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# Late Eocene impact ejecta: geochemical and isotopic connections with the Popigai impact structure

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#### Abstract

Late Eocene microtektites and crystal-bearing microkrystites extracted from DSDP and ODP cores from the Atlantic, Pacific, and Indian oceans have been analyzed to address their provenance. A new analysis of Nd and Sr isotopic compositions confirms previous work and the assignment of the uppermost microtektite layer to the North American tektites, which are associated with the 35.5 Ma, 85 km diameter Chesapeake impact structure of Virginia, USA. Extensive major element and Nd and Sr isotopic analyses of the microkrystites from the lowermost layer were obtained. The melanocratic microkrystites from Sites 216 and 462 in the Indian and Pacific oceans possess major element chemistries, Sr and Nd isotopic signatures and Sm-Nd, T<sub>CHUR</sub>, model ages similar to those of tagamite melt rocks in the Popigai impact structure. They also possess Rb-Sr, T<sub>UR</sub>, model ages that are younger than the tagamite  $T_{CHUR}$  ages by up to ~1 Ga, which require a process, as yet undefined, of Rb/Sr enrichment. These melanocratic microkrystites are consistent with a provenance from the 35.7 Ma, 100 km diameter Popigai impact structure of Siberia, Russia, while ruling out other contemporaneous structures as a source. Melanocratic microkrystites from other sites and leucocratic microkrystites from all sites possess a wide range of isotopic compositions ( $\varepsilon$ (<sup>143</sup>Nd) values of -16 to -27.7 and  $\varepsilon$ (<sup>87</sup>Sr) values of 4.1-354.0), making the association with Popigai tagamites less clear. These microkrystites may have been derived by the melting of target rocks of mixed compo sition, which were ejected without homogenization. Dark glass and felsic inclusions extracted from Popigai tagamites possess  $e^{143}$ Nd) and  $e^{87}$ Sr) values of -26.7 to -27.8 and 374.7 and 432.4, respectively, and T<sub>CHUR</sub> and T<sub>UR</sub> model ages of 1640–1870 Ma and 240–1830 Ma, respectively, which require the preservation of initially present heterogeneity in the source materials. The leucocratic microkrystites possess diverse isotopic compositions that may reflect the melting of supra-basement sedimentary rocks from Popigai, or early basement melts that were ejected prior to homogenization of the Popigai tagamites. The ejection of melt rocks with chemistries consistent with a basement provenance, rather than the surface  $\sim 1$  km of sedimentary cover rocks, atypically indicates a nonsurficial source to some of the ejecta. Microkrystites from two adjacent biozones possess statistically indistinguishable major element compositions, suggesting they have a single source. The occurrence of microkrystites derived from a single impact event, but in different biozones, can be explained by: (1) diachronous

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biozone boundaries; (2) post-accumulation sedimentary reworking; or (3) erroneous biozonation. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: microtektites; Eocene; impacts; Popigay Structure; ejecta; isotopes

## 1. Introduction

The correlation of ejecta layers in the stratigraphic record with source impact structures allows a comparison to be made between impact structure sizes, target lithologies, and ejecta distribution with changes in the biota across the layer. Consequently, the environmental implications of various impact events can be judged. The best known of the globally distributed impact ejecta layers is that associated with the Chicxulub event at the end of the Cretaceous [1]. This event produced a relatively large (ca. 195 km diameter) impact structure [2], as well as an iridium-enriched ejecta layer with global distribution [1]. The extent and quantity of fragmented and impact-fused target rocks ejected from an impact site are related to the impact energy and, hence, the final diameter of the impact structure. A global distribution of ejecta requires sufficiently high velocities to eject impact-shattered and molten target rocks beyond Earth's atmosphere. This is known as atmospheric blow-out. The ejecta then return gravitationally at a wide range of latitudes, where they are further distributed by stratospheric winds prior to deposition. Melosh ([3], p. 212) has calculated that impact structures with diameters as small as 3 km are capable of producing atmospheric blow-out.

We assess the ejecta associated with impact



Fig. 1. The five known late Eocene impact structures and Ocean Drilling Program/Deep Sea Drilling Program sites containing upper Eocene impact ejecta. Major element data are presented in Figs. 3 and 4 for all sites labelled. Open circles define core sites for which isotopic data are presented in this study. The sizes of the shaded circles on land are proportional to the impact structure diameter (data from [4]). The ages of the impacts have been approximately established based on stratigraphic relationships for the Logoisk structure [5,6] and by isotopic dating of impact melt rocks for the other structures. Numbers on the left hand panel refer to references: 1, [7]; 2, Site 612 tektites [8]; 3, [9]; 4, [10]. All are <sup>40</sup>Ar-<sup>39</sup>Ar plateau ages except 3, which is a K-Ar age. Ages 3 and 4 were recalculated using the decay constants of [11].

events occurring toward the end of the Eocene. The correlation of late Eocene impact ejecta with a particular source impact structure is complicated by a number of impacts of requisite age, all of which were, at least in theory, large enough to have generated widespread ejecta fields. Of the more than one hundred and fifty impact structures so far recognized on Earth [4], five impacts are known from the late Eocene (Fig. 1): Popigai, Siberia (100 km diameter); Chesapeake Bay, USA (85 km); Mistastin, Canada (28 km); Logoisk, Belarus (17 km); and Wanapitei, Canada (7.5 km).

Late Eocene impact ejecta have been identified in ocean sediments from the Indian, Pacific, and Atlantic Oceans, the Caribbean Sea/Gulf of Mexico and the Weddell Sea, off Antarctica [12-13] (Fig. 1). The Eocene ejecta comprise glassy and semicrystalline spheres, droplets and fragments, generally < 1 mm in diameter. The glassy impact ejecta are known as microtektites, while their crystalline equivalents are known as microkrystites or crystal-bearing spherules [14,15]. Glassy late Eocene ejecta have also been found in Texas, Georgia, Massachusetts (Martha's Vineyard) and Barbados [16-19]. The more continuous sedimentation rates and higher biostratigraphic resolution afforded by marine microfossil assemblages in the oceanic realm allow workers to define more precisely the biozones in which the ejecta layers are found. It is for this reason that this work focusses on the marine ejecta layers.

#### 2. Stratigraphic controversy

Biostratigraphic correlations from a number of Ocean Drilling Program (ODP) and Deep Sea Drilling Program (DSDP) cores have been used to construct a *composite* stratigraphic section. Some workers interpret this composite section to contain at least three biostratigraphically distinguishable upper Eocene ejecta layers [20,21]. These comprise a microtektite layer lying above two microkrystite layers (Fig. 2B). The inference is that each layer was ejected at a different time from a distinct impact crater. Others believe that there are only two geochemically and stratigraph-



Fig. 2. A schematic interpretation of the number and stratigraphic location of upper Eocene impact ejecta layers in marine sediment cores. Each composite section represents the position and number of impact ejecta layers inferred from multiple cores at different drill sites, according to the authors specified.

ically distinguishable layers, an upper microtektite layer and only one lower microkrystite layer [14] (Fig. 2C). Although individual cores typically contain a single microtektite layer, or a microtektite layer above a single microkrystite layer, the proponents of three layers in the *composite* section propose that the microkrystites should be subdivided into two layers based on subtle major element differences and because the layers occur in different biozones in different cores: the uppermost in the *Turborotalia cunialensis* Zone (formerly ascribed to the *Globorotalia cerroazulensis* Zone; [22]), and the lowermost in the *Porticulasphaera semiinvoluta* Zone.

Some workers have suggested that they can resolve six layers in the composite stratigraphic section, using a graphic correlation technique of microfossil assemblages [23,24] (Fig. 2A). Others argue that the first and last occurrences of these fossils can be diachronous, and so the correlation of layers separated by only a few thousand years exceeds the limits of resolution of this technique [25]. The actual number of discrete ejecta layers present in the ocean record, therefore, remains contentious. We address this controversy here using major element and isotope geochemistry.

## 3. Strewn fields

Since its discovery by seismic methods [26], the

Table 1

ι	Jpper	Eocene	microtek	ctites an	d micro	krystites	sample	ed f	or t	he is	sotopic	determi	inations	in 1	this	stud	y
						~											~

Number	Color	Homogeneity	Opacity	Magnetism	Vesicularity	Shape			
						Splash	Spherical	Fragment	
Site-core-section	, depth: 612–2105. 11	5–117 cm (North	American layer)	(sample 612)					
$\gg 1000$	colorless	homogeneous	TR	non-magnetic	vesicular			$\gg 1000$	
462-36-01, 145-	147 cm ( <i>cunialensis</i> Z	one)							
Melanocratie fre	action (sample 462M1	)							
112	amber-medium brown	homogeneous	TL to TR	nd	non-vesicular	3	103	6	
61	medium brown	mottled	OP	nd	non-vesicular	1	58	2	
6	dark brown	homogeneous	OP	nd	non-vesicular	0	6	0	
462-36-02, 6-8	cm ( <i>cunialensis</i> Zone)								
Melanocratie fre	action (sample 462M2	)							
169	amber–medium brown	homogeneous	TL	nd	some vesicular	5	147	17	
70	medium brown	homogeneous	OP	nd	non-vesicular	0	66	4	
10	dark brown	homogeneous	OP	nd	non-vesicular	0	10	0	
216-16-02, 5-7	cm ( <i>semiinvoluta</i> Zone	)							
Clear fraction (	sample 216C)								
140	colorless	homogeneous	TR	non-magnetic	vesicular	17	38	85	
28	amber–medium brown	homogeneous	TR	non-magnetic	vesicular	0	18	10	
Leucocratic frac	tion (sample 216L)								
174	opalescent	homogeneous	TL	non-magnetic	non-vesicular	16	35	123	
96	white	homogeneous	OP to TL	non-magnetic	non-vesicular	7	55	34	
11	amber	homogeneous	TL to TR	non-magnetic	non-vesicular	0	9	2	
Melanocratie fre	action (sample 216M1	and 216M2-2 spl	its)	-					
657	medium brown	mottled	OP	nd	non-vesicular	11	629	22	
292-38-02, 82-8	6 cm ( <i>semiinvoluta</i> Zo	ne)							
Leucocratic frac	ction (sample 292L)								
3	opalescent	mottled gray	TL to OP	non-magnetic	non-vesicular	0	3	0	
161	white-cream	homogeneous	TL to OP	non-magnetic	non-vesicular	0	7	154	
Melanocratie fre	action (sample 292M)								
206	dark to light brown	homogeneous	TR	non-magnetic	vesicular	0	7	199	
315-10-6, 41-47	cm (biostratigraphic a	conation not accur	ately established)	1					
Melanocratie fre	action (sample $315M$ )								
638 <sup>a</sup>	medium brown to	some mottled	TR to OP	non-magnetic	non-vesicular	11	360	267	

OP, opaque; TL, translucent; TR, transparent; nd, not determined.

<sup>a</sup>Includes 547 microspherules from the < 54 micron sieve fraction.

Chesapeake Bay impact structure has been considered the potential source of the uppermost layer of late Eocene microtektites and tektites, especially those located at on-land USA locations and in nearby offshore drill cores [27]. These glassy impact ejecta collectively define the 'North American strewn field'. It has been speculated that the Popigai impact structure may be the source for the microkrystite layer(s) [28–30], though definitive geochemical links have not been made to date. Major and trace element analyses of the ejecta layers typically fail to identify distinct source impact structures. This is because they generally display considerable overlap in their ranges of chemical compositions, and typically fail to exhibit geochemical traits that can be uniquely linked with a particular source crater.

Studies of Rb-Sr and Sm-Nd in tektites and in

microtektites have established that samples from known strewn fields have distinctive radiogenic Sr and Nd isotopic ratios [31,32]. The <sup>143</sup>Nd/ <sup>144</sup>Nd compositions and  $T_{CHUR}$  model ages are consistent with the provenance of tektites from distinct crustal materials of well-defined mean crustal ages. A favorable comparison between isotopic data for ejecta impact melts (microtektites and microkrystites) and melt rocks that remain in contemporaneous impact structures can, thus, be used to link a particular impact ejecta layer to a potential source crater.

We have extracted glassy to semicrystalline ejecta droplets and fragments from eight ODP/ DSDP sites for analysis (Fig. 1). These include dark colored (melanocratic) microkrystites from both the lower T. cunialensis and Po. semiinvoluta Biozones and melanocratic, leucocratic (light colored) and colorless microkrystite fractions from a single layer in the Po. semiinvoluta Zone. In addition, we have sampled the uppermost 'North-American' microtektite layer. Sr and Nd isotopic analyses were determined for impact ejecta from five of these ODP sites and from all of the three layers proposed by D'Hondt et al. [21] (Fig. 2B; Table 1). We have also determined the isotopic compositions of two impact melt rocks from the Mistastin impact structure and a composite sample of pale brown glass clasts from the melt rocks of the Popigai impact structure, for which there are no previous analyses.

### 4. Analytical techniques

Microtektites and microkrystites were extracted from friable ODP/DSDP sediment samples by soaking and wet sieving with detergent. The > 130 mm diameter fraction was retained. The fraction comprising > 54 mm diameter microkrystites was retained from the Site 315A sample, owing to insufficient coarser grained material. Indurated samples were disaggregated using an ultrasonic probe. The microtektites and microkrystites were handpicked and separated into groups with similar physical characteristics. A representative fraction of each was mounted in crystal bond and polished for electron microprobe analysis. The remaining fractions, each comprising 168–657 microtektites/microkrystites, were leached in dilute acids, following Stecher et al. [32], in order to remove localized, secondary ferromanganese and iron sulfide coatings. Samples as small as 0.4 mg were analyzed. The Nd was measured with high sensitivity, as NdO<sup>+</sup>. Typical amounts per mass spectrometer analysis were 10 ng Nd, yielding ion beam currents of  $\sim 0.7 \times 10^{-11}$  A for several hours.

Major elements were determined on a JEOL 744 electron microprobe at an accelerating potential of 15 keV, a current of 3 nA and a peak-count duration of 30-40 s using wavelength dispersive spectrometers. Loss of volatile alkali elements was minimized during an analysis by peak counting for these before the less volatile elements. Each composition presented here comprises the average of three analyses per single microtektite/krystite. Each average was normalized to 100%, to allow comparison with the renormalized data of D'Hondt et al. [21]. Pre-normalization totals average 91.7-99.9 wt%. Carbon and oxygen identification in the ejecta glasses was established using an analytical scanning electron microscope with the Be window removed. Although the principal volatile component is probably water, probably owing to hydration in the ocean, some glasses contain significant quantities of dissolved carbon (probably as  $CO_2$ ).

#### 5. Major element geochemistry

The 189 microkrystite/microtektite major element analyses performed for this study augment a selection of the existing data, compiled principally from D'Hondt et al. [21] (n=115), with an additional 24 analyses from other sources [32,33]. The North American microtektites are clearly geochemically distinct from the analyses of the older Eocene microkrystites, with higher Al<sub>2</sub>O<sub>3</sub>, Fe/Mg, K<sub>2</sub>O and TiO<sub>2</sub> and lower CaO and MgO contents for corresponding SiO<sub>2</sub> values than the microkrystites (Fig. 3). This suggests that the microkrystites have a distinct provenance from the North American tektites/microtektites (i.e., from a different impact structure, with a dif-



Fig. 3. Major element data for North American layer (NAL) microtektites from four locations (ocean drill sites 612, 94 and RC9-58, and Barbados) (triangles), *cunialensis* layer microkrystites from Sites 462, 315A and RC9-58 (black dots) and *semiinvoluta* layer microkrystites from Sites 216 and 292 (gray circles). Data compiled from [21,32,33] and 189 analyses from this study (analyses available upon request from the senior author). The North American microtektite data are geochemically distinct from the microkrystites indicating distinct provenances. The two microkrystite layers are compositionally indistinguishable from one another.

ferent target composition). However, the microkrystites from the *T. cunialensis* and *Po. semiinvoluta* Zones, while displaying a large degree of major element compositional variation, display almost the same compositional range. Some workers (e.g. [21]) believe that the crystal-bearing microkrystites from the *T. cunialensis* and *Po. semiinvoluta* Zones can be distinguished on the



Fig. 4. A comparison of melanocratic (dark colored), mesocratic (intermediate colored) and leucocratic (includes light colored and clear) microkrystites from Site 462 with the composition of the impact melt rocks from the Logoisk [36], Popigai (this study), Mistastin [37] and Wanapitei [5,40] impact structures. The melanocratic microkrystites are very similar to the compositionally homogeneous tagamite impact melt rocks from Popigai (no geochemical data for impact melt rocks from the buried Chesapeake impact structure have been found).

basis of the results of an R- and O-mode factor statistical analysis that they performed on their major element data. However, this technique only provides a qualitative identification of differences, without a strict test of how much the two layers differ, or if, in fact, they show a statistically significant difference at all [34]. The application of more quantitative statistical analyses to the data, including univariate and multivariate analyses of variance and linear discriminant analysis [35], indicates that the two microkrystite layers do not differ significantly enough to distinguish confidently one microkrystite layer from the other. Consequently, we concur with Glass (e.g. [15]) that the microkrystites could have a single source. On the basis of major element compositions, we can only distinguish two ejecta layers: the North America microtektite layer and a single underlying microkrystite layer.

Microkrystites sampled from a single layer commonly display a wide variation in opacity and color. These can range from opaque black, through translucent brown, cream and white to transparent and colorless. These physical differences correspond to their chemical compositions. Melanocratic microkrystites contain higher Al<sub>2</sub>O<sub>3</sub>, FeO, K<sub>2</sub>O, Na<sub>2</sub>O and TiO<sub>2</sub> and lower MgO and CaO than more leucocratic examples from the same layer (Fig. 4).

The major element compositions of the impact melt rocks from the Logoisk, Mistastin, Popigai and Wanapitei impact structures are also plotted on Fig. 4. Melt rocks up to 11.1 m thick in the Logoisk structure comprise aphanitic and holohyaline lenses, dykes and veins in suevite and allogenic breccia [36]. Clasts derived from the granite-gneiss target at Logoisk comprise approximately 20-40% of the rock by volume [36]. In the Mistastin and Popigai craters, the melt rocks comprise sheets at least 15 and 780 m thick, respectively [37,38]. The melt rocks are wholly located within the impact structures and contain variable amounts of partially melted clasts derived from the target area, principally comprising variably shocked quartz, plagioclase and pyroxene. The Popigai melt rocks, which are known as tagamites, locally contain rounded glass clasts and gneiss clasts from the basement, in



Fig. 5. Nd and Sr isotopic compositions of upper Eocene microtektite/microkrystite and impact melt rocks from upper Eocene impact structures. Sample numbers refer to the source ODP/DSDP drill site, suffixed by M, L or C (melanocratic, leucocratic or clear microkrystite fraction). Data sources: North American tektite field [32,33,41]; Wanapitei [39]; Popigai tagamites and gneisses [32,42]. (a) The melanocratic microkrystites are similar to Popigai tagamites and dissimilar to other upper Eocene impact melts. (b) The Nd model ages  $(T_{CHUR}^{Nd})$  of melanocratic microkrystites are similar to Popigai tagamites, though the Rb-Sr system has been affected by a younger Rb/Sr fractionation event, yielding younger  $T_{UR}^{Sr}$  ages. Leucocratic and clear microkrystites possess a distinct isotopic character. Glass and felsic clasts extracted from the tagamites show preservation of isotopic heterogeneity in the source materials.

addition to mineral and lithic clasts. At Wanapitei, the crater structure is submerged so access to melt rocks is restricted to local glacial drift deposits [39], from which these analyses were derived [9,40]. The compositional data for the Logoisk, Mistastin and Wanapitei impact melt rocks clearly plot outside the compositional ranges for all of the microkrystites (Fig. 4).

Thirty-four X-ray fluorescence major element analyses were obtained for tagamites from the Popigai impact structure for this study. These melanocratic, semicrystalline to holohyaline rocks possess a far more restricted compositional range than the melt rocks from the Mistastin or Wanapitei impact structures (Fig. 4). The Popigai tagamites plot almost entirely within the range of the melanocratic microkrystites. However, the leucocratic or clear microkrystites are chemically distinct from the tagamites.

#### 6. Isotopic results

The Sm–Nd and Rb–Sr concentrations and isotopic compositions are shown in Table 2 and in Fig. 5a. The  $\varepsilon$ (<sup>143</sup>Nd) for all samples (except one) fall in the range of -16 to -28. The  $\varepsilon$ (<sup>87</sup>Sr) fall in the range of  $(2-4)\times10^2$  (with three exceptions). The  $\varepsilon$ (<sup>143</sup>Nd) and  $\varepsilon$ (<sup>87</sup>Sr) values generally show that the parent materials of the microkrystite

Table 2	;		
Nd, Sm, Sr, Rb, and isotopic analyses of microkrystite/microtektite samples from ODP Sites 216, 292, 315A, 462 and 612, the Mistastin im	the Mistastin	impact structure, a	and
clasts in Popigai tagamites			

name layer	pled No.	te Weigi	ht Rb	Sr	PN	Sm	144Nd 144Nd	$\frac{147}{144}$ Sm	$\varepsilon$ <sup>143</sup> Nd	-/+	f(Sm/ND)	TCHUR	-/+	$^{87}\mathrm{Sr/}^{86}\mathrm{Sr}$	<sup>8/</sup> Rb/ <sup>80</sup> Sı	$\varepsilon^{87} Sr$	-/+	<i>f</i> (Rb/Sr)	$\mathrm{T}_{\mathrm{UR}}^{\mathrm{Sr}}$
(see		(mg)	(udd)	(udd)	(udd)	(udd)	_					(Ma)							(Ma)
Lable 1)																			
612 NAI	UF VF	2.80	43.1	41.1	11.09	2.12	0.51151	0.11550	-6.6	0.4	-0.4126	640	40	0.72383	3.041	274.4	1.0	35.77	460
462M1 cuni	MK	0.97	85.2	127.8	24.1	1 4.37	0.51065	0.10940	-23.4	0.5	-0.4440	2100	50	0.72944	1.932	354.0	1.8	23.40	066
462M2 cuni	MK	1.18	84.1	117.0	21.7.	7 3.66	0.51062	0.10150	-24.0	0.7	-0.4839	1970	60	0.72944	2.084	354.0	I.I	25.20	880
216M1 <sup>a</sup> semi	MK	2.98	pu	81.2	14.15	3 2.43	0.51073	0.10357	-21.8	0.5	-0.4735	1830	50	0.72860	pu	342.1	0.7	pu	pu
216M2 <sup>a</sup> semi	MK	2.51	42.3	81.0	13.85	5 2.36	0.51066	0.10320	-23.2	0.4	-0.4754	1940	40	0.72845	1.514	340.0	I.0	17.30	1180
216L semi	LK	9.31	39.9	122.7	15.25	5 2.67	0.51082	0.10570	-20.0	0.5	-0.4625	1720	40	0.71923	0.942	209.1	0.6	10.39	1210
216C semi	C	2.16	64.1	168.7	22.2	3.82	0.51103	0.10410	-16.0	0.4	-0.4710	1350	30	0.71769	1.101	187.2	I.0	12.31	910
292M semi	MK	0.42	60.1	579.5	126.8	14.50	0.51091	0.0692	-18.4	0.5	-0.6483	1120	30	0.70479	0.300	4.1	I.0	2.63	(94)
292L semi	LF	0.39	41.2	140.0	55.52	\$ 10.47	0.51046	0.11400	-27.3	0.6	-0.4204	2560	60	0.72090	0.852	232.8	4.2	9.31	1500
15M ?	MK	69.0	52.7	181.7	131.3	11.51	0.51043	0.05300	-27.7	0.4	-0.7305	1510	20	0.71900	0.840	205.8	0.6	9.16	1350
<b>Glass clasts</b> , na	I	1.29	185.2	68.0	86.0	12.06	0.51048	0.08490	-26.7	0.7	-0.5686	1870	50	0.73090	7.901	374.7	1.6	94.54	240
opigai																			
Felsic clasts, na	I	2.22	31.1	71.9	34.62	3.65	0.51042	0.06370	-27.8	0.4	-0.6759	1640	20	0.73496	1.256	432.4	1.2	14.19	1830
opigai																			
Mistastin na M57-60	IS	4.93	27.2	599.1	52.7(	5 9.36	0.51049	0.10720	-26.5	0.6	-0.4548	2320	50	0.70746	0.131	42.0	1.1	0.59	(4290)
Mistastin na	IS	4.94	53.0	472.9	56.82	2 10.17	0.51048	0.10830	-26.8	0.6	-0.4496	2370	50	0.70930	0.324	68.1	0.7	2.92	1400
LM55-EE																			

samples have clear crustal affinities. The concentrations of Sm–Nd are typical for crustal materials, except for high values in two samples. Except for uncertainties in the weight of the samples, we have no identified cause for the high concentrations. We describe briefly the results.

The two melanocratic microkrystite samples from the cunialensis Biozone at Site 462 (samples 462M1 and 462M2) possess similar  $\epsilon$ <sup>(143</sup>Nd) values of -23.4 and -24.0, and identical  $\varepsilon$ (<sup>87</sup>Sr) values of 354.0. Two splits of a larger melanocratic microkrystite sample from Site 216 in the semiinvoluta Zone (216M1 and 216M2) have isotopic compositions almost identical to the melanocratic microkrystites from the cunialensis Biozone at Site 462 [ $\epsilon$ (<sup>143</sup>Nd) of -21.8 to -23.2 and  $\epsilon$ (<sup>87</sup>Sr) of +340.0 to +342.1] (Fig. 5a). In contrast, a leucocratic microkrystite sample (216L) and a clear microtektite sample (216C) show less radiogenic  $\epsilon$ <sup>(87</sup>Sr) values and  $\epsilon$ <sup>(143</sup>Nd) values of -20 and -16. These leucocratic and clear samples are isotopically different from the melanocratic samples, even though all three types were analyzed for Site 216. All the analyzed fractions are isotopically distinct from the North American microtektites (Fig. 5a). Fragments of glassy to crystalline melanocratic microkrystites from Site 292 (samples 292M) possess a slightly less radiogenic  $\varepsilon$ <sup>(143</sup>Nd) value of -18.4 and essentially nonradiogenic  $\varepsilon$ <sup>(87</sup>Sr) value of +4.1 relative to the melanocratic microkrystites from Sites 462 and 216. Melanocratic microkrystites from Site 315A (sample 315M) possess an  $\varepsilon$ <sup>(143</sup>Nd) value of -27.7 and an  $\varepsilon$ <sup>(87</sup>Sr) value of 205.8, distinct from the isotope values for melanocratic microkrystites from the other sites. The Site 292 fragments and 315A microkrystites possess f(Sm/Nd) fractionation values significantly more negative than those of the other samples.

A sample combining several globular glass clasts and a sample comprising several felsic clasts from the Popigai tagamites were analyzed. The glass clasts sample possesses an  $\varepsilon$ <sup>(143</sup>Nd) value slightly more depleted, and an  $\varepsilon$ <sup>(87</sup>Sr) value slightly less enriched than the homogeneous Popigai tagamites. The substantial Rb content, coupled with the moderate Sr content, results in very high Rb/Sr and f(Rb/Sr) values, which are

not accompanied by extremely radiogenic  ${}^{87}$ Sr/ ${}^{86}$ Sr, and, therefore, the data yield a young T<sub>UR</sub> model age. This requires Rb–Sr remobilization at a time very much younger than the age of the parent material at the site of the impact. The felsic clast sample possesses a similar  $\varepsilon$ ( ${}^{143}$ Nd) value to that of the glass clast sample, and an  $\varepsilon$ ( ${}^{87}$ Sr) value slightly more enriched than the tagamites.

Melanocratic microkrystites from Sites 462 and 216 possess Nd model ages ( $T_{CHUR}$ ) of 1830–2100 Ma. By contrast, the sample of melanocratic fragments from Site 292 shows a much younger  $T_{CHUR}$  of 1120 Ma. This sample also displays a low f(Sm/Nd) value of -0.648. Site 315A also possesses a low f(Sm/Nd) value of -0.7305, as well as a  $T_{CHUR}$  of 1510 Ma, significantly younger than that of melanocratic microkrystites from Sites 216 and 462.

The leucocratic and colorless microkrystites from Site 216 possess  $T_{CHUR}\ of\ 1720$  and 1350 Ma. Leucocratic fragments from Site 292 possess  $T_{CHUR}$  of 2560 Ma. The Sr model ages ( $T_{UR}$ ) are generally much younger than the T<sub>CHUR</sub> ages. For cases where the chemical fractionation for Rb/Sr  $(f_{\rm Rb/Sr})$  is small, or the Sr is non-radiogenic, the T<sub>UR</sub> model ages are not well defined, e.g. sample 292M and Mistastin sample LM57-69 (cf. discussion in [32]). Consequently, although microtektites have been assigned by some workers to a particular strewn field on the basis of Sr isotopic data alone (e.g. Site 689B microtektites [43]), the variability of the T<sub>UR</sub> ages suggests that correlations based on Sm-Nd data may prove more reliable.

We have also analyzed a bulk sample of North American layer microtektite fragments from Site 612 (sample 612) to confirm whether the isotopic composition is indeed distinct (Table 2; Fig. 5a,b). The new data on a bulk sample are consistent with the previous results that have been determined for individual macroscopic tektites from the same site [32].

## 7. Discussion

The melanocratic microkrystites from Sites 462 and 216 are indistinguishable from one another

although they are from the Pacific and Indian oceans, respectively, as well as being from different biozones. They were clearly derived from the melting of target rocks with similar isotopic compositions. Melanocratic microkrystites from Site 292 and Site 315A (samples 292M and 315M) possess an isotopic signature distinct from those of the Site 216 and 462 microkrystite samples (216M and 462M), which implies their provenance either from contemporaneous, different impact structures or from a single impact structure with heterogeneous target rocks.

The differences in isotopic compositions between the leucocratic and melanocratic microkrystites (e.g. samples 216L and 216M) in the same biozone imply that, while they are found in a single layer, the target material must have been heterogeneous, consistent with the differences in their major element compositions (Fig. 4).

We now wish to compare these results with the isotopic composition of melt rocks located within the possible source impact structures. Melt rocks from the Wanapitei impact structure possess significantly less radiogenic Nd  $[\epsilon(^{143}Nd) = -31.4 \pm 0.4]$  and more radiogenic Sr  $[\epsilon(^{87}Sr) = 958.7 \pm 0.4$  to  $972.3 \pm 2.8]$  than any of the microkrystites analyzed here (Fig. 5A). Impact melt rocks from the Mistastin impact structure, in this study, exhibit similar  $\epsilon(^{143}Nd) = -26.5$  to -26.8 and  $\epsilon(^{87}Sr) = 42.0$  to 68.1, but appear distinct from the results of all the microkrystites.

Seven samples of the Popigai tagamites have been analyzed [42]. They define very narrow ranges of Nd and Sr compositions that are almost indistinguishable from those of the melanocratic microkrystites from the Indian and Pacific oceans at Sites 462 and 216 (Fig. 5a). This and the identical major element compositions of the melanocratic microkrystites and Popigai tagamites, strongly suggest Popigai as the source for the melanocratic microkrystites from these sites.

Three samples of the target rock gneisses at Popigai have also been analyzed [42]. These display a very large degree of isotopic variation and the data scatter widely about the isotopic compositions of the tagamites (Fig. 5a). The Popigai tagamites were considered to have been derived through the melting and homogenization of these isotopically diverse source rocks [42], though the isotopic composition and contribution of other rocks in the target region has not yet been assessed. The similarity of the composition of microkrystite samples 216M1, 216M2, 462M1 and 462M2 with the Popigai tagamites is consistent with these microkrystites having been derived from this homogenized tagamite melt. Although there is considerable variation in the  $\varepsilon$ <sup>(143</sup>Nd) and  $\varepsilon$ <sup>(87</sup>Sr) values of the nine samples analyzed from the microkrystite layer(s), the values do not closely coincide with impact melt rocks from the other possible source impact structures. Indeed, where the microkrystite samples do coincide with impact melt rocks in an upper Eocene impact structure, they coincide with the tagamites from Popigai. In addition, all the microkrystite samples analyzed plot within the three values of the basement gneisses to Popigai. Consequently, it is possible that the variation in the isotopic composition of the microkrystites could be the result of the melting and ejection of the isotopically heterogeneous basement rocks at Popigai. The differences between the Popigai tagamites and the glass and felsic clast samples that were derived from the target rocks provides direct evidence for a significant degree of preserved isotopic heterogeneity. The Proterozoic Sm-Nd model ages of the microkrystites are consistent with microkrystites from all sites having been derived by impact melting of the Archean to Proterozoic basement rocks to Popigai. An origin of the microkrystites from the Chesapeake impact structure can be ruled out, since the Sm-Nd model ages of all of the microkrystites are much older than the model ages of rocks from the eastern seaboard of the USA and the North American tektites. Similarly, a Wanapitei or Mistastin source can be dismissed on the basis of the microkrystites possessing clearly distinct  $\varepsilon$ <sup>(143</sup>Nd) and  $\varepsilon$ <sup>(87</sup>Sr) values and major element compositions from the impact melts from the Wanapitei [39] or Mistastin impact structures (Fig. 5). We have no isotopic data for the basement rocks or impact melts to the 17 km diameter Logoisk impact structure in Belarus, so we cannot exclude it as a potential source for the microkrystites, on this basis. However, the dissimilar major element compositions of the impact melt rocks



Fig. 6. A backscattered electron image of a microkrystite from Site 216 which contains crystallites of diopside. The black regions of the crystallites represent areas from which the crystallites have been removed by dissolution.

and microkrystites do not support a Logoisk provenance (Fig. 4). In addition, the smaller size of this structure is less likely to have generated the observed, volumetrically significant quantities of impact ejecta distributed up to 130° west and 45–50° south of this potential source structure. Despite the range in the  $\varepsilon$ <sup>(143</sup>Nd) and  $\varepsilon$ <sup>(87</sup>Sr) values for the microkrystites, no samples possess signatures that overlap with impact melt rocks from other possible source impact structures considered.

The Rb-Sr model ages (T<sub>UR</sub>) of the microkrystites are lower than those for the Popigai tagamites (Fig. 5b). A lowering of the Rb-Sr model ages is simple to explain for sedimentary rocks, namely, by increasing Rb concentrations relative to Sr due to the growth of clay minerals. However, the basement rocks from which we propose the bulk Popigai tagamites and microkrystites were derived are gneisses, located beneath an unconformable Paleozoic sequence. The gneisses are not heavily weathered, nor are extensive basal conglomerates present at the unconformity (V.L. Masaitis, personal communication, 2000). Thus, clay formation prior to the impact event is not likely the cause of lowered Rb-Sr model ages. At this time, there are no isotopic data on the sedimentary rock cover to the gneisses. This cover includes a very wide array of rock types, e.g. Triassic basalts and basaltic tuffs, limestones, dolomites, sandstones and siltstones. If the younger  $T_{UR}$  model ages of the melanocratic microkrystites relative to those for the tagamites are caused by differential incorporation and ejection of melts derived from the sedimentary cover, then we would require that the source regions of the sedimentary cover have the same crustal formation age (e.g. Proterozoic  $T_{CHUR}$  model ages) as the basement source rocks of the tagamites. The clarification of the process responsible for lowering the Rb–Sr model ages for the melanocratic microkrystites, if they are derived from the Popigai impact structure, must await elemental and isotopic data for the sedimentary cover.

The Rb/Sr ratio can also be increased by alteration of the microkrystites. Many of the microkrystites contain void space (Fig. 6). The extent to which this process can account for the observed younger Rb-Sr model ages remains to be investigated.

The two microkrystite horizons from Site 462 were sampled across a 9–13 cm zone straddling two sequential core sections from the same hole. This break in the core does not allow positive stratigraphic distinction of the two samples, and it is, therefore, equivocal whether the microkrystites were derived from one or two different source impacts [44]. The two samples possess similar isotopic compositions whose spread is comparable to that expected for analytical precision alone. This evidence suggests that they were derived from a common target and, thus, they do not represent two temporally or spatially discrete impact events. This is consistent with their similar major element compositions.

Sample 292M is from the same biozone (*Po. semiinvoluta*) as samples 216M1 and 216M2, despite its different isotopic signature. This suggests that either the sample 292M microkrystites were derived from an impact other than Popigai, Chesapeake, Mistastin and Wanapitei, that also fortuitously occurred during the deposition of the *Po. semiinvoluta*-bearing sediment, or that the target rocks that were melted, ejected and deposited at Sites 216 and 292 possess a variable isotopic composition. We propose that the variability in the melanocratic microkrystite isotopic

signatures probably represents the preservation of evidence of an isotopically heterogeneous target, despite their having been generated from a single impact at Popigai. It should be noted that although the isotopic compositions of many tektite strewn fields are limited, the North American microtektites and tektites generated by the Chesapeake impact event, also display a wide variation, particularly in their  $\varepsilon$ <sup>(87</sup>Sr) values [32,33,41].

The variable isotopic signatures of the melanocratic microkrystites do not clearly resolve whether the microkrystites sampled from the Po. semiinvoluta and T. cunialensis Biozones are from the same or different source impact structures. However, contrary to earlier studies [14,22] we find no statistical difference between the major element composition of the microkrystites from the two biozones. Our observation is consistent with the assertion of Glass and Burns [15], that the microkrystites from the semiinvoluta and cunialensis Biozones most probably represent a single impact event, older than the impact that produced the North American layer. Assuming that the biozonation is correct, the occurrence of ejecta derived from a single impact in two separate biozones indicates that the first appearance of the index fossil on which the biozonation is based (T. cunialensis) was not globally synchronous. Indeed, there is evidence to suggest that this biozone boundary could be diachronous by up to 0.7 Ma [26]. The simultaneous deposition of ejecta across the oceans may, thus, result in the deposition of microkrystites in different biozones.

The leucocratic microkrystites were probably derived through bulk melting of target rocks with a different major element and isotopic composition to those rocks that formed the melanocratic microkrystites. The distinct isotopic signature of the leucocratic microkrystites has a number of possible origins. The leucocratic microkrystites might simply represent melting of the isotopically diverse basement rocks (Fig. 5), which were ejected prior to the major melting and homogenization of the tagamites. Alternatively, the leucocratic microkrystites could have been derived by melting of the supra-basement Cambrian to Triassic rocks at Popigai, which have a different isotopic signature to the basement as they have a sedimentary provenance outside the Anabar basement gneiss massif (V.L. Masaitis, personal communication, 1998). In this context, the wide range of Sr compositions may reflect melting of sediments with a high isotopic diversity, owing to variable Rb/Sr enrichment by sedimentary processes. Variation in the microkrystite isotopic signatures could also be the result of the assimilation of various quantities of target region Triassic basaltic rocks and upper Proterozoic to Cambrian limestones and dolomites in the target region.

Finally, the basement gneisses at Popigai were covered by up to 1 km of sedimentary rocks at the time of impact. Melanocratic microkrystites derived by melting of this basement indicate excavation and ejection from >1 km depth beneath the Eocene land surface. This is unlike the so-called 'Moldavite' tektites that originated from the 15 Ma old, 24 km diameter Ries crater in Germany, which appear to have been derived solely through melting and jetting of only the uppermost 30 m of surficial Tertiary sediment [45,46]. Excavation and ejection models for melt removal from the Popigai transient cavity, which assume a similar Ries-type surficial source (e.g. [47]), will have to be modified to accommodate the deeper source depth at Popigai.

### 8. Summary

Evidence has been presented in support of a geochemical link between microkrystites from ODP/DSDP marine cores and a potential source impact structure. Our isotopic and geochemical data indicate, for the first time, that the microkrystites have a provenance from the Popigai impact structure of Siberia, while negating other broadly contemporaneous impact structures as their source. The Site 216 and Site 462 melanocratic microkrystites from the Indian and Pacific oceans, respectively, possess isotopic signatures most similar to the bulk, homogenized tagamites at Popigai. Other samples possess major element compositions similar to Popigai tagamites, but display a range of isotopic signatures. This range may be due to the isotopic diversity of the source region, coupled with a lack of isotopic homogenization of the melts during the impact process. The isotopic diversity is revealed by analyses of basement gneisses from Popigai, while the lack of homogenization during the impact process is supported by isotope data on globular glass clasts and felsic inclusions extracted from the Popigai tagamites.

The highly variable isotope signatures of microkrystites from the *semiinvoluta* Biozone at Sites 216 and 292 suggest that distinguishing whether the microkrystites from the T. cunialensis and Po. semiinvoluta Biozones are from a single or multiple source is not possible using these isotope data. However, the major element data indicate that they are statistically indistinguishable. The isotopic signatures of the leucocratic and clear microkrystites are different from the signatures of homogenized Popigai tagamites and the ejected homogenized melts at Sites 216 and 462. The leucocratic microkrystites may represent melts of the Phanerozoic cover to the Archean and Proterozoic basement or basement melts that were ejected prior to homogenization of the tagamite melts.

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