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学 位 論 文 内 容 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 (TSP) samples were collected over the western North Pacific (13°14'N-53°37'N and 140°46'E-179°54'W) in May-July, 2010, the central Pacific (1°59'N-35°N and 171°54'E-90°58'W) during September-October, 1999, and in the North-South Pacific (3°05'N-34° 02'N, 6°59'S-25°46'S and 144°52'E-173°55'W) were studied for dicarboxylic acids (C_2-C_{11}), 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 (C_2) acid, we found a depletion of C_2 in four samples, which were collected in the high latitudes (48°N-54°N) of the western North Pacific where succinic (C_4) acid is dominant. We consider that photochemical degradation of unsaturated fatty acids emitted from the ocean surface produced C_4 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 show predominance of malonic (C_3) acid over C_4, suggesting photochemical degradation of C_4 to C_3. 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 from Japan to Mexico showed a predominance of oxalic (C_2) acid followed by malonic (C_3) and succinic (C_4) acid, where oxalic acid comprises 74% of total diacids. Abundances of oxalic acid showed 3 order of magnitude higher values in the upwelling zone of the central Pacific than that of the non-upwelling zone. Positive correlation of oxalic acid with biological volatile organic compounds (BVOCs) oxidation products (ωC_2+Gly+Pyr) suggest that secondary production

of oxalic acid occurs in aqueous aerosol phase via the oxidation of marine derived VOCs 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 aerosol samples. During this cruise, abundance of C_2 in the Western North Pacific illustrated 5 to 10 time higher values than in the central North and in the South Pacific, respectively. However, the aerosol samples, which were collected in the western North Pacific, demonstrated that glyoxylic (ωC_2) acid and methylglyoxal (MeGly) were dominant with C_2. We found a strong correlation between C_2 and ωC_2 (r=0.87) and C_2 and MeGly (r=0.97) in the same samples but their correlations are significantly weak for the rest of the samples. These results together with 7-day calculated back-ward air masses trajectory indicate that both of ωC_2 and MeGly are of photochemical oxidation product of polluted-continental air mass, which are then serve as precursor of C_2 during long-range atmospheric transport.