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东海大气气溶胶中二元羧酸的分布特征及来源

Distributions and sources of dicarboxylic acids in atmospheric aerosols over the East China Sea

关键词：二元羧酸 乙二酸 气溶胶 东海

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摘要：大气气溶胶中的二元羧酸因其在全球气候变化中的潜在作用而受到广泛关注.利用2011年5月12日—6月6日在东海采集的气溶胶样品,分析其中水溶性二元羧酸及常量离子浓度,探讨东海气溶胶中二元羧酸的时空分布特征及来源.结果显示东海大气气溶胶中乙二酸、丙二酸和丁二酸的浓度分别为26.0~1475.5 ng·m^{-3}、0.1~61.4 ng·m^{-3}和0.1~132.4 ng·m^{-3},乙二酸在这3种二元羧酸中的贡献最大,为88.3%.东海气溶胶中二元羧酸浓度的昼夜变化不显著.空间分布整体呈现近海高、远海低的趋势.气团的来源和迁移路径以及气象因素影响气溶胶中二元羧酸的分布,气团来自污染较重的陆源时气溶胶中二元羧酸的浓度较高,气团来自清洁的海洋源时,二元羧酸的浓度则较低;阴雾天气时气溶胶中二元羧酸浓度相对较高,降雨发生时二元羧酸的浓度较低.二元羧酸与常量离子的相关性分析表明,自然源和人为源释放的挥发性有机物质在液相中氧化生成二元羧酸是东海大气气溶胶中二元羧酸的主要源,而汽车尾气和生物质燃烧的一次排放、海洋源以及碱性粗颗粒吸收气体二元羧酸不是主要源.液相中乙醛酸氧化形成的乙二酸和长链二元羧酸氧化形成的乙二酸对东海气溶胶中乙二酸的贡献分别为41%和59%.

Abstract: Dicarboxylic acids in the atmospheric aerosols have received great attention because of their potential roles in influencing the global climate. 26 total suspended particles (TSP) samples were collected over the East China Sea during the period from 12 May to 6 June 2011. Concentrations of water-soluble dicarboxylic acids and major ions in TSP were measured to investigate their temporal and spatial distribution characteristics and to identify potential sources. The concentrations of oxalic acid, malonic acid and succinic acid were 26.0~1475.5 ng·m^{-3}, 0.1~61.4 ng·m^{-3} and 0.1~132.4 ng·m^{-3}, respectively. Oxalic acid was the most abundant dicarboxylic acid, accounting for 88.3% of the sum of the three dicarboxylic acids. No significant difference between daytime and nighttime samples was observed for the concentrations of dicarboxylic acids. The concentrations of dicarboxylic acids decreased with increasing distance from the coast. The spatial distributions of dicarboxylic acids in aerosols were associated with sources and transport pathways of air mass and meteorological factors. High concentrations of dicarboxylic acids were observed when the air mass passed over polluted regions. Low concentrations were observed when the air mass came from the clean marine atmosphere. The concentrations of dicarboxylic acids also increased in cloudy and fog days, while decreased in rainy days. Correlation analysis between dicarboxylic acids and major ions suggested that aqueous-phase oxidation of volatile organic compounds from natural/anthropogenic sources was likely the major formation pathway of dicarboxylic acids in the East China Sea. Dicarboxylic acids were mainly emitted from vehicular exhaust/biomass burning, while marine sources and dicarboxylic acids in the gas phase adsorbed onto alkaline coarse particles made little contribution. Aqueous secondary productions of oxalic acid via the oxidation of glyoxylic acids and decays of higher diacids contributed to 41% and 59% of the level of oxalic acid in the East China Sea, respectively.

Key words: dicarboxylic acids oxalic acid aerosol the East China Sea