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The distributions and direct radiative effects of marine aerosols over East Asia in springtime

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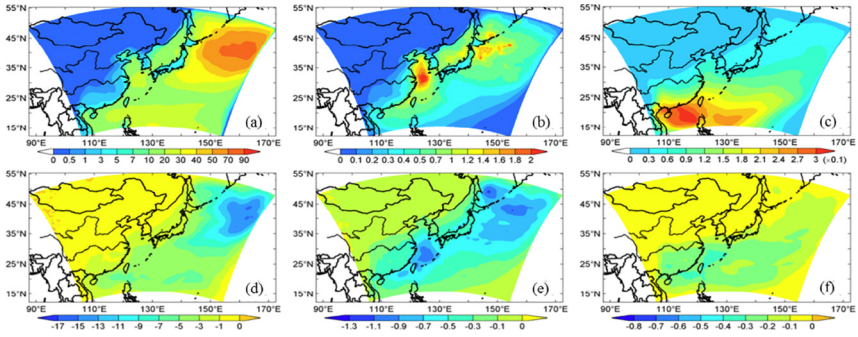
HIGHLIGHTS

- The distributions and direct radiative effects of marine aerosols were explored with RIEMS-Chem.
- The maximum mean levels of sea salt and MPOA exceeded 70 μg m^{-3} and 2 μg m^{-3} in the western Pacific Ocean.
- The all-sky DRE by marine aerosols was −3.5 W/m^2, accounting for 27% of the DRE by total aerosols over the ocean.
- The DRE by MPOA was comparable to that by sea salt in regions of plankton bloom, such as the East China Sea.

GRAPHICAL ABSTRACT

The model simulated mean near-surface concentrations (unit: μg m^{-3}) of (a) sea salt, (b) MPOA, (c) DMS-produced aerosols, and clear-sky direct radiative effects (unit: W/m^2) at the top of the atmosphere by (d) sea salt, (e) MPOA, (f) DMS aerosols in spring 2014.



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ABSTRACT

The characteristics, distributions, and direct radiative effects (DRE) of marine aerosols in the western Pacific Ocean over East Asia during the period from 17 March to 22 April 2014 were investigated by an online-coupled regional atmospheric chemistry/aerosol-climate model RIEMS-Chem (Regional Integrated Environmental Model System with Chemistry). The emissions and relevant processes of sea salt, marine primary organic aerosol (MPOA), sulfate and Methyl sulfonic acid (MSA) produced from dimethylsulfide (DMS) were parameterized and coupled with RIEMS-Chem. The model results for total aerosol masses (PM_{10} and $PM_{2.5}$), inorganic and carbonaceous aerosols, gas precursors, and aerosol optical depth (AOD) were compared with various observational data sets including a research cruise Dongfanghong II from the Yellow Sea to the open oceans, near-surface aerosol and gas concentrations from the Acid Deposition Monitoring Network in East Asia (EANET) and China National Environmental Monitoring Center (CNEMC), and AOD from the Aerosol Robotic Network (AERONET). Model comparisons demonstrated a generally good skill of the RIEMS-Chem in representing the temporal and spatial variations of these variables. The distributions of marine aerosols were characterized by the maximum sea salt concentration up to 70 μg m^{-3} in the ocean northeast of Japan, the maximum concentration of MPOA >2 μg m^{-3} in the East China Sea and in portions of the northwest Pacific (NWP) region, and the maximum DMS-produced aerosol concentration >0.3 μg m^{-3} in the southern parts of the ocean. It was noteworthy that marine aerosols can be easily transported to the inland areas of south China. The clear-sky DREs by sea

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salt ranging from −9 to −17 W/m^2 occurred in the open oceans northeast of Japan, comparable to the DREs of −10 ~ −20 W/m^2 by anthropogenic aerosols, whereas the DREs by MPOA were strongest (up to −1.3 Wm^{-2}) in the East China Sea and the oceans northeast of Japan due to active phytoplankton blooms there and comparable in magnitude to the DREs by sea salt (around −3 Wm^{-2}) in the East China Sea. The maximum DRE by the DMS-produced aerosols was −0.4 Wm^{-2} mainly in the northern parts of the South China Sea. Sea salt exhibited an increasing radiative importance from the China marginal seas to the open oceans, accounting for 10% and 33% of the DREs by all aerosols, respectively. Under all-sky conditions, the sum of DREs by all the marine aerosols were estimated to be −2.2 W/m^2, −3.5 W/m^2, −2.3 W/m^2, and −4.3 W/m^2 averaged over the entire domain, ocean, East China Sea, and the NWP region, accounting for 20%, 27%, 13%, and 36% of the DREs by all aerosols, respectively, which demonstrated the important role of marine aerosols in modulating shortwave radiation in springtime in the western Pacific Ocean which was just downwind of the Asian continent with large amounts of anthropogenic and dust emissions.

1. Introduction

Aerosols affect radiation transfer by scattering or absorbing solar and infrared radiation, by acting as cloud condensation nuclei to modify cloud properties, and by heating atmosphere to alter cloud formation, which are known as aerosol direct radiative effect, indirect effect, and semi-direct effect, respectively (Ramanathan et al., 2001; IPCC, 2013). The climatic effects by total aerosols are potentially as important as those caused by greenhouse gases but with more uncertainties and with an opposite sign. Different from the radiative effect of greenhouse gases, which is homogeneous across the earth, aerosols tend to exert a stronger radiative effect on the regional scale, and to date our knowledge on regional radiative effect of aerosols is still limited (Vogel et al., 2009).

East Asia is one of the regions experiencing high aerosol loading in the world. Due to continuous economic growth and large energy consumption in the past decades (Smith et al., 2011; Li et al., 2017), aerosols from anthropogenic activities have been kept at a high level. In addition, East Asia is also one of the major natural aerosol sources in the world. Dust events frequently occur in springtime and exert significant influences on air quality and climate system (Shao and Dong, 2006; Han et al., 2012). The vast western Pacific Ocean in East Asia contributes greatly to marine aerosols including sea salt, organic aerosols (OC), sulfate produced from dimethylsulfide (DMS), especially during the periods of high wind speed and active phytoplankton activity (Gong, 2003; Thomas et al., 2010; Gantt et al., 2009, 2012a; Rap et al., 2013). To establish a better understanding of the impacts of anthropogenic aerosols on climate system, it is of importance to separate the anthropogenic aerosols from the natural ones.

The anthropogenic and mineral dust aerosols have been extensively investigated globally and regionally (e.g. Ginoux et al., 2001; Shao and Dong, 2006; Han et al., 2012). However, so far the studies on aerosols from oceans, especially over the western Pacific ocean, were still very limited (e.g. Zakey et al., 2008; Gantt et al., 2011; Yue and Liao, 2012; Rap et al., 2013). Given that about 70% of the earth's surface is covered by oceans, the global emissions of marine aerosols are significantly larger than that of anthropogenic aerosols (Chin et al., 2009), and the investigation of marine aerosols should be of significant importance for understanding of the climatic effects of both anthropogenic and natural aerosols.

There have been a number of studies on the behaviors and radiative effects of marine aerosols (Grini et al., 2002; Takemura et al., 2002; Liao et al., 2004; O'Dowd et al., 2004; Meskhidze and Nenes, 2006; Kloster et al., 2006; Ayash et al., 2008; Ma et al., 2008; Vignati et al., 2010), mostly on sea salt aerosol at the global scale. Heald et al. (2014) estimated with GEOS-Chem a global annual mean DRE of −1.1 W/m^2 by sea salt at the top of the atmosphere (TOA) under clear-sky condition, whereas Yue and Liao (2012) derived a stronger global mean TOA radiative effect of −2.0 W/m^2. Using a regional climate model RegCM, Zakey et al. (2008) reported an annual mean clear-sky direct radiative forcing of −1.0–0 W/m^2 at TOA by sea salt over the Mediterranean Sea and −2.0 ~ −1.0 W/m^2 over the northern Atlantic Ocean. Lundgren et al. (2013) estimated a weaker clear-sky DRE of ~−0.1 W/m^2 at TOA in the Mediterranean Sea in summer. Guo et al. (2015) estimated an annual mean clear-sky DRE of −1.4 W/m^2 at TOA by sea salt over East Asia. Compared with sea salt, there were few studies on the radiative effects of DMS-derived aerosols (sulfate and methylsulfonic acid oxidized from DMS). Graf et al. (1997) indicated that DMS-sulfate induced a global annual mean DRE of −0.17 W/m^2 at TOA, with a weaker value in the northern hemisphere (−0.15 W/m^2) and a stronger one in the southern hemisphere (−0.19 W/m^2). Rap et al. (2013) estimated a global annual mean DRE of −0.35 W/m^2 at TOA due to DMS-sulfate under clear-sky condition and of −0.23 W/m^2 under all-sky condition. Thomas et al. (2010) and Mahajan et al. (2015) estimated the global annual mean all-sky DREs at TOA (by both direct and indirect effects) due to DMS-produced aerosols to be −2.0 W/m^2 and −1.8 W/m^2, respectively. Marine primary organic aerosol (MPOA) is another important marine aerosol component generated by phytoplankton biogeochemical processes (O'Dowd et al., 2004; Gantt et al., 2011). Previous studies explored the potential impact of MPOA on cloud microphysical properties and indirect radiative effects (Novakov et al., 1997; O'Dowd et al., 2004; Meskhidze et al., 2011; Ovadnevaite et al., 2011; Gantt et al., 2012b). However, the investigation of direct radiative effects of MPOA was very limited at both global and regional scales.

East Asia is an area with high aerosol loadings from complex sources. While anthropogenic aerosols have been explored extensively, the behaviors and radiative effects of marine aerosols remain poorly understood due to limitations in both observation and modeling studies. To our knowledge, this study is the first attempt to characterize and quantify the distributions and direct radiative effects of major aerosol components from marine origins in the western Pacific Ocean. This paper is organized as follows: the emission models for marine aerosols and precursors coupled with the online coupled chemistry-climate model RIEMS-Chem, model options and observational data sets are described in Section 2. Section 3 conducts a detailed model comparison for gas, aerosol components, and aerosol optical depth against a series of observations from the ground-based and cruise measurements. The distributions and direct radiative effects of marine aerosols relative to total (anthropogenic) aerosols are investigated and quantified in section 4. Section 5 summarizes the main model results and conclusions.

2. Model and data

2.1. Model description

The online-coupled regional atmospheric chemistry/aerosol-climate model RIEMS-Chem was used in this study. The host regional climate model RIEMS includes a series of modules and parameterizations to represent major physical processes, such as land surface (the Biosphere-Atmosphere Transfer Scheme, BATS; Dickinson et al., 1993), planetary