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# Post-depositional enrichment of black soot in snow-pack and accelerated melting of Tibetan glaciers

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**Abstract**
The post-depositional enrichment of black soot in snow-pack was investigated by measuring the redistribution of black soot along monthly snow-pits on a Tien Shan glacier. The one-year experiment revealed that black soot was greatly enriched, defined as the ratio of concentration to original snow concentration, in the unmelted snow-pack by at least an order of magnitude. Greatest soot enrichment was observed in the surface snow and the lower firn-pack within the melt season percolation zone. Black carbon (BC) concentrations as high as 400 ng $g^{-1}$ in the summer surface snow indicate that soot can significantly contribute to glacier melt. BC concentrations reaching 3000 ng $g^{-1}$ in the bottom portion of the firn pit are especially concerning given the expected equilibrium-line altitude (ELA) rise associated with future climatic warming, which would expose the dirty underlying firn and ice. Since most of the accumulation area on Tibetan glaciers is within the percolation zone where snow densification is characterized by melting and refreezing, the enrichment of black soot in the snow-pack is of foremost importance. Results suggest the effect of black soot on glacier melting may currently be underestimated.

**Keywords:** black soot, enrichment, glacier melting

## 1. Introduction

Black soot deposited on snow surfaces reduces albedo by absorbing more sunlight, which may exacerbate warming (Hansen and Nazarenko 2004, Flanner and Zender 2005, Flanner *et al* 2007, 2009, McConnell1 *et al* 2007) and glacier melting (Warren and Wiscombe 1985, Hansen and Nazarenko 2004, Qian *et al* 2011). Black carbon (BC) concentrations of only 10 ng $g^{-1}$ or more can significantly reduce snow albedo (Warren and Wiscombe 1985, Hansen and Nazarenko 2004, Flanner *et al* 2009), and BC concentration of 15 ng $g^{-1}$ in snow is sufficient to reduce albedo by 1% (Warren and Wiscombe 1980). As BC concentrations reach 500 ng $g^{-1}$, the albedo reduction is in the range of 5–17% (Warren and Wiscombe 1985) causing a 50% increase of snow melting (Conway *et al* 1996).

As the Asian water tower (Immerzeel *et al* 2010) and the third largest reservoir of ice after the poles, the Tibetan

 

Plateau (TP) holds 46 298 glaciers with total glacial area of 59 406 km$^2$, and total glacial volume of 5590 km$^3$ in China (Yao *et al* 2004). The largest glaciated regions are extremely important freshwater reservoirs, including those in the Tien Shan, Karakoram and Kunlun mountains which release about 137.7 $\times$ 10$^8$ m$^3$ of meltwater to the lower reaches of the Tarim Basin each summer (Yao *et al* 2004). Glaciers have been melting at an accelerated rate (Yao *et al* 2004, Kehrwald *et al* 2008), and temperature on the TP has increased at twice the rate of the observed global warming over the last three decades (Xu *et al* 2009). Black carbon was recently proposed as a major cause for the observed dramatic changes by both heating the high elevation atmosphere (Ramanathan *et al* 2007) and decreasing albedo after being deposited on glaciers (Xu *et al* 2009).

It was identified that Asia is currently the largest source of black soot emissions in the world (Novakov *et al* 2003, Bond *et al* 2007, Ohara *et al* 2007), and the TP and adjacent mountain regions are affected by heavy air pollution containing a large fraction of black and organic carbon (OC, here refers to water insoluble organic carbon). The extensive soot aerosols can be lifted to high altitudes (Ramanathan *et al* 2007) and reach the glaciers incorporated in snowflakes. Ice cores drilled at high elevations have shown a rapid increase of black soot in the monsoon region since the 1990s (Ming *et al* 2008, Xu *et al* 2009) or an even earlier increase since 1975 (Kaspari *et al* 2011). Studies revealed that the effects of black soot dominate in the winter and spring due to its high concentration both in air (Marinoni *et al* 2010) and snow cover in the TP region (Flanner *et al* 2007, Xu *et al* 2009). However, the enrichment of black soot in snow-pack with summer melting may more efficiently absorb sunlight. Surface snow samples from widely spaced glaciers on the TP revealed a large and variable range of BC and OC concentrations; from several parts per billion to parts per million from site to site (Xu *et al* 2006, Ming *et al* 2009, Huang *et al* 2011). This variability mainly reflects differences in black soot concentrations between fresh snow and aged snow, each with different degrees of enrichment. Thus, to quantify the effect of black soot on the melting of TP glaciers, sampling of BC and OC concentrations in different seasons is needed, since the surface snow contains varying amounts of both BC and OC which differ in their sunlight absorbing effectiveness in absorbing sunlight and therefore have varying impacts on melting.

## 2. Experiment and methods

To investigate post-depositional black soot enrichment in snow-pack, snow/firn-pits were sampled monthly in the percolation zone, where some meltwater penetrates down and refreezes in the glacier, of Urumqi glacier no.1 (UG1, 43°06′N, 86°49′E), Tien Shan Mountains, above the ELA at 4130 m asl (figure 1). Snow samples were collected from the pit wall from the glacier surface to the interface of firn and ice. The floor of the firn-pack at this site is composed of superimposed ice (melted and refrozen snow/firn) that is typically formed in the glacial percolation zone. A total of 13

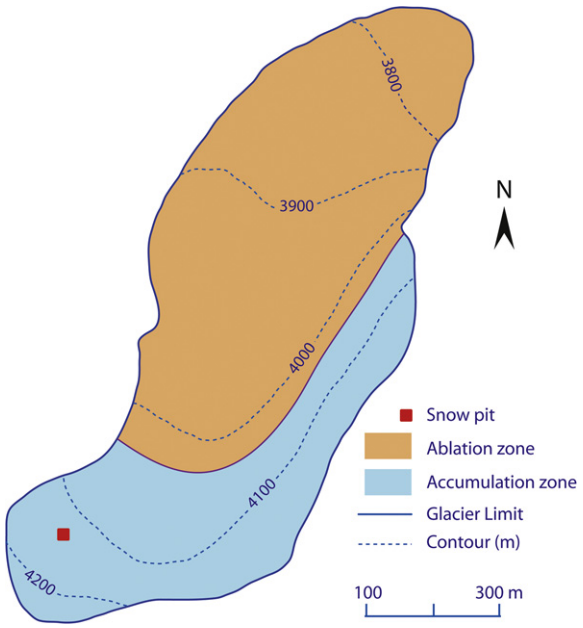


**Figure 1.** Map showing the sites of snow-pits on the east branch of glacier no.1 of the Urumqi River, Tien Shan Mountains.

monthly samplings of the firn pit wall were performed and 77 snow samples were obtained from 28 July 2004 to 26 July 2005. All the collected samples were preserved in glass bottles with a volume of 300 ml, pre-cleaned using a liquid mixture of $H_2O$–$K_2Cr_2O_2$–$H_2SO_4$ (25:1:46) and pure water (Milli-$Q$, 18.2 MΩ). Sample bottles were wrapped with aluminum foil and transported frozen to the State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environmental and Engineering Research Institute in Lanzhou.

In the laboratory, snow samples were weighed and melted at room temperature in a class 100 clean room, and immediately filtered through quartz fiber filters which were pre-heated in an oxygen stream for at least 2 h in a tube oven with temperature of 800 °C. The water samples were filtered twice, and both the containers and filtration unit were rinsed three times with ultrapure water (Milli-$Q$, 18.2 MΩ; Millipore) to ensure complete transfer of particles to the filters (Xu *et al* 2006). To avoid possible positive BC artifacts that would result from carbonate production of $CO_2$, carbonates were removed before analysis by dripping 50 $\mu$l of 0.1 M HCl onto the sample spot three times (Lavanchy *et al* 1999).

BC and OC on the filters were measured by using the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance protocol (Chow *et al* 2004). The analytical uncertainty was assessed to be 15% for BC and 16% for OC (Xu *et al* 2009).

## 3. Result and discussion

Snow and black soot stratigraphies are shown in figure 2. BC and OC concentrations were largely variable within each snow-pit, rather than uniformly mixed. BC concentrations varied from 11 to 3000 ng g$^{-1}$, while OC varied from 49 to 8370 ng g$^{-1}$. Each variable range exceeds two orders

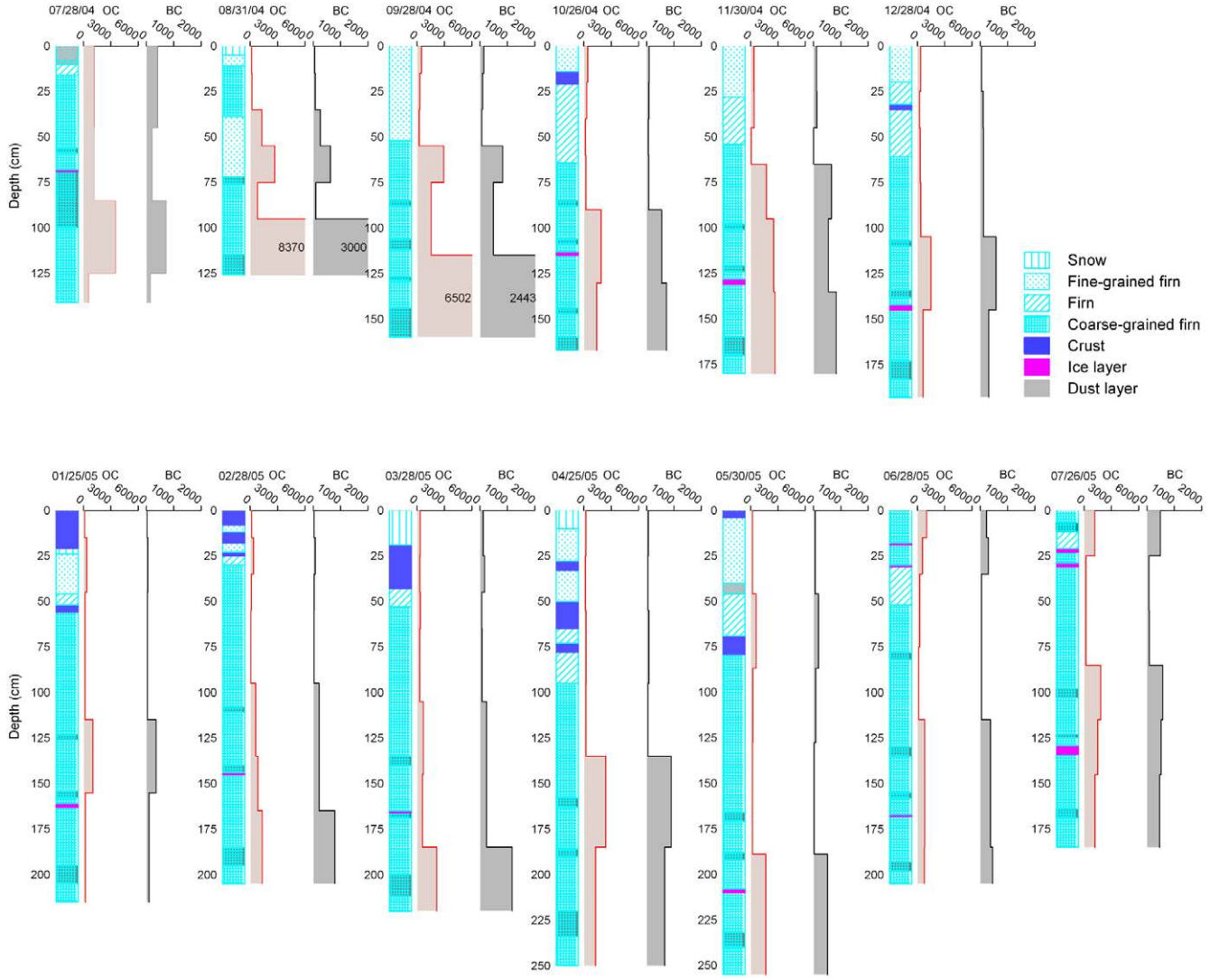


**Figure 2.** Visual stratigraphy of snow-packs shown together with OC and BC concentrations.

of magnitude, and outstrips the maximum difference of BC amounts in surface snow on TP glaciers from site to site (Xu *et al* 2006, Ming *et al* 2009). It is shown in the stratigraphies that the lower part of each snow-pit, which experienced a longer time of densification or melting, contained higher BC and OC concentrations, but higher concentrations were also found in surface snow during summer. The data show that black soot was redistributed in the snow-pack due to melting, resulting in enrichment in unmelted snow-packs both in surface snow and in the bottom firn. For the purpose of this letter, we use the bottom firn to refer to the coarse-grained firn at the lowest depths of the firn pit directly above the interface with the glacier ice (figure 2).

BC and OC concentrations in surface snow and in the bottom firn for each snow-pit are extracted and shown in figure 3, together with snow-pack thickness, monthly average air temperature and precipitation. Although the amount of precipitation increased rapidly, the snow-pack thinning reveals that strong snow melting occurred during summer as temperature increased. This melt resulted in a remarkable BC and OC accumulation in surface snow, with the exception of the sample collected on 31 August 2004 which was collected while snow was falling. Immediately below the fresh snow, the enriched black soot layer from melting snow was found at a depth of 35–55 cm where BC concentrations were as high as 255 ng $g^{-1}$. In general, BC and OC concentrations in surface snow during summer can reach 250–500 and 1000–1200 ng $g^{-1}$, respectively. Snow melting was weaker during autumn and spring as revealed by the increased snow-pack thickness, and accordingly BC concentrations in surface snow decreased to 60–150 ng $g^{-1}$ for BC and 200–500 ng $g^{-1}$ for OC. Although precipitation was infrequent during winter, the snow-pack thickness was maintained due to very low temperatures. The winter concentrations in surface snow, 27–31 ng $g^{-1}$ for BC and 136–153 ng $g^{-1}$ for OC, may therefore be similar to the original concentrations incorporated in snowflakes. This implies that the BC and OC in the surface snow have no enrichment in winter.

The highest BC and OC concentrations were found in the bottom firn during late summer and early autumn, when BC and OC concentrations reached 2440–3000 ng $g^{-1}$ and

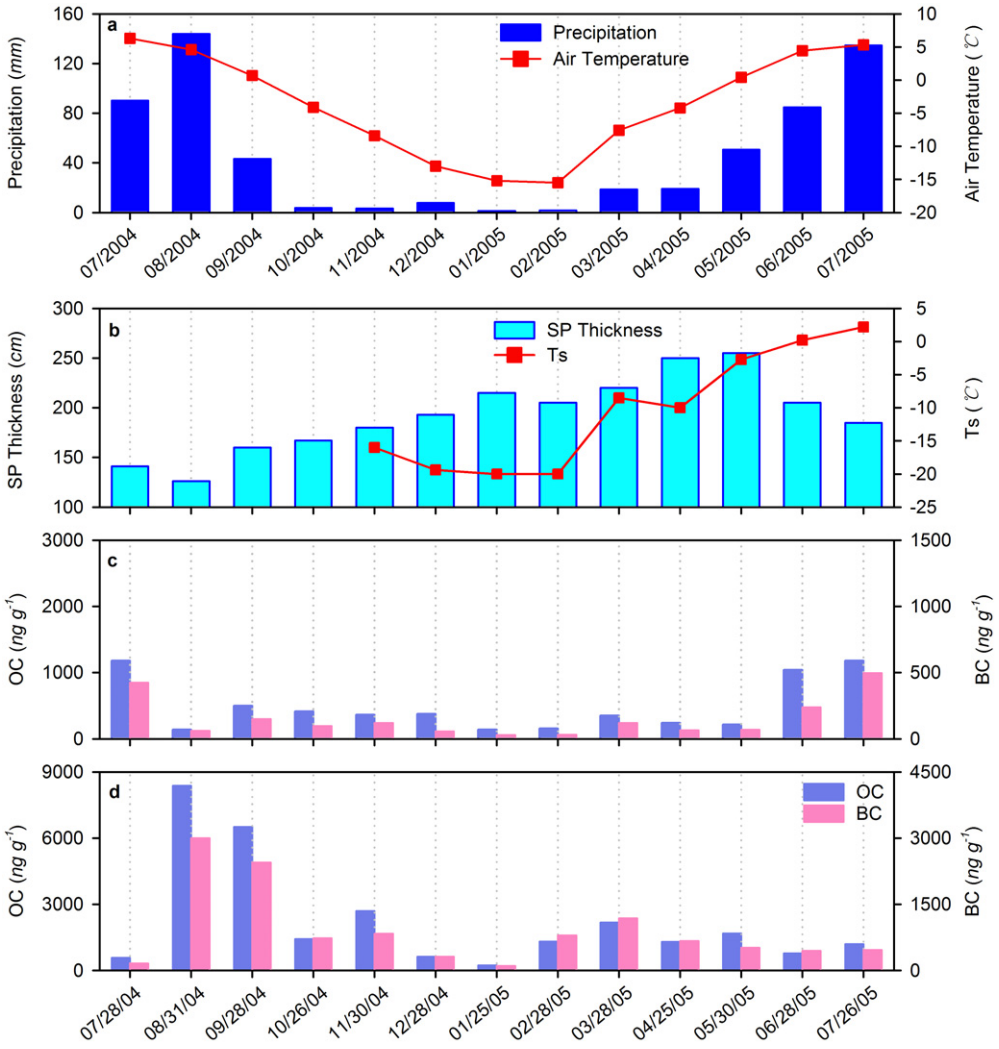


**Figure 3.** (a) Monthly precipitation and average air temperature at an elevation of 3539 m asl; (b) snow-pack (SP) thickness and surface snow temperature ($T_s$) measured during sampling times; (c) BC and OC concentrations in surface snow, note that the surface snow sample taken on 31 August 2004 was fresh snow which diluted BC and OC concentrations; (d) BC and OC concentrations in the bottom firn.

6500–8370 ng $g^{-1}$, respectively (figure 3). This indicates the downward percolation of meltwater redistributed black soot in the snow-pack. BC concentration levels in surface snow are dependent upon melt strength; strong melt can transport soot particles downwards through the snow-pack resulting in decreased BC concentrations in surface snow (Conway *et al* 1996), while weaker melt can result in elevated BC concentrations (Wang *et al* 2012). Results presented here are from snow-pits located in the percolation zone, where the summer surface snow melt is not strong enough to elute surface BC. At certain depths, typically within the depth interval of 30–110 cm, meltwater has flushed BC through the snow-pack. The result is a decrease in BC concentrations at the intermediary depths (figure 2). At the lowermost layers of the snow-pit, further downward percolation of meltwater is obstructed by the superimposed ice which formed in the previous year, causing a buildup of BC. It is noted that the black soot enrichment in the bottom firn lags that of the surface snow by two months; this may be the percolation time of meltwater in the snow-pack. During mid-autumn, the bottom firn filled with meltwater carrying black soot. When the meltwater refroze and formed superimposed ice due to the temperature decrease, the subsequent snow-pit and the bottom firn above the glacier ice contained less black soot, as observed in figure 3(d). Data indicate that the glacier ice (here it is the superimposed ice) underlying the snow-pack formed in the middle of autumn contained a large amount of BC and OC, which exceeded 2400 and 6500 ng $g^{-1}$, respectively. Less snow meltwater and black soot were found in the bottom firn between the mid-autumn to mid-summer months.

Seasonal enrichment of BC and OC in surface snow and the bottom firn are shown in figure 4, plotted as the ratio of the seasonal average concentrations divided by the average concentration of OC or BC in surface snow during the coldest period of January and February. The months of January and February were chosen to calculate enrichment since these months are least influenced by melting and can therefore best represent original snow concentrations. Surface enrichment was highest during summer (June–August); it was 13 times higher for BC and eight times higher for OC.

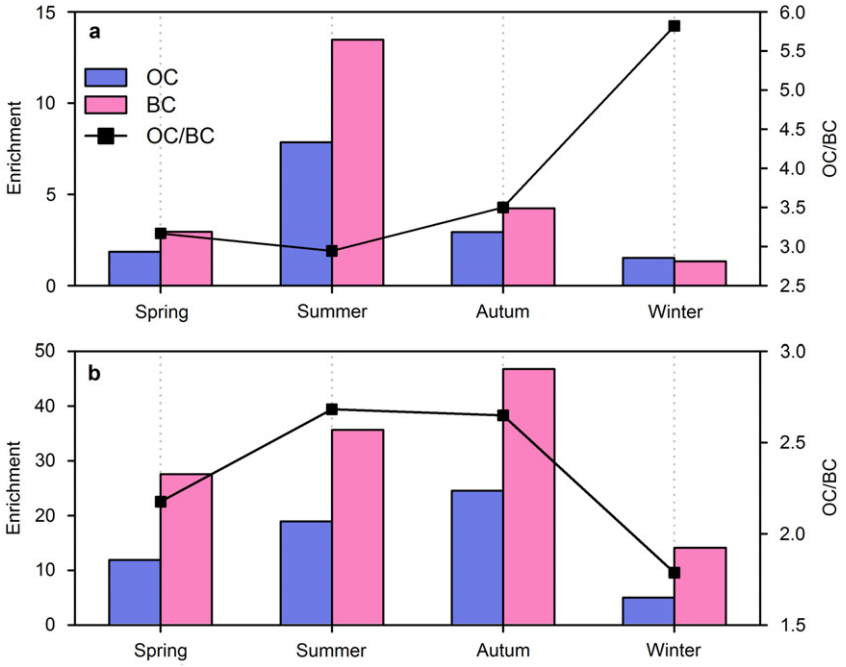


**Figure 4.** Seasonal enrichment of BC and OC, and the changes of OC/BC ratios in surface snow (a) and the bottom firn (b). Enrichment is calculated as the ratio of seasonal average concentration to the January and February average surface snow concentration.

Enrichment in surface snow during spring (March–May) and autumn (September–November) was also remarkable; it was 3–4 times higher for BC and 2–3 times higher for OC. Relative to OC, BC is more inclined to enrichment in unmelted snow as revealed by the changes of OC/BC ratios. The highest OC/BC ratio of 5.8 was found in winter (December–February) surface snow, but it decreased to 2.9 in summer when the surface melting was the most intense. This deficit of OC in unmelted snow reveals that OC is more easily washed away with meltwater compared to BC, resulting in a large reduction of the OC/BC ratio. BC enrichment in the bottom firn was prominent in all seasons, but there were large seasonal variations for both BC and OC, following the same descending order of autumn > summer > spring > winter. The remarkable enrichments of BC in the bottom firn were caused partly by the long melt time period, characterized by higher OC/BC ratios. The percolation of meltwater should be considered as the most important factor, especially in summer and autumn when a large amount of meltwater filled the bottom firn as identified by the increase of OC/BC ratios.

In addition to the rapid increase of black soot emissions in Asia (Novakov *et al* 2003, Bond *et al* 2007, Ohara *et al* 2007) and subsequent deposition on glaciers incorporated in snowflakes in the TP region (Ming *et al* 2008, Xu *et al* 2009, Kaspari *et al* 2011), the mechanism of mutual reinforcement between snow melting and increased black soot concentrations in snow-pack, as shown from these results, further suggests that black soot is an important factor in the melting of TP glaciers. The radiative effect of the up to 400 ng $g^{-1}$ of BC concentration, which is an enrichment of an order of magnitude or more in surface snow during summer, must not be ignored. Unlike the large dry-snow zones of the polar ice sheets, where snow densification is characterized by compaction, most of the accumulation area on TP glaciers is a percolation zone where snow quickly densifies by melting and refreezing, except at very high altitudes. Thus the surface enrichment of black soot in the snow-pack is a prevalent phenomenon for TP glaciers. The melting-induced densification causes black soot to accumulate in the bottom firn of a snow-pack, because the superimposed ice under the snow-pack prevents further meltwater infiltration. This enrichment is critically important for the future, because the ELA's of all glaciers on TP is expected to continue rising due to the coupled impacts of greenhouse-gas warming and black soot enrichment in surface snow. This will result in the dirty ice, formed at present in the accumulation zone underlying the snow-pack, to be exposed with an ELA increase. In this respect, the present and future impacts of black and organic carbon on glacier melting may be underestimated.

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