

***SEA-SALT AEROSOL PARTICLES DURING 2008 VOCALS:
PRODUCTION, DISTRIBUTION, AND MODIFICATION BY GAS, AEROSOL,
AND IN-CLOUD PROCESSES***

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ABSTRACT

Bulk sea-salt aerosol (SSA) mass concentration in the marine boundary layer (MBL) was determined as Na^+ and Cl^- using the PILS-IC technique deployed on the DOE G-1 aircraft during the 2008 VOCALS field campaign. Although the SSA concentrations were in rough agreement with the predicted values that are typically uncertain to a factor of 3, they were rather insensitive to wind speed. This suggests that the MBL steady state SSA concentration (observed over a wind speed range 2 to 10 m s^{-1}) was governed by the residence time of an air parcel entraining the SSA particles as well as the SSA production rate, the two processes having the opposite wind speed dependencies. Regarding SSA chemistry, a lowered Cl^- to Na^+ ratio in SSA particles compared to that of sea-water was observed. This SSA modification, referred to as Cl^- deficit, results from acidification of SSA by strong acids, primarily HNO_3 and H_2SO_4 , followed by desorption of HCl . Because the uptake of gaseous HNO_3 by SSA accounted for only a portion of the Cl^- deficit, H_2SO_4 was also implicated. While in-cloud H_2SO_4 production from oxidation of SO_2 by H_2O_2 and O_3 are potentially important, coagulation (in clear air and in cloud) of SSA particles with strongly acidic H_2SO_4 aerosols advected into the MBL from coastal sulfur emission sources characteristic of the VOCALS study region also needs to be investigated. The relative importance of acidification of SSA by H_2SO_4 through gas-phase, aqueous-phase, and coalescence processes are compared using time constants evaluated for a range of concentrations of O_3 , H_2O_2 (not measured), and SO_2 (< 0.2 ppb) and coagulation coefficients appropriate for aerosol particles and cloud droplets.

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