Tracking Local and Regional Influences of Potassium at the Korean Peninsula under Asian Continental Outflows during Spring

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ABSTRACT

The atmospheric stability of biomass-burning tracers such as K^+ and levoglucosan is important in their application in assessing the impact of biomass-burning emissions in areas downwind of Asian continental outflows. It was found that levoglucosan in tested aerosol samples degraded ~48% at a suburban site in Daejeon, Republic of Korea within 31 h of exposure to ambient atmospheric conditions during spring, while K^+ was stable under the same conditions. Analysis of aerosol samples collected at the same site during spring 2013 revealed that SO_4^{2-} concentrations were moderately correlated with K^+ concentrations, but not with levoglucosan concentrations. Air-mass backward-trajectory analysis showed that high SO_4^{2-} concentrations at the sampling site were attributable mainly to long-range transported aerosols from the Asian continent. The poor correlation between levoglucosan and SO_4^{2-} suggests that negligible amounts of levoglucosan might be transported from the Asian continent to downwind area of the Korean Peninsula during spring mainly due to the rapid degradation of levoglucosan during the long-range atmospheric transport. This result implies that levoglucosan is not a suitable tracer of biomass burning in area downwind of the Asian continental outflows during spring season. Levoglucosan/K^+ ratios on the Asian continent and downwind on the Korean Peninsula indicate that ~39% of atmospheric K^+ in Daejeon during spring 2013 was attributable to long-range transported aerosols from the Asian continent.

Keywords: Biomass-burning tracer, Levoglucosan, Potassium, Long-range transport

1 INTRODUCTION

Biomass burning, which includes wildfires, permitted burnings, agricultural waste burnings, and domestic biofuel combustion, emits large amounts of gaseous and particulate pollutants to the atmosphere (Andreae and Merlet, 2001; Jung et al., 2016; Zhou et al., 2017; Du et al., 2018; Wu et al., 2018). Biomass burning is an important contributor to carbonaceous aerosols over the Asian continent (Cheng et al., 2014; Jung et al., 2014; Shang et al., 2017; Liu et al., 2019; Fan et al., 2020; Wang et al., 2020). Liu et al. (2019) reported that 20.9% of organic carbon (OC) originated from biomass burning in Nanjing, China during winter 2015. Fan et al. (2020) reported that biomass burning was the predominant source of primary OC at night (16.0% ± 6.9%) in Tianjin, China, during winter 2015. Jung et al. (2014) estimated that 45% and 12% of OC and elemental carbon (EC), respectively, originated from biomass burning in Daejeon, Korea, during the rice harvest period in 2012.

Because the Korean Peninsula is located downwind of Asian continental outflows, aerosol particles comprise a mixture of locally and distally emitted pollutants during winter and spring (Jung et al., 2015; Kim et al., 2018; Bhardwaj et al., 2019). Severe haze episodes over the Korean Peninsula generally result from long-range transported aerosols, with additional aerosols produced locally under stagnant atmospheric conditions (Jung et al., 2019). However, the relative local and regional contributions to severe haze episodes over the Korean Peninsula are not well understood.
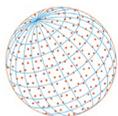
Qualitative estimates of the contribution of biomass burning to ambient aerosol concentrations require the use of biomass-burning tracers. Commonly used tracers are levoglucosan and K^+ (Andreae and Merlet, 2001; Simoneit, 2002; Giannoni et al., 2012). Levoglucosan is produced by pyrolysis of cellulose at temperatures above 300°C (Simoneit et al., 1999; Achad et al., 2018), and is not emitted from other sources. Although a small fraction of atmospheric K^+ originates from sea salt and resuspended soil (Pio et al., 2008), most is emitted by biomass burning in the form of KCl, K_2SO_4, and KNO_3 (Li et al., 2003). Emission rates of levoglucosan and K^+ depend on various factors such as the type of biomass (e.g., softwood, hardwood, or crop residues), where it is burnt (e.g., traditional stoves or field burning), the type of burning (e.g., smoldering or flaming), and the burning season (Fu et al., 2012; Cheng et al., 2013; Jung et al., 2014).

Quantitative determination of the contribution of biomass burning to ambient aerosols requires an understanding of the stability of K^+ and levoglucosan in the atmosphere and in aerosol samples collected on air-sampling filters, with K^+ being considered more stable than levoglucosan. Laboratory experiments indicated that levoglucosan was degraded as a function of cumulative OH exposure with a typical lifetime of 0.7–2.2 days, via photo-oxidative degradation with increasing ambient temperature (Hennigan et al., 2010). Hoffmann et al. (2010) reported that levoglucosan in deliquescent particles is readily oxidized by OH radicals during daytime with mean degradation rates of about 7.2 and 4.7 ng m^{-3} h^{-1} in summer and winter, respectively, for a polluted continental plume. There have been no previous studies of the stability of biomass-burning tracers downwind of Asian continental outflows.

In this study, the stabilities of levogulcosan and K^+ were investigated experimentally using aerosol samples collected at a suburban site in Daejeon, Republic of Korea during spring 2013. Based on correlation analyses between concentrations of biomass-burning tracers and SO_4^{2-}, potential source regions of levoglucosan and K^+ were investigated, and local and regional contributions of the tracers were quantified.

2 EXPERIMENTAL METHODS

2.1 Aerosol Sampling and Analysis

Daily PM$_{10}$ (particulate matter with an aerodynamic diameter of 10 μm or less) sampling was carried out at a suburban site (36°23′19″N, 127°22′21″E) in Daejeon, Korea, during spring 2013 (15 April–23 May). PM$_{10}$ aerosol particles were collected on pre-baked quartz fiber filters (20 × 25 cm, Pall-Life Sciences, Ann Arbor, MI, USA) using a high-volume air sampler (PM10HVPLUS, Tisch Environment Inc., Cleves, OH, USA) at a flow rate of 67.8 m^3 hr^{-1}. The sampler was installed on a rooftop ~15 m above ground-level at the Korea Research Institute of Standards and Science (KRISS). Before and after sampling, filter samples were wrapped with aluminum foil and stored in 'zip-lock' plastic bags (3M, SCC1500) at −20°C.

Ultrapure water used in this study was prepared using a Labpure S1 filter with a UV lamp, with resistivity and total organic carbon (TOC) content of 18.2 MΩ cm and 1 ppb, respectively (ELGA, PureLab Ultra). To determine levoglucosan and water-soluble ion concentrations, a portion (2.01 cm^2) of each filter sample was extracted with 10 mL ultrapure water under ultrasonication (for 30 min). Water extracts were then filtered through a disk filter (Millipore, Millex-GV, 0.45 μm) and stored in a refrigerator at 4°C before analysis.

Levoglucosan was analyzed by an improved high-performance anion-exchange chromatography (HPAEC) method with pulsed amperometric detection (PAD), using a CarboPak MA1 analytical column (4 × 250 mm) with an eluent of 360 mM sodium hydroxide solution at a flow rate of 0.4 mL min^{-1}. The analytical method has been explained in detail by Jung et al. (2014). The detection limit of levoglucosan was 3.0 ng m^{-3}. The recovery of levoglucosan was higher than 95%.

Concentrations of Na$^+$, K$^+$ and SO$_4^{2-}$ ions in water extracts were determined by ion chromatography (Thermo, Dionex ICS-5000), involving an IonPac CS-12A column (4 × 250 mm) for Na$^+$ and K$^+$ with an eluent of 38 mM methanesulfonic acid (MSA) at a flow rate of 1.0 mL min^{-1}; and an IonPac AS−15 column (4 × 250 mm) for SO$_4^{2-}$ with an eluent of 40 mM KOH at a flow rate of 1.2 mL min^{-1}. The detection limits of Na$^+$, K$^+$ and SO$_4^{2-}$ were 0.01 μg m^{-3}. Because marine source of K$^+$ quantified using the mass ratio of K$^+$ to K$^+$ in seawater (0.036) only contributed 0.3% to the total K$^+$, marine source of K$^+$ was not considered in this study.
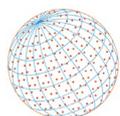
Concentrations of OC and EC in filter samples were determined using a thermal optical OC/EC analyzer (Sunset Laboratory Inc. Forest Grove, OR, USA) with laser-transmittance-based correction for pyrolysis. Details are reported elsewhere (Jung *et al.*, 2014). In brief, a portion (1.5 cm^2) of filter was placed in a quartz dish inside the thermal desorption oven of the analyzer. OC and EC concentrations were determined using a temperature program developed by the US National Institute for Occupational Safety and Health (NIOSH) in an inert atmosphere (100% He) or in an oxidizing atmosphere (98% He + 2% O$_2$), respectively. Detection limits of both OC and EC, which are defined as three times the standard deviation of field blanks, were 0.04 μg C m^{-3}.

2.2. Levoglucosan and K$^+$ Stability Experiments

To investigate the stability of biomass-burning tracers such as levoglucosan and K$^+$, aerosol samples collected on quartz filters were exposed to either an inert nitrogen atmosphere in the dark at 50 °C or to the ambient atmosphere. Four PM$_{10}$ samples collected at the same site during winter 2012 and spring 2013 were used in this experiment. Thermal stability of the tracers was assessed by placing filter portions (3.14 cm^2) in 50 mL amber glass vials filled with pure nitrogen, with the vials then being tightly capped (Fig. 1(a)). The capped vials were then placed in an oven at 50°C for 9 h. Samples were analyzed for levoglucosan and K$^+$ before and after thermal treatment as described in Section 2.1.

Other filter portions were placed in clear glass vials as shown in Fig. 1(b), with these vials then being placed on a rooftop and exposed to ambient conditions. Two sets of experiments were conducted during 22–23 May 2014. Meteorological conditions during the experiment were obtained from a nearby observatory (~5 km from the sampling site) of Korea Meteorological Agency, as summarized in Table 1. In one experiment, the samples were exposed for 9 h during daytime (9:00–18:00 LT), with an average ambient temperature and relative humidity (RH) of 25 ± 2.3°C and 59% ± 5.8%, respectively, with an average wind speed of 0.9 ± 0.4 m s^{-1}. In another experiment, the samples were exposed for 31 h under similar conditions (Table 1). Aerosol samples were analyzed for levoglucosan and K$^+$ before and after exposure as described in Section 2.1.

3 RESULTS AND DISCUSSION

3.1 Stability of Biomass-burning Tracers

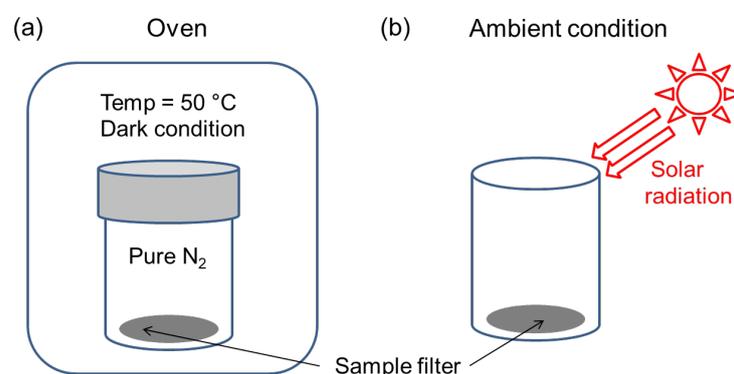Results of stability test for K$^+$ and levoglucosan are shown in Fig. 2. Although a small decrease

Fig. 1. Experimental setup for tracer stability experiments[+]. (a) Thermal-stability experiment. (b) Ambient exposure experiment.

Table 1. Experimental conditions for the stability tests of biomass-burning tracers on filter samples.

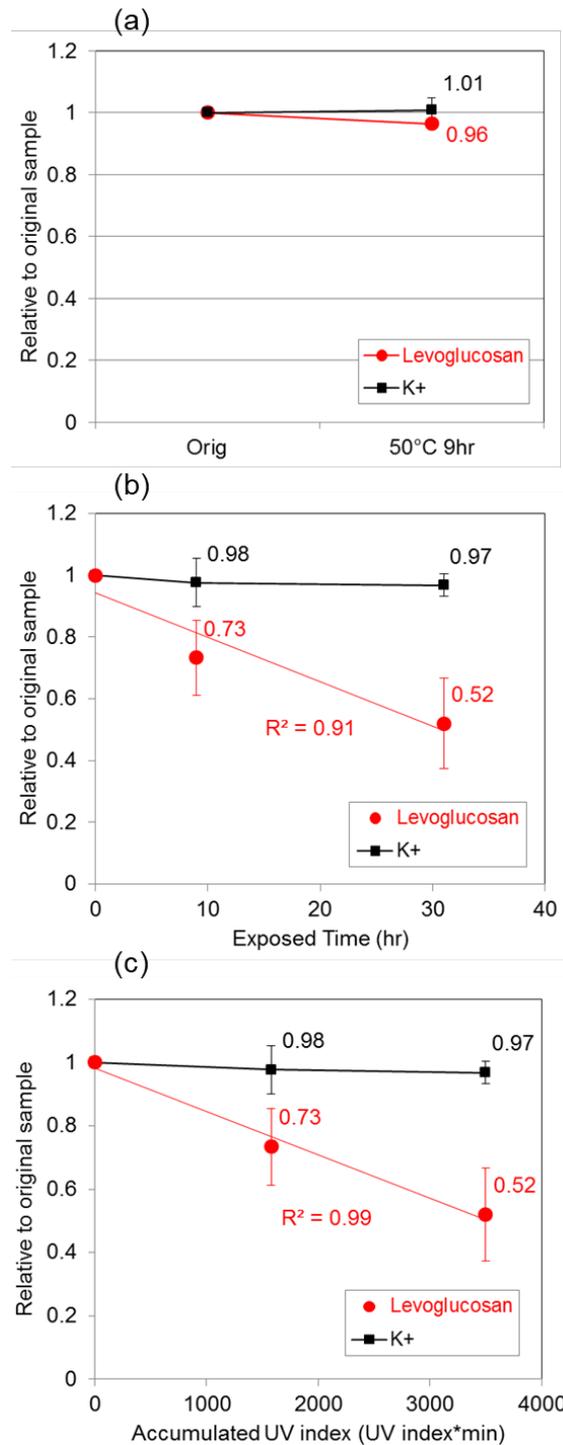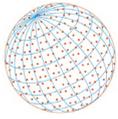|	Exposing time (h)	Ambient temperature (°C)	RH (%)	Wind speed (m s^{-1})
		Min-Max (Average ± S.D.)		
Case #1	9	20–27 (25 ± 2.3)	52–71 (59 ± 5.8)	0–2.2 (0.9 ± 0.4)
Case #2	31	18–30 (23 ± 3.5)	30–76 (60 ± 14)	0–2.2 (0.8 ± 0.5)

Fig. 2. Changes in levoglucosan and K⁺ contents of aerosol samples collected during fall 2012 and spring 2013 after exposure to (a) 50°C for 9 h; and (b, c) to the ambient atmosphere during spring 2014.

in filter levoglucosan content was observed, both K⁺ and levoglucosan appear thermally stable at 50°C for 9 h in a nitrogen atmosphere (Fig. 2(a)). Filter K⁺ contents were also stable when the filters were exposed to ambient conditions for more than two days in spring (Figs. 2(b) and 2(c)), while the levoglucosan content decreased to 75% and 52% of the original level after 9 h and 31 h exposure, respectively; i.e., almost half of the levoglucosan collected on a quartz filter was removed after 31 h exposure to ambient conditions during spring. Because almost half the levoglucosan
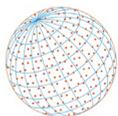
was removed after 31 h exposure and clear decreasing trend was observed, additional experiment with more hours of exposure was not conducted. These results indicate that levoglucosan is unstable in the atmosphere during spring and can be easily degraded whereas K^+ is stable.

During the ambient-conditions exposure experiments, temperatures did not exceed 30°C, with levoglucosan being stable at up to 50°C (Fig. 2(a)). The decrease in levoglucosan content during exposure to ambient conditions was therefore not due to thermal degradation. The decrease of filter levoglucosan contents was negatively correlated with the accumulated UV index rather than exposure time (Figs. 2(b) and 2(c)). Previous studies have reported that degradation of levoglucosan is caused mainly by photo-oxidative reactions with OH radicals (Hennigan et al., 2010; Hoffmann et al., 2010).

It is well known that the production of OH radicals in the atmosphere during daytime is strongly related to solar UV intensity and O_3 concentration (Seinfeld and Pandis, 1998). Highest O_3 concentrations were observed during spring season in Korea, followed by summer and fall (Jung et al., 2017). These are the likely causes of the negative correlation between the decrease in levoglucosan content and UV exposure. It was found that atmospheric levoglucosan at the sampling site can easily be removed through photo-oxidative degradation during sunny spring conditions.

Levoglucosan has been widely used to quantify the contribution of biomass burning to carbonaceous aerosols (Zhang et al., 2008; Jung et al., 2014; Chen et al., 2018). Using the levoglucosan to OC ratio, it was estimated that 18%–38% of the $PM_{2.5}$ OC was attributed to biomass burning in Beijing, China in 2002–2003 (Zhang et al., 2008) whereas 45% ± 12% of the $PM_{2.5}$ OC was attributed to biomass burning in Daejeon, Korea during the fall 2012 (Jung et al., 2014). If photo-oxidative degradation of levoglucosan is active at the both sites, the contribution of biomass burning to OC might be underestimated. Thus, care is required in using levoglucosan as a biomass-burning tracer.

3.2 Sources of Biomass-burning Tracers

Fig. 3 shows scatter plots of OC versus K^+ and levoglucosan concentrations in Daejeon during spring 2013. Large amounts of OC are emitted from biomass burning, and it is generally correlated well with biomass burning tracers such as K^+ and levoglucosan (Cheng et al., 2014; Jung et al., 2014; Liu et al., 2019; Fan et al., 2020). A positive correlation (R^2 = 0.35) was observed between OC and K^+ concentrations during the entire sampling period (Fig. 3(a)) whereas no correlation was observed between OC and levoglucosan concentrations (Fig. 3(b)). In contrary, very good correlation was observed between OC and levoglucosan concentrations at the same site during late fall season with a R^2 of 0.87.

Fig. 4 shows scatter plots of SO_4^{2-} versus K^+ and levoglucosan. Most SO_4^{2-} is secondarily produced from SO_2 in the atmosphere, while most K^+ and levoglucosan are directly emitted by biomass
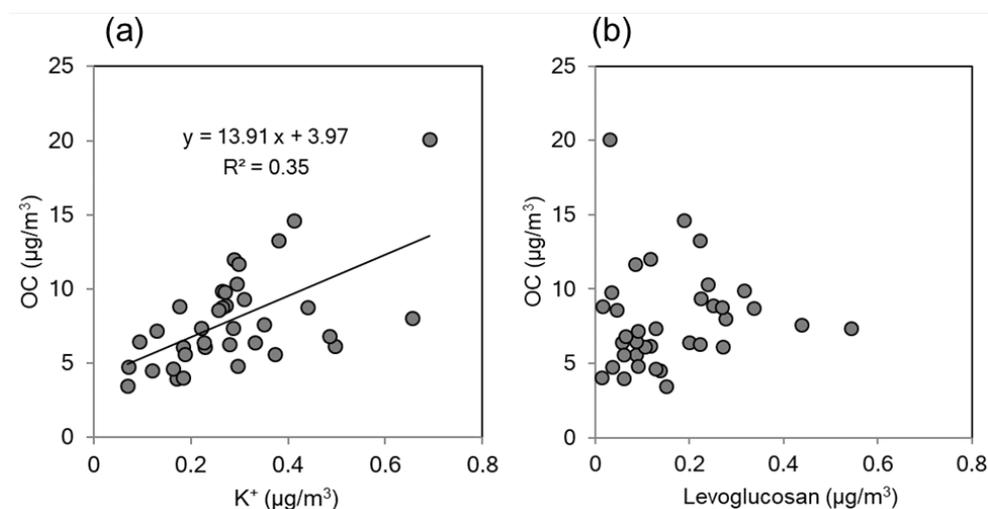

Fig. 3. Scatter plots of (a) OC versus potassium (K^+); and (b) OC versus levoglucosan in aerosol samples collected at a suburban site in Daejeon, Korea, during spring 2013.
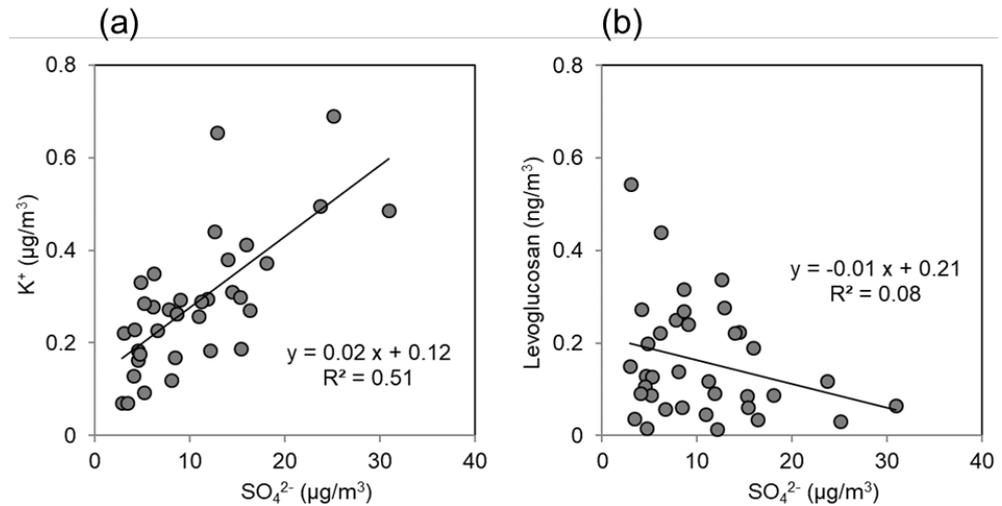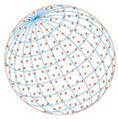
Fig. 4. Scatter plots of (a) SO_4^{2-} versus K^+; and (b) SO_4^{2-} versus levoglucosan during spring 2013.

burning (Seinfeld and Pandis, 1998; Pio *et al.*, 2008). Thus, no significant correlation between SO_4^{2-} and levoglucosan as shown in Fig. 4(b) can be explained by their different production mechanisms. Interestingly, even though production mechanisms are different between SO_4^{2-} and K^+, moderate positive correlation ($R^2 = 0.51$) was observed between SO_4^{2-} and K^+.

Air-mass backward trajectories during high- ($SO_4^{2-} > 15$ µg m^{-3}) and low-SO_4^{2-} ($SO_4^{2-} < 5$ µg m^{-3}) periods are shown in Fig. 5. During high-SO_4^{2-} periods (Fig. 5(a)), most air masses arriving at the sampling site originated from eastern China, whereas during low-SO_4^{2-} periods they originated mainly from the northern or eastern Korean Peninsula. High SO_4^{2-} concentrations are thus related to long-range transport of aerosols from the Asian continent. A sharp increase in SO_4^{2-} concentration (to 22.6 ± 8.1 µg m^{-3}) was also observed at Deokjeok Island off the west coast of the Korean Peninsula due to air masses originating from East China during spring 2009 (Jung *et al.*, 2015). The contribution of biogenic sulfur emitted from the ocean to total sulfur budget was determined to be 5.8% over the North Yellow Sea during July–August 2006 (Yang *et al.*, 2009). Thus, this study
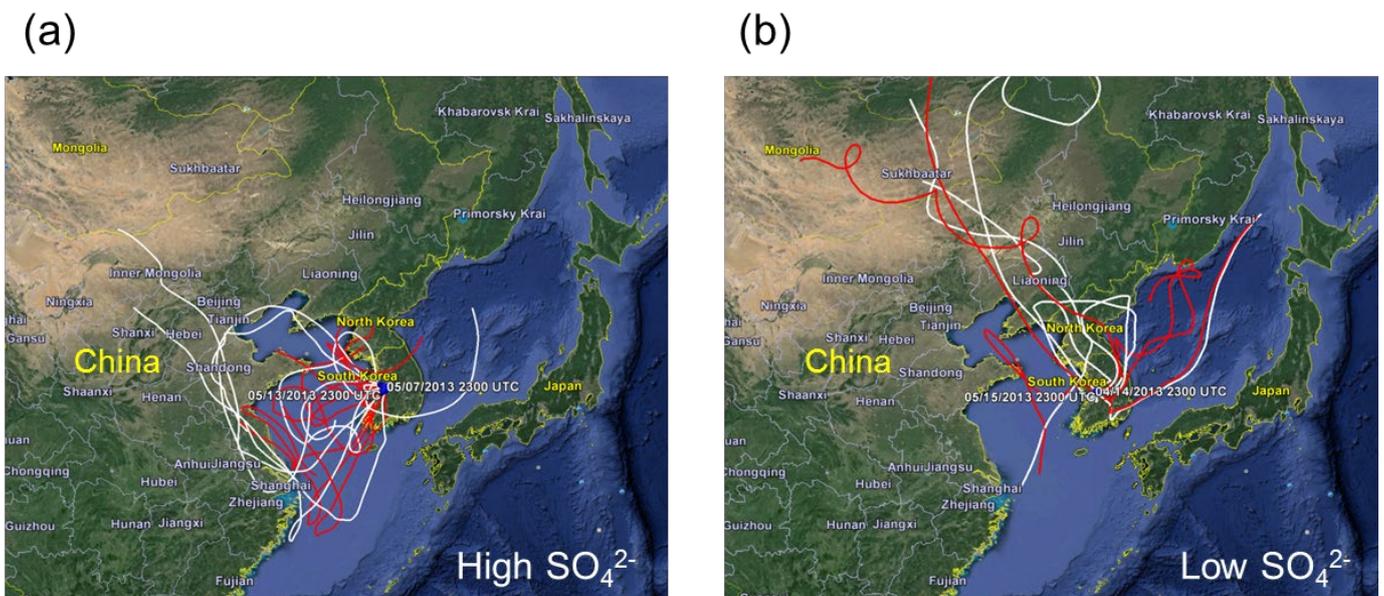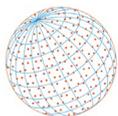

Fig. 5. Air-mass backward trajectories from the sampling site during (a) SO_4^{2-}-rich periods (24, 25 April; 8, 12–14, 20, and 22 May 2013); and (b) SO_4^{2-}-poor periods (15, 19, and 20 April; and 1, 16, and 19 May 2013). Red and white trajectories represent air masses arriving at altitudes of 200 and 500 m, respectively.

assumed that the marine source of SO_4^{2-} was not significant during long-range transport period. Thus, high SO_4^{2-} concentrations at the sampling site can be used as an indicator of long-range transport of aerosols from the Asian continent (Jung et al., 2016).

Atmospheric K^+ and SO_4^{2-} concentrations are moderately correlated (Fig. 4(a)) and elevated SO_4^{2-} levels are related to long-range transport from the Asian continent (Fig. 5(a)), so it is likely that some K^+ is also transported from the Asian continent. We conclude, therefore, that atmospheric K^+ at the sampling site is a mixture of locally and distally produced aerosols. However, because atmospheric levoglucosan can easily be degraded during sunny spring conditions, most levoglucosan at the sampling site might be produced locally.

3.3 Quantification of Distal Versus Local K^+

A scatter plot of K^+ versus levoglucosan concentrations at different SO_4^{2-} concentrations is shown in Fig. 6, indicating that the levoglucosan/K^+ ratio (i.e., the plot gradients) decreases with increasing SO_4^{2-} concentration. The levoglucosan/K^+ ratio during the post-harvest biomass-burning season in 2012 was obtained to be 1.16 ± 0.34 (Jung et al., 2014), similar to the ratio when SO_4^{2-} concentrations are low (Fig. 6). Levoglucosan/K^+ ratios during the post-harvest season may thus represent locally produced spring ratios. Because local biomass-burning occurred in nearby suburban areas, this study assumed that the degradation of levoglucosan from local biomass-burning was negligible.

Average levoglucosan/K^+ ratios at urban sites in Beijing and Chengdu, China, during spring are reported to be 0.28 and 0.19, respectively (Tao et al., 2014; Wang et al., 2014), as indicated by lines of those gradients in Fig. 6. Levogucosan/K^+ ratios in Beijing and Chengdu during spring were thus similar to those in Daejeon when the SO_4^{2-} concentration was high. It follows that as high SO_4^{2-} concentrations are related to long-range aerosol transport from the Asian continent (Fig. 5), lower levoglucosan/K^+ ratios may also be related to long-range transport of K^+ from the Asian continent.

A scatter plot of levoglucosan/K^+ ratios versus SO_4^{2-} concentrations as a function of ambient temperature is shown in Fig. 7. While levoglucosan/K^+ ratios decrease with increasing SO_4^{2-} concentrations (Fig. 6), they also decrease with increasing temperature at fixed SO_4^{2-} concentrations (Fig. 7). This can be explained by enhanced photo-oxidative degradation of levoglucosan with increasing ambient temperature, as also reported by Hennigan et al. (2010).

There is little correlation between levoglucosan and SO_4^{2-} concentrations (Fig. 4(b)), and levoglucosan/K^+ ratios decrease with increasing SO_4^{2-} concentration (Fig. 7), confirming that negligible levoglucosan is transported over long distances from the Asian continent to the Korean Peninsula during spring. We therefore assume that long-range transport of levoglucosan from the Asian continent to the Korean Peninsula is negligible in spring. Based on this assumption, distally and locally emitted K^+ were quantified roughly as follows.
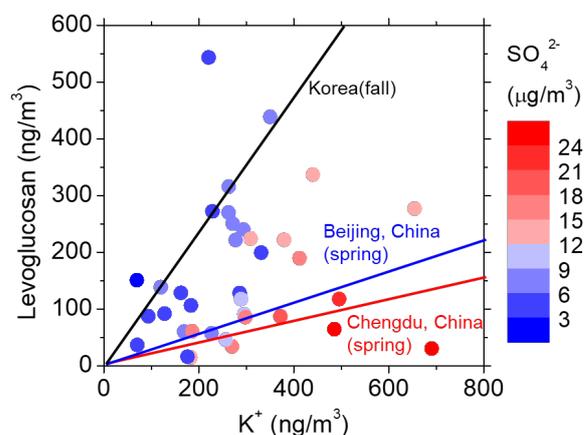

Fig. 6. Scatter plots of K^+ versus levoglucosan as a function of SO_4^{2-} concentration. Red and blue lines represent levoglucosan/K^+ ratios at Beijing and Chengdu, China, during spring 2003 and spring 2011, respectively (Tao et al., 2014; Wang et al., 2014).
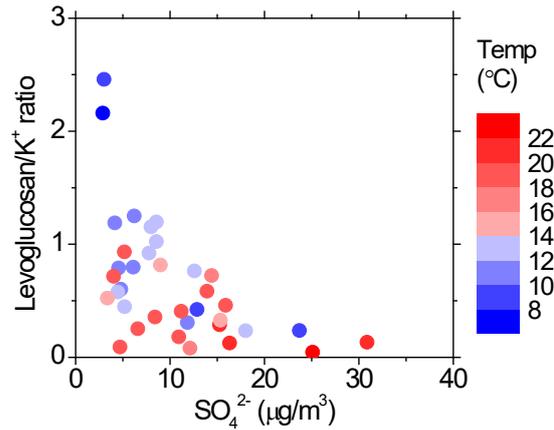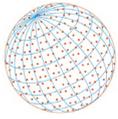
Fig. 7. Scatter plot of levoglucosan/K$^+$ ratio versus SO$_4^{2-}$ concentration as a function of ambient temperature.

$$K^+\_LTP(Day, i) = K^+(Day, i) - \frac{K^+}{Levo}(Local\ AVG) \cdot Levo(Day, i) \tag{1}$$

$$K^+\_Local(Day, i) = K^+(Day, i) - K^+\_LTP(Day, i) \tag{2}$$

where K$^+$(Day, i) represents the total K$^+$ concentration at day i; K$^+$/Levo(Local AVG) represents the local average K$^+$/levoglucosan ratio, assumed to be the average ratio obtained at the same sampling site during the post-harvest season (Jung *et al.,* 2014); Levo(Day,i) represents the levoglucosan concentration measured at day i; K$^+$_LTP(Day,i) and K$^+$_Local(Day,i) represent distally and locally produced K$^+$, respectively.

Temporal variations in distally and locally produced K$^+$ and SO$_4^{2-}$ concentrations are shown in Fig. 8, which indicates similar patterns for distally produced K$^+$ and SO$_4^{2-}$ concentrations. On average, ~39% of K$^+$ at the sampling site during spring 2013 was estimated to be sourced through long-range aerosol transport from the Asian continent. When SO$_4^{2-}$ concentrations were above 20 μg m^{-3}, the contribution of distally produced K$^+$ to total K$^+$ increased to ~84%. The positive correlation between K$^+$ and OC (Fig. 3(a)) suggests that a large fraction of OC emitted from biomass burning in the Asian continent can be also transported to the Korean Peninsula in Asian continental outflows.
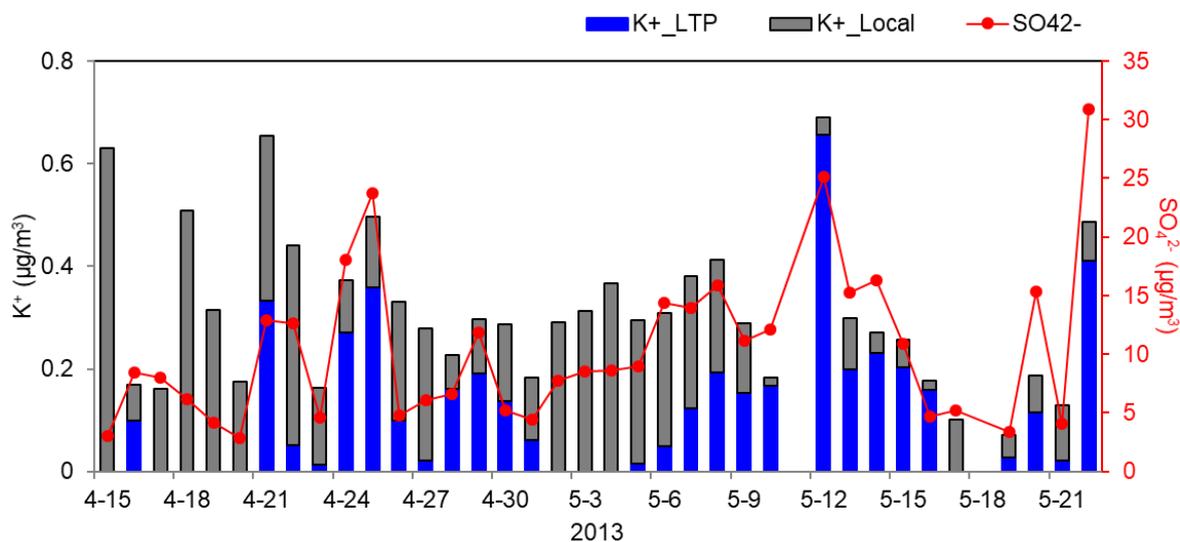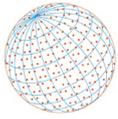

Fig. 8. Temporal variations in distally and locally produced K$^+$ and SO$_4^{2-}$ concentrations during spring 2013.

4 SUMMARY AND CONCLUSIONS

Levoglucosan and K$^+$ have been widely used as tracers for biomass burning during recent decades. Their stabilities are important issues in the qualitative and quantitative determination of the impact of biomass burning, especially downwind of Asian continental outflows. We found that levoglucosan is unstable when exposed to ambient atmospheric conditions during spring, while K$^+$ is stable. The degradation of levoglucosan in aerosol samples collected at a suburban site on the Korean Peninsula might be enhanced by long-range aerosol transport from the Asian continent. Caution should therefore be needed in the use of levogulcosan as a biomass-burning tracer downwind of the Asian continent during spring season.

Laboratory experiments, regression analysis, and air-mass backward-trajectory analysis indicate the following for the sampling site on the Korean Peninsula during spring 2013.

- High atmospheric SO_4^{2-} concentrations may be mainly attributed to long-range aerosol transport from the Asian continent.
- Atmospheric K$^+$ concentrations may comprise a mixture of distal and local emissions.
- Most levoglucosan is degraded during long-range atmospheric transport from the Asian continent to the Korean Peninsula.
- Most levoglucosan at the sampling site was produced locally.

Based on the assumption that negligible amounts of levoglucosan can be transported long distances in Asian continental outflows during spring, local and distal source contributions of K$^+$ were estimated using levoglucosan/K$^+$ ratios for the Asian continent and the Korean Peninsula, with results indicating that ~39% of K$^+$ might be transported from the Asian continent.

ACKNOWLEDGEMENTS

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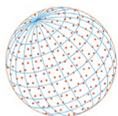
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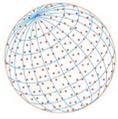
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