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学 位 論 文 審 査 の 要 旨

博士（環境科学）

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学 位 論 文 題 名

Studies on ice core records of dicarboxylic acids, ω -oxocarboxylic acids, pyruvic acid, α -dicarbonyls and fatty acids from southern Alaska since 1665 AD: A link to climate change in the Northern Hemisphere

(南アラスカにおけるジカルボン酸、 ω -オキシカルボン酸、ピルビン酸、 α -ジカルボニルおよび脂肪酸の 1665 年以降のアイスコア記録に関する研究：北半球における気候変動とのリンク)

Alaskan ice core (180 m long, 343 years) collected at Aurora Peak has been analyzed for a homologous series of normal, branched chain, unsaturated, multifunctional dicarboxylic acids, ω -oxocarboxylic acids, pyruvic acid, glyoxal and methylglyoxal using gas chromatography (GC/FID) and GC/mass spectrometry (GC/MS) to understand historical changes in water soluble organic aerosols. Similarly, homologous series of straight chain fatty acids ($C_{12:0}$ - $C_{30:0}$) have been detected by using GC/FID and GC/MS system.

Predominance of oxalic acid was found followed by adipic and succinic acid. Molecular distributions of ω -oxocarboxylic acids are characterized by the predominance of 9-oxononanoic, followed by 4-oxobutanoic and glyoxylic acids. Historical concentrations of diacids, oxoacids and α -dicarbonyls are formed by the oxidation of precursor compounds emitted from the biological and biomass burning activities and they are controlled under climate oscillations and similar meteorological parameters.

Molecular distributions of fatty acids are characterized by even carbon number predominance with a peak at palmitic acid followed by oleic and myristic acids. The historical trends of short-chain fatty acids, together with correlation analysis with inorganic ions and organic tracers, suggest that short-chain fatty acids (except for $C_{12:0}$ and $C_{15:0}$) were mainly derived from sea surface micro layers. In contrast, long-chain fatty acids ($C_{20:0}$ - $C_{30:0}$) are originated from terrestrial higher plants, soil organic matter and dusts, which are linked with Greenland Temperature Anomaly. Hence, this study suggests that Alaskan fatty acids are strongly influenced by Pacific Decadal Oscillation/North Pacific Gyre Oscillation and/or extra tropical North Pacific surface climate and Arctic oscillation.

Organic tracers in ice core were derivatized with N,O-bis-(trimethylsilyl) trifluoroacetamide with 1% trimethylsilyl chloride and pyridine and peaks were determined using a GC/MS system. Levoglucosan, dehydroabietic acid and vanillic acid showed higher concentrations with many sporadic peaks in 1660s-1830s, 1913, and 2005. There are a few discrepancies of higher spikes

among them after 1980s with sporadic peaks in 1994-2007 for dehydroabietic acid. Historical trends of levoglucosan, dehydroabietic acid and vanillic acid showed that biomass burning activities associated with resin and lignin phenols from boreal conifer trees and other higher plants and grasses were significant before 1840s and after 1980s in the source regions of southern Alaska.

Nitrite (NO_2^-), nitrate (NO_3^-), sulfate (SO_4^{2-}) and methanesulfonate (CH_3SO_3^-) were determined in the Alaskan ice core using an ion chromatograph. They have common periods for higher spike during the years of 1665-2008. They are attributed to the same source regions and similar pathways. Interestingly, we found multi-decadal scale atmospheric transport from lower to higher latitudes in the North Pacific, which is reflected in historical concentration trends of anion species.

All the committee members agreed that this dissertation provides a new data set that is useful for the community of atmospheric and ice core chemistry and relevant environmental sciences and that Mr. Pokhrel is honest and enthusiastic as a researcher. In addition to the excellent achievements in the research, his academic records throughout the Ph. D course are excellent. Based on these evidences, the committee reached to a conclusion that Ambarish Pokhrel deserves to become a Doctor of Environmental Science.