Concentrations and source variations of *n*-alkanes in a 21 m ice core and snow samples at Belukha glacier, Russian Altai mountains

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ABSTRACT. Ice-core and snow samples collected on Belukha glacier, Russian Altai mountains, were analyzed for *n*-alkanes by gas chromatography. On the basis of the total concentrations (T-HCs), carbon preference index (CPI) values and the plant wax contributions (WaxC_n), it is suggested that mountain glaciers on the Asian continent received higher loading of *n*-alkanes from natural and anthropogenic sources than the Greenland ice sheet. It appears that the loading and variation of *n*-alkanes on glaciers in the Altai are approximately the same as those of the mountain in general, because the concentrations, CPI values and WaxC_n percentages of *n*-alkanes in Sofiyskiy glacier, also in the Russian Altai, are at the same levels as or slightly greater than those in ice-core and snow samples from Belukha glacier. It seems that the *n*-alkanes on Belukha glacier are derived mainly from higher plant wax and petroleum exhaust other than from diesel engines. Vertical profiles of T-HCs and CPI values of *n*-alkanes show that the non-WaxC_n portions and T-HCs have recently increased with a decrease in CPI values on Belukha glacier. A similar trend of *n*-alkanes was reported for Sofiyskiy glacier, indicating that the influence of human activities has gradually increased around the Altai.

1. INTRODUCTION

Various organic and inorganic compounds are contained in glaciers and ice sheets on all the continents, including Greenland and Antarctica. Chemical compositions of ice cores from these areas have provided information on the history of global and/or regional environmental changes (Delmas, 1992; GRIP members, 1993; Legrand and Mayewski, 1997; Kang and others, 2002, 2004; Watanabe and others, 2003).

The compounds in ice are derived from natural and anthropogenic sources on the ground or produced by reactions in the atmosphere. For example, polycyclic aromatic hydrocarbons (PAHs) are emitted together with soot (elemental carbon) chiefly from fossil fuel combustion and biomass burning (Seinfeld and Pandis, 1998; Simoneit, 2002). *n*-alkanes, which are kinds of straight-chain saturated hydrocarbons, are emitted from anthropogenic (fossil fuel combustion) and natural (plant wax and soil) sources (Seinfeld and Pandis, 1998). After being introduced into the atmosphere, they are transported by wind and scavenged from the air by snowfall and dry fallout. *n*-alkanes found in the atmosphere as aerosol constituents have been used as source tracers in discussing the intensities of possible organic sources (Simoneit and others, 1977; Matsumoto and Hanya, 1980; Simoneit and Mazurek, 1982; Schneider and others, 1983; Simoneit, 1984, 1989; Kawamura and Kaplan, 1986; Simoneit and others, 1991; Lin and Lee, 2004). PAHs also have been known as chemical tracers of fossil fuel combustion and biomass burning (Kawamura and others, 1994; Seinfeld and Pandis, 1998; Masclet and others, 2000). Thus, the transported organic compounds such as *n*-alkanes and PAHs, which were the tracers used to reconstruct historical environmental changes, should be preserved in glaciers and ice sheets. However, the analytical data on *n*-alkanes and PAHs in ice-core and snow samples are very few (e.g. Kawamura and Suzuki, 1991; Kawamura and others, 1994; Masclet and others, 2000; Xie and others, 2000). Miyake and others (2005) have reported that ice-core, snow and snowfall samples collected on Sofiyskiy glacier, Russian Altai mountains, were analyzed for *n*-alkanes.

In the present study, ice-core and snow samples were collected from Belukha glacier, Russian Altai mountains, and analyzed for *n*-alkanes. Here we report the possible organic sources and their changes in intensity with time on the basis of compositional features of *n*-alkanes such as carbon preference index (CPI) and the plant wax contributions (WaxC_n). Furthermore, we compare the data of Belukha glacier with those of Sofiyskiy glacier (Miyake and others, 2005) to discuss the difference in environment between the two sites and the environmental variation around the Altai mountains area.

2. SAMPLING SITE AND METHODS

An ice core 18.74 m long was taken in July 2001, between depths of 2.20 and 20.94 m below the surface on the accumulation area of the western plateau of Belukha glacier (49°49′ N, 86°34′ E; 4100 m a.s.l.), located on the west side of the summit of Belukha mountain (4506 m a.s.l.) in the Russian Altai (Fig. 1). Fujita and others (2004) carried out the

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glaciological and meteorological observations at the glacier from 2001 to 2003. They reported that the glacial depth of the plateau ranged from 150 to 190 m, surface flow velocities were ~1–2 m a^{-1} on average, the annual range of snow temperature was –30 to 0°C, and annual accumulations of snow were 2.26 m for the year from summer 2001, and 1.60 m for the year from summer 2002. Moreover, the disturbances of climatic signals from melt-water on the glacier were reported as small because no heavy-melting feature was found in the snow temperature record and the snow-pit observations of July 2002 (Fujita and others, 2004). Snow samples were collected on 13 July 2002, from a 2.42 m pit near the drilling site for the ice core. A series of snow samples were obtained by cutting the snow wall of the pit into sections 2–16 cm long. The ice-core and snow samples were kept frozen during the transport to the National Institute of Polar Research, Tokyo, Japan, and they were preserved at –20°C until analysis.

For chemical (δD and $\delta^{18}O$ of water, ionic and organic compositions and microparticles) and biological (micro-organism and pollen) analyses, the ice core was cut into 4–8 cm sections (Iizuka and others, 2004) and melted at room temperature. Since only a small amount of water (10–20 and 40–110 mL from individual sections of ice-core and snow samples, respectively) could be used for *n*-alkanes analysis due to a limited sample amount, the water samples taken from 30–37 sections of the ice core were combined into six samples of about 3 m length in each case. In a similar manner, water samples from 7–13 sections of the snow from a pit were combined into three samples representing the 0.28–1.09 m depth interval in the snow pit (see Table 1).

After filtration of the sample water through a glass-fiber filter (Whatman, GF/F, diameter: 25 mm), the filter was spiked with an internal standard (Merck, Tetracosane-d_{50}: $C_{24}D_{50}$). Particulate materials on the filter were then analyzed for *n*-alkanes by the method described by Miyake and others (2005). The particle materials were extracted three times with 5 mL of organic solvents (methanol/dichloromethane (2:1) and dichloromethane) in an ultrasonic cleaner for 5 min each time. The extracts were combined, filtered and concentrated. The condensate was fractionated by a column chromatograph using a silica gel (Merck, Silica gel 80), which was sufficiently pre-cleaned with hexane to remove organic contaminants. The *n*-alkanes were eluted with 4 mL of hexane and then dried by a rotary evaporator and nitrogen stream. The *n*-alkanes fraction was dissolved in dichloromethane (~20 μL), and then measured by a gas chromatograph with a flame ionization detector

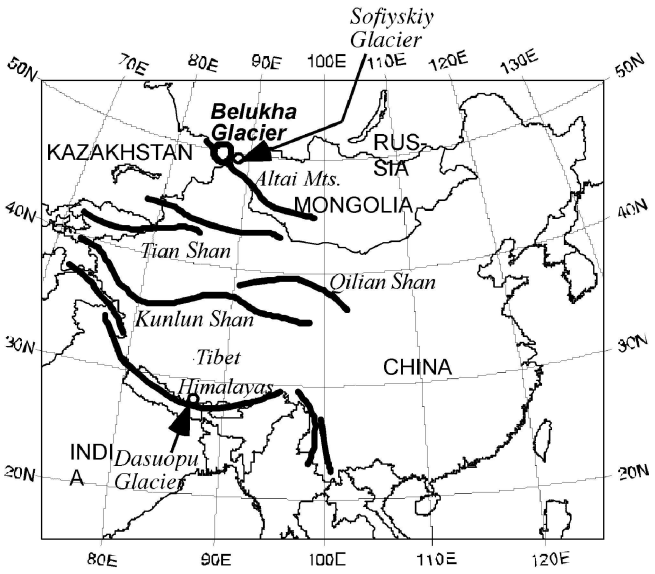

Fig. 1. Locations of Belukha and Sofiyskiy glaciers, Russian Altai, and Dasuopu glacier, Himalaya.

(GC-FID, Shimadzu, GC-14B) and a fused silica capillary column (GL Sciences, TC-1: 0.25 mm i.d. × 30 m length, 0.25 μm of film thickness). The gas chromatograph conditions were as follows: injector temperature: 300°C; detector temperature: 300°C; column temperature program: 80–300°C at 3°C min^{-1}; and the hold time: 10 min at 300°C. Detection limits for individual *n*-alkanes were approximately several to >10 pg g^{-1} for several-hundred mL samples. Analytical replicate precision of the gas chromatograph for individual *n*-alkanes ($n = 3$) was below ~5%. The blank levels of organic solvents for *n*-alkanes were <10% of the peak levels of the samples.

The main sources of *n*-alkanes in the samples were estimated based on the predominant constituents, CPI values of homologue distributions and the contribution ratios of biogenic ($WaxC_n$) or anthropogenic (non-$WaxC_n$) sources (Miyake and others, 2005). The CPI values, the preference of *n*-alkanes with odd carbon numbers over those with even carbon numbers, have been used to deduce the *n*-alkane sources (Simoneit and others, 1977; Simoneit and Mazurek, 1982). In the present study, CPI values were calculated according to:

$$\mathrm{CPI} = \frac{C_{21} + C_{23} + C_{25} + C_{27} + C_{29} + C_{31}}{C_{22} + C_{24} + C_{26} + C_{28} + C_{30} + C_{32}}. \quad (1)$$

Table 1. Total concentrations (T-HCs), carbon preference index (CPI) values and percentages of the plant wax contributions ($WaxC_n$) of *n*-alkanes in ice core (01BC-1–6) and snow from a pit (02BP-1–3) at Belukha glacier

	Ice core						Snow		
	01BC-1	01BC-2	01BC-3	01BC-4	01BC-5	01BC-6	02BP-1	02BP-2	02BP-3
Depth (m)	2.20–5.21	5.21–8.17	8.17–11.11	11.35–14.64	14.64–17.79	17.79–20.94	0–0.28	0.28–1.37	1.37–2.42
T-HCs[1]	1.43	1.10	0.99	0.84	0.81	0.72	0.52	1.42	0.72
CPI[2]	1.28	1.33	1.52	2.29	2.38	1.86	1.23	1.19	3.07
$WaxC_n$[3]	15	16	22	40	41	30	15	11	52

[1]Total concentration of *n*-alkanes (T-HCs) from C_{21} to C_{32} (unit: ng g^{-1}).
[2]Carbon preference index (CPI) = $C_{21-31(odd)}/C_{22-32(even)}$.
[3]$WaxC_n = [C_n] - [(C_{n+1}) + (C_{n-1})]/2$ (unit: %).

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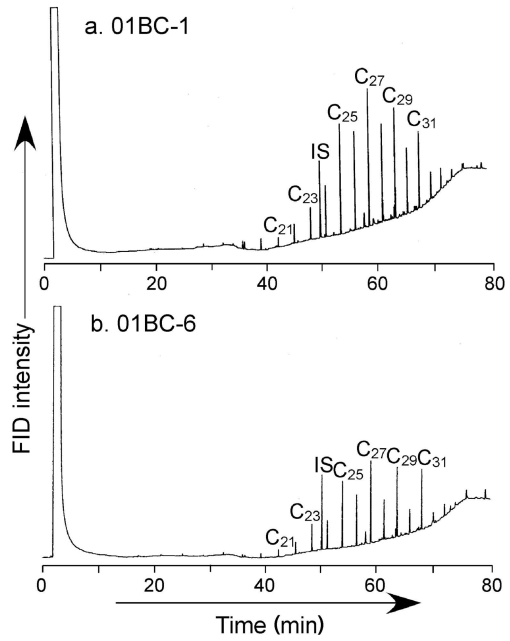

Fig. 2. Typical chromatograms of *n*-alkanes from ice-core samples 01BC-1 (a) and 01BC-6 (b) at Belukha glacier. C_n is carbon number of *n*-alkanes, and IS is internal standard (Tetracosane d_{50}: $C_{24}D_{50}$).

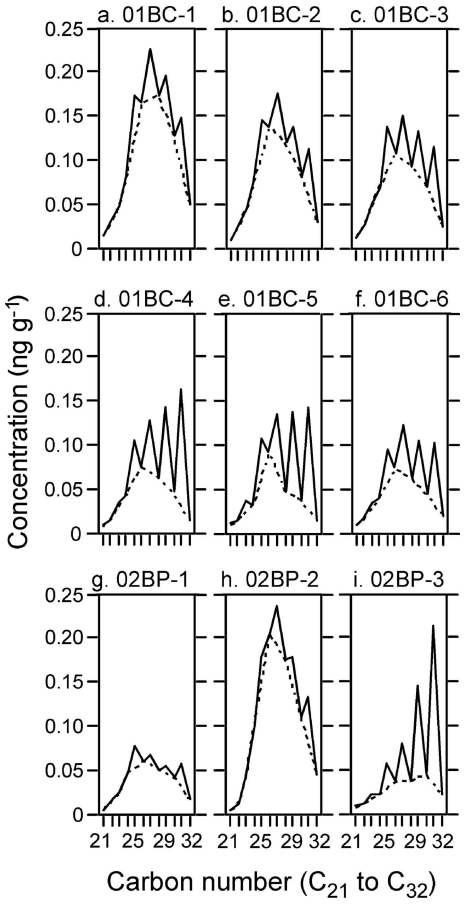

Fig. 3. The homologue distributions of *n*-alkanes in ice core (a–f) and snow (g–i) on Belukha glacier. Portions above and below the dashed line indicate the estimated concentrations of *n*-alkanes from plant wax ($WaxC_n$) and petroleum (non-$WaxC_n$), respectively.

The biogenic or anthropogenic source contributions to *n*-alkanes were also evaluated more quantitatively by the method of Simoneit and others (1991). They assumed that *n*-alkanes in the sample were derived from two sources, i.e. vascular plant wax and fossil-fuel combustion products, and that the CPI values of petroleum-derived *n*-alkanes were ~1.0 (Simoneit and Mazurek, 1982; Kawamura and Kaplan, 1986; Simoneit, 1989). The wax-derived portion in the *n*-alkanes with carbon numbers of *n* ($WaxC_n$) was estimated by subtracting an average concentration of next higher (C_{n+1}) and lower (C_{n-1}) homologues from the concentration of C_n. Negative values of $WaxC_n$ were taken as zero (Simoneit and others, 1991):

$$\mathrm{WaxC}_n = [\mathrm{C}_n] - \frac{[(\mathrm{C}_{n+1}) + (\mathrm{C}_{n-1})]}{2}. \qquad (2)$$

The non-$WaxC_n$ values were calculated by subtracting $WaxC_n$ from total concentrations of *n*-alkanes (T-HCs) with carbon number of *n* (T-HCs_n) (Miyake and others, 2005).

3. RESULTS AND DISCUSSION

3.1. T-HCs, CPI and $WaxC_n$ of *n*-alkanes at Belukha glacier

Figure 2 shows chromatograms of *n*-alkanes in ice-core samples 01BC-1 (Fig. 2a) and 01BC-6 (Fig. 2b). The T-HCs values of *n*-alkanes in the ice-core and snow samples collected on Belukha glacier are listed in Table 1, together with the CPI values and the percentages of $WaxC_n$. The T-HCs values in ice-core and snow samples were in the ranges 0.72–1.43 and 0.52–1.42 ng g^{-1}, respectively. The CPI values of *n*-alkanes in these samples were in the range 1.19–3.07.

Table 2 lists the ranges of T-HCs, CPI and $WaxC_n$ of *n*-alkanes in ice and snow samples from Belukha glacier (the present study); from Sofiyskiy glacier (49°47′10″ N, 87°43′43″ E; 3450 m a.s.l.; Miyake and others, 2005), also located in the Altai mountains, about 80 km east of our study site; in snow samples at Dasuopu glacier, Himalaya (28°23′ N, 85°43′ E; 7000 m a.s.l.; Xie and others, 2000); and in ice-core samples at Site-J, Greenland (66°51.9′ N, 46°15.9′ W; 2300 m a.s.l.; Kawamura and Suzuki, 1991).

The T-HCs in Belukha and Sofiyskiy glaciers were similar to the snow samples in the Himalaya, but much higher than those in ice in Greenland. This would probably suggest that *n*-alkanes are loaded more heavily onto the mountain glaciers on the Asian continent than onto the Greenland ice sheet, because annual snow accumulation rates are 1.60–2.26 m (0.56–0.87 m w.e. a^{-1}) at Belukha glacier from 2001 to 2003 (Fujita and others, 2004), 1.1–2.1 m (average: 0.99 m w.e. a^{-1}) at Sofiyskiy glacier from 1994 to 1999 (Kameda and others, 2003) and about 2 m at Dasuopu glacier from 1996 to 1997 (Xie and others, 2000). This is greater than the average ice accumulation of 0.42 m ice a^{-1} at Site-J (Shoji and others, 1991). Further study is needed to accurately date the core for discussing *n*-alkanes loading to glaciers. The CPI values of *n*-alkanes in snow and ice samples in Asian continental glaciers are wider than in Greenland ice samples. It seems that *n*-alkanes in the continental glaciers derive from various sources and these glaciers are nearer to source areas than Greenland.

It appears that the concentrations, CPI values and $WaxC_n$ percentages of *n*-alkanes in ice-core and snow samples on Sofiyskiy glacier are the same as or slightly

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Table 2. T-HCs, CPI values and WaxC_n percentages of *n*-alkanes in ice-core and snow samples at Belukha glacier and other glaciers

Site		T-HCs ng g^{-1}	CPI	WaxC_n %
Belukha glacier, Russian Altai[1]				
(C_{21}–C_{32})	Ice core ($n = 6$)	0.72–1.43	1.28–2.38	15–41
	Snow ($n = 3$)	0.52–1.42	1.19–3.07	11–52
Sofiyskiy glacier, Russian Altai[2]				
(C_{21}–C_{32})	Ice core ($n = 5$)	1.08–3.53	1.27–2.26[3]	15–40
	Snow ($n = 4$)	1.70–3.58	1.27–2.90[3]	15–50
	Snowfall ($n = 6$)	0.67–9.86	1.04–4.60[3]	7.3–66
Dasuopu glacier, Himalaya[4]				
(C_{15}–C_{33})	Snow ($n = 6$)	0.57–1.52	1.05–1.88	NC[6]
Site-J, Greenland[5]				
(C_{14}–C_{35})	Ice core (n= 2)	0.240–0.510	1.34–1.48	NC[6]

[1] Present study.
[2] Miyake and others (2005); 49°47′10″ N, 87°43′43″ E; 3450 m a.s.l.
[3] CPI values of *n*-alkanes are calculated from C_{21} to C_{32} in the table, although calculated from C_{21} to C_{30} in the literature (Miyake and others, 2005).
[4] Xie and others (2000); 28°23′ N, 85°43′ E; 7000 m a.s.l.
[5] Kawamura and Suzuki (1991); 66°51.9′ N, 46°15.9′ W; 2300 m a.s.l.
[6] Not calculated.

greater than on Belukha glacier. Therefore, it seems that the loading and variation of *n*-alkanes are approximately the same as in the Altai mountains in general. However, the concentrations, CPI values and WaxC_n percentages of *n*-alkanes have a wider range in snowfall samples than in ice-core and snow samples at Sofiyskiy glacier. It seems that *n*-alkanes in snow precipitation events on the glacier have been influenced by different sources. The elevation of Belukha glacier (4100 m a.s.l.; Fujita and others, 2004) is higher than that of Sofiyskiy glacier (3450 m; Fujii and others, 2002). The major types of vegetation surrounding the Altai are tundra and steppe, including conifer forest, and the tree lines on the north- and south-facing slopes around Sofiyskiy glacier are ~2500 m a.s.l. (Nakazawa and others, 2004). It seems that the higher T-HCs in snowfall samples on Sofiyskiy glacier can be attributed to the greater loading of *n*-alkanes from vegetation and soil than on Belukha glacier.

The WaxC_n values for Belukha glacier are 15–41% for ice-core samples and 11–52% for snow samples. These ranges are almost the same as those for Sofiyskiy glacier. Therefore, *n*-alkanes derived from plant wax in the Altai account for 10–50% of the total *n*-alkanes, and more than half of *n*-alkanes derived from non-plant wax, mainly petroleum. In urban aerosols, it is reported that the plant wax contributions to the total concentrations of *n*-alkanes are estimated to be only 5.9–32% (Lin and Lee, 2004), lower than those of ice and snow samples in the Altai.

3.2. Homologue distributions of *n*-alkanes

Figure 3a–i show the homologue distributions of *n*-alkanes in ice-core and snow samples on Belukha glacier. In this figure, the portion below the dashed line indicates the estimated concentration of *n*-alkanes from petroleum sources (=non-WaxC_n), because *n*-alkanes derived from petroleum generally show a CPI value of about 1, as mentioned above. As shown in Figure 3, *n*-alkanes have main peaks at C_{31} or C_{29} or C_{27} in most samples.

In general, main peaks with carbon numbers greater than C_{27} reflect the significant incorporation of higher plant wax and some fungi, whereas those with lower carbon numbers indicate major inputs from microbial activity (algae and bacteria: C_{17}, C_{18}) or petroleum (diesel exhaust: C_{20}, C_{21}) (Simoneit, 1984, 1989). The contribution of petroleum to the total concentrations of *n*-alkanes was mentioned above, based on CPI values. Probably, the *n*-alkanes on Belukha glacier are mainly derived from higher plant wax and petroleum (gasoline and power-plant but not diesel) exhaust. However, from their homologue distributions in these samples, we speculate that contributions of *n*-alkanes from microbial activity and diesel-engine exhaust to the total *n*-alkanes concentrations are relatively small compared with other petroleum exhaust and higher plant.

3.3. Vertical profile of *n*-alkanes and environmental variation around the Altai

Figure 4 shows the vertical profiles of T-HCs and the CPI values of *n*-alkanes in the ice-core samples. The WaxC_n and non-WaxC_n portions of T-HCs are also shown. We can see that T-HCs gradually increased from the past toward the present by an increase in non-WaxC_n of *n*-alkanes. This is concordant with a decrease in CPI values in the upper layers of the ice core, suggesting that the intensity of anthropogenic sources of *n*-alkanes has recently increased.

Fujita and others (2004) reported that the snow accumulation rates on Belukha glacier were 1.60–2.26 m a^{-1} from 2000 to 2003. According to these accumulation rates, the top layer (2.20–5.21 m depth) of our ice core is estimated to have accumulated during 1999 and 2000. Miyake and others (2005) reported that the annual deposition fluxes of *n*-alkanes on Sofityskiy glacier have recently increased, especially from 1994 to 2000. It seems that the loading of *n*-alkanes from anthropogenic sources should have increased recently around the Altai, reflecting an increase in anthropogenic activities in this area. It is difficult to identify the source regions of *n*-alkanes in ice-core and snow samples from Belukha glacier. However, we speculate that *n*-alkanes on the glacier are derived mainly from the vicinity and areas to the west, because westerlies are predominant in this area (Seppälä, 2004).

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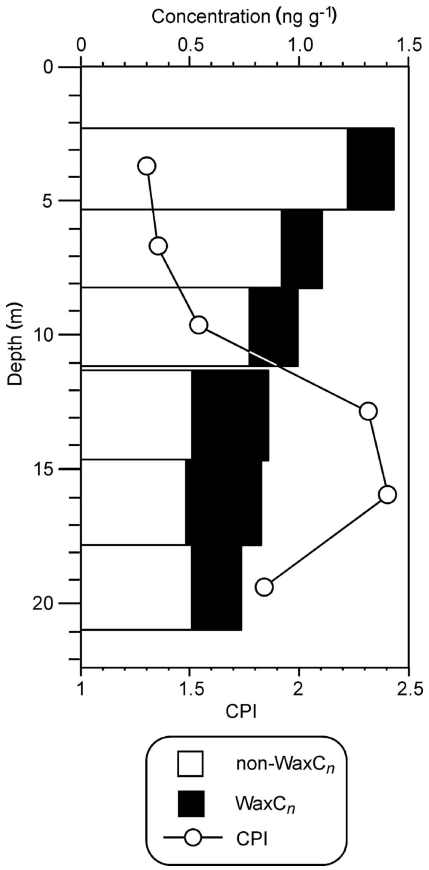

Fig. 4. Vertical profiles of total concentrations (T-HCs; axis scale at top) and CPI values (axis scale at bottom) of *n*-alkanes in ice-core samples on Belukha glacier. The CPI values are shown as open circles. The $WaxC_n$ (filled column) and non-$WaxC_n$ (open column) portions of T-HCs are also shown. The width of the filled rectangle is concentration of $WaxC_n$.

4. CONCLUSIONS

1. The T-HCs and CPI values of *n*-alkanes in ice and snow samples collected from Belukha glacier ranged from 0.52 to 1.43 ng g^{-1} and from 1.19 to 3.07, respectively. By comparing these values to those of ice and snow samples from the Altai, Himalaya and Greenland, it is suggested that the glaciers on the Asian continent receive higher loads of *n*-alkanes from natural and anthropogenic sources than the Greenland ice sheet.
2. It appears that the loading onto and variation of *n*-alkanes on Belukha and Sofiyskiy glaciers are approximately equal to those in the Altai mountains overall, because the concentrations, CPI values and $WaxC_n$ percentages of *n*-alkanes in Sofiyskiy glacier are the same as or slightly greater than in ice-core and snow samples on Belukha glacier.
3. From their homologue distributions in these samples, the *n*-alkanes at Belukha glacier are mainly derived from higher plant wax and petroleum (not including diesel) exhaust.
4. The non-$WaxC_n$ portion and T-HCs have recently increased, and CPI values have decreased. A similar trend of *n*-alkanes was also reported for Sofiyskiy glacier, indicating that the influence of human activities has gradually increased around the Altai.

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