North Siberian Lakes: A Methane Source Fueled by Pleistocene Carbon

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The sizes of major sources and sinks of atmospheric methane (CH_4), an important greenhouse gas, are poorly known. CH_4 from north Siberian lakes contributes ~1.5 teragrams CH_4 $year^{-1}$ to observed winter increases in atmospheric CH_4 concentration at high northern latitudes. CH_4 emitted from these lakes in winter had a radiocarbon age of 27,200 years and was derived largely from Pleistocene-aged carbon.

The highest concentration and greatest seasonal amplitude of atmospheric CH_4 occurs at 65° to 70°N. Concentrations are highest in March to April and lowest in summer (*1*). Photochemical oxidation of CH_4 contributes to the low summer levels (*2*) but does not explain why the seasonal amplitude of atmospheric CH_4 is twice as high in the Northern as in the Southern Hemisphere, given large summer effluxes from North American bogs and tundra (*3*, *4*) and modest CH_4 fluxes from Siberian wetlands (*5*). Between August and April, 5.8 Tg (1 Tg = 10^{12} g) of CH_4 accumulate in the atmosphere north of 60°N (*6*). High-latitude winter fluxes measured in a muskeg and a peatland were only 10 to 12% of the annual total (*4*, *7*), an insufficient flux to explain a winter maximum in atmospheric CH_4. Here we provide evidence for a large winter CH_4 source from Siberian lakes.

In the Pleistocene, most of the northern Siberian plains were unglaciated and accumulated ~400,000 Tg of organic C in sediments (*8*) (mainly derived from plant roots), similar to the total C in the terrestrial biosphere (*9*). These sediments contained abundant ice (40 to 70% of soil volume) (*10*–*12*), which began melting during the Holocene to form thermokarst (thaw) lakes that now make up ~30% of the landscape. These lakes migrated across the north Siberian plains during the Holocene (*10*), releasing to the atmosphere an average of 170 to 220 g C m^{-2} $year^{-1}$, including ~16 g CH_4 m^{-2} $year^{-1}$; we estimate that half of this CH_4 was derived from Pleistocene C (*13*). Siberian lake sediments produce CH_4 bubbles in lakes throughout the year (*14*), particularly near shores with active erosion. During winter, the bubbles form koshkas, which are flat bubbles of CH_4 in lake ice separated by ice films that periodically sublimate and release CH_4 to the atmosphere. In areas where CH_4 ebullition (bubbling) is most active, channels through the ice remain open all winter.

To evaluate the significance of this source, we incubated Pleistocene sediments from an eroding lakeshore with lake water. The yield was 65 ± 3 mg CH_4 g^{-1} sediment at 15°C (mean ± SE, n = 3) over 12 months, equivalent to 5% of the C originally present in the soil; 26 ± 2 mg CH_4 g^{-1} were emitted at 3.5°C, and 19 ± 2 mg CH_4 g^{-1} were emitted at 0°C. These data indicate that the C in Pleistocene sediments is sufficiently labile to support methanogenesis and that, although methanogenesis is temperature-sensitive, it occurs at substantial rates at 0° to 3.5°C.

To determine whether methanogenesis in lake sediments is currently fueled by Pleistocene-aged organic matter, we measured stable and radiocarbon isotopes of CH_4 emitted by ebullition from two representative thaw lakes near Cherskii, Republic of Sakha (Yakutia), Russia (69°N, 161°E). CH_4 collected from these lakes in winter (April) had an average ^{14}C age of 27,200 years (Table 1). This age indicates that Pleistocene sediments deposited 20,000 to 40,000 ^{14}C years ago (*11*) contributed 68 to 100% of CH_4 flux from these lakes. In contrast, CH_4 emitted in the summer (July) had an average ^{14}C age of 9,200 years, indicating that Pleistocene C fueled 23 to 46% of summer methanogenesis and thus that more CH_4 was produced in the younger surface sediments, which are warmer in summer than winter (*10*). Thus, about half of current annual methanogenesis is fueled by Pleistocene C. In contrast, CH_4 from Alaskan lakes was only 200 years old (*15*) because Alaska lacks extensive Pleistocene sediments.

The $\delta^{13}C$ value of CH_4 collected from Siberian lakes was −71 to −73 (Table 1). This value is less than that produced in summer by Alaskan tundra lakes ($\delta^{13}C$ = −61 ± 2) (*15*) or North American wet tundra ($\delta^{13}C$ = −66 to −63) (*15*, *16*). These values imply that the Siberian winter-collected CH_4 was not as oxidized as in these other environments, or that there was an isotopic difference in substrate or a different pathway of methanogenesis (*17*). The hydrogen isotopic composition of the CH_4 was variable, but most samples from the Siberian lakes were low (δD = −370), indicative of a biotic source for CH_4, low oxidation rates in the water column, and CH_4 production by fermentation (*17*, *18*).

We measured CH_4 ebullition fluxes from two thaw lakes using large funnels suspended beneath the ice (*19*). CH_4 fluxes were generally highest from October to January (Fig. 1), when deep sediments had their annual thermal maximum (*10*). Fluxes were highly variable within a season; fluxes were highest at

Table 1. Isotopic data for CH_4 collected from sediments in two thaw lakes in Pleistocene sediments in the Kolyma lowlands. Results are given as percent modern C, or 100 times the ratio of $^{14}C/^{12}C$ in the sample divided by the $^{14}C/^{12}C$ ratio in 1895 wood (corrected for $^{13}C/^{12}C$ differences) (*27*, *28*).

Lake no.	Lake depth (m)	$\delta^{13}C$	$^{14}CH_4$ age	Modern C (% of CH_4-C)	δD
			Summer		
13	5	−69.5	*	*	
	7	−69.6	11,731 ± 360	23.2 ± 1.0	
	10	−71.8	8,330 ± 240	35.3 ± 1.0	
	10†	−72.1	8,370 ± 180	35.1 ± 0.8	
	10†	–	8,330 ± 100	35.3 ± 0.4	
Average:		−70.8 ± 0.7	9,200 ± 800	32 ± 3	
			Winter		
13	2	−75.1	*	*	−169
	10	−80.0	38,000	0.2	−430
14	4†	−72.7	28,670 ± 850	2.8	−479
	4†	–	27,230 ± 730	3.4	–
	10	−65.3	15,000 ± 150	15.3	−391
Average:		−73.3 ± 3.1	27,200 ± 4700	5 ± 3	−370 ± 70

*Not determined. †Subsamples analyzed separately.

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Table 2. Summary of CH_4 fluxes from Siberian lakes during times of increasing (August to April) and decreasing (May to July) atmospheric CH_4, based on values presented in the text.

Flux component	Area (% of lake)	Area-averaged CH_4 flux (g CH_4 m^{-2} $year^{-1}$)		
		May–July	August–April	Annual
Ebullition flux				
Lake center*	80	0.3	1.0	1.3
Lake shore*	20	1.0	3.1	4.1
Overflow	100	0	0.5	0.5
Diffusion	100	0.5	0.4	0.9
Lake total (g CH_4 m^{-2} $year^{-1}$)		1.8	5.0	6.8
Siberian total (Tg CH_4 $year^{-1}$)		0.5	1.5	2.0

*Averaged over the entire lake area. Ebullition flux near the lake shore is a conservative estimate that does not include CH_4 released through koshkas.

times of low atmospheric pressure, as in north temperate lakes (*20*). The average CH_4 ebullition flux in centers of lakes (80% of the lake area) was 4.7 ± 2.3 mg CH_4 m^{-2} day^{-1} (90 measurements). Near eroding lake shores, fluxes were so high that they frequently overturned the collection funnels; the flux (2 cm^3 min^{-1}, n = 25; 90% CH_4) emitted from open holes in the ice (300 ha^{-1}) was 56 mg CH_4 m^{-2} day^{-1}. In addition to open holes, there were koshkas (400 to 1000 ha^{-1}) containing 1 to 100 liters of 50% (25 to 75%) CH_4 (n = 8) that vent CH_4 several times each winter and provide an additional unquantified CH_4 source. Sediments in these lakes released 22 g CH_4 m^{-2} (38 ± 12 liter m^{-2} of 80% CH_4, n = 3) in September, when we disturbed the sediments (*15*), but <5 g CH_4 m^{-2} a month later. This large CH_4 release over 1 month provides independent evidence for a large CH_4 ebullition flux.

CH_4 can also move to the atmosphere in winter in overflow, when the weight of winter snow pushes the ice below the equilibrium water level of the lake. The CH_4 concentration in overflow water decreases from 1.7 mg CH_4 $liter^{-1}$ (*21*) to <0.01 mg CH_4 $liter^{-1}$. The 30 cm of overflow that typically accumulate on lakes of the forest zone would thus release 0.52 g of dissolved CH_4 m^{-2} $year^{-1}$.

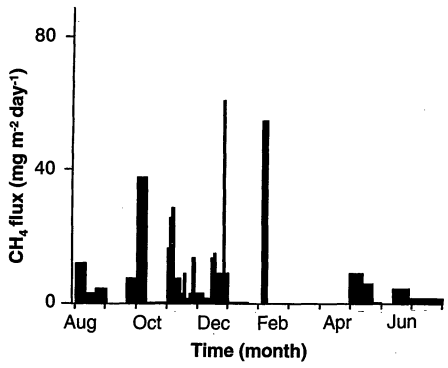

Fig. 1. Time course of CH_4 flux from the center of a thaw lake near the forest-tundra border 100 km south of the Arctic Ocean near the Kolyma River Valley (69°N, 161°W) in northern Siberia (*19*).

The average summer diffusive flux measured in 19 lakes along a climate transect inland from the Arctic Ocean was 7.6 ± 1.4 mg CH_4 m^{-2} day^{-1} (60 measurements) (*21*), a value similar to that in Alaskan lakes (6.8 ± 1.3 mg m^{-2} day^{-1}) (*22*). The 19 lakes had 3.1 ± 0.7 mg m^{-2} of dissolved CH_4 in March, indicating that winter accumulation of CH_4 is typical of north Siberian lakes.

We estimate the total annual flux of CH_4 for the lakes in our study region to be at least 7 g CH_4 m^{-2} $year^{-1}$ (Table 2), ~50% of the potential flux we estimated (16 g CH_4 m^{-2} $year^{-1}$) from regional C inputs to lakes. Approximately 75% of this flux occurs in winter. If these fluxes are typical of Siberian lakes, these lakes would emit ~1.5 Tg CH_4 in winter (2 Tg CH_4 annually). This is small relative to global sources (*18*) but is 25% of the high-latitude winter accumulation of CH_4 in the atmosphere. If high-latitude warming trends (*23*) continue, thawing of permafrost would increase, and methane flux from Siberian thaw lakes would act as a positive feedback to climate warming.

REFERENCES AND NOTES

1. I. Fung *et al.*, *J. Geophys. Res.* **96**, 13033 (1991).
2. M. A. K. Khalil and R. A. Rasmussen, *ibid.* **88**, 5131 (1983).
3. W. S. Reeburgh and S. C. Whalen, *Ecol. Bull.* **42**, 62 (1992).
4. S. C. Whalen and W. S. Reeburgh, *Global Biogeochem. Cycles* **6**, 139 (1992); *ibid.* **2**, 399 (1988).
5. V. A. Samarkin, E. M. Rivkina, E. A. Bocharnikova, in *Methane Biogeochemistry in Arctic Marsh Soils*, D. A. Gilichinsky, Ed. (Russian Academy of Sciences, Pushchino, Russia, 1992); T. R. Christensen, S. Jonasson, T. V. Callaghan, M. Havström, *J. Geophys. Res.* **100D**, 21035 (1995).
6. The atmosphere north of 60°N has about 380 Tg CH_4 [8% of the total global atmospheric CH_4 pool of 4800 Tg (*18*)] and increases during winter by 1.5% (25 ppb) (*1*), amounting to 5.8 Tg.
7. N. B. Dise, *Biogeochemistry* **17**, 71 (1992).
8. We estimate that 400,000 Tg C accumulated in Pleistocene loess from soil volume (1 × 10^6 km^2, 40 m depth) (*10*, *11*) and C density (1 g cm^{-3} of soil × 1.08 ± 0.04% organic C; n = 46 sites throughout Siberia; 0 to 50 m depth) (*12*).
9. D. S. Schimel, *Global Change Biol.* **1**, 77 (1995).
10. S. V. Tomirdiaro, *Lessovo-ledovaia formatsia Vostochnoi Sibiri v pozdnem pleistotsene i golotsene* (Nauka, Moscow, 1980).
11. Y. K. Vasil'chuk, *Izotopno kislorodnyi sostav podzemnykh l'dov* (Soveta po kriologii zemli Rosiiskoi Akademii Nauk i Severo-Vostochnogo nauchno proizvodstvennogo Kompleksa Ekotsentr, Moscow, 1992), vol. 1.
12. S. V. Gubin, *Paleograficheskie aspekty pochvoobrazovaniia na primorskoi nizmennosti severa lakutii* (Akademiia Nauk, Institut Pochvovedeniia i Fotosintesisa, Pushchino, Russia, 1987).
13. We estimate 120 g C m^{-2} $year^{-1}$ input of Pleistocene C to lakes, assuming that 400 kg C m^{-2} were initially in Pleistocene sediments (*8*) and that 0.3 × 10^6 km^2 of lakes (*24*) migrated across 90% of the plains during the last 10,000 years (*10*). We estimate Holocene C input to lakes assuming that the erosion rate for lakes 1 km^2 in area is 0.5 to 1.0 m $year^{-1}$ (*10*) (that is, 0.05 to 0.1% of lake area) and that the lakes receive 100 kg m^{-2} organic C from vegetation (3 to 10 kg m^{-2}), peat (0 to 200 kg m^{-2}), soil (7 to 30 kg m^{-2}), and upper 3 m of permafrost (20 to 60 kg m^{-2}) (*10*, *11*, *25*) = 50 to 100 g C m^{-2} of lake area. We do not know the contribution of in-lake production plus dissolved organic C inputs to CH_4 fluxes, so we used values measured in Alaskan oligotrophic tundra lakes lacking major erosional inputs: 0.7 g CH_4 m^{-2} $year^{-1}$ (*22*). If 5 to 10% of this C input were converted to CH_4 in anaerobic sediments (*26*), this would yield an annual flux of ~16 (9 to 23) g CH_4 m^{-2} $year^{-1}$.
14. S. A. Zimov and S. M. Govorushko, *Mate. Glatsiol. Issled.* **37** (1979).
15. C. S. Martens, C. A. Kelley, J. P. Chanton, *J. Geophys. Res.* **97D**, 16689 (1992).
16. P. D. Quay, S. L. King, J. M. Lansdown, W. O. Wilber, *Global Biogeochem. Cycles* **2**, 385 (1988); M. Wahlen *et al.*, *Science* **245**, 286 (1989).
17. M. F. Whiticar, E. Faber, M. Schoell, *Geochim. Cosmochim. Acta* **50**, 693 (1986).
18. R. J. Cicerone and R. S. Oremland, *Global Biogeochem. Cycles* **2**, 299 (1988).
19. We measured CH_4 flux in conical 8-m^2 plastic traps anchored beneath the ice surface, which funneled CH_4 bubbles into collection flasks. Samples were collected every 0.5 to 10 days, depending on flux. Fluxes from seven other chambers in two lakes were similar to those shown but gave incomplete seasonal data due to winter ice damage. Water depth was 10 m. CH_4 content of recently produced bubbles was >80% CH_4, as determined (±1% accuracy) by gas chromatography (GC). Each sample was analyzed on both a TSVET-530 and a Shimadzu 14A GC with a thermal conductivity detector and a flame ionization detector, respectively. We used data from the thermal conductivity detector if CH_4 concentration was >1%; otherwise, we used data from the flame ionization detector. By running all samples through both detectors, we cross-calibrated the two analytical methods. CH_4 concentration of samples that remained in collection flasks >24 hours declined due to diffusion into the water column. In these cases, we estimated CH_4 content as 0.8 × gas volume.
20. M. D. Mattson and G. E. Likens, *Nature* **347**, 718 (1990).
21. We sampled 19 lakes in June, August, September, and March along a climate gradient (4° to 12°C July mean temperature) from the Arctic Ocean to boreal forest (68° to 70°N) in the Kolyma Lowland for dissolved CH_4 at 1-m-depth intervals through the water column. Lake depth and sediment types were representative of north Siberian lakes. Samples were analyzed for CH_4 within 24 hours (*19*). We used the boundary-layer model and meteorological assumptions of Kling *et al.* (*22*) to estimate diffusive flux during ice-free months when atmospheric CH_4 was increasing (August to September) or decreasing (June to July) based on measurements of surface-dissolved CH_4 concentration in June, August, and September. Data are available to *Science* Online subscribers at www.sciencemag.org.
22. G. W. Kling, G. W. Kipphut, M. C. Miller, *Hydrobiologia* **240**, 23 (1992).
23. W. L. Chapman and J. E. Walsh, *Bull. Am. Meteorol. Soc.* **74**, 33 (1993); K. R. Briffa, P. D. Jones, F. H. Schweingruber, S. G. Shiyatov, E. R. Cook, *Nature* **376**, 156 (1995).
24. S. E. Mostakhov, in *Mezhdunarodnaia conferentsiia po merzlotovedeniiu, Podzemnye vody kriolitosfery*, N. I. Tolstikhin, Ed. (Iakutskoe Knizhnoe Izdatel'stvo, Iakutsk, Russia, 1973), vol. 5.
25. K. Van Cleve and V. Alexander, *Ecol. Bull.* **33**, 375 (1981); E. Gorham, *Ecol. Appl.* **1**, 182 (1991); G. L.

Michaelson, C.-L. Ping, J. M. Kimble, *Arct. Alp. Res.* **28**, 414 (1996).
26. J. D. Hamilton, C. A. Kelly, J. W. M. Rudd, R. H. Hesslein, N. T. Roulet, *J. Geophys. Res.* **99D**, 1495 (1994).
27. M. Stuiver and H. Polach, *Radiocarbon* **19**, 355 (1977).
28. Samples were collected directly into glass serum vials from ebullition flux (*15*). Before analysis, water was added to vials to maintain high internal pressures, the sample was shaken vigorously to get dissolved CH_4 out of solution, each sample was transferred to a 6-liter stainless steel container with a syringe, and analytical grade-zero air was added. CH_4 was separated from each sample and combusted. A subsample of the resulting CO_2 was analyzed for $^{13}C/^{12}C$ by mass spectrometry, and the remaining CO_2 was catalytically reduced to graphite (*29*), and its $^{14}C/^{13}C$ values measured by atomic mass spectrometry at the Lawrence Livermore National Laboratory.
29. S. C. Tyler, *J. Geophys. Res.* **91**, 13232 (1986); D. C. Lowe, C. A. Brenninkmeijer, S. C. Tyler, E. J. Dlugkencky, *ibid.* **96**, 15455 (1991); J. S. Vogel, *Radiocarbon* **34**, 344 (1992).
30. We thank G. W. Kling, W. S. Reeburgh, M. S. Torn, and M. Wahlen for constructive criticism and the Russian Fund for Fundamental Research, the International Science Foundation, and U.S. Department of Energy for financial support.

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Sizes and Ages of Seamounts Using Remote Sensing: Implications for Intraplate Volcanism

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Satellite altimetry was used to identify and characterize Pacific intraplate seamounts. The gravimetric amplitudes of seamounts appear to be related to the age difference between the sea floor and seamounts; by inverting this relation, pseudo ages can be obtained for undated seamounts. These pseudo ages imply that excursions in seamount volcanism generally correlate with times of formation of large oceanic plateaus.

The Pacific plate may support more than 50,000 seamounts taller than 1 km, yet ~50% of these undersea volcanoes are uncharted because of sparse bathymetric coverage (*1*, *2*). Even fewer (<1%) have been sampled for radiometric dating (*3*), making assessment of temporal fluctuations in intraplate volcanism uncertain. Because electromagnetic sensing devices cannot penetrate the oceans, we are unable to image the sea floor remotely and instead must rely on surface ships equipped with sonar. At the present rate of data acquisition, complete bathymetric coverage may take centuries. However, the density contrast between seawater and the sea floor basalt gives rise to gravity anomalies. These minute variations in Earth's gravitational pull cause seawater to be attracted to seamounts, leading to a sea surface (which approximates the geoid) whose shape reflects these underlying features (*4*). Thus, since the early 1980s, satellite altimetry has provided broad coverage of the sea surface or geoid undulations (*5*). Early attempts to map the seamount distribution were largely limited by the coarseness of the satellite coverage [the typical track spacing was >100 km (*6*)], and many seamounts went undetected. Because seamounts are typically much smaller than 100 km, it was difficult to estimate what part of the seamount had been traversed by the satellite, leading to large uncertainties in estimates of seamount height and diameter (*7*).

Recently, the U.S. Navy declassified its Geosat satellite altimetry, which has been combined with the European Space Agency

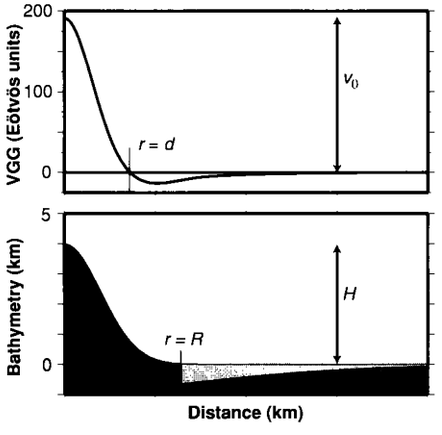

Fig. 1. Theoretical VGG over an isostatically compensated seamount of radius R and height H (*14*). The amplitude v_0 and zero-crossing distance d are the two clearest characteristics of the anomaly.

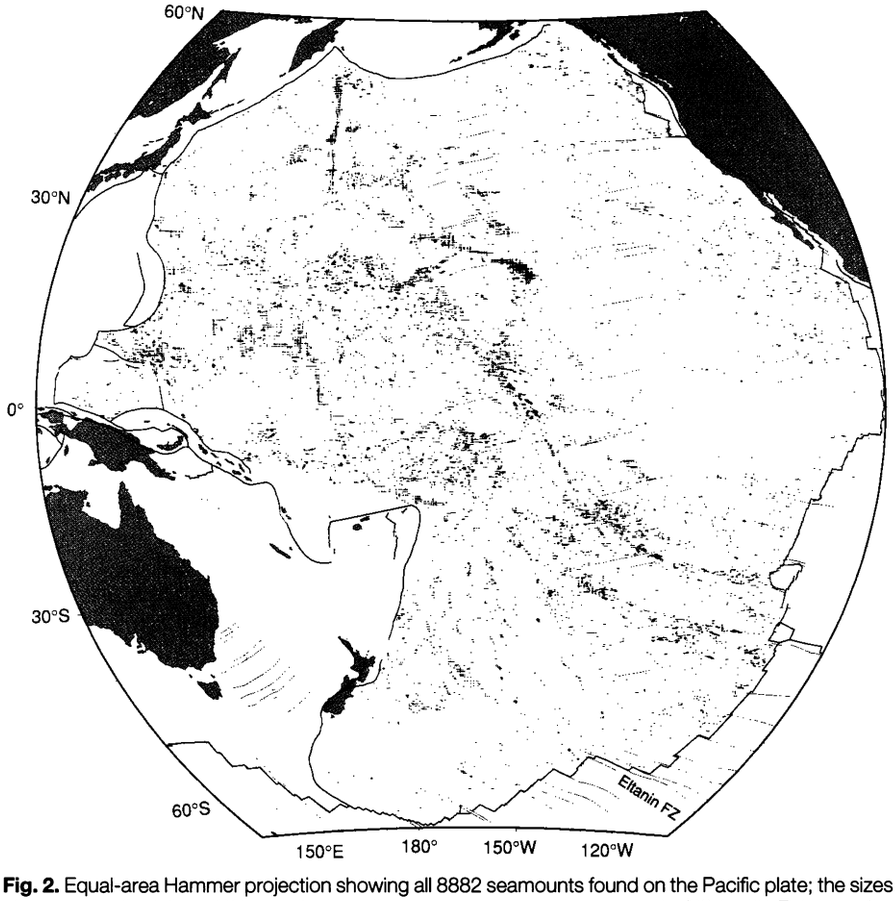

Fig. 2. Equal-area Hammer projection showing all 8882 seamounts found on the Pacific plate; the sizes of crosses reflect the VGG amplitudes. Blue crosses are small seamounts (30 to 60 Eötvös units, generally <2.5 km tall), red crosses are large seamounts (>120 Eötvös units, generally >3.5 km tall), and green crosses are of intermediate size. The Eltanin fracture zone is indicated.