A 220 kyr record of Pb isotopes at Dome C Antarctica from analyses of the EPICA ice core

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[1] Pb isotopic compositions and Pb and Ba concentrations are reported in EPICA Dome C ice core samples dating to 220 kyr BP, indicating that Pb isotopic compositions in Antarctic ice vary with changing climate. $^{206}Pb/^{207}Pb$ ratios decrease during glacial periods, with the lowest values occurring during colder climatic periods (stages 2, 4 and 6) and the Holocene. Low Pb concentrations (<1 pg/g) were found during the Holocene and the last interglacial (climate stage 5.5) while higher Pb concentrations (>10 pg/g) were found during cold climatic periods. Ba, a proxy for mineral dust, was used to determine that dust usually accounts for ~70% of Pb in Dome C ice, while the remaining ~30% was attributed to volcanic emissions. Pb isotopic compositions at Dome C differ from those reported in pre-industrial ice from other Antarctic locations, due to greater proportions of dust Pb at Dome C. **Citation:** Vallelonga, P., P. Gabrielli, K. J. R. Rosman, C. Barbante, and C. F. Boutron (2005), A 220 kyr record of Pb isotopes at Dome C Antarctica from analyses of the EPICA ice core, *Geophys. Res. Lett.*, *32*, L01706, doi:10.1029/2004GL021449.

1. Introduction

[2] The deep ice cores from East Antarctica have provided an insight into climatic conditions and aerosol fallout fluxes and provenance during the Holocene and previous glacial and interglacial periods [*EPICA Community Members*, 2004]. High Pb concentrations [*Hong et al.*, 2003, and references therein] observed during cold climate stages 2, 4 and 6 in the Vostok [*Petit et al.*, 1999] and "old" (1978) Dome C [*Lorius et al.*, 1979] ice cores were attributed to increased dust levels, which have been determined to most probably originate from the loess deposits of South America [*Delmonte et al.*, 2004].

[3] *Rosman et al.* [1999] reported Pb isotopic compositions in the earlier Dome C ice core and noted that Holocene Pb isotopic compositions were more-radiogenic (higher $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios) than those of the last glacial maximum (LGM), suggesting that Pb isotope ratios in Antarctic ice may be affected by changing climate. Recent analyses of Pb isotopes at coastal locations in Antarctica have determined pre-industrial Holocene Pb isotopic signatures [*Matsumoto and Hinkley*, 2001; *Planchon et al.*, 2003; *Vallelonga et al.*, 2002b] and revealed that proportions of dust and volcanic Pb vary across Antarctica. The provenance of Antarctic dust (by Pb isotopes) is restricted by a dearth of Pb isotope data representing dust sources in the Southern Hemisphere.

[4] We present the first Pb isotopic compositions from the EPICA Dome C ice core and the longest Pb isotopic record available from an ice core. These data[1] allow variations in Pb deposition and dust Pb content at Dome C to be evaluated for the past two glacial cycles.

2. Methods

[5] Thirty samples have been analysed from 26 sections of the EPICA ice cores drilled at Dome C (75°06′S, 123°21′E; 3233 m asl; mean temperature −54°C, Figure 1). From the surface to 763 m depth, samples were from the EDC96 core (drilled 1996–1999), while deeper samples were from another EPICA Dome C core (drilled 1999–2003). Sections analysed were from depths of 229 to 2193 m, corresponding to the period 6.9 to 217 kyr BP using the EDC2 timescale [*EPICA Community Members*, 2004]. Core sections were typically 55 cm long and 5 cm in radius. About 30% of the cross-section of each core section (cut longitudinally) was available for Pb isotope analyses. While two samples were usually produced from a 55 cm core section, a preliminary data set of thirty samples is presented here.

[6] Samples were prepared (decontaminated) at the Laboratory of Glaciology and Geophysics of the Environment in Grenoble, France, in a clean bench supplied with High Efficiency Particulate (HEPA) filtered air and located within a cold room (Temperature −15°C). The decontamination procedure [*Candelone et al.*, 1994; *Gabrielli et al.*, 2004] involved the chiselling of concentric layers of an ice core to remove external contamination introduced during drilling, transport and/or storage. Decontaminated samples were melted and aliquotted into LDPE bottles that were then sent, frozen, to Curtin University in Perth, Australia, for Pb and Ba analysis.

[7] The preparation and analysis of ice core samples by thermal ionization mass spectrometry has been described in detail by *Vallelonga et al.* [2002a, and references therein]. Samples were melted and ~5 g was transferred to a Teflon beaker. HNO_3, HF, H_3PO_4 and an isotopically enriched tracer solution (containing ^{205}Pb and ^{137}Ba for isotope dilution) were added before the sample was evaporated to dryness. The sample was then transferred in 4 μl of silica gel to a zone-

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[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2004GL021449.

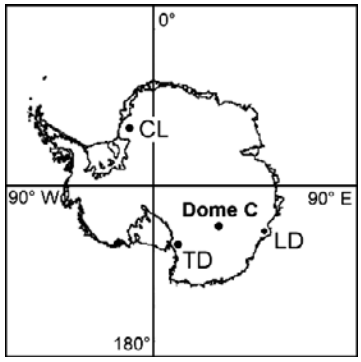

Figure 1. Map of Antarctica indicating the locations of Dome C and other sites for which Pb isotope data have been reported: Law Dome (LD; 66°46′S, 112°48′E), Taylor Dome (TD; 77°48′S, 158°43′E) and Coats Land (CL; 77°34′S, 25°22′W).

refined rhenium filament and again evaporated to dryness. Samples were then loaded into the mass spectrometer.

[8] Samples were analysed using a thermal ionization mass spectrometer (model VG354, Fisons Instruments). All Pb isotopes (^{204}Pb, ^{205}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb) were measured using a Daly detector, with ~70 isotope ratios collected per sample. ^{205}Pb, which is not present in terrestrial Pb, was used as an isotopic tracer as it enabled the determination of Pb concentrations and Pb isotope ratios in one analysis. Ba can be used as a proxy for mineral dust because it is much more abundant in Earths' crust compared to volcanic emissions [*Matsumoto and Hinkley*, 2001]. All Pb/Ba ratios were calculated by weight.

[9] The accuracy of the reported Pb and Ba concentrations is estimated to be ±15% (95% confidence interval). This relatively large uncertainty (for isotope dilution) is a result of the ±7% (2σ) precision to which μL-quantities of the tracer solution could be dispensed. The accuracy of the isotope ratio measurements was monitored by routine analysis of a 200 pg sample of Standard Reference Material 981 (NIST, Gaithersburg, USA). A correction for isotopic fractionation of 0.24 ± 0.06 % per mass unit was applied to the measured ratios. Pb isotope ratio uncertainties (Table 1) account for the sampling and storage of ice cores as well as sample analysis.

3. Results and Discussion

[10] Pb and Ba concentrations in ice from the EPICA Dome C core have varied by almost two orders of magnitude over the past 220 kyr with the greatest concentrations constrained to cold climatic periods (Table 1). Lower Pb and Ba concentrations were observed during the Holocene (0.4 pg Pb/g, 11 pg Ba/g) and the last interglacial (0.5 pg Pb/g, 14 pg Ba/g), while higher Pb and Ba concentrations occurred during climate stages 2 (LGM, 14 pg Pb/g, 380 pg Ba/g), 4 (63 kyr BP, 12 pg Pb/g, 290 pg Ba/g) and 6 (the penultimate glacial maximum, 11 pg Pb/g, 240 pg Ba/g), as shown in Figure 2. The results presented here agree with those reported in other Antarctic ice cores [*Hong et al.*, 2003, and references therein], which were linked to enhanced aerosol production by such mechanisms as lowered sea levels, stronger winds and reduced hydrological washout of aerosols, favouring the long-distance transport of dust from continents of the Southern Hemisphere to Antarctica [*Delmonte et al.*, 2004].

Table 1. Lead and Ba Concentrations and Pb Isotopic Compositions in Antarctic Dome C Ice

Depth (m)	Age (kyr BP)	^{206}Pb/^{207}Pb	±[a]	^{208}Pb/^{207}Pb	±[a]	^{206}Pb/^{204}Pb	±[a]	Pb[b] (pg/g)	Ba[b] (pg/g)
229.1	6.9	1.190	0.016	2.471	0.020	19.2	1.2	0.19	7.4
229.4	6.9	1.195	0.022	2.467	0.032	19.0	1.7	0.22	6.3
316.3	10.3	1.194	0.024	2.460	0.026	19.0	1.7	0.19	6.7
379.2	12.8	1.220	0.009	2.486	0.011	19.7	0.8	0.70	13
405.9	14.2	1.217	0.009	2.481	0.011	19.3	0.5	0.82	17
432.6	15.6	1.193	0.013	2.483	0.012	18.7	1.4	0.52	16
461.5	17.4	1.226	0.002	2.488	0.009	19.0	0.4	6.1	107
471.4	18.2	1.257	0.004	2.507	0.008	19.8	0.6	29.5	176
489.0	19.7	1.190	0.011	2.493	0.065	18.3	0.8	15.0	460
515.6	21.9	1.199	0.014	2.465	0.019	18.5	0.3	14.3	467
574.2	27.0	1.207	0.008	2.465	0.014	17.7	0.4	16.1	362
598.1	29.1	1.203	0.017	2.468	0.057	18.4	1.5	8.6	219
654.0	33.7	1.205	0.003	2.477	0.013	18.7	0.3	8.2	185
709.0	38.2	1.226	0.007	2.470	0.019	17.6	1.6	2.7	37
818.1	47.2	1.206	0.004	2.467	0.008	18.6	0.3	5.4	127
983.4	60.7	1.214	0.006	2.484	0.004	18.3	0.6	9.8	225
1010.9	63.5	1.196	0.003	2.464	0.006	18.5	0.4	13.9	360
1093.1	71.6	1.227	0.013	2.504	0.024	18.1	2.2	1.4	19
1258.4	86.0	1.222	0.006	2.477	0.007	19.0	0.5	3.5	61
1423.4	102.3	1.225	0.003	2.477	0.010	18.9	0.5	3.2	43
1533.1	114.1	1.229	0.010	2.487	0.009	19.3	0.3	0.36	11
1643.1	122.8	1.216	0.014	2.497	0.023	18.9	1.0	0.73	16
1643.4	122.8	1.208	0.010	2.475	0.018	19.2	0.7	0.52	14
1753.4	131.9	1.237	0.006	2.488	0.010	18.6	1.2	2.6	30
1863.1	152.2	1.204	0.003	2.468	0.006	18.6	0.2	18.3	370
1863.4	152.2	1.193	0.003	2.459	0.009	18.6	1.0	18.9	447
1973.1	174.4	1.200	0.004	2.470	0.009	18.7	0.3	7.3	192
1973.4	174.5	1.192	0.005	2.462	0.009	18.6	0.2	9.7	150
2094.4	198.9	1.199	0.018	2.489	0.025	19.4	1.5	0.27	9.5
2193.4	217.1	1.214	0.009	2.476	0.006	19.4	1.0	2.0	32

[a]Uncertainties in the isotope ratios are 95% confidence intervals.
[b]Concentrations are accurate to ±15% (95% confidence interval).

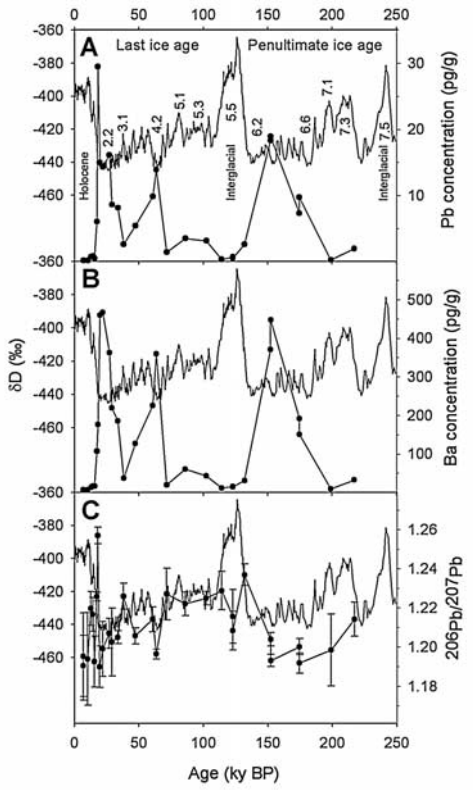

Figure 2. 220 kyr record of δ deuterium [*EPICA Community Members*, 2004] and Pb concentration (a), Ba concentration (b) and $^{206}Pb/^{207}Pb$ (c) from the EPICA Dome C ice core. Marine isotope stage numbers [*Delmonte et al.*, 2004] are also shown in Figure 2a.

[11] If it is assumed that dust deposited in Antarctic ice has a Pb/Ba ratio identical to that reported for the upper continental crust (~0.03 [*McLennan*, 2001]), then the average Pb/Ba ratio observed at Dome C (~0.05) indicates that mineral dust contributes ~70% of Pb in ice. Higher Pb/Ba ratios indicate that Pb from other sources, most likely volcanic emissions, occasionally contributed up to 65% of Pb at Dome C. More-radiogenic Pb isotopic compositions occur in samples with higher Pb/Ba ratios, suggesting that these samples contain greater proportions of volcanic Pb. The clearest sign of volcanic Pb in Dome C ice is an LGM sample (471.4 m, 18.2 kyr BP) with exceptionally high $^{206}Pb/^{207}Pb$ and Pb/Ba ratios, indicating a dust Pb proportion of ~20%. This sample corresponds to a fluoride peak in the EDC96 core [*Schwander et al.*, 2001, and references therein] which matches a larger, contemporaneous, fluoride peak identified in the Byrd ice core. A contemporaneous fluoride peak in the two cores indicates a volcanic emission source local to Antarctica.

[12] Pb isotopic compositions observed at Dome C over the past 220 kyr vary with changing climatic conditions (Figure 2). During the last glacial period, $^{206}Pb/^{207}Pb$ ratios decreased from 1.22 (average 71.6–102 kyr BP) to 1.21 (average 33.7–63.4 kyr BP) and then averaged 1.20 (19.6–29.1 kyr BP) during the LGM. They were greater during the glacial-interglacial transition (1.22, 12.8–18.2 kyr BP) compared to the LGM (1.20, 19.6–29.1 kyr BP), but they are not precise enough to determine any change since the end of the last transition. These trends also appear likely to have occurred during the penultimate glacial period, with ratios lower during the penultimate glacial period (1.20, 152–217 kyr BP), high during the transition (1.24, 132 kyr BP) and lower during the last interglacial (1.22, 114–123 kyr BP).

[13] The Pb isotope variations observed in Dome C ice appear to be climate related but the relationship between $^{206}Pb/^{207}Pb$ and temperature (δD) is not simple. Pb in Dome C ice originates from mineral dust and volcanism. If there is more than one source of mineral dust deposited in Antarctica, the Pb isotopic composition in the ice is likely to change as different source regions are diversely affected by climate change. For example, *Delmonte et al.* [2004] noted that the higher latitudes of southern South America, New Zealand and Australia were more likely to have experienced a colder and drier climate during the LGM, compared to interglacial conditions, while the low latitudes of Chile and southwestern southern Africa experienced a wetter climate compared to interglacial conditions. If all dust sources were found to have identical Pb isotopic compositions, then changing proportions of dust sources would not alter the isotopic composition of Pb in Dome C ice.

[14] Pb isotopic compositions in EPICA Dome C ice are within the range $^{206}Pb/^{207}Pb = 1.19–1.26$ and $^{208}Pb/^{207}Pb = 2.46–2.51$ (Figure 3), representing a mixture of Pb from mineral dust and volcanism. Data reported for Dome C [*Rosman et al.*, 1994, 1999], Taylor Dome [*Matsumoto and Hinkley*, 2001] and Coats Land [*Planchon et al.*, 2003] snow and ice are also shown in Figure 3. Most of the EPICA Dome C samples display lower $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios than those observed at Taylor Dome and Law Dome. At the latter locations, where the proportion of dust Pb in ice is consistently less than 30%, Pb isotopic compositions are similar to those reported for local volcanic Pb sources such as Antarctic Peninsula basalts [*Hole et al.*, 1993], Ross Island basanitoids [*Sun and Hanson*, 1975] and Marie Byrd Land volcanics [*Hart et al.*, 1997]. While *Matsumoto and Hinkley* [2001] noted that Taylor Dome ice displays similar Pb isotopic compositions to some ocean island volcanoes of the Southern Hemisphere, the extent to

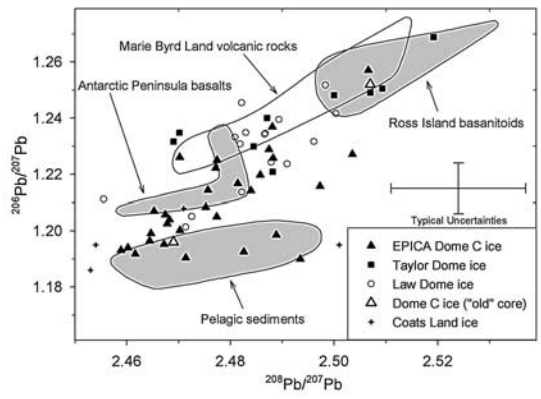

Figure 3. Pb isotopic compositions in snow and ice from Dome C and other Antarctic sites. Also shown are Pb isotopic compositions of South Atlantic and South Pacific Ocean pelagic sediments [*Chow and Patterson*, 1962], Antarctic Peninsula basalts [*Hole et al.*, 1993], Ross Island basanitoids [*Sun and Hanson*, 1975] and Marie Byrd Land volcanics [*Hart et al.*, 1997].

which such volcanoes influence Antarctica is still to be determined and so only Antarctic volcanic systems have been shown in Figure 3.

[15] *Delmonte et al.* [2004] reported that dust deposited at Dome C during cold climate periods originated from the same source(s), most probably the loess deposits of southern South America. Pb isotopic compositions are not yet available for southern South American loess, so we have used pelagic sediments [*Chow and Patterson*, 1962] of predominantly Aeolian origin to represent South American loess. Sediments were collected near South America and are likely to contain South American dust Pb. While the less-radiogenic EPICA Dome C samples have similar Pb isotopic compositions to the pelagic sediments, the provenance of Dome C dust Pb cannot be determined reliably until Pb isotope data have been reported for potential dust sources.

[16] The findings of the present study are in contrast to those of *Matsumoto and Hinkley* [2001] who reported high Pb concentrations (~5 pg/g from 2.2 to 10 kyr BP) in Taylor Dome ice during the Holocene and determined that volcanic Pb emissions are the dominant source of Pb deposited in Antarctica. The results presented here differ from those of Taylor Dome in two respects: lower Pb concentrations (~0.4 pg/g) are observed in Dome C ice from 6.9 to 15.6 kyr BP, and average Pb/Ba ratios (~0.05) indicate that ~70% of Pb deposited at Dome C originates from dust. These differences might be accounted for by considering ice core Pb to be a mixture of continental dust and Antarctic aerosols containing a relatively high proportion of radiogenic Pb of volcanic origin, where the proportion of volcanic Pb at Taylor Dome is greater than that at Dome C.

4. Conclusion

[17] These preliminary results support and extend existing data showing that Pb isotopic compositions and Pb concentrations in Antarctic ice vary with changes in climate. The deeper sections of the EPICA Dome C core must be analysed to confirm the cyclical patterns observed for the past 220 kyr, while Pb isotopic composition data pertaining to the loess deposits of the Southern Hemisphere are necessary to further constrain the potential sources of dust particles deposited in central East Antarctica. Finally, reliable and detailed records of Pb and Ba concentrations over the Holocene-LGM transition are required from other Antarctic locations to account for the differences observed between Taylor Dome and Dome C.

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References

Candelone, J.-P., S. Hong, and C. F. Boutron (1994), An improved method for decontaminating polar snow or ice cores for heavy metal analysis, *Anal. Chim. Acta*, *299*, 9–16.

Chow, T. J., and C. C. Patterson (1962), The occurrence and significance of lead isotopes in pelagic sediments, *Geochim. Cosmochim. Acta*, *26*, 263–308.

Delmonte, B., I. Basile-Doelsch, J.-R. Petit, V. Maggi, M. Revel-Rolland, A. Michard, E. Jagoutz, and F. E. Grousset (2004), Comparing the Epica and Vostok dust records during the last 220,000 years: Stratigraphical correlation and provenance in glacial periods, *Earth Sci. Rev.*, *66*, 63–87.

EPICA Community Members (2004), Eight glacial cycles from an Antarctic ice core, *Nature*, *429*, 623–628.

Gabrielli, P., et al. (2004), Determination of Ir and Pt down to the sub-femtogram per gram level in polar ice by ICP-SFMS using preconcentration and a desolvation system, *J. Anal. At. Spectrom.*, *19*, 831–837, doi:10.1039/b316283d.

Hart, S. R., J. Blusztajn, W. E. LeMasurier, and D. C. Rex (1997), Hobbs Coast Cenozoic volcanism: Implications for the West Antarctic rift system, *Chem. Geol.*, *139*, 223–248.

Hole, M. J., P. D. Kempton, and I. L. Millar (1993), Trace-element and isotopic characteristics of small-degree melts of the asthenosphere: Evidence from the alkalic basalts of the Antarctic Peninsula, *Chem. Geol.*, *109*, 51–68.

Hong, S., Y. Kim, C. F. Boutron, C. P. Ferrari, J. R. Petit, C. Barbante, K. J. R. Rosman, and V. Y. Lipenkov (2003), Climate-related variations in lead concentrations and sources in Vostok Antarctic ice from 65,000 to 240,000 years BP, *Geophys. Res. Lett.*, *30*(22), 2138, doi:10.1029/2003GL018411.

Lorius, C., L. Merlivat, J. Jouzel, and M. Pourchet (1979), A 30,000-yr isotope climatic record from Antarctic ice, *Nature*, *280*, 644–648.

Matsumoto, A., and T. K. Hinkley (2001), Trace metal suites in Antarctic pre-industrial ice are consistent with emissions from quiescent degassing of volcanoes worldwide, *Earth Planet. Sci. Lett.*, *186*, 33–43.

McLennan, S. M. (2001), Relationships between the trace element composition of sedimentary rocks and upper continental crust, *Geochem. Geophys. Geosyst.*, *2*, doi:10.1029/2000GC000109.

Petit, J.-R., et al. (1999), Climate and atmospheric history of the past 420,000 years from the Vostok ice core, Antarctica, *Nature*, *399*, 429–436.

Planchon, F. A. M., K. Van de Velde, K. J. R. Rosman, E. W. Wolff, C. P. Ferrari, and C. F. Boutron (2003), One hundred fifty-year record of lead isotopes in Antarctic snow from Coats Land, *Geochim. Cosmochim. Acta*, *67*, 693–708.

Rosman, K. J. R., W. Chisholm, C. F. Boutron, J.-P. Candelone, and C. C. Patterson (1994), Anthropogenic lead isotopes in Antarctica, *Geophys. Res. Lett.*, *21*, 2669–2672.

Rosman, K. J. R., W. Chisholm, C. F. Boutron, and J.-P. Candelone (1999), Lead isotopes as tracers of pollution in snow and ice, *Korean J. Polar Res.*, *10*, 53–58.

Schwander, J., J. Jouzel, C. U. Hammer, J.-R. Petit, R. Udisti, and E. W. Wolff (2001), A tentative chronology for the EPICA Dome Concordia ice core, *Geophys. Res. Lett.*, *28*, 4243–4246.

Sun, S. S., and G. H. Hanson (1975), Origin of Ross Island basanitoids and limitations upon the heterogeneity of mantle sources for alkali basalts and naphelinites, *Contrib. Mineral. Petrol.*, *52*, 77–106.

Vallelonga, P., K. Van de Velde, J.-P. Candelone, C. Ly, K. J. R. Rosman, C. F. Boutron, V. I. Morgan, and D. J. Mackey (2002a), Recent advances in measurement of Pb isotopes in polar ice and snow at sub-picogram per gram concentrations using thermal ionisation mass spectrometry, *Anal. Chim. Acta*, *453*, 1–12.

Vallelonga, P., K. Van de Velde, J.-P. Candelone, V. I. Morgan, C. F. Boutron, and K. J. R. Rosman (2002b), The lead pollution history of Law Dome, Antarctica, from isotopic measurements on ice cores: 1500 AD to 1989 AD, *Earth Planet. Sci. Lett.*, *204*, 291–306.

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