/total Fe³⁺ values in meteorites that fell approximately at those times [9].

Palaeoclimate data from the varnish: Palaeoclimate information derived from weathering products within Forrest 009 can be independently assessed by examining the chemical microstratigraphy of its varnish. The information that varnish may contain regarding past climates has been the subject of a number of previous studies and is still the source of considerable controversy. The most recent work [1] has concluded that variations in the concentration of Ba and Mn though a sequence of desert varnish records, by mechanisms that are not currently understood, changes in ambient humidity (or 'wetness') during accretion. As these authors stress, one of the principal difficulties in using the technique more widely, especially in areas outside the USA, is the paucity of reliable dates for the duration of subaerial exposure of rock surfaces.

Most of the varnish on Forrest 009 is relatively Ba- and Mn-rich, but the innermost and especially outermost parts are poor in both cations (Fig. 5). Interpreting these data using the model of [1] would suggest that Forrest 009 spent most of its terrestrial history in a relatively humid environment and conditions became more arid relatively recently. The relatively high degree of oxidation and abundance of magnetically-ordered Fe³⁺ in Forrest 009, and the two other meteorites that fell at a similar time (Fig. 1), certainly does support the idea that Forrest 009 experienced humid conditions early in its terrestrial history. The fact that Fe-oxide/oxyhydroxide veins do not cut the varnish (Fig. 2b, c) also confirms that the weathering products formed rapidly in comparison to the accretion rate of the varnish. Although there is a qualitative agreement between palaeoclimatic data from the interior of the meteorite and from the chemical stratigraphy of the varnish, we fully recognise the limited value in drawing conclusions from a single sample. One way in which these conclusions could be strengthened is by dating the carbonates that fill fractures in the outer parts of the varnish (Alex Bevan, personal communication 2002).

Nurina 004 lacks a clearly-developed chemical microstratigraphy to use for palaeoclimate reconstruction and also has evidence for abrasion (Fig. 4d), again mitigating against firm conclusions being drawn from the varnish. However, the abundance of Fe-oxide/oxyhydroxide veins that cross-cut the varnish does demonstrate that the meteorite was still undergoing chemical weathering even after accretion of tens of micrometres of varnish. This could indicate that the weathering rate of the meteorite was relatively slow, suggesting that it fell during an arid period. This conclusion is supported by Mössbauer spectroscopy data from meteorite weathering products [9] and agrees with independent climate records (Fig. 1).

6. Implications

Results of this study show that meteorite finds have a number of advantages over terrestrial rock substrates for the purpose of using desert varnish to study climate change. Firstly, a large number of finds have already been collected from hot deserts, principally the Sahara (1508 finds), Nullarbor (>280 finds) and Roosevelt County USA [28]. Secondly, many of these meteorites have been dated using ¹⁴C techniques, which have an error of $\pm \sim 1-2$ kyr. It is absolutely certain that varnish accretion started after the fall of the meteorite; sampling geomorphologic sites can have more uncertainties. Finds in the Nullarbor span the last 33 kyr and Saharan finds range over a comparable time period (~35 kyr). As the flux of meteorites is continuous, so long as a sufficient number of varnished finds are available from one area a complete chronology can be established. By contrast, finding suitable terrestrial rocks in hot deserts for radiocarbon dating is difficult [1]. Another advantage is that the varnished surface of all meteorite finds will come from an identical microenvironment, a few centimetres above the ground surface, and so differences in topography will not be a significant variable in the nature or rate of varnish accumulation between sites. Ordinary chondrites of a given type also vary little in original mineralogy or bulk chemical composition, regardless of where they fall. Thus, differences in rock substrate can be discounted as a potential factor affecting the chemistry of varnish from different localities. Lastly, as the volume and mineralogy of weathering products within the equilibrated ordinary chondrites is determined by climate soon after the fall of the

meteorite, these properties can be used to cross-check palaeoclimate data derived from overlying varnish.

We recognise that studying desert varnish on meteorite finds has some inherent disadvantages in comparison to terrestrial rocks. Not all finds will have a varnish and that on some may be unsuitable for the purpose (for example it may have been partially eroded, as is the case in Nurina 004). As they are low-lying, some meteorites may have been partially covered by soil or sediment for a portion of their terrestrial residence, although this does not appear to have happened with regard to the two meteorites studied here and may in fact be of use. For example [29] interpreted the absence of varnish from the surface of Korra Korrabes, a Namibian find, to indicated that it may have been covered by sand for much of its terrestrial history. Lastly, most authors emphasise the importance of selecting samples for study that have the most complete varnish record. Given the relatively small size of individual meteorites and the limited numbers of finds available, the potential for such selectivity is restricted. However, as the outer surface of meteorites is typically pitted, the result of ablation during atmospheric entry, many microbasins will be available for study even on a small sample.

7. Conclusions.

This work has shown that desert varnish on extraterrestrial rocks can provide detailed information on changes in the climate of terrestrial environments. The advantages of using meteorites for understanding climate change over terrestrial rocks is the ease of dating their duration of residence on the Earth's surface and the presence of weathering products containing a climate signal within the meteorites themselves. As large numbers of meteorites have already been recovered from hot deserts in Africa, America and Australia, this approach has world-wide applicability. Further work is currently in progress to test this model, using finds from the Acfer region of the Sahara.

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	Wt % Correlation coefficients										
		SiO_2	Al_2O_3	Fe ₂ O ₃	NiO	MnO	MgO	BaO	CaO	Na ₂ O	K ₂ O
SiO ₂	30.8	1.00	0.77	0.45	-0.04	-0.12	0.60	-0.11	-0.29	0.04	0.78
Al_2O_3	19.8	0.77	1.00	0.60	0.00	0.00	0.49	0.00	-0.42	0.03	0.57
Fe_2O_3	9.1	0.45	0.60	1.00	-0.01	0.01	0.47	-0.10	0.33	0.03	0.55
NiO	0.2	-0.04	0.00	-0.01	1.00	0.22	-0.05	0.27	0.00	0.00	-0.08
MnO	12.4	-0.12	0.00	0.01	0.22	1.00	-0.04	0.94	-0.14	0.00	-0.16
MgO	1.9	0.60	0.49	0.47	-0.05	-0.04	1.00	-0.10	-0.18	0.01	0.58
BaO	1.0	-0.11	0.00	-0.10	0.27	0.94	-0.10	1.00	-0.14	n.a.	-0.43
CaO	4.1	-0.29	-0.42	0.33	0.00	-0.14	-0.18	-0.14	1.00	-0.07	-0.27
Na ₂ O	0.1	0.04	0.03	0.03	0.00	0.00	0.01	n.a.	-0.07	1.00	0.03
K_2O	2.1	0.78	0.57	0.55	-0.08	-0.16	0.58	-0.43	-0.27	0.03	1.00
Total	81.5										

Table 1Mean chemical composition of the Forrest 009 varnish and correlation coefficients of the elements.

Number of analyses were 124 for all elements apart for Na₂O (78 analyses) and BaO (45 analyses). n.a. denotes not applicable because Na₂O and BaO were analysed in separate runs. Data in bold are significant correlations ($R^2 > 0.55$).



Fig 1. Diagram illustrating data from nine L and LL ordinary chondrites finds from the Nullarbor studied by [9]. For each find the degree of oxidation and ratio of paramagnetic Fe^{3+} to total Fe^{3+} (quantified by Mössbauer spectroscopy) is shown, together with the ¹⁴C terrestrial age. The shaded areas represent humid periods identified by [9] from other proxies including lake levels and periods of speleothem growth. Error bars on each datapoint represent the errors in ¹⁴C age determinations (typically ± 1.3 kyr).



Fig 2. BSE images of Forrest 009. Olivine and pyroxene grains are both light grey in these images and are difficult to distinguish owing to their similar mean atomic number, but feldspar grains are dark grey. (a) The interior of the meteorite showing primary silicate grains (mainly olivine and pyroxene) cut by two Fe-oxide/oxyhydroxide veins (oriented NE-SW in the image). Silicates between the two veins have been partially dissolved, leaving a network of small pores (arrowed). (b) Desert varnish (D) within a microbasin in the external surface of the meteorite. Note how the varnish thickens into the microbasin, although parts on the right and left hand side may have been partially removed during thin section preparation. (c) Desert varnish (D) coating the external surface of the meteorite. Note how many of the Fe-oxide/oxyhydroxide veins (white) are oriented roughly perpendicular to the former outer surface of the meteorite and terminate abruptly at the base of the varnish. (d) The interface between an olivine grain (medium grey, upper right) and heavily oxidised Fe,Ni metal (white, lower half of image). Much of the interior of the olivine has been dissolved to leave pores (black). These pores are bridged by narrow Fe-oxide/oxyhydroxide veins, which produce a 'skeletal' texture and demonstrate that silicate dissolution postdates at least one stage of vein formation.



Fig. 3. Image and corresponding X-ray maps of the outer part of Nurina 004. Lighter grey tones in the X-ray maps correspond to greater numbers of counts. (a) BSE image showing the interior of the meteorite, which contains Fe-oxide/oxyhydroxide veins (V) and pseudomorphs of metal and sulphide grains (P). This image features a large 'pocket' filled with rounded grains (delineated by arrows). The outer surface of the meteorite (top edge of the image) is coated with desert varnish. (b) Al K α X-ray map showing that with the exception of small grains of feldspar in the interior of the meteorite most Al occurs within the varnish that coats its external surface. Some very Al-rich areas within the pocket (arrowed) are discrete grains of varnish. Desert varnish also occurs between rounded grains within the 'pocket'. (c) Si K α X-ray map also allows grains of olivine (OI) in the interior of the meteorite to be distinguished from orthopyroxene (Px). (d) Fe K α X-ray map illustrating the abundance of Fe-oxide/oxyhydroxide veins in the interior of the meteorite (white/light grey). Fe-oxide/oxyhydroxide also occurs between rounded grains in the 'pocket'.



Fig. 4. BSE images of desert varnish from Nurina 004. (a) Finely lamiated varnish overlying heavily weathered clinopyroxene (CPX) grains. (b) A complex interface between varnish (upper part of the image, base and top delineated by arrows) and the meteorite substrate (lower part). Most of the interior of the meteorite is composed of compositionally zoned olivine (OI) that has been partially replaced by euhedral Fe-oxide/oxyhydroxide crystals that are concentrated just beneath the varnish. A Fe-oxide/oxyhydroxide vein (V) can be traced from the interior of the meteorite into the varnish and may be associated with a Fe-rich patch in upper parts of the varnish (Fe). (c) A vein of Fe-oxide/oxyhydroxide (V) extends from the interior of the meteorite (base of the image) into the varnish (D) and connects with a Fe-rich band in the interior of the varnish (Fe). (d) Area of varnish (D, upper part of image) overlying a 'pocket' in the outer part of the meteorite that is filled with rounded grains and cemented by Feoxide/oxyhydroxide. Most of the rounded grains are quartz, but some are of desert varnish. Varnish also fills a fissure at the left hand side edge of the pocket and a Fe-oxide/oxyhydroxide vein (V) runs up the centre of the fissure. Note that this vein, and other lower parts of the varnish is truncated at an 'unconformity' (arrowed).



Fig. 5. BSE image of a complete sequence of desert varnish from the centre of the 'microbasin' in Forrest 009. The black and white line marks the path of an electron probe traverse. Compositional data (right hand side) indicate that the centre of the varnish layer is Ba- and Mn-rich whereas the outer edges are Ba- and Mn-poor. Note that the outer layers of the varnish are being exfoliated by displacive growth of Ca-carbonate. Other fractures, which are oriented perpendicular to the layering, contain Ca-carbonate in upper parts of the varnish but appear to have been partially 'healed' lower down.