FATTY ACIDS IN THE BEACON GROUP OF SOUTHERN VICTORIA LAND IN ANTARCTICA

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Abstract: Fatty acids in the Beacon Group samples, shale, coal and silicified wood, from the McMurdo Sound region of southern Victoria Land in Antarctica were studied to elucidate their features in relation to chemical fossils of ancient organisms. The concentrations of fatty acids for the saponification extracts (0.5 M potassium hydroxide methanol, 80°C, 2 h) and the harsher saponification extracts (0.5 M potassium hydroxide methanol, 160°C, 3 h) ranged from 0.41 to 2.6 and from 0.043 to 2.3 μg/g of dry sample, respectively. The major constituents are comprised only of short-chain acids ($<C_{20}$), including normal alkanoic, branched and unsaturated acids, along with the dominance of even-carbon numbers. The high abundances of unsaturated and branched acids suggest strongly the contribution of bacterial lipids. The fatty acids found in the Beacon Group samples are thought to have been derived not only from ancient organisms but also from Recent organic matter and/or living microorganisms, including bacteria, fungi and algae. The evidence of ancient vascular plants was not indicated in the samples studied so far.

1. Introduction

The Beacon Group, composed of sandstone, siltstone, conglomerate and carbonaceous beds, including various fossils, constitutes the major formation of the Transantarctic Mountains, which are the largest mountain chains in Antarctica, extending 4000 km with the highest peak of 4528 m. The Beacon Group, ranging in age from early or pre-Devonian to Jurassic, was formed in lacustrine and shallow marine environments of Gondwana Land. In the McMurdo Sound region the Beacon Group is represented by 2100 m or thicker sedimentary sequences, predominantly pale yellow quartz sandstone, accompanied by carbonaceous siltstone, feldspathic sandstone, minor limestones and such fossils as coals and silicified woods, and is overlying unconformably the igneous and metamorphic rocks of the basement complex (WARREN and GUNN, 1961; MCKELVEY and WEBB, 1962; WEBB, 1963; TOWNROW, 1966; BALLANCE and WATTERS, 1971; BARRETT and KYLE, 1975; GEVERS and TWOMEY, 1982; FUNAKI, 1983, 1984).

A high abundance of the Beacon Group may have contributed to the distribution of organic matter in the regions surrounding the Transantarctic Mountains. For instance, SACKETT *et al.* (1974) suggested that most of the surface sedimentary organic matter in the Ross Sea was derived from those sedimentary rocks, and metamorphic and igneous rocks that are being glacially eroded on the Antarctic

continent and transported seaward. We have discussed that long-chain normal alkanoic acids extending C_{40} found in soils from the Dry Valleys are likely to be derived from the Beacon Group (MATSUMOTO *et al.*, 1981). Thus the Beacon Group can be expected to contain chemical fossils of ancient vascular plants including long-chain normal alkanoic acids. No one has, however, studied organic constituents in the Beacon Group. Here, for the first time we report the analyses of fatty acids in the Beacon Group samples from the McMurdo Sound region of southern Victoria Land in Antarctica and discuss their distribution patterns.

2. Materials and Methods

2.1. Sampling sites and samples

WEBB (1963) discussed the Beacon Group in the Dry Valleys: The formations exposed at the Plane Table, located in Asgard Range, are the Beacon Heights Orthoquartzite of middle-Devonian age (Fig. 1). The Beacon Heights Orthoquartzite is the thickest and most widespread formation of this region, and consists of a great thickness of almost pure quartz sandstone. Contorted beds of green-gray siltstone and shale are interbedded with orthoquartzites at only a few localities. A thick sequence of flatlying Beacon Group with lower or middle Triassic plant fossils and carbonaceous beds is found at Mount Fleming (GUNN and WARREN, 1962; FUNAKI, 1983). At the Allan Hills, sedimentary rocks referred to the Beacon Group, having a coal bed and fossil plants, dominantly *Glossopteris* of Permian to Triassic, are exposed (BALLANCE and WATTERS, 1971; FUNAKI, 1983).

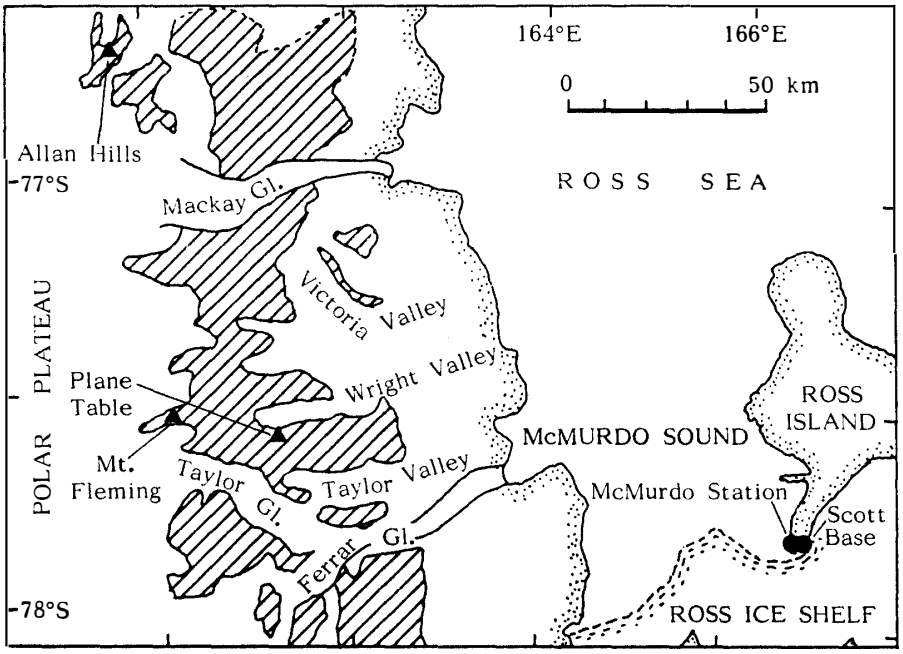

Fig. 1. Sampling locations of the Beacon Group of the McMurdo Sound region in southern Victoria Land, Antarctica.
▨: *Area of the Beacon Group outcrop (BARRETT and KYLE, 1975).*

In the 1976–77 and 1983–84 austral summers, the Beacon Group samples, shale, silicified wood and coal, were collected from the outcrops of Mount Fleming, the Plane Table of Asgard Range and the Allan Hills of the McMurdo Sound region in southern Victoria Land (Fig. 1). Some descriptions of the samples are given in Table 1. The samples were enveloped with teflon sheets or polyethylene bags and kept at temperature of $-20°C$ until analyses. The sample powders ($\leq$200 mesh) were prepared by the use of a ball mill (Fritsch Puloerisette 00501, West Germany).

Table 1. Total organic carbon (TOC) and total nitrogen (TN) results for the Beacon Group samples from the McMurdo Sound region of southern Victoria Land in Antarctica.

Sample	TOC (mg/g of dry base)	TN (mg/g of dry base)	Atomic ratio C/N	Color
Mt. Fleming-1 (shale)	1.4	0.50	3.3	Light bluish gray
Mt. Fleming-2 (shale)	1.7	4.0	0.50	Dark greenish gray
Mt. Fleming-3 (shale)	7.4	0.23	38	Greenish gray
Mt. Fleming-4 (shale)	62	11	6.6	Dark olive gray
Mt. Fleming-5 (shale)	19	1.6	14	Dark greenish gray
Mt. Fleming-6 (silicified wood)	14	4.2	3.9	Gray
Plane Table-1 (shale)	1.5	0.49	3.6	Light greenish gray
Plane Table-2 (shale)	0.21	0.068	3.6	Light gray
Plane Table-3 (shale)	0.22	0.12	2.1	Gray
Allan Hills (coal)	628	13	56	Black

2.2. *Analyses*

Total organic carbon (TOC) and total nitrogen (TN) were determined using a Yanako MT2 CHN Corder after the treatment of samples with 6 M hydrochloric acid to remove inorganic carbon. The analytical methods for fatty acids were described in detail elsewhere (MATSUMOTO *et al.*, 1979, 1981). In short, powdered samples (20–30 g) were first refluxed with 0.5 M potassium hydroxide methanol for 2 h at 80°C followed by centrifugation (1×10^3 g). The supernatant liquids and the residues were acidified separately and extracted 3 times with ethyl acetate, repeatedly. The ethyl acetate extracts were evaporated to dryness, dissolved in 50 μl benzene/ethyl acetate (1/1) and chromatographed on a silica gel column (160$\times$5 mm i.d., 100 mesh, 5% water). After the elution of hydrocarbons with 2 column volumes of hexane, fatty acids were eluted with 3 column volumes of benzene/ethyl acetate (95/5). Fatty acid fractions were methylated with 14% boron trifluoride methanol solution (80°C, 2 h).

The residues were further subjected to harsher saponification with 0.5 M potassium hydroxide methanol for 3 h at 160°C in the pyrex tubes using the air-tight brass syrinders. The saponified samples were treated following the same procedures noted above.

Fatty acids were determined using a combined Shimadzu LKB 9000 gas chromatograph-mass spectrometer equipped with a coiled glass column (2 m$\times$3 mm i.d.) packed with 1.5% OV-101 on 80–100 mesh Chromosorb W AW DMCS. The column temperature was programmed from 120 to 290°C at 8°C/min. The flow rate of helium carrier gas was 30 ml/min. The temperatures of injection block, molecular

separator and ion source were maintained at 300, 310 and 330°C, respectively. Mass fragmentograms were made at 20eV with an accelerating voltage of 20eV. Mass spectra were taken at 70eV. The analytical uncertainty was within ±12%.

3. Results and Discussion

3.1. General features, TOC and TN

Paleomagnetic investigations have showed that wide areas of the basement complex and the Beacon Group in the McMurdo Sound region were heated and remagnetized by the Ferrar dolerite intrusion in the Jurassic period (FUNAKI, 1983, 1984). The Beacon Group in Asgard Range, including the Plane Table may also have been heated by the Ferrar dolerite intrusion (FUNAKI, personal communication). The results of chemical analysis of coal samples from the Allan Hills showed that the fuel ratios are considerably high (16.9–20.4) and thus the areas were probably not heated (FUNAKI, 1983). Some parts of the Beacon Group at Mount Fleming may have been heated by dykes of the Ferrar dolerite and partially remagnetized parallel to the Jurassic geomagnetic field (FUNAKI, 1983, 1984).

The TOC and TN values for the Beacon Group samples (shale) from Mount Fleming ranged from 1.4 to 62 mg/g of dry sample and 0.23 to 11 mg/g, respectively, and are somewhat higher than those of the samples from the Plane Table, reflecting probably the thermal history of the areas studied (Table 1). The TOC content of the silicified wood from Mount Fleming was considerably low (14 mg/g), indicating that most organic carbon was replaced with mineral constituents, whereas that of the coal sample from the Allan Hills showed high TOC value of 628 mg/g. The atomic ratios of C/N also revealed a wide range of variation among the samples, suggesting that the quality of the samples is considerably different from each other.

3.2. Fatty acids

3.2.1. Saponification extract

Fatty acids ranging in carbon chain lengths from C_{10} to C_{26}, including normal alkanoic, branched and unsaturated, were found in the mass fragmentogram (m/z 74) of the fatty acid fraction from a Beacon Group sample (Mount Fleming–2), together with the dominance of even-carbon numbered normal alkanoic acids (Fig. 2). The major constituents of the acids (more than 10%) were nC_{16}, unsaturated-C_{18}, nC_{18}, nC_{14} and unsaturated-C_{16} in decreasing abundances (Table 2). High abundances of unsaturated acids suggest the contribution of living organisms and/or Recent organic matter, because unsaturated acids are more labile than saturated acids and thus could not have existed in sedimentary rocks during the geological times.

Carbon chain lengths of the acids ranged from C_{10} to C_{28} (Table 3). Unfortunately, very long-chain normal alkanoic acids were not detected in the Beacon Group samples studied so far. The concentrations of the acids ranged from 0.41 to 2.6 μg/g, which are similar to those of soils from the Dry Valleys (MATSUMOTO *et al.*, 1979, 1981, 1984). Fatty acid composition showed that short-chain normal alkanoic acids are the most dominant except for the Mount Fleming–3 sample. The high abundances of branched (7.5–29.2%) and unsaturated (7.8–53.1%) acids suggest again the contri-

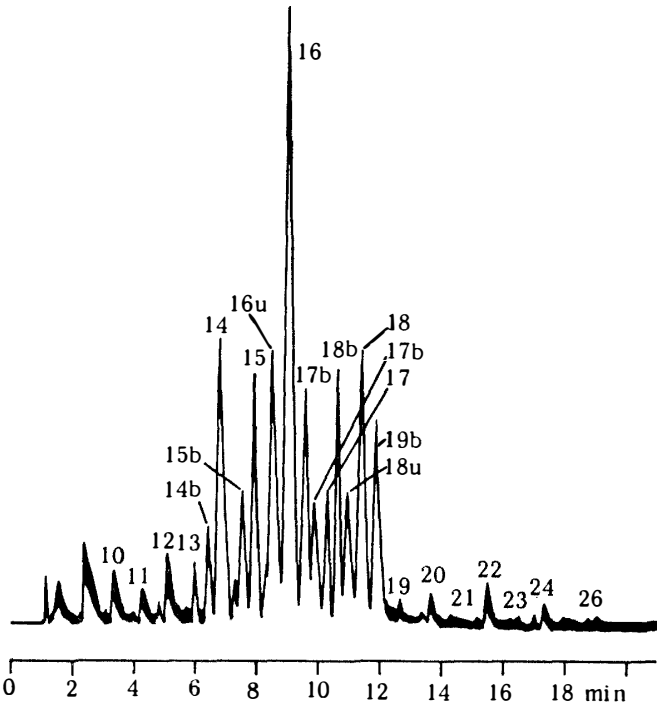

Fig. 2. Mass fragmentogram (m/z 74) of the fatty acid fraction from a Beacon Group sample (Mt. Fleming-2).
Arabic figures on the peaks indicate the carbon chain lengths of fatty acids. b: Branched. u: Unsaturated. Peaks with no figures were not identified.

bution of Recent organic matter and/or living microorganisms, especially bacteria, because branched acids are abundant in bacterial lipids (LEO and PARKER, 1966; KANEDA, 1967; TORNABENE and ORÓ, 1967). The long-chain normal alkanoic acids are less abundant (0.8–6.0%), and thus the Short/Long ratios are considerably high (10–32, Table 3).

When the intrusion of the Ferrar dolerite at the Plane Table occurred in Jurassic age, fatty acids in the Beacon Group should have been lost. Thus fatty acids found in our samples from the Plane Table are probably attributed to microbial activity after the Jurassic period. In addition, the similarity of the fatty acid compositions between the samples from the Plane Table and Mount Fleming suggest that the acids in the latter can also be explained by microbial activity. A large variety of the fatty acid composition among the samples are considered to be the differences in the relative contribution of the kinds of microorganisms and subsequent different alteration of them. It is well known that microorganisms, including bacteria, fungi and algae are widely distributed in the Dry Valleys (*e.g.*, CAMERON and CONROW, 1969; CAMERON and DEVANEY, 1970; FRIEDMANN and OCAMPO, 1976; PARKER, 1978).

The CPI_A values of fatty acids, which indicate the ratios of even- to odd-carbon numbered normal alkanoic acids, for living organisms and Recent environmental samples show higher values, while those of ancient samples of sedimentary rocks including shales, and petroleum reservoir waters reveal near unity (BRAY and EVANS,

Table 2. Fatty acid composition of the Beacon Group samples from southern Victoria Land in Antarctica (%).

Carbon chain length	Mt. Fleming-1	Mt. Fleming-2	Mt. Fleming-3	Mt. Fleming-4	Mt. Fleming-5	Mt. Fleming-6	Plane Table-1	Plane Table-2	Plane Table-2	Allan Hills
Normal										
10	1.52	1.17	0.24	0.88	0.33	3.58	1.33	4.60	1.74	4.13
11	0.50	0.82	0.29	0.64	0.34	2.17	0.73	1.70	1.38	2.26
12	3.29	1.76	0.63	1.60	0.74	8.95	3.31	7.96	2.92	6.79
13	1.57	1.47	0.36	1.11	0.85	2.00	1.45	2.00	1.45	2.26
14	9.57	6.98	1.00	5.06	3.85	12.70	12.74	19.43	8.67	11.31
15	7.59	6.19	4.27	5.16	4.43	7.19	5.56	5.01	6.78	6.40
16	24.66	15.03	10.05	17.18	13.29	29.85	21.34	22.83	23.48	28.90
17	3.29	3.01	4.12	3.52	13.29	2.46	3.15	2.14	3.33	2.72
18	11.36	6.53	3.97	8.37	4.49	8.66	10.78	10.80	14.10	12.20
19	0.56	0.38	0.49	0.19	0.79	0.34	0.45	0.40	0.82	0.57
20	1.39	0.66	0.28	0.88	0.45	0.87	1.30	0.77	1.45	1.24
21	0.33	0.08	0.06	0.12	0.07	0.17	0.25	0.12	0.29	0.28
22	1.42	0.84	0.28	1.13	0.55	1.01	1.78	1.53	1.91	1.93
23	0.33	0.12	0.04	0.16	0.05	0.18	0.41	0.12	0.24	0.23
24	1.17	0.44	0.10	0.39	0.18	0.65	1.09	0.61	1.23	0.85
25	0.38	0.16	ND	0.12	0.07	0.21	0.31	0.14	0.36	0.17
26	0.46	0.14	ND	0.12	0.06	0.21	0.52	0.22	0.39	0.23
27	0.02	ND	ND	ND	0.01	0.04	0.07	ND	ND	ND
28	0.05	ND	ND	ND	0.03	0.06	0.21	0.04	0.14	ND
Branched										
12	0.21	0.56	0.14	0.35	0.22	0.81	0.56	ND	0.51	0.68
13	0.66	0.50	0.14	ND	0.32	0.82	0.53	0.24	0.58	0.85
14	2.84	2.36	0.50	1.83	1.33	3.13	2.45	1.41	2.53	2.26
15	3.17	3.30	0.65	3.29	4.48	3.34	2.74	2.16	3.21	2.60
17a	1.65	5.72	8.69	7.06	7.68	1.36	0.75	1.51	2.34	0.57
17b	ND	2.80	4.50	5.16	6.46	1.40	1.66	2.06	2.32	0.57
18	ND	6.15	3.78	6.11	4.26	ND	ND	1.21	0.89	ND
19	ND	4.76	2.34	0.60	4.49	ND	ND	ND	ND	ND
Unsaturated										
16	8.69	13.16	21.60	10.99	8.53	3.08	5.78	4.95	5.96	2.97
18	13.32	14.91	31.48	17.98	18.36	4.76	18.75	6.04	10.98	7.03

ND: Not detected.

Table 3. Fatty acids found in the saponification extracts of the Beacon Group samples from southern Victoria Land in Antarctica.

Sample	Conc. μg/g	Range	Composition (%)				Short*1 / Long*2	CPI_A*5
			Short*1	Long*2	Branched*3	Unsaturated*4		
Mt. Fleming-1	0.92	C_{10}-C_{28}	63.9	5.5	8.5	22.0	11	3.8
Mt. Fleming-2	0.91	C_{10}-C_{26}	43.3	2.4	26.2	28.1	18	2.7
Mt. Fleming-3	2.6	C_{10}-C_{24}	25.4	0.8	20.7	53.1	32	1.7
Mt. Fleming-4	0.49	C_{10}-C_{26}	43.7	2.9	24.4	29.0	15	3.2
Mt. Felming-5	2.2	C_{10}-C_{28}	42.4	1.5	29.2	26.9	28	1.2
Mt. Fleming-6	1.6	C_{10}-C_{28}	77.9	3.4	10.9	7.8	23	4.5
Plane Table-1	0.73	C_{10}-C_{28}	60.9	5.9	8.7	24.5	10	4.4
Plane Table-2	0.50	C_{10}-C_{28}	76.8	3.6	8.6	11.0	21	5.9
Plane Table-3	0.41	C_{10}-C_{28}	64.7	6.0	12.4	16.9	11	3.8
Allan Hills	1.2	C_{10}-C_{26}	77.6	4.9	7.5	10.0	16	4.5

*1 Short chain n-alkanoic acids (nC_{10}-nC_{19}).
*2 Long chain n-alkanoic acids (nC_{20}-nC_{28}).
*3 Branched acids (bC_{12}-bC_{19}).
*4 Unsaturated acids (uC_{16} and uC_{18}).
*5 Carbon preference index.

1961; COOPER and BRAY, 1963; KVENVOLDEN, 1966, 1970). A possible explanation of this is that the normal even-carbon numbered alkanoic acids from organisms are converted in sedimentary environments into odd-carbon numbered normal alkanes and alkanoic acids (COOPER and BRAY, 1963; MORRIS and CULKIN, 1975), thus:

$$\underset{\text{(even-number)}}{R\text{-}CH_2\text{-}COOH} \longrightarrow R\text{-}\dot{C}H_2 + CO_2 + H^{\cdot}$$

$$R\text{-}\dot{C}H_2 + RH \longrightarrow \underset{\text{(odd-number)}}{R\text{-}CH_3} + R^{\cdot}$$

$$R\text{-}\dot{C}H_2 + (O) \longrightarrow \underset{\text{(odd-number)}}{R\text{-}COOH} + H_2O\,.$$

The sedimentation ages of the Beacon Group samples are quite old and thus the expected CPI_A values are close to unity. The CPI_A values of our samples are, however, considerably high ranging from 1.2 to 5.9 (Table 3). These results suggest again the contribution of Recent organic matter and/or living microorganisms to the sedimentary rocks. On the other hand, the evidence of fatty acids relating to ancient vascular plants was not fonud in the Beacon Group samples.

3.2.2 Harsher saponification extract

The major constituents of the harsher saponification extracts of the Beacon Group samples are nC_{14}, nC_{16} and nC_{18} acids. The carbon chain lengths ranged from C_{10} to C_{28}, which are similar to those of the saponification extracts (Table 4). The concentrations of fatty acids ranged from 0.043 to 2.3 μg/g, which are considerably lower than those of the saponification extracts except for the coal sample (Table 5). The composition of the acids denotes that short-chain normal alkanoic acids constituted more than 60% of the total acids in all the samples studied. The percentages of

branched and unsaturated acids are much lower than those of the saponification extracts described above. The proportions of long-chain normal alkanoic acids are also low (5.2–10.4%), and thus the Short/Long ratios are considerably high (8.0–15) as in the case of the saponification extracts. The CPI_A values are somewhat higher than those of the saponification extracts (Table 4). These tightly bound fatty acids are probably associated with organic and inorganic matrices in the Beacon Group samples.

Table 4. Fatty acids found in the harsher saponification extracts of the Beacon Group samples from southern Victoria Land in Antarctica.

Sample	Conc. μg/g	Range	Composition (%)				Short*1 / Long*2	CPI_A*5
			Short*1	Long*2	Branched*3	Unsaturated*4		
Mt. Fleming-1	0.45	C_{10}–C_{28}	73.9	8.3	6.3	11.5	8.9	4.3
Mt. Fleming-2	0.056	C_{10}–C_{26}	88.0	6.8	5.2	0.0	13	7.0
Mt. Fleming-3	0.063	C_{10}–C_{24}	63.9	7.3	28.8	0.0	8.8	3.1
Mt. Fleming-4	0.043	C_{10}–C_{26}	83.4	10.4	6.2	0.0	8.0	5.0
Mt. Fleming-5	NM	—	—	—	—	—	—	—
Mt. Fleming-6	0.097	C_{10}–C_{22}	85.6	8.0	6.4	0.0	11	8.0
Plane Table-1	0.13	C_{10}–C_{24}	79.0	5.2	8.2	7.6	15	3.9
Plane Talbe-2	NM	—	—	—	—	—	—	—
Plane Table-3	0.14	C_{10}–C_{26}	73.0	7.7	7.9	11.4	9.5	4.1
Allan Hills	2.3	C_{10}–C_{26}	86.2	8.8	5.0	0.0	9.8	5.8

*1 Short chain n-alkanoic acids (nC_{10}–nC_{19}).
*2 Long chain n-alkanoic (nC_{20}–nC_{28}).
*3 Branched acids (bC_{12}–bC_{19}).
*4 Unsaturated acids (uC_{16} and uC_{18}).
*5 Carbon preference index.
NM: Not measured.

Table 5. Relative abundances of fatty acids for the saponification and the harsher saponification extracts (%).

Sample	Saponification	Harsher saponification
Mt. Fleming-1	67.2	32.8
Mt. Fleming-2	94.2	5.8
Mt. Fleming-3	97.6	2.4
Mt. Fleming-4	91.9	8.1
Mt. Fleming-5	No datum	No datum
Mt. Fleming-6	94.3	5.7
Plane Table-1	84.9	15.1
Plane Table-2	No datum	No datum
Palne Table-3	74.5	25.5
Allan Hills	34.3	65.7

4. Conclusion

Fatty acids ranging from C_{10} to C_{28}, including normal alkanoic, branched and unsaturated acids were found in the Beacon Group samples from the McMurdo

Sound region of southern Victoria Land in Antarctica. The major constituents are all short-chain acids, *i.e.*, nC_{16}, unsaturated-C_{18}, nC_{18}, nC_{14} and unsaturated-C_{16} acids in decreasing abundances. The high abundances of unsaturated and branched acids, along with the considerably high CPI_A values suggest that fatty acids in the Beacon Group samples were derived not only from ancient organisms but also from living organisms, especially bacteria and/or Recent organic matter. The evidence of fatty acids of ancient vascular plants was not found.

Acknowledgments

The authors express their thanks to Antarctic Division, DSIR, New Zealand, the Japan Polar Research Association, the National Science Foundation of U.S.A., the U.S. Navy, the National Institute of Polar Research and the Event-21 members for their supports in the Antarctic researches. We thank Associate Prof. K. YANAI of the National Institute of Polar Research for providing silicified wood and coal samples. Thanks are due to Prof. H. NAKAHARA and his colleagues of Tokyo Metropolitan University for kindly permitting the use of a ball mill.

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(Received December 28, 1984; Revised manuscript received May 15, 1985)