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学 位 論 文 審 査 の 要 旨

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学 位 論 文 題 名

Geochemical study on low molecular weight dicarboxylic acids and related compounds in the marine aerosols from the Pacific Ocean
(太平洋エアロゾル中の低分子ジカルボン酸と関連有機物に関する地球化学的研究)

To better understand the sources and atmospheric processing of organic aerosols in the remote marine atmosphere, aerosol samples were collected over the western North Pacific (13°14'N-53°37'N and 140°46'E-179°54'W) during May-July 2010, central Pacific (1°59'N-35°N and 171°54'E-90°58'W) during September to October 1999, and North-South Pacific (3°05'N-34° 02'N, 6°59'S-25°46'S and 144°52'E-173°55'W) during February to March 1994 and were studied for dicarboxylic acids (C₂-C₁₁), ω-oxoacids, pyruvic acid and α-dicarbonyls as well as organic and elemental carbon and water-soluble organic carbon. Over the western North Pacific, diacids are most abundant followed by ω-oxoacids and α-dicarbonyls. Although the molecular compositions of diacids are generally characterized by the predominance of oxalic (C₂) acid, Mozammel Hoque found a depletion of C₂ in four samples, which were collected in the high latitudes (48°N-54°N) of the western North Pacific where succinic (C₄) acid is dominant. He proposed that photochemical degradation of unsaturated fatty acids emitted from the ocean surface produced C₄ over the high latitudinal western North Pacific, where *Chlorophyll a* maximized during the cruise. Moreover, seven samples collected in mid and high latitudes of the western North Pacific showed a predominance of malonic (C₃) acid over C₄, suggesting a photochemical degradation of C₄ to C₃. Spatial distributions of diacids, ω-oxoacids and α-dicarbonyls together with total carbon were characterized by their higher abundances in the coastal western North Pacific followed by high and low latitudinal regions, signifying that continental aerosols are transported long distances to the remote marine atmosphere. However, in the central Pacific aerosols collected between Japan and Mexico, he found a predominance of oxalic (C₂) acid followed by malonic (C₃) and succinic (C₄) acid, where oxalic acid comprises 74% of total diacids. Abundances of oxalic acid were found to be 3 times higher in the upwelling zone of the central Pacific than those of the non-upwelling zone. Positive relation ($r^2=0.79$) obtained between oxalic acid and oxidation products (ωC₂+Gly+Pyr) of biogenic volatile organic compounds (BVOCs) suggests that secondary production of oxalic acid occurs in aqueous aerosol phase via the oxidation of marine derived BVOCs in the upwelling central Pacific. Atmospheric abundances of

short chain (C_2 - C_4) diacids during the North-South Pacific cruise, showed 4 times higher values in the North Pacific than in the South Pacific. During this cruise, abundances of C_2 in the western North Pacific illustrated 5 and 10 time higher values than in the central North and South Pacific, respectively. However, the aerosol samples that were collected in the western North Pacific, demonstrated the predominance of glyoxylic (ωC_2) acid and methylglyoxal (MeGly) together with C_2 . The applicant found a strong correlation between C_2 and ωC_2 ($r^2=0.89$) and C_2 and MeGly ($r^2=0.96$) in the same samples that were collected in the western North Pacific but their correlations are significantly weak for the rest of the samples. These results, together with 7-day backward air masses trajectory, indicate that ωC_2 and MeGly are both photochemical oxidation products of aromatic hydrocarbons in polluted-continental air masses, which can serve as precursors of C_2 during long-range atmospheric transport.

All the committee members agreed that this dissertation provides a new data set that is useful for the community of atmospheric chemistry and relevant environmental sciences and that Mr. Hoque 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 Mozammel Hoque deserves to become a Doctor of Environmental Science.