

Sea spray aerosol concentration modulated by sea surface temperature

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Natural aerosols in pristine regions form the baseline used to evaluate the impact of anthropogenic aerosols on climate. Sea spray aerosol (SSA) is a major component of natural aerosols. Despite its importance, the abundance of SSA is poorly constrained. It is generally accepted that wind-driven wave breaking is the principle governing SSA production. This mechanism alone, however, is insufficient to explain the variability of SSA concentration at given wind speed. The role of other parameters, such as sea surface temperature (SST). remains controversial. Here, we show that higher SST promotes SSA mass generation at a wide range of wind speed levels over the remote Pacific and Atlantic Oceans, in addition to demonstrating the wind-driven SSA production mechanism. The results are from a global scale dataset of airborne SSA measurements at 150 to 200 m above the ocean surface during the NASA Atmospheric Tomography Mission. Statistical analysis suggests that accounting for SST greatly enhances the predictability of the observed SSA concentration compared to using wind speed alone. Our results support implementing SST into SSA source functions in global models to better understand the atmospheric burdens of SSA.

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ver two-thirds of the Earth is covered by the ocean. The material exchange between the ocean and the atmosphere affects the balance of the Earth's energy on a global scale (1). Sea spray aerosol (SSA) is the major particulate material directly emitted from the ocean. Studies have shown that SSA dominates the aerosol mass in the marine boundary layer (MBL). Such dominance renders SSA an important player in climate change (2). However, the exact processes by which the SSA is introduced to the atmosphere still remains to be learned, making the SSA budget highly uncertain (3).

It is generally established that SSA is produced by mechanical processes (4-6). Wind stress induces breaking waves that entrain bubbles into the surface ocean (7). Film and jet drops formed during bubble bursting are the main sources of SSA particles (8). The wind-driven mechanism is supported by the positive correlation between wind speed and SSA concentration from field observations (9, 10). Therefore, wind speed is used as the sole parameter to characterize SSA in many models (1, 4, 11).

In addition to wind speed, sea surface temperature (SST) may play a large role in SSA production (12–15). SST affects the drop formation process by modifying the physical properties of the surface ocean water. An increase of SST reduces the kinematic viscosity and surface tension of the ocean, thereby enhancing the entrainment efficiency and rising speed of bubbles (12, 16). As a result, the number size distribution of the bubbles may change, leading to varying SSA properties (14, 15, 17).

Limited laboratory and field studies regarding the effects of SST on SSA production have shown disparate results. Some argue that SSA production is independent of SST (18) or suppressed by increasing SST (14, 15, 19, 20) from 0 to 10 °C, while other laboratory (12, 21-23) and field measurements (3, 5) suggest that SSA production increases monotonically with water temperature. Furthermore, recent observations in the remote Atlantic Ocean shows that increasing SST enhances the modal mean diameter of SSA (24). On the other hand, model simulations have demonstrated that incorporating SST into SSA source functions generally improves the SSA prediction (3, 25, 26). The inconsistency in the previous work suggests that the impacts of SST on SSA formation remain unclear.

In this study, we conducted unprecedented aircraft measurements of SSA concentration on a global scale during the Atmospheric Tomography Mission (ATom). These measurements consist of a series of flights spanning three seasons (summer, fall, and winter) over remote oceans (Fig. 1 and SI Appendix, Fig. S1). Our observations again confirm that wind speed is the dominant factor controlling the concentration of SSA. Further, we show that increasing SST enhances the mass concentration of SSA.

Significance

Climate models use pre-industrial atmosphere as the reference to evaluate the impacts of human activities on the Earth's radiation balance. Sea spray aerosols (SSA) are the key component in the relatively pristine preindustrial conditions that substantially affect model calculations. Currently, the abundance of SSA is poorly constrained. In particular, studies on the influence of sea surface temperature on SSA production have shown disparate results. This uncertainty arises from limited field measurements, especially over remote oceans. Our global aircraft measurements over the remote Pacific and Atlantic Oceans show that higher sea surface temperature enhances the production of SSA. Updating the current parameterization in global models using our observational constraints will improve the estimate of atmospheric SSA budget and human-

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