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Effect of phytoplankton biomass in seawater on chemical properties of sea spray aerosols

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ABSTRACT

This study is to investigate the effect of biological seawater properties on sea spray aerosols (SSA). Concentrations of chlorophyll-a and bacteria were measured at coastal site in Korea in fall and summer seasons. Also, aerosol mass spectrometer (AMS) was used to determine chemical constituents (organics, sulfate, nitrate, ammonium, and chloride) of non-refractory submicrometer aerosols sprayed from seawaters using a bubble bursting system. The average concentration of chlorophyll-a in seawater in fall was $1.75 \pm 0.78 \mu\text{g/l}$, whereas it significantly increased to $5.11 \pm 2.16 \mu\text{g/l}$ in summer. It was found that the fraction of organics in the submicrometer SSA was higher in summer (68%) than fall (49%), and that the organic fraction in the SSA increased as the concentration of chlorophyll-a increased in seawater, suggesting that the high phytoplankton biomass in seawater could lead to the enhancement of organic species in the SSA.

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1. Introduction

Marine aerosol is one of the largest contributors to ambient aerosols on earth (Andreae, 1995). They affect the earth's radiation balance (Slingo, 1990; Murphy et al., 1998), cloud formation by acting as cloud condensation nuclei (CCN) (O'Dowd et al., 2004; Fuentes et al., 2010; Moore et al., 2011; Ovadnevaite et al., 2011; Westervelt et al., 2012), and biological cycle between the ocean and the air (O'Dowd et al., 2004; Facchini et al., 2008). A wave breaking from the sea surface under high wind speed results in the production of film and jet droplets in the ambient atmosphere, leading to the formation of primary marine aerosols. The droplets contain inorganic salts, organics (e.g., biological materials), clay minerals, and other materials (Wells and Goldberg, 1992; O'Dowd et al., 1997; Fuentes et al., 2010). Secondary marine aerosols are formed by gas-to-particle conversion of precursor gases emitted from the sea (e.g., dimethyl-sulfide (DMS) and volatile organic species) (Charlson et al., 1987; O'Dowd et al., 2002). Thus, chemical characteristics of primary and secondary marine aerosols should be dependent on properties of the seawater. Also, continental aerosols can be transported over the ocean and mixed with marine aerosols (Zorn et al., 2008; Crippa et al., 2013). Thus, various production and mixing processes

may lead to a complex mixing state of marine aerosols (Ceburnis et al., 2008).

A number of studies have provided evidence for the presence of a significant fraction of organics in submicrometer marine aerosols (Cavalli et al., 2004; O'Dowd et al., 2004, 2008; Miyazaki et al., 2010; Rinaldi et al., 2010; Prather et al., 2013; Quinn et al., 2015). Organic aerosols (OA) observed at coastal areas can be produced from nearby marine and continental sources (O'Dowd et al., 1997; Vaattovaara et al., 2006; Crippa et al., 2013) and/or transported over a long distance. Facchini et al. (2008) found that organics accounted for up to ~74% of the primary marine aerosols smaller than $0.5 \mu\text{m}$ during a phytoplankton bloom in the North Atlantic. The global emissions of primary marine organic aerosols (PMOA) was calculated to be approximately 10 Tg year^{-1} (Spracklen et al., 2008; Gantt et al., 2009; Vignati et al., 2010), and the change in aerosol indirect forcing due to PMOA was estimated to be 0.1 W m^{-2} (Meskhidze et al., 2011; Gantt et al., 2012). PMOA can include toxic marine bacteria (e.g., *Lyngbya majuscula*) that originate from seawater during a phytoplankton bloom (Osborne et al., 2001). In addition, dissolved organic species in seawater can be included in PMOA during the drying process in the ambient atmosphere (Ogawa and Tanoue, 2003; Hansell et al., 2009; Gantt and Meskhidze, 2013). Their contribution may not exceed 30% of the total mass concentration of biological materials smaller than 700 nm in seawater (Ogawa and Tanoue, 2003). However, it has been difficult to determine the

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fraction of PMOA in the total OA, and their effects on climate change are still uncertain due to the lack of a detailed chemical characterization of PMOA.

Phytoplankton, bacteria, and viruses can be major types of biological materials in seawater that contribute to PMOA (Yamasaki et al., 1998; Stramski et al., 2004). Typical concentration levels for phytoplankton, bacteria, and viruses in seawater are 10^4 – 10^5 /ml, 10^5 – 10^6 /ml, and 10^6 – 10^7 /ml, respectively (Maranger and Bird, 1995; Yoo, 2008). Variations in these concentrations have been found to follow a seasonal pattern depending on the physicochemical parameters such as seawater temperature, nutrient level, degree of turbulent mixing, and solar irradiation (Hyun and Kim, 2003). It was also reported that during a phytoplankton boom, the elevation of biological materials in seawater occurred as a function of time in the order 1) chlorophyll-a, 2) bacteria, and 3) viruses (Cochlan et al., 1993; Shin et al., 2003).

The aerosol mass spectrometer (AMS) (Aerodyne Inc., USA) has been a useful tool to determine the chemical characteristics of non-refractory submicrometer particles in real time and to identify the type of organics (Topping et al., 2004; Bates et al., 2005; Takami et al., 2005; Coe et al., 2006; Phinney et al., 2006; Zorn et al., 2008; Shank et al., 2011). Various characteristic marker fragments in the AMS mass spectra have been used to identify OA types, such as hydrocarbon-like OA (HOA), oxygenated OA (OOA), biomass burning OA (BBOA), cooking OA (COA), methanesulfonic acid OA (MSA-OA), and primary biological OA (PBOA) (e.g., amino acid OA (AA-OA)) (Zorn et al., 2008; Mohr et al., 2012; Ng et al., 2011; Schneider et al., 2011). Schneider et al. (2011) derived markers of primary biological particles using laboratory-generated amino acids and carbohydrates that can be major chemical components (60–80%) of biological cells (e.g., bacteria and viruses). They reported that the fragments at m/z 30 (CH_4N^+) and 42 ($\text{C}_2\text{H}_4\text{N}^+$) and fragments at m/z 60 ($\text{C}_2\text{H}_4\text{O}_2^+$), 61 ($\text{C}_2\text{H}_5\text{O}^+$), and 73 ($\text{C}_3\text{H}_5\text{O}_2^+$) in the AMS mass spectra can be used as markers for amino acid and carbohydrate particles, respectively. In the pristine rainforest of the central Amazon Basin, Brazil, the amino acid-OA fraction was found to be 7.5% of the total OA (Schneider et al., 2011). The fragmentation patterns of real marine bacteria should be required to identify the primary biological particles that originate from seawater.

In this study, concentrations of biological materials such as chlorophyll-a (>700 nm) and bacteria (200 nm–1 μm) in seawater were measured to examine biological properties of the seawater. The AMS was used to determine chemical constituents (organics, sulfate, nitrate, ammonium, and chloride) of aerosols sprayed from seawaters using a bubble bursting system (i.e., sea spray aerosol (SSA)). This will enable us to determine the relationship between concentrations of chlorophyll-a and bacteria in seawater and chemical properties of the SSA. Also, AMS mass spectra of organic aerosols sprayed from natural seawater and deionized water containing laboratory-cultured marine

bacteria (e.g., *Vibrio littoralis*, *Flavobacterium* sp., and *Pseudomonas aeruginosa*) were used to determine marine organic mass spectra.

2. Experimental methods

Seawater samples were obtained from the South Sea of Korea (Boseong) once per day in the fall of 2012 (10/29/2012–11/16/2012), two times per day in the summer of 2015 (8/24/2012, 8/31/2015), and two times per day in the fall of 2015 (10/21/2015, 10/28/2015). Fig. 1 shows the location of the seawater sampling. All seawater samples were collected from sea surface at a depth of ~30 cm in coastal area several meters away from shore. The surface seawater was collected into a sterilized bottle (20 l). Each sample was placed in a portable icebox to minimize any biological activity during transportation. The biological analysis was conducted within 3 h after sampling.

The schematic of experimental setup to measure properties of seawater and SSA is shown in Fig. 2. The mass concentration of chlorophyll-a ($\mu\text{g/l}$) in the seawater sample filtered by a glass fiber membrane (Whatman, Germany) with a pore size of 700 nm was measured using a UV spectrophotometer (Optizen 2120UV, MECASYS, Korea). To quantify the bacteria in the seawater, an epifluorescent microscope was used (Niven et al., 1995; Maranger and Bird, 1995). This provides concentrations of free-living bacteria in the size range of 200 nm–1 μm (bacteria/ml) in seawater samples. A detailed description of the biological measurements can be found elsewhere (Park et al., 2014). A total dissolved solid (TDS) meter (Oakton Instruments, Singapore) was used to determine mass concentration (ppm) of total dissolved solids (TDS) in seawater. The pH and conductivity of seawater samples were measured by pH meter (TOA DKK Corp., Japan) and TDS meter (Oakton Instruments, Singapore), respectively.

Deionized (DI) water samples containing three types of marine bacteria (*Pseudomonas aeruginosa*, *Vibrio littoralis*, and *Flavobacterium* sp.) and seawater samples (sampled at Boseong, Korea) were used to produce sea spray aerosols (SSA) by using the bubble bursting system as shown in Fig. 2. *Vibrio littoralis* and *Flavobacterium* sp. were cultured in marine broth (Difco, USA) at 37 °C, and *Pseudomonas aeruginosa* was cultured in nutrient broth (Difco, USA) at 25 °C. The marine broth media contain minerals and nutrients that are similar to those in seawater. The microfiltration membrane (0.45 μm pore) was used to remove any particles larger than 0.45 μm in each sample.

To determine chemical constituents of the primary marine aerosols (organics, sulfate, nitrate, ammonium, and chloride), each seawater sample was aerosolized by the bubble bursting chamber to mimic the production of primary marine aerosols on the sea surface (i.e., sea spray aerosols). The bubble bursting chamber (Park et al., 2014) consisted of a sintered glass filter with 20–40 μm pore sizes (Millipore, USA) inserted at the bottom. A clean compressed air was introduced

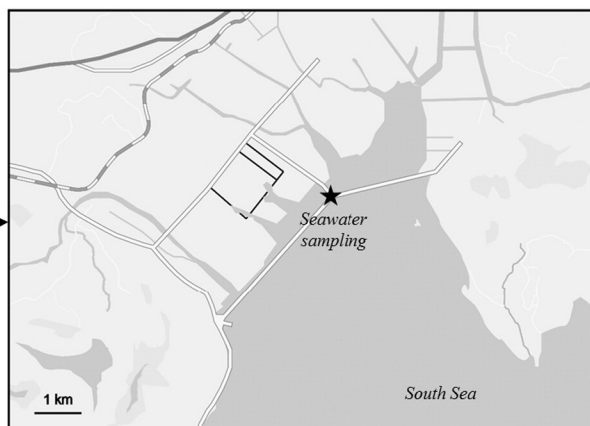


Fig. 1. Map of seawater sampling location.

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