

Climatic and environmental implications from organic matter in Dasuopu glacier in Xixiabangma in Qinghai-Tibetan Plateau *

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Abstract A series of organic compounds in snow and ice were identified from Dasuopu glacier in Xixiabangma in Qinghai-Tibetan Plateau. Organic compounds derived from natural organisms include n-alkanes ranging from C_{15} to C_{33} , normal monocarboxylic acids of C_6 — C_{18} , n-alkan-2-ones of C_{24} — C_{31} and esters. The unstable compounds widely present in lower troposphere disappear in the middle-upper troposphere. Lots of other organic compounds from petroleum residues were also unexpectedly identified from the glacier, covering pristane, phytane, extended tricyclic terpanes of C_{19} — C_{29} , C_{24} tetracyclic terpane, $\alpha\beta$ hopane compounds of C_{27} — C_{35} , and cholestanes of C_{27} — C_{29} . The remote Xixiabangma region is unambiguously polluted from anthropology activities. The petroleum residues were proposed to be mainly from the Mideast and India, not from China. The organic pollutants from oil fires ignited during the well-known Gulf War which broke out in 1991 were also recorded in the glacier. Such organic indexes as nC_{29}/nC_{17} , nC_{29}/nC_{27} , nC_{29}/nC_{24} , C_{30} -ketone/ C_{30} -ester, C_{24} -tetracyclic-/ C_{24} -tricyclic-terpane and CPI_n (n standing for n-alkanes) exhibit the same strong seasonal variation as $\delta^{18}O$, providing information on the atmospheric circulation of the plateau. These ratios decrease in summer half year when the monsoon dominated, and increase in winter half year when the westerly stream jet dominated.

Keywords: Qinghai-Tibetan Plateau, snow and ice in glacier, organic compound, petroleum residue, climate and environment.

Lots of high-resolution records for global climatic and environmental changes are extracted from snow and ice on the earth, the study of which constitutes the frontier of modern international glaciology. Inorganic aspects including isotopes, ions, gases, microparticles, pH, electric conductivity have received much attention. The climatic implication from light carboxylic acids also reaches successful achievements. Yet the trace heavy-molecular-weight organic compounds with a great variety of species, especially biomarkers, have not systematically been investigated in glaciers. The strong study basis in inorganic aspects of icecores and the extensive application of advanced techniques in molecular organic geochemistry make it possible for scientists to have an attempt probe on organic geochemistry in snow and ice. Such an objective makes up the following.

Qinghai-Tibetan Plateau and Asian monsoon are the two leading factors in the climatic and environmental changes in the North Hemisphere. Dasuopu glacier in the Xixiabangma Peak is proposed

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to be the leading medium in the study of Asian monsoon through icecores in the leading region of Himalayas^[1]. All the organic samples in the paper were collected from Dasuopu glacier. The glacier (28°21'N, 85°46'E) is located in the north slope of the Himalayas, and 1 km west of the Xixiabangma Peak, with the maximum altitude of 7 200 m a. s. l. Owing to the special location, the glacier offers great advantages in studying climatic and environmental changes. The glacier is favourable for annual dating because of a greater annual accumulation in the middle latitude region, and sensitive to the changes of middle-upper troposphere atmosphere because of its elevation of 7 000 m a. s. l.^[2] A new ice-forming genesis of refreezing-recrystallization occurs in the glacier^[3] with a good preserve of firm layers^[4]. The glacier is believed to be the most prospecting one for the air-bubble study in the plateau^[5]. In the Himalayan region, the period from November to March belongs to arid and cold season with little precipitation but strong wind, whereas the period from June to September is the warm and damp season characterized by much precipitation. The rest is the transitional period^[6].

1 Sample pretreatment and instrument analysis conditions

Three deep icecores total 475.99 m in length were drilled in Dasuopu glacier by a joint scientific investigation team from China, USA, Russia, Peru and Nepal from July to October in 1997. The investigation setting several world records in scientific expedition was evaluated to be one of Chinese science and technology developments in 1997. Altogether 11 samples for organic analysis were acquired from snow and ice layers, 2 of which were taken from the layers 0—75 and 75—88 cm in depth in the snowpit 6 400 m a. s. l. Six other samples were taken at intervals of 40 cm from another 240-cm-deep snowpit 7 000 m a. s. l. Two more samples were respectively taken from 0—10.9 and 10.9—15.1 m in depth in 2 combined shallow icecores 7 000 m a. s. l. Meltwater volume of these samples ranges from 5.00—24.91 L. The corresponding samples for isotope and ion analyses were also taken, at intervals of 4—5 cm in depth.

All snow and ice naturally melted at 5 800 m a. s. l. and concentrated HCl was added to bring pH to 2. The trace organic matter in meltwater was then concentrated *in situ* by resin adsorption columns, and the volume of meltwater for each sample was coincidentally measured. All utensils were purified in laboratory. Chemical solvent was distilled at least 4 times to clean out the pollutants. The mixture of G.D.X-102 and G.D.X-105 chemical reagents were extracted with a purified solvent for 96 h. After the concentration of organic matter, the 2 ends of the columns were sealed up and taken back to laboratory at low temperature. Organic matter adsorbed on the resin was slowly extracted with 30-mL dichloroform 3 times in the ultrapurified laboratory. Na₂SO₄ activated for 24 h at 450°C was used to dewater the extracted material. Then the organic extract was concentrated on a rotary evaporator under reduced pressure and then transferred to a small cell vial. Following the evaporation of the remaining solvent, the total extractable organic matter was quantified, and then preserved in a refrigerator before any analysis was taken.

To avoid further loss, the isolation of trace organic compounds is omitted. For GC and GC-MS analysis, 60- μ L chloroform was added, and 1 μ L of this was injected into the instruments. Procedural blanks have also been analysed to assess the possible laboratory contaminant. Fig. 1 shows one of the chromatographies of the procedural blanks.

GC-MS was carried out in the Faculty of Earth Sciences, China University of Geosciences Wuhan, China. A Hewlett-Packard 5973A mass spectrometer interfaced directly with a 6890⁺-gas

chromatograph equipped with an HP-5MS-fused silica capillary column (30 m × 0.25 mm × 0.25 μm) was used. The operating conditions are as follows: temperature program 70—280 °C at 3 °C/min, being held isothermally at 280 °C for 20 min and using helium as carrier gas.

Oxygen isotope was measured on a MAT-252 mass spectrometer with 0.5‰ precision. Ion analysis was finished in the Climatic Change Research Center of University of New Hampshire. For analysis method, refer to Buck et al.^[7]

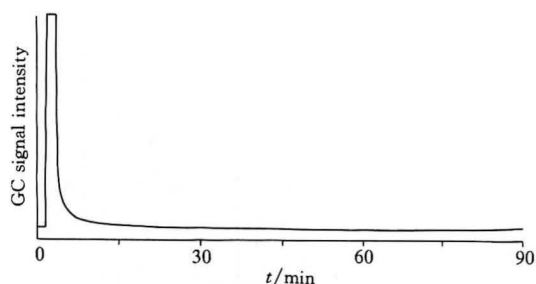


Fig. 1. GC analysis of one procedural blank to assess the laboratory contaminant.

2 Organic compounds derived from natural organisms

The concentration of the total extractable organic matter in the glacier ranges from 33.7 to 57.3 μg/L with an accuracy of ± 4.6 μg/L, averaged at 45.8 μg/L without distinct seasonal variation. All organic compounds originating from natural organisms are included below.

Normal alkanes in the glacier have the carbon number distribution of C₁₅—C₃₃, with 2 main peaks at C₁₇ and C₂₉ and a distinct odd-even carbon predominance above C₂₂ (fig. 2(a)). The carbon preference index (CPI_n) value is 1.05—1.88. This kind of distribution of n-alkanes is believed to be a mixed source. The homologs > nC₂₃ are chiefly derived from epicuticular waxes of vascular plants. The homologs < nC₂₃ should be derived from oceanic algae and microbes. Some compounds from petroleum residues are overlapped by those from natural biogenesis n-alkanes.

In many glaciological studies a certain degree of emphasis was placed on acetate and formate, the sensitive indicators for climatic changes. No systematic studies were made in the other organic acids. The distribution of the n-monofatty acids identified in Dasuopu glacier ranges from C₆ to C₁₈ with a maximum at C₁₆ and a strong even-odd carbon predominance above C₁₀ (fig. 2(b)). The CPI is 0.54—0.68. This distribution pattern definitively expresses the characteristic of a microbial source. Confined to the analytic condition and the omitting of derivation, some other heavy-molecular-weight fatty acids present in the glacier might fail to be detected.

A series of n-alkan-2-ones from C₂₄ to C₃₁ seldom reported in aerosols, were detected in the glacier (fig. 2(c)). These n-alkan-2-ones have a maximum at C₂₉ and a strong odd-even carbon predominance. The CPI is 2.12—3.07. They should be derived from higher plants. Besides, 6,10,14-trimethylpentadecan-2-one and nonacosan-15-one were found. Three compounds of esters were detected in the glacier.

All the compounds discussed above were also identified from the lower troposphere^[8–12]. Yet all the unstable compounds widespread in the lower troposphere such as unsaturated fatty acids, sesqui- and di-terpenoids disappear in the glacier, indicating the difference in organic composition between the lower and middle-upper troposphere.

3 Environmental implications of petroleum residues in the glacier

3.1 Petroleum residues in the glacier

Petroleum residues detected in the glacier cover isoprenoids (pristane and phytane), terpanes,

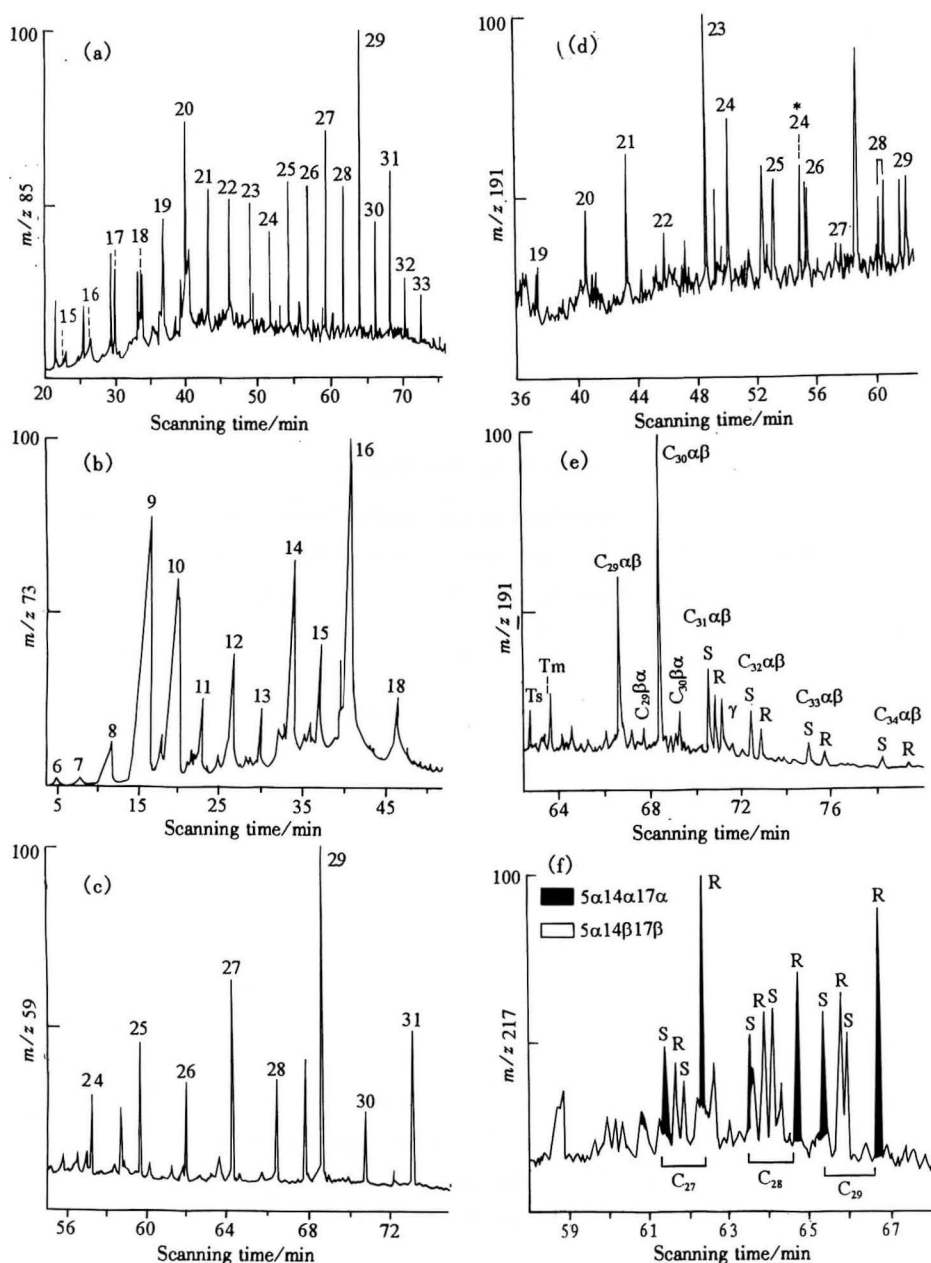


Fig. 2. Mass chromatograph of typical organic compounds identified from the glacier. (a) n-alkanes; (b) n-monofatty acids; (c) n-alkan-2-ones; (d) extended tricyclic terpanes (* for tetracyclic terpane); (e) hopanes (γ for gammacerane); (f) steranes. The number over the peak stands for carbon number, and the vertical axis is the relative content.

steranes and alkylcyclohexanes. A series of extended tricyclic terpanes were identified with carbon number distribution of C_{19} — C_{29} , maximizing at C_{23} (fig. 2(d)). Besides, C_{24} tetracyclic terpane was also found. Altogether 16 compounds of hopanes and gammacerane were unexpectedly detected (fig. 2(e)). All steranes identified in the glacier are shown in fig. 2(f). Alkylcyclohexanes from petroleum residues were detected with the carbon number distribution of C_{15} — C_{27} and a maximum at C_{23} , without carbon predominance.

3.2 Environmental implications

The compounds in the glacier including alkylcyclohexanes, pristane, phytane, hopanes and steranes and a proportion of n-alkanes were of geological source, derived from petroleum residues, most of which have been detected from aerosols in the lower troposphere^[13-17]. Simoneit^[15] has identified a series of organic compounds from the terrestrial petroleum residues in the aerosols over the ocean, 5000 km away from land. Organic contaminants in the atmosphere are different from the CO₂ pollution. The latter may have a slight localized effect but may result in environmental problems on a global scale. The organic contaminants have not only a serious local effect but also a global effect. Organic matter from petroleum residues in the glacier unambiguously shows the strong pollutant from human activities in the remote mountainous area of Xixiabangma as well as in the middle-upper troposphere.

It is clearly shown that the organic pollutants in the glacier were not from China according to the origin of air masses and the characteristics of organic constituents. In contrast to other countries in the world, China owns a greater percentage of coal in the energy consumption structure. Different from petroleum, coal will produce a lot of polycyclic aromatic hydrocarbons (PAHs) when burnt, a typical product of which is 2,9-dimethylpicene^[19-21]. Therefore, aerosols in China usually contain a greater proportion of PAHs, which is testified by the study on aerosols from Beijing, Guangzhou and Guiyang. The lack of PAHs in Dasuopu glacier further proves that organic pollutants do not originate from China. Forest fires also contribute little to the glacier because of the lack of PAHs.

Considering to the origin of air masses in the region, petroleum residues were from the Mideast, one of the most oil-producing areas in the world, and India. A greater annual concentration of organic matter from petroleum was found (table 1) in the shallow icecore dated to the year of 1988—1992 when the Gulf War broke out and resulted in a greater loss of petroleum in Persian Gulf^[22]. The products of oil combustion during the war migrated to the whole globe. It was reported that black snow was observed in the Kalakunlun Mountains^[18] as well as in Japan. The effect of the war is further strengthened by a greater annual concentration of NO₃⁻ and SO₄²⁻ during the years of 1988—1992 in the glacier^[2]. However, organic compounds from petroleum exhibit only a slightly increased concentration in winter (dominated by westerly stream jet), indicating the possibility of other sources. Existing reports reveal that gasoline and diesel do not contain triterpanes and steranes as identified in the glacier, but abundant compounds such as pristane, phytane, alkylcyclohexanes, hopanes and steranes were identified from the exhaust of automobile and other diesel cars^[14], which may also be one of the organic sources in the glacier. Besides, geological dusts abundant in terpanes and steranes might also contribute a small amount of paraffin to the glacier.

Table 1 Yearly-averaged organic ratios of 1988—1992/1997

	nC ₂₉ -alkane	nC ₂₀ -alkane	nC ₁₇ -alkane	n-alkane CPI	Pristane
1988—1992/1997	3.00	1.72	1.43	1.18/1.46	2.46
	C ₂₃ alkylcyclohexane	C ₂₃ tricyclic terpane	C ₃₀ hopane	C ₂₇ ααR cholestane	C ₂₉ ααRc holestane
1988—1992/1997	2.76	1.69	3.14	1.56	1.15

4 Climatic implication of organic compounds in the glacier

4.1 Dating in the 240 cm-deep snowpit profile

One of the advantages of studying organic matter from icecores and snowpits is to be able to ex-

pore organic variation with time, which is a prerequisite for extracting its climatic implication. The variation of $\delta^{18}\text{O}$ in the 240-cm-deep snowpit 7000 m a.s.l. can be used for dating. It is very difficult, sometimes even unnecessary, to precisely quantify every organic compound. Yet it is of great

significance to intercompare, through the GC-MS signal intensity, the relative concentration of organic matter within the same class under the identical analysis conditions. The relative concentration of every organic compound in the paper is transferred to the corresponding GC-MS signal value per milliliter meltwater.

As one of the most extensively investigated indicators, $\delta^{18}\text{O}$ is believed to be affected not only by temperature, but also by the air mass source in the plateau^[23]. The existing reports^[24,25] show that $\delta^{18}\text{O}$ in the summer rainfall is controlled by the volume of rainfall in the south of the plateau. Namely, the summer rainfall has lower $\delta^{18}\text{O}$ just as the snowpit exhibited. In the 240-cm-deep snowpit 7000 m a.s.l., summer half year of 1997 (0–120 cm in depth) and winter half year of 1996–1997 (120–200 cm in depth) can be clearly distinguished through the variation of $\delta^{18}\text{O}$ (fig. 3). The deepest sample from 200 to 240 cm in depth belongs to summer half year of 1996, and the surface of the snowpit corresponds to the date of September 2 in 1997.

N-alkanes from petroleum residues were usually superimposed on the natural biogenic background. A numerical evaluation based on CPI has been used by Simoneit et al.^[14–16] and also in the paper to distinguish plant wax from fossil fuel alkanes. The calculation result is shown in fig. 4. The main peak of $n\text{C}_{17}$ surpassing that of $n\text{C}_{29}$ in summer indicates the strong summer monsoon while $n\text{C}_{29} > n\text{C}_{17}$ is due to the westerly stream jet in winter. Thereby the relative concentration between $n\text{C}_{17}$ and $n\text{C}_{29}$ can be used as an indicator of atmospheric circulation changes.

Similar to $\delta^{18}\text{O}$, organic indexes such as $n\text{C}_{29}/n\text{C}_{27}$, $n\text{C}_{29}/n\text{C}_{17}$, $n\text{C}_{22}^+/n\text{C}_{21}^-$, $n\text{C}_{29}/n\text{C}_{24}$, $n\text{C}_{20}/n\text{C}_{17}$, $\text{C}_{30}\text{-ketone}/\text{C}_{30}\text{-ester}$, $\text{C}_{29}\text{-ketone}/n\text{C}_{29}$ and CPI_n also show a sharp variation between the 2 half years (fig. 3). The sharp variation of these indexes, coinciding with $\delta^{18}\text{O}$, is closely related to organic source, temperature and the air masses.

$n\text{C}_{29}$ and $n\text{C}_{22}^+$ represent the long-chained n-alkanes derived from higher plants in the south of the Himalayas, while $n\text{C}_{17}$ and $n\text{C}_{21}^-$ stand for the short-chained n-alkanes from

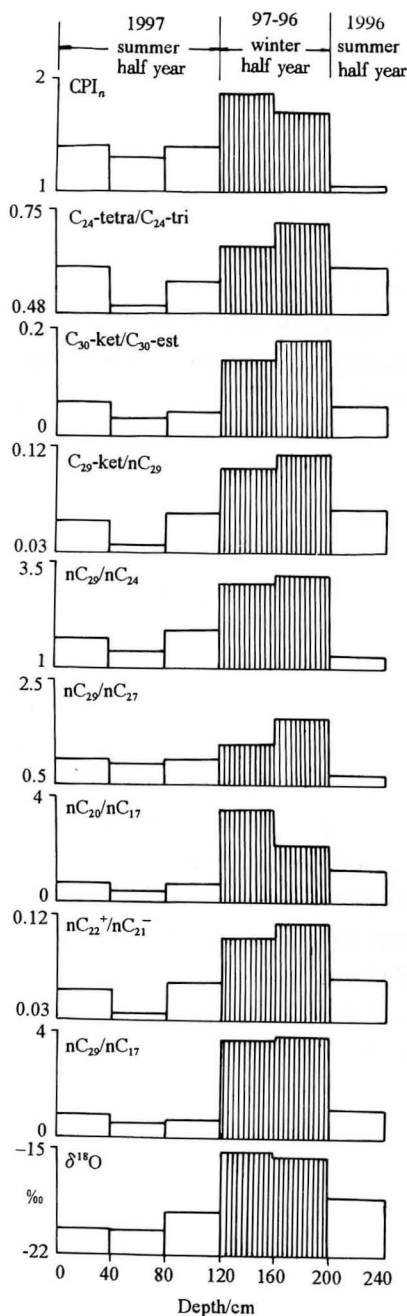


Fig. 3. The corresponding relation between $\delta^{18}\text{O}$ and organic indexes in the snowpit 7000 m a.s.l. tetra: Tetracyclic terpane; tri: tricyclic terpane; ket: n-alkan-2-one; est: ester.

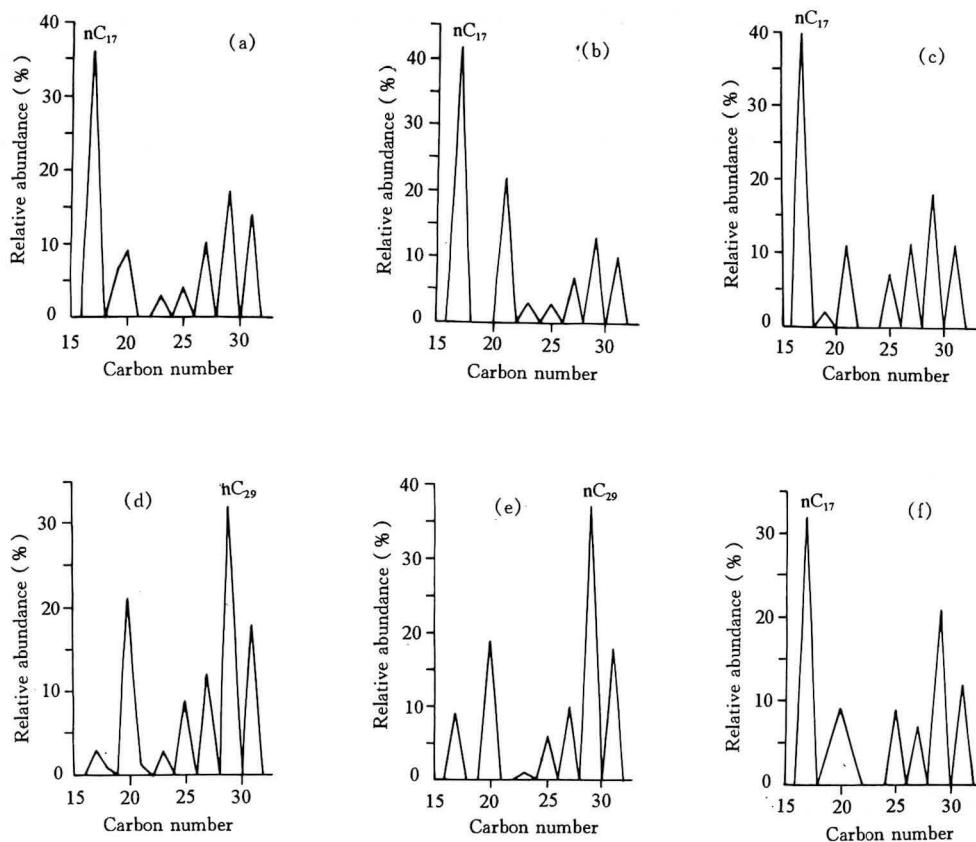


Fig. 4. Distribution of n-alkanes from natural organisms in the 240 cm-deep snowpit profile in the glacier. (a) 0—40 cm, summer half year; (b) 40—80 cm, summer half year; (c) 80—120 cm, summer half year; (d) 120—160 cm, winter half year; (e) 160—200 cm, winter half year; (f) 200—240 cm, summer half year.

oceanic organisms. Thus the ratios of nC_{29}/nC_{17} and nC_{22}^+/nC_{21}^- indicate the relative contribution of the 2 sources. Higher plants in the south are deciduous with little seasonal difference, while oceanic organisms are more flourishing in summer, and what is more, a strong summer monsoon dominated the region in the summer half year. So the seasonal variation of nC_{29}/nC_{17} and nC_{22}^+/nC_{21}^- is mainly caused by organism flourishing and atmospheric circulation. CPI_n is related to its derivation and the temperature organic matter experiences. The greater the temperature, the lower the CPI_n , which is an irreversible process. So CPI_n , along with the maturity parameters, records the temperature organic matter underwent before its precipitation. Naturally, CPI_n exhibits a lower value in the summer half year and a greater value in the winter half year, displaying a seasonal variation. CPI_n variation may also be attributed to the difference of organic source.

Ketones may be derived from lipids, cellulose and semicellulose, and ester mainly from wax in plant lipids. Because of its extreme fragility, the plant wax is more easily sloughed off from plant surfaces and becomes airborne in the heavy rainfall in summer. The ratio of C_{30} -ketone/ C_{30} -ester ineluctably exhibits this kind of seasonal variation.

All in all, although the extractable organic concentration keeps stable in profile, some organic indexes show seasonal variation and are demonstrated to be related to the organism characteristics as

well as atmospheric circulation. Therefore these indexes are suggested to imply climatic information. When the monsoon dominates in summer, n-alkanes show $nC_{17} > nC_{29}$ and the organic ratios such as nC_{29}/nC_{17} , nC_{22}^+/nC_{21}^- , nC_{29}/nC_{27} , nC_{29}/nC_{24} , nC_{20}/nC_{17} , C_{30} -ketone/ C_{30} -ester, C_{29} -ketone/ nC_{29} and CPI_n decrease. When the westerly stream jet dominates in winter, n-alkanes display $nC_{29} > nC_{17}$, and the organic ratios increase. The sharp seasonal variation of the organic matter can also be used to date the shallow icecores in the glacier, just as $\delta^{18}O$ does.

5 Conclusions

Organic compounds derived from subtropical higher plants and marine organisms include n-alkanes from C_{15} to C_{33} , maximizing at C_{17} and C_{29} , normal monocarboxylic acids of C_6 — C_{18} , n-alkan-2-ones of C_{24} — C_{31} and esters. The lack of unstable organic compounds in the glacier reveals the organic difference between the lower and middle-upper troposphere.

Lots of other organic compounds from petroleum residues were also unexpectedly identified from the glacier, including pristane, phytane, extended tricyclic terpanes of C_{19} — C_{29} , C_{24} tetracyclic terpane, $\alpha\beta$ hopane compounds of C_{27} — C_{35} , and steranes of C_{27} — C_{29} . The remote Xixiabangma region is unambiguously polluted from human activities. These pollutants were transported from the Mideast and India. The oil combustion from the Gulf War was also recorded in the glacier.

Organic indexes such as nC_{29}/nC_{17} , nC_{29}/nC_{27} , C_{30} -ketone/ C_{30} -ester, nC_{17} and CPI_n exhibit the same strong seasonal variation as $\delta^{18}O$, providing information on climatic change. When the monsoon dominates in summer, n-alkanes show $nC_{17} > nC_{29}$ and organic ratios such as nC_{29}/nC_{17} , nC_{29}/nC_{27} , nC_{22}^+/nC_{21}^- , nC_{29}/nC_{27} , nC_{29}/nC_{24} , nC_{20}/nC_{17} , C_{30} -ketone/ C_{30} -ester, C_{29} -ketone/ nC_{29} and CPI_n decrease. When the westerly stream jet dominates in winter, the main peak of nC_{29} surpasses that of nC_{17} and the organic ratios increase.

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