学 位 論 文 審 査 の 要 旨

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学 位 論 文 題 名

Molecular compositions and seasonal variations of organic compounds in atmospheric aerosols from subarctic Alaska

（亜北極アラスカにおける大気エアロゾル中の有機化合物組成と季節変動）

Organic constituents are recently highlighted because they account for a substantial portion of atmospheric particles, up to 50% in mass. Global models predict that biomass burning emissions and secondary organic aerosols (SOA) formation from biogenic volatile organic compounds (BVOCs) are the two major sources of organic aerosols (OA). Most of them are water-soluble and they can act as cloud condensation nuclei and ice nuclei, affecting climate by altering the hygroscopic properties of aerosols. Total of 32 total suspended particle (TSP) samples were collected from Fairbanks, Alaska in June 2008 to June 2009 using a low volume air sampler at a flow rate of 16.7 L/min. Here, we report the molecular compositions and seasonal variations of anhydro-sugars (levoglucosan, galactosan and mannosan), lignin and resin acids (4-hydroxybenzoic, syringic, vanillic and dehydroabietic acids) and biogenic SOA tracers (isoprene, *α*-/*β*-pinene and *β*-caryophyllene SOA tracers) which were measured using solvent extraction/TMS-derivatization technique followed by gas chromatography-mass spectrometry (GC-MS) determination.

Anhydro-sugars and lignin & resin acids are specific tracers from biomass burning emissions. Levoglucosan was observed as a dominant biomass burning tracer (average 66.8 ng m^{-3}) followed by its two isomers: mannosan (15.0 ng m^{-3}) and galactosan (ave. 9.88 ng m^{-3}). All anhydro-sugars showed similar temporal and seasonal trends with high abundant in winter and autumn. All lignin and resin products exhibited similar temporal and seasonal variations with levoglucosan in winter maximum and spring minimum. Dehydroabietic acid was detected as a most abundant (ave. 8.51 ng m^{-3}) tracer among all lignin and resin products. The analyses of air mass back trajectories and fire spots demonstrated that biomass burning tracers may be associated from residential heating and cooking in regional area, especially in cold season. Levoglucosan contributed 2.92% to WSOC and 1.62% to OC. The highest values of WSOC (6.26%) and OC (3.63%) in

wintertime indicate that contribution of biomass burning to Alaskan aerosols is important in winter period.

The concentration level of isoprene oxidation products was higher (ave. 4.14 ng m^{-3}) than *α*-/*β*-pinene- (2.01 ng m^{-3}) and *β*-caryophyllene-SOA product (1.53 ng m^{-3}). Isoprene-SOA tracers showed higher concentrations in summer (ave. 8.77 ng m^{-3}), whereas *α*-/*β*-pinene- and *β*-caryophyllene-SOA tracers exhibited highest levels in spring (3.55 ng m^{-3}) and winter (4.04 ng m^{-3}), respectively. *β*-Caryophyllinic acid and levoglucosan showed a positive correlation, indicating that biomass burning may be a major source for *β*-caryophyllene. Using a tracer-based method, the amounts of secondary organic carbon (SOC) were estimated for isoprene (ave. 159 ngC m^{-3}), *α*-/*β*-pinene (35 ngC m^{-3}) and *β*-caryophyllene (66.3 ngC m^{-3}). Based on SOA tracers, this study suggests that isoprene is a more important precursor for the production of biogenic SOA than *α*-/*β*-pinene and *β*-caryophyllene in subarctic Alaska.

All primary sugars and sugar alcohols except xylose and erythritol showed higher concentrations in summer and spring than winter and autumn due to the abundance of vegetations, pollens and developing flower buds in spring/summer period in Alaska region. Sucrose was most dominated tracer (30.5 ng m^{-3}) among all the primary sugars and sugar alcohols. Positive linear correlation between nss-Ca^{2+} and arabitol indicates that sugar alcohols partly derived from soil microbes. On the other hand, xylose and erythritol exhibited higher concentrations in winter and lower in spring, which is similar with those of levoglucosan, indicating that they are originated from biomass burning. Based on air masses backward trajectories, it can be predicted that primary biological aerosol particle (PBAPs) tracers mainly derived from regional boreal and tundra forest or local vegetation due to the high biological activities in spring and summer period.

The present study demonstrated clear seasonal trends for biomass burning tracers with high abundance in winter/autumn, whereas biogenic SOA tracers, primary sugars and sugar alcohols were dominated in summer/spring. In this study, mixed air masses origin were observed in all seasons when air masses mainly come with short distance over oceanic regions and some of from Alaska regional area to sampling place. This result suggests that regional emission is the important source for organic aerosols in Alaska atmosphere. Positive matrix factorization (PMF) analysis demonstrated that biomass burning is the important source (49.2%) in Alaska aerosols. Based on forward trajectories from sampling area, it should be proposed that Alaska pollutants can be transported to the Arctic which can affect the Arctic climate. Thus, the current study will be useful to better understand the effect of organic aerosols on the Arctic or subarctic atmosphere.

In addition to the excellent academic knowledge in the research, his academic records throughout the Ph. D course are excellent. Based on these evidences, the committee reached to a conclusion that Md. Mozammel Haque deserves to become a Doctor of Environmental Science.