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Levoglucosan evidence for biomass burning records over Tibetan glaciers☆

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ABSTRACT

Intense biomass burning (BB) events are widespread in tropical and subtropical Asia. However, the impact of BB aerosols on the Tibetan Plateau (TP), especially on Tibetan glaciers, is poorly understood. In this study, BB signals are revealed using the specific molecular tracer levoglucosan in snow and ice samples from different Tibetan glaciers. Tibetan glaciers mainly act as receptors of BB emissions from surrounding regions. Significant differences in levoglucosan concentrations in glacier samples collected from two slopes on the same mountain range indicate that high mountains can act as natural barriers to block the transport of smoke aerosols to the TP. Levoglucosan concentrations show a decreasing trend from west to east on glaciers impacted by the Indian summer monsoon on the southern edge of the TP, while the opposite pattern was observed on glaciers under the prevailing westerlies along the northern edge. The emission sources, the controlling climate system, as well as deposition and degradation during transport determined the spatial distribution regimes of levoglucosan concentration on Tibetan glaciers.

1. Introduction

Biomass burning (BB) emissions are one of the main sources of atmospheric aerosols (Simoneit et al., 1999; van der Werf et al., 2010; Venkataraman et al., 2006), contributing over two thirds of the total carbonaceous aerosols in regions with intense BB (e.g. the Indian Peninsula) (Engling and Gelencser, 2010; Gustafsson et al., 2009; Venkataraman et al., 2005). Smoke particles from BB emissions are considered to be a major cause of the South Asia Brown Clouds over the Indian peninsula (Engling and Gelencser, 2010; Gustafsson et al., 2009). Smoke plumes from intense BB events can be transported to the upper troposphere and lower stratosphere by strong thermal convection processes (Fu et al., 2008; Kahn et al., 2008; Vadrevu et al., 2012), where they can potentially be transported to remote high elevation regions like the Tibetan Plateau (TP).

The main body of the TP is enclosed by high-elevation mountains (e.g. the Himalayas to the south and the Kunlun to the north, see Fig. S1) (Xu et al., 2015), which can substantially interrupt the transport of atmospheric pollutants from surrounding regions (Marinoni et al., 2010; Qiu, 2013; Xu et al., 2009a, 2014). However, the TP is located very close to regions such as South and Southeast Asia (Fig. 1) which are considered as some of the world's most intense BB emission sources (Venkataraman et al., 2005, 2006). Both observational and modeling results have indicated that BB emissions could easily be transported to the high-elevation marginal regions of the TP such as the Himalayas (Engling et al., 2011; Qiu, 2013; Xu et al., 2014). Smoke aerosols released from intense BB events could be even transported up and over the Himalayas (Cong et al., 2015; Xu et al., 2014, 2015), and penetrate central TP regions such as the Namco Lake Basin (Xia et al., 2011) and the Qinghai Lake Basin (Wang et al., 2015b).

Carbonaceous aerosols can significantly reduce snow albedo on glacier surfaces, and are considered as important contributors to the accelerating melt of Tibetan glaciers, besides rising temperature, in recent years (Ming et al., 2008; Wang et al., 2015a; Xu et al., 2009a). This has had a significant impact on water resources for the

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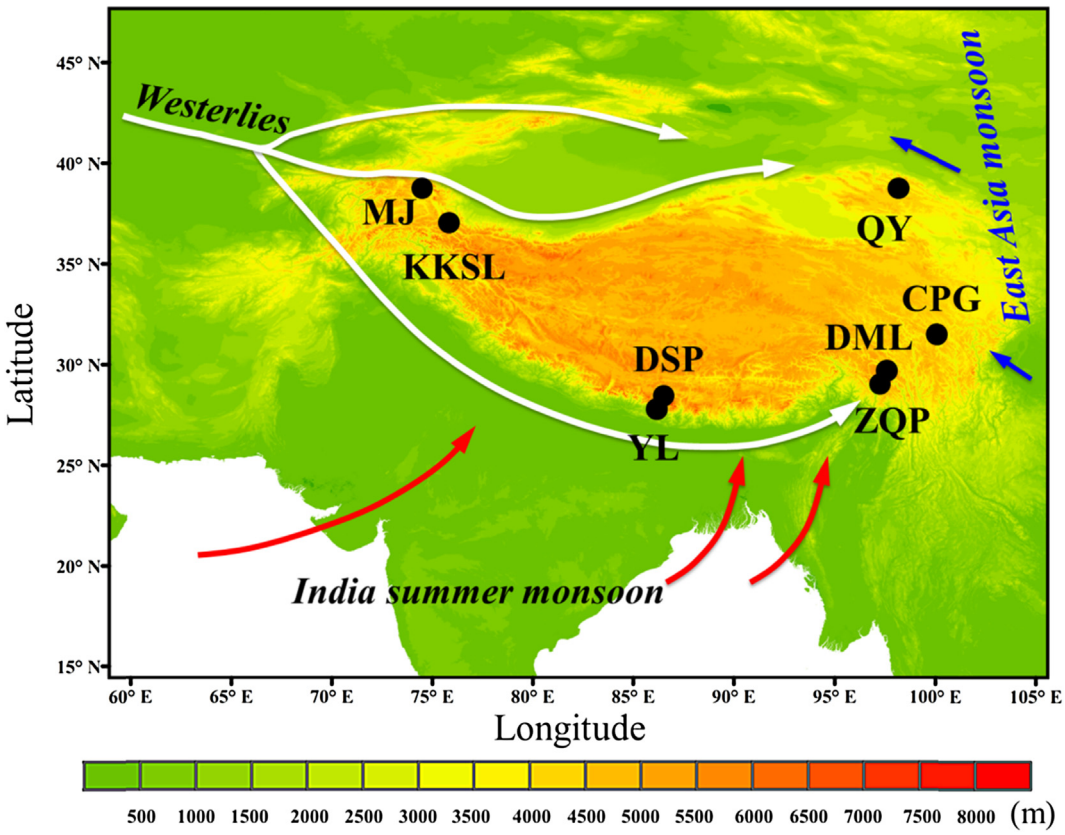

Fig. 1. Atmospheric circulation features around the TP, and sampling sites (the black dots) in this study.

surrounding regions (Ming et al., 2013; Xu et al., 2009b). Geochemistry evidence indicates that carbonaceous aerosols deposited on Tibetan glaciers are mainly from fossil fuel emissions (Wang et al., 2015a; Xu et al., 2009b). Other studies found that Tibetan glaciers were contaminated by BB aerosols at different levels (Xu et al., 2013; Yao et al., 2013; You et al., 2014). Levoglucosan is a specific biomarker for BB, because it can only be generated by combustion of cellulose and hemicellulose when the combustion temperature is higher than 300 °C (Simoneit et al., 1999). The atmospheric life time of levoglucosan varies from several hours to more than ten days under different conditions, in particular depending on the concentrations of the hydroxyl radical (.OH) (Hennigan et al., 2010; Hoffmann et al., 2010). Experimental studies have reported that levoglucosan can react with. OH when exposed in conditions of high relative humidity conditions (Hennigan et al., 2010), but it can normally remain stable for several days under most atmospheric conditions during the transport process (Fraser and Lakshmanan, 2000; Hu et al., 2013; Mochida et al., 2003). Studies have also reported that levoglucosan displays almost negligible degradation in snow and ice layers when under frozen deposition conditions (Gambaro et al., 2008; Kehrwald et al., 2012). Therefore, it was suggested that levoglucosan could be used as an ideal tracer for BB in snow and ice records from polar and high altitude sites (Gambaro et al., 2008; Kawamura et al., 2012; Kehrwald et al., 2012; You et al., 2014).

High levoglucosan concentrations were detected in aerosol samples at sites in the marginal regions of the TP (Cong et al., 2015), which were shown to have been affected by smoke plumes from BB more seriously than the Arctic (Fu et al., 2009; Stohl et al., 2007) and Antarctic (Zangrando et al., 2016) atmospheric aerosol samples. Tibetan glaciers have also been more seriously contaminated by BB emissions when compared with polar regions (Yao et al., 2013; You et al., 2014, 2016). However, knowledge of the spatial distribution of BB aerosols on Tibetan glaciers is still unclear due to limited observations. In this study, the spatial distribution of levoglucosan is investigated using snow and ice samples from glaciers in different TP regions. Based on these results, the sources and process of BB aerosols transported to Tibetan glaciers are discussed.

2. Materials and methods

2.1. Sample collection and preparation

Surface snow samples were collected from seven glaciers (Yala Glacier (YL), Dasuopu Glacier (DSP), Zuoqiupu Glacier (ZQP), Demula Glacier (DML) on the southern TP; the Muji Glacier (MJ), Kuokuosele Glacier (KKSL) and Qiyi Glacier (QY) on the northern TP) within different climate control regions of the TP (Table 1 and Fig. 1) from May to November 2012. Additional snow-pit samples were collected from the MJ Glacier with a vertical resolution of 5 cm and the ZQP Glacier with a vertical resolution of 20 cm (Table S3). The polyethylene terephthalate (PET) bottles were washed twice by soaking in ultrapure water for at least 24 h and rinsing three times with ultrapure water. These were dried in the shade in a clean room (You et al., 2014). Disposable polyethylene gloves were used for snow sample collections (You et al., 2014, 2016). Snow samples were compacted in pre-cleaned PET bottles and were kept frozen at a temperature of −20 °C before chemical analysis. A shallow ice core with a length of 13.50 m was drilled at an altitude of 5463 m above sea level (a.s.l) in the accumulation area from the Cuopugou (CPG) Glacier on the eastern TP on November 14th, 2014. Ice core sections were transported frozen to Lhasa, and were stored in a cold