

Article

Deposition of Potassium on Chimney Wall from Wood Stove Smoke: Implication for the Influence of Domestic Biomass Burning on Atmospheric Aerosols

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Abstract: Based on the field studies of biomass burning plumes in Alaska, we hypothesized that potassium (K) may be significantly scavenged, during wood stove burning, as deposits on the inner wall of the chimney where the temperature decreases with the height. To test this hypothesis, we analyzed chimney deposit samples collected from the inner wall of a chimney (6 m long) for the measurement of major ions and anhydrosugars including levoglucosan (Lev). Concentrations of K were found to be highest in the lower part of the chimney with a decreasing trend with height, whereas Lev showed an opposite trend with the lowest concentrations near the bottom of the chimney and an increase with height. We detected an anti-correlation between the two components in the chimney deposits, confirming that K is largely scavenged as a deposit within the chimney while Lev is significantly emitted to the ambient air. We propose that, using K/Lev mass ratios, the relative contributions of open fires and domestic wood burning to ambient aerosols can be evaluated.

Keywords: domestic burning; wood stove; potassium (K); levoglucosan (Lev); deposition of K on chimney wall; K/Lev ratio; chimney deposit; soot; anti-correlation between K and Lev



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1. Introduction

Biomass burning (BB) is an important source of atmospheric aerosols and gases, causing the deterioration of air quality, adverse effect on human health, and significant influences on climate change [1,2]. Forest fires, agriculture waste burning, and domestic burning (wood stove for house heating and cooking) are three major components of BB, which control the levels of atmospheric particles on a local, regional, and global scale. On a global scale, BB could account for >85% of primary emissions of organic aerosols [1]. Carbonaceous aerosols of BB origin not only cause human respiratory diseases and premature death [3–5] but also affect global climate changes by modifying radiative forcing of the atmosphere, in which BB products can act as cloud condensation nuclei by forming fog and clouds as well as wet precipitations [6]. The BB products emitted from mid-latitudes including southeast Asia, Europe, and North America can be long-range transported to the Arctic, where black carbon and brown carbon, the major products of BB, can contribute to accelerating global warming via snow melting in Greenland and in the High Arctic [7].

In the chemical studies of ambient atmospheric particles affected by BB, potassium (K) has often been used as a tracer of BB [2,8,9]. In fact, K⁺ has been reported as the most abundant ion in the smoke emitted from the burning of different woods in wood

stoves and fireplaces [10]. K is one of major elements in biomass, which is emitted to the atmosphere during the burning of wood, leaves, and agricultural wastes such as wheat/rice straws [11,12]. Meanwhile, levoglucosan (Lev) is derived from the pyrolysis (>400 °C) of cellulose and hemicellulose [13] and has been used as a good organic tracer of biomass burning [11,14,15]. Lev is also known to be emitted to the air from the combustion of lignite or brown coal [16,17]. Although there are generally good correlations between K and Lev in BB-influenced ambient aerosols, the correlation coefficients are highly variable depending on the regions studied [11,18,19].

During a one-year observation of ambient aerosols from Fairbanks, Alaska, we found a large peak of Lev, but no correlation was detected between K and Lev [20]. We hypothesized that the Fairbanks aerosols are less involved with forest fires but are largely associated with domestic wood burning, in which we hypothesized that K can be significantly removed from the smokes on the inner wall of chimney connected to wood stove. However, there is no study on the chemical analyses of chimney deposit samples to examine the above hypothesis.

Here, for the first time, we analyzed the chimney deposits collected from a modern wood-fired stove with a secondary combustion system for chemical compositions, including K and Lev, in order to evaluate the preferential removal of K from the BB smokes within the wood-fired chimney where the temperature gradient is significant from the stove top (ca. 200–250 °C) to the chimney exit (ca. 0–10 °C) adjacent to the ambient air. Based on the vertical distributions of inorganic ions and organic tracers in the chimney deposits, we discuss the possible scavenging of K as a deposit during the upward transport of BB products (smoke and burnt products) within the chimney. We present the first evidence in support of the hypothesis of a preferential removal of K within the wood stove system, which can greatly reduce the correlation coefficient between K and Lev in the ambient aerosols. This situation of a domestic wood stove system provides a completely different chemical composition of BB plumes than that of open fires such as forest fires and field burning of agricultural waste, which are becoming more common on the regional and global scales.

2. Samples and Methods

In this study, we collected chimney deposit samples using a wood stove (Morso model 1126CB, Denmark) connected to a 6 m long chimney (Figure 1) after burning dried wood (main wood was oak). The stove was used every evening and occasionally in the morning from December 2017 to March 2018. The inner diameter of the chimney is approximately 20 cm with a double-walled steel structure for insulation. This stove facilitates the operation of the secondary burner to achieve a clean burn (<https://www.stoveworlduk.co.uk/what-is-secondary-burn-on-a-wood-burning-stove>) (accessed on 10 February 2023). The combustion efficiency of the wood stove model is 80.7%, according to the company (<https://www.andersen-stove.jp/brand/morso/detail/?id=1126CB>) (accessed on 10 February 2023). Temperatures measured by a bimetallic thermometer on the stove-top during stove use were generally in a range of 180–250 °C, but temperatures inside of the stove oven were roughly estimated to be 500–800 °C. Chimney deposit samples (n = 18, see Figure 2) were collected in May 2018 from the inner wall of the chimney using a stainless steel spoon and brush for selected heights. Small portions (ca. 1 mL) of each deposit were placed in a clean 1.5 mL glass vial (preheated at 450 °C) with a Teflon-lined screw cap. The total amount of chimney deposits collected during the winter season was estimated to be approximately 0.8 L (or about 200 g). The total amount of wood burned in the stove during this season was approximately 500 kg. The amount of ash recovered from the stove was approx. 3 kg.



Figure 1. A wood stove and chimney (left panel) used in this study and fires in the stove oven (right panel).



Figure 2. An example of chimney deposit (soot) sample collected from the inner surface of the chimney. In order to obtain access to the deposit, the second part of the chimney device was separated from the whole chimney system, and then soot samples were collected at selected heights using a spoon for the lower levels and an elongated brush tool for the higher levels.

Small amounts of stack deposit samples (2–3 mg) were subjected to various chemical analyses. Total carbon (TC) and total nitrogen (TN) were measured with an elemental analyzer (EA, Flash 2000, Thermo) coupled to a gas chromatograph with a packed column [21,22]. Water-soluble organic carbon (WSOC) and water-soluble total nitrogen (WSTN) were measured using Shimadzu TOC meter (model TOC-V_{CSH}) after the extraction

with organic-free pure water (15 mL) under ultrasonication for 15 min followed by filtration with a 0.22 μm pore size syringe filter [23].

Major cations and anions were determined by using ion chromatography (Model 761 compact IC, Metrohm, Switzerland) after the extraction of chimney deposit (2 mg) with ultrapure water ($>18\text{ M}\Omega\text{ cm}$, 10 mL \times 2 times) under ultrasonic agitation for 15 min followed by filtration with a syringe filter (Millex-GV, 0.45 μm pore size, Millipore, Burlington, MA, USA). Anions were isolated on a Shodex column (SI-90 4E, Showa Denko, Tokyo, Japan) with a suppressor using a mixture of 1.8 mM Na_2CO_3 and 1.7 mM NaHCO_3 as an eluent at a flow rate of 1.2 mL min^{-1} . For cations, a Metrosep C2-150 column (Metrohm) was used with 4 mM tartaric acid + 1 mM 2,6-pyridinedicarboxylic acid as an eluent at a flow rate of 1.0 mL min^{-1} . The injection loop volume was 200 μL for both columns. All ions were quantified using a standard calibration curve [24].

Levoglucosan and other sugar compounds were measured by GC/MS after the solvent extraction, followed by trimethylsilyl (TMS) derivatization. Briefly, an aliquot of chimney deposit (10 mg) was extracted with dichloromethane/methanol (2:1; *v/v*, 5 mL) under ultrasonication for 10 min for three times. Sugar compounds in the total extracts were dried and then derivatized to TMS ethers using 60 μL of *N,O*-Bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl (TMS) chloride in the presence of pyridine (10 μL) at 70 $^\circ\text{C}$ for 3 h. After the reaction, derivatives were diluted by the addition of 140 μL of *n*-hexane containing 1.43 ng μL^{-1} of the internal standard (C_{13} *n*-alkane) prior to analysis by gas chromatography/mass spectrometry (GC/MS) using a Hewlett-Packard model 6890 GC coupled to Hewlett-Packard model 5973 mass-selective detector (MSD).

The GC separation was performed on a DB-5MS fused silica capillary column (30 m \times 0.25 mm i.d., 0.5 μm film thickness) with helium as the carrier gas at a flow rate of 1.0 mL min^{-1} . The GC oven temperature was programmed from 50 $^\circ\text{C}$ (2 min) to 120 $^\circ\text{C}$ at 30 $^\circ\text{C min}^{-1}$ and then to 300 $^\circ\text{C}$ at 6 $^\circ\text{C min}^{-1}$ with a final isotherm hold at 300 $^\circ\text{C}$ for 16 min. The sample was injected at 280 $^\circ\text{C}$ in a splitless mode. The mass spectrometer was operated in an electron impact (EI) mode at 70 eV and scanned from 50 to 650 daltons. Mass spectral data were acquired and processed using the Hewlett-Packard Chemstation software. Details of the GC/MS analyses can be found in Fu et al. [25].

3. Results and Discussion

3.1. TC, TN, WSOC and WSTN

The concentrations (weight %) of total carbon (TC) and nitrogen (TN) in the chimney deposits ranged from 42.5 to 82.0% (av. $58.2 \pm 9.2\%$) and from 1.4 to 6.1% ($3.58 \pm 1.7\%$), respectively (Table 1). Carbon is the dominant element in the chimney deposits, accounting for more than half of the total mass. This result is reasonable because the color of the chimney deposit is indeed black (Figure 2), suggesting that the major form of carbon is soot (black or elemental carbon). We found that the concentrations of water-soluble organic carbon (WSOC) (0.35–0.16 wt %, av. $0.67 \pm 0.30\text{ wt \%}$) are 86 times lower than TC (Table 1), indicating that major fraction of TC is water-insoluble and that chimney deposits are largely composed of elemental carbon or black carbon, as well as pyrolytic carbon that is produced by high temperature pyrolysis of wood without sufficient oxygen. Soot is largely deposited on the inner surface of the chimney, whereas water-soluble and/or semi-volatile organics are emitted to the atmosphere as gases or particles without serious deposition.

Similarly, concentrations of water-soluble total nitrogen (WSTN) are significantly lower (0.03–0.66 wt %, av. $0.19 \pm 0.20\text{ wt \%}$) than those of TN (av. 3.5 wt %) with the WSTN/TN ratios of 1.76–11% (av. $4.5\% \pm 2.7\%$). However, the average ratio is about 3 times higher than that of WSOC/TC ratio (av. $1.18 \pm 0.57\%$, see Table 1). These comparisons suggest that water-soluble nitrogens such as NH_4^+ , NO_2^- , and NO_3^- are not negligible, but their concentrations are much lower than other ions (e.g., SO_4^{2-} , K^+ , Ca^{2+}) (Table 1).

Table 1. Summary of carbon, nitrogen, and ion analyses of the chimney deposits from domestic wood stove (n = 18).

Components	Minimum	Maximum	Average	Standard Deviation
TC (wt %)	42.5	81.9	58.1	9.18
TN (wt %)	1.38	6.14	3.58	1.75
WSOC (wt %)	0.35	1.61	0.67	0.30
WSTN (wt %)	0.03	0.66	0.19	0.20
WSOC/TC (%)	0.53	2.87	1.18	0.57
WSTN/TN (%)	1.76	11.0	4.55	2.75
Anions (mg/g)				
F ⁻	0.05	0.24	0.10	0.05
MSA ⁻	0.24	16.4	4.18	4.61
Cl ⁻	0.08	0.97	0.34	0.29
NO ₂ ⁻	0.00	2.75	0.35	0.74
Br ⁻	0.00	0.07	0.01	0.02
NO ₃ ⁻	0.00	0.39	0.06	0.10
PO ₄ ³⁻	0.00	1.53	0.42	0.38
SO ₄ ²⁻	18.9	99.0	46.0	22.6
Total anions	20.3	116	51.4	25.6
Cations (mg/g)				
Na ⁺	0.14	2.96	0.83	0.88
NH ₄ ⁺	0.00	7.87	1.46	2.38
K ⁺	7.70	43.6	17.5	9.90
Ca ²⁺	3.52	29.0	14.5	8.27
Mg ²⁺	0.34	3.40	1.63	0.94
Total cations	14.7	78.6	35.9	18.2

TC: Total carbon, TN: Total nitrogen, WSOC: Water-soluble organic carbon, WSTN: Water-soluble total nitrogen, MSA: Methanesulfonic acid.

These results from smokestack deposition are very different from the ambient aerosols, where WSOC is the major fraction of TC; i.e., the WSOC/TC ratio is >70% [24]. In the ambient aerosols, a secondary formation of water-soluble organic matter, such as low molecular weight dicarboxylic acids, is significant due to the photochemical oxidation of volatile organic compounds [26]. We also found that mass concentration ratios of water-soluble total nitrogen (WSTN) in TN are significantly lower (1.76–11.0%, av. $4.55 \pm 2.75\%$) than those reported for ambient aerosols (>80%) [24,27]. These results indicate that majority of TN in the stack deposits is water-insoluble, although we detected water-soluble nitrogen species, such as ammonium, nitrite, and nitrate at trace levels. It is interesting to note that NO₂⁻ is about five times more abundant than NO₃⁻ in the chimney deposits (Table 1), which is opposite in the ambient aerosols where NO₂⁻ concentrations is very low or undetectable due to photochemical oxidation [28]. This result suggests an insufficient oxidation of nitrite to nitrate in the wood stove/chimney system, possibly due to limited available oxygen.

Interestingly, we found different vertical profiles of TC and TN with the stack height. Figure 3 shows the vertical distribution of TC and TN concentrations. TC increased from the bottom of the chimney to the height of 80 cm and then decreased towards the top of the chimney with some variations (Figure 3a). The increase in TC within one meter from the bottom of the stack may be related to the deposition of carbonaceous components including soot, as a result of the rapid decrease in the temperature inside the stack (Figure 3b). In contrast, TN concentrations increased continuously from the bottom to higher levels of the stack, except for the bottom sample, although the concentrations of TN were lower than those of TC (Figure 3b). Such an increase may be related to gas-to-particle conversion of nitrogen species such as NO₃⁻, NH₄⁺, and amines. These species showed an increase in the concentrations in the stack deposit, especially in the case of NH₄⁺.

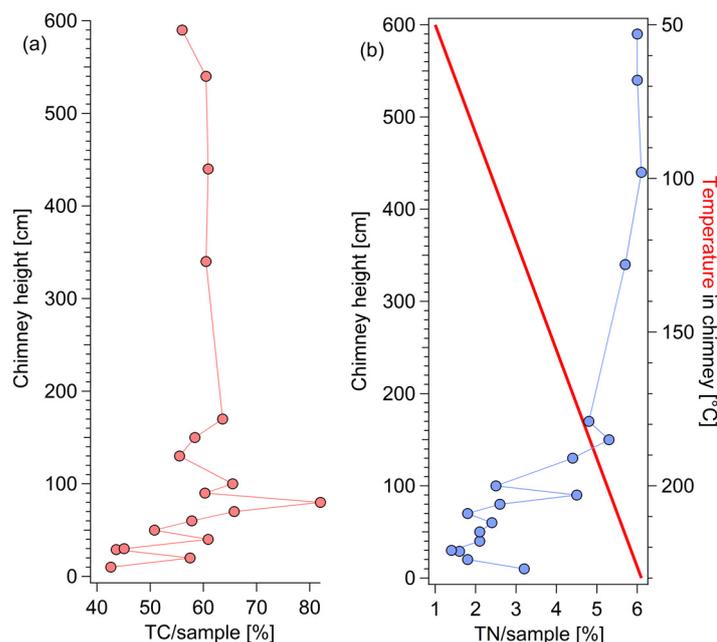


Figure 3. Vertical profiles of the mass concentrations (%) of (a) total carbon (TC) and (b) total nitrogen (TN) in the chimney deposit samples with estimated temperature (red line) in the chimney inner wall. On the top of the chimney, there is a chimney cap cover (ca. 40 cm high), thus the temperature of the chimney top can be estimated to be around 50 °C or more in winter when the outside temperature is 0 °C or less in winter overnight.

3.2. Major Ions and Levoglucosan

The anion analyses of the chimney deposits showed that sulfate (SO_4^{2-}) is the dominant anion followed by methanesulfonate (MSA^-) (Table 1). In contrast, nitrate (NO_3^-), which is abundant in ambient aerosols [29], is rather minor anion, which is less abundant than nitrite (NO_2^-) in the deposits (Table 1). Halogens (F^- , Cl^- and Br^-) were also detected as well as phosphate (PO_4^{3-}). Interestingly, K^+ was found to be the most abundant among the cations detected in the chimney deposits, followed by Ca^{2+} . Although Mg^{2+} , NH_4^+ , and Na^+ were detected, their concentrations are one order of magnitude lower than K^+ (Table 1). The chemical composition of chimney deposits is very different from that of the smoke or ambient aerosols derived from biomass burning in terms of composition of major ions; $\text{SO}_4^{2-} > \text{K}^+ > \text{NH}_4^+ > \text{NO}_3^-$ (chimney deposits, this study) vs. $\text{K}^+ > \text{NH}_4^+ > \text{SO}_4^{2-} > \text{NO}_3^-$ (e.g., $\text{PM}_{2.5}$ and PM_{10} from East Africa in the dry season) [11]. Potassium is known to be one of essential nutrients, along with N and P, for plant growth and is abundant in plants (e.g., 0.05% in wood, 0.19% in bark and 0.34% in leaves) [30].

We measured various sugar compounds together with carbon/nitrogen contents and major ions. Here we focus on the data of levoglucosan (1,6-anhydro- β -D-glucose) and potassium detected in the chimney deposit samples. Lev concentrations ranged from 0.28 to 2.96 ng/mg-deposit sample with a mean (\pm standard deviation) of 7.1 ± 4.0 ng/mg-deposit, while K ion concentrations ranged from 7.7 to 43.6 $\mu\text{g}/\text{mg}$ -deposit with a mean of 17.4 ± 9.9 $\mu\text{g}/\text{mg}$ -deposit. Figure 4a,b shows vertical profiles of Lev and K concentrations in the chimney deposit samples. Lev concentrations increased from the bottom to the top of the chimney. In contrast, those of K showed the maximum concentration near the bottom of the chimney connected to the stove oven and gradually decreased towards the upper layers of the chimney. Consequently, the mass concentration ratios of K/Lev decreased from the bottom to the top of the chimney (Figure 4c). A rapid decrease in the K/Lev ratios near the bottom of the chimney is associated with a significant deposition of K as a result of the rapid decrease in temperature that occurs between the top of stove and the lower levels of the chimney.

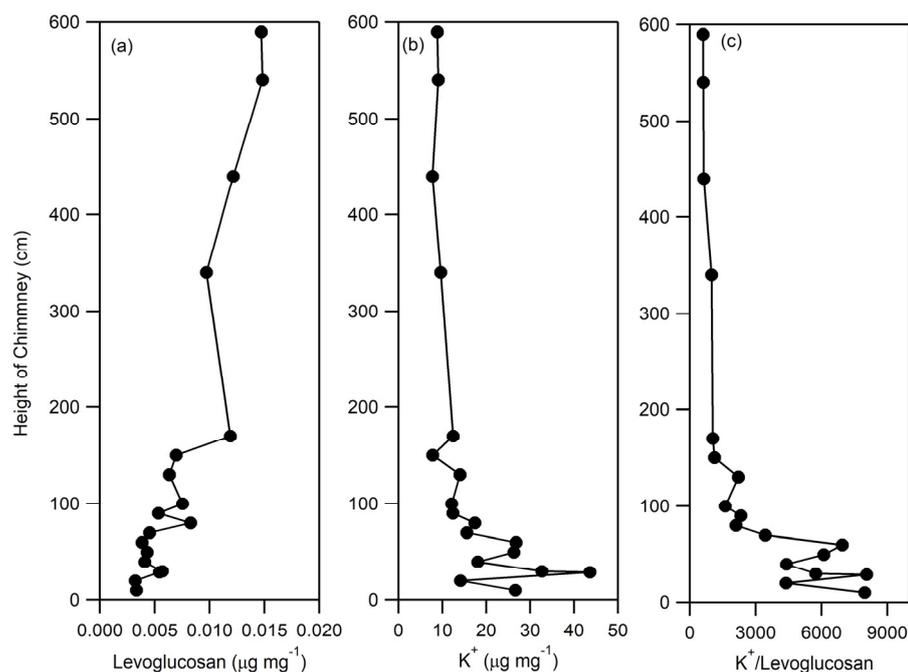


Figure 4. Vertical profiles of the concentrations ($\mu\text{g mg}^{-1}$) of (a) levoglucosan and (b) K^+ in the chimney deposits and (c) mass concentration ratios of K^+ /levoglucosan.

Figure 5 shows an anti-correlation between Lev and K in the chimney deposit samples. This relationship is caused by the different behaviors of the two species during upward transport in the chimney, which are exposed to a significant temperature gradient after the combustion of wood in the stove oven. Such a different behavior between K and Lev in the chimney can be explained by the different physicochemical properties of the two species; i.e., the boiling point of K ($760\text{ }^\circ\text{C}$) (<https://www.britannica.com/science/potassium>) (accessed on 10 February 2023) is much higher than that of Lev ($385\text{ }^\circ\text{C}$) [31]. K can be deposited in the stack as salts such as K_2SO_4 [32]. On the other hand, Lev can be largely emitted to the atmosphere from the top of the stack. This situation is very different from the open burning, where there is no wall effect. Although the depositional removal of K in the wood stove occurs seriously during the migration of the smoke from the bottom to the top of the chimney, the degree of removal may be variable depending on the type of stove and the burning conditions of the wood [33]. Similar depositional removal of K also occurs during traditional domestic wood burning for cooking [8] and biomass burning in boilers for electricity generation using wood pellets [34].

We found that the concentrations of K in the chimney deposit samples are three orders of magnitude higher than those of levoglucosan. Lev is an incomplete combustion product (thermal oxidation or pyrolysis) of wood, whereas K is stable against the thermal oxidation. The enrichment of K over Lev in the chimney deposits can be explained by the much higher boiling point of K than Lev, as mentioned above. A large difference in the physico-chemical properties between K and Lev indicates that Lev is largely present as gas in the burnt plumes, whereas K is present as a particle in the flames; the latter can be easily removed from the plumes by deposition on the chimney wall as the combustion plumes rise. The average concentration ratio of K/Lev (range: 600–8050, mean: 3350 ± 2620) is 2–3 orders of magnitude higher than that of K/Lev in the ambient aerosols (3–33, Kundu et al., 2010) [35]. The K/Lev ratios (av. 0.19) reported in the winter Fairbanks aerosols [20] are similar to those in wood stove smoke ($<0.25\text{--}1$) [36], whose ratios are much smaller than those (3–33) of the ambient aerosols as mentioned above, although the types of the samples ($\text{PM}_{2.5}$ and soot from chimney) are not same.

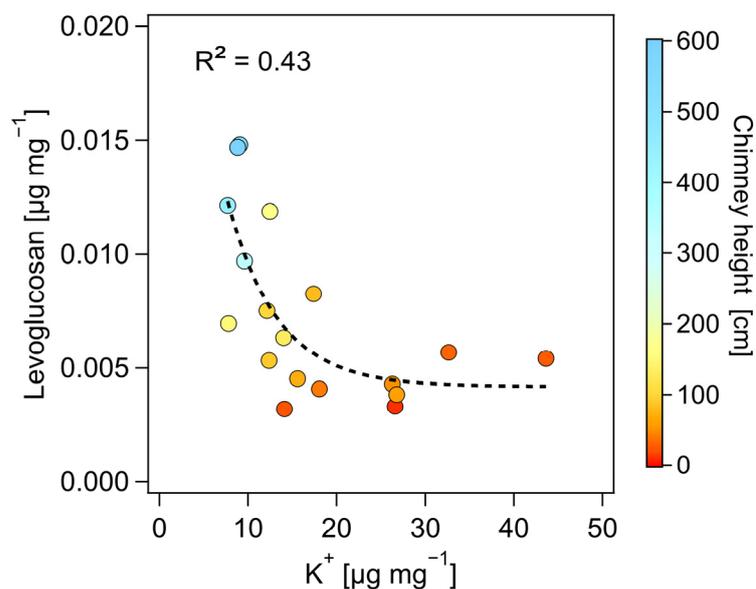


Figure 5. Relationship between levoglucosan and K^+ in the chimney deposit samples. The color scale reflects the height of deposit sample in chimney.

In addition to K^+ , we detected high abundances of Ca^{2+} , followed by Mg^{2+} and Na^+ in the chimney deposits (Table 1). Among the anions, sulfate is the most abundant species, followed by methanesulfonic acid (MSA). These sulfur species are much more abundant than nitrogen species, such as nitrate, in the chimney deposits, largely due to the higher vapor pressure of nitric acid than sulfuric acid and MSA. Interestingly, ammonium is more abundant than nitrate. The formation of ammonium salts with inorganic acids such as sulfuric acid inside the stack may be the reason for the high abundance of ammonium. On the other hand, in the open fires (forest fires and field burning of agricultural residues) there is no surface such as the inner wall of the chimney to adsorb/remove the mineral and/or salt particles from the smoke plumes. Therefore, many metals and metal ionic species as well as carbonaceous components are emitted directly into the atmosphere without any significant removal/deposition process on the surfaces during open fires.

In summary, the preferential depositional removal of K within the chimney of domestic wood stoves is the main reason for the lower K/Lev ratios reported for the ambient aerosols, such as the Fairbanks aerosol from Alaska [20]. This study can explain the wide range of K^+ /Lev ratios or weak correlations between the two components reported in ambient aerosols affected by different types of BB. Because wood stove smoke contains less abundant K^+ relative to Lev with lower K^+ /Lev ratios [36], it is very likely to detect the influence of domestic burning by analyzing the K^+ /Lev ratios in ambient aerosols and estimating the relative contributions of open burning (3–7) [35] vs. wood stove burning (ca. 0.2–3) [20,36].

4. Summary and Conclusions

Based on the chemical analyses of chimney deposits (soot) from a domestic wood stove, we tested the hypothesis on a selective scavenging of potassium (K^+) over levoglucosan (Lev) from the smoke plumes. We found that potassium (K^+) is heavily deposited in the lower part of chimney as a soot, whereas levoglucosan (Lev) is largely emitted into the air via the chimney. The preferential removal of K^+ in the stack can be explained by the different boiling points of K (760 °C) and Lev (385 °C). Lev is more likely to be emitted to the environment than K^+ during domestic burning with a chimney system. This study confirms that the behavior of K is largely different between open burning and domestic burning, demonstrating the lower relative abundances of K^+ than Lev in the latter plumes. Based on the current experimental results, we propose that open burning vs. domestic

burning can be evaluated using K^+ /Lev ratios in the ambient aerosols. This study also argues that more attention should be paid when using K as a BB tracer in the study of ambient aerosols, as domestic wood stove burning is becoming more popular in developed countries and sometimes disturbs the K^+ /Lev ratios in the atmospheric particles.

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