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Carbon Speciation in Sea Spray Aerosol by Scanning Transmission X-ray Microscope with Near-Edge X-ray Absorption Fine Structure (STXM/NEXAFS) Spectroscopy

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1 Introduction

Sea spray aerosol (SSA) is one of the most dominant natural aerosol particles. Recently, SSA generation experiments have been conducted to clarify carbon species and their concentration processes in fine SSA because organic carbon (OC) can control hygroscopic behavior of fine SSA (below 2.5 μm) [1]. On the other hand, carbon speciation based on field observation was scarce, although atmospheric reaction is an important source of OC in aerosol particles. In this study, carbon speciation in SSA above the Antarctic and the equatorial Pacific Oceans were conducted by scanning transmission X-ray microscope with near-edge X-ray absorption fine structure (STXM/NEXAFS) spectroscopy. Biological activity in surface seawater at the Antarctic Ocean is much higher than that at the equatorial Pacific Ocean. Therefore, we can evaluate emission sources of carbon such as suspension of organic carbon from seawater, chemical reaction in atmosphere and biological emission

2 Experiment

Size-fractionated aerosol particles above the Antarctic and the equatorial Pacific Oceans were collected on molybdenum grid with formvar coating by a high volume air sampler with cascade impactors during research cruise of *R/V Hakuho-Maru* (KH-14-6: GEOTRACES).

Functional group of carbon were analyzed by STXM/NEXAFS at BL-13A, Photon Factory. STXM can determine distribution of carbon functional group in single SSA because incident X-ray beam is focused by a Fresnel zone plate to a spot size about 30 nm. The scanning of the sample were conducted by piezo-controlled scanning stage at fixed X-ray position. Transmission X-ray was detected by photomultiplier. All experiments were conducted under 1×10^4 Pa Helium condition. Details of the schematic optics are described in Takeichi *et al.* [2]. Pre (280 eV) and post-edge (300 eV) of carbon K-edge maps were obtained to decide target particle from the optical density (OD). Image-stacks were conducted around the target particle, which is obtained by transmitted X-ray map with small energy step (0.1 eV to 0.3 eV). Carbon K-edge NEXAFS spectra were obtained by the differences of OD at each energy in interest positions/regions using the software aXis2000.

3 Results and Discussion

In this study, 32 SSA particles were analyzed. Dominant carbon functional group in about two thirds of measurement particles were carboxylate (-COOH) and carbonate ($-CO_3$), which were distributed on the aerosol edge (Fig. 1). These results are comparable to those of SSA generation experiments with STXM analysis [3]. It is considered that OC in fine SSA particles is derived from seawater rather than secondary formation by atmospheric reactions. Other particles contained two or three of C=C, C≡C, C-O-C, -COOH, and $-CO_3$, which were distributed not only on aerosol edge but also in aerosol core. These particles found in high biological activity region: the Antarctic Ocean. Therefore, formation processes of these OC particles may be related with biological activity in surface seawater. In addition of these particles, soot particles were also detected, which were transported from continental region. Thus, various particles can be detected by carbon speciation via field sampling. Further investigation is needed to clarify OC species and their emission sources accurately.

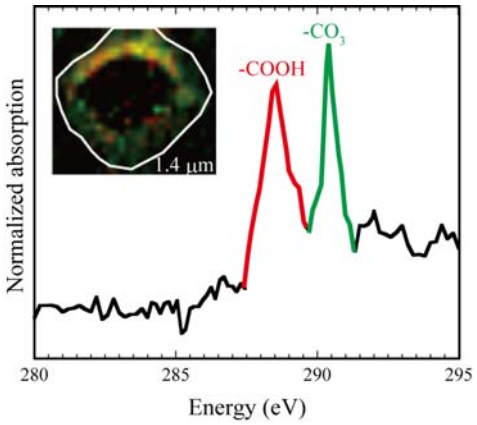

Fig. 1 Representative carbon K-edge NEXAFS spectrum and a map of carbon distribution in fine SSA.

References

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