



Ship-plume sulfur chemistry in the ITCT 2K2 experiment

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Estimation of the lifetime is a conventional way of examining the atmospheric evolution of pollutants. In this study, the ship-plume lifetimes of SO_2 were estimated for ITCT 2K2 (Intercontinental Transport and Chemical Transformation 2002) ship-plume case using a ship-plume photochemical/dynamic model. The magnitude of ship-plume SO_2 lifetimes is mainly affected by two main factors: (i) the levels of in-plume hydroxyl radicals (OH) and (ii) pH of sea-salt particles (pH_{ss}). The former is governed by stability condition of marine boundary layer (MBL) when the ship-emitted NO_x flux is fixed. The latter determines magnitude of aqueous-phase oxidation coefficients. SO_2 is oxidized inside deliquescent sea-salt particles via two main reactions: (1) bisulfite (HSO_3^-) + dissolved H_2O_2 at $\text{pH}_{ss} < 6.5$ and (2) sulfite (SO_3^{2-}) + dissolved O_3 at $\text{pH}_{ss} > 6.5$. According to the multiple ship-plume photochemical/dynamic model simulations, the estimated SO_2 lifetimes over the entire ship plumes ranged from 8.7 to 17.9 hrs under the neutral (D) to stable (F) MBL conditions, when pH_{ss} was assumed to be ≤ 7 . These values are clearly shorter than the background SO_2 lifetime of 23.2 hrs. Moreover, dependence of SO_2 loss budget on pH_{ss} was investigated. The changes of the SO_2 loss budget with pH_{ss} clearly show a shift in the dominant SO_2 loss processes, e.g., when $\text{pH}_{ss} < 6.5$, approximately 60% of the SO_2 is lost by the gas-phase oxidation by OH radicals, whereas 96% of SO_2 is destroyed by the massive heterogeneous conversion of SO_2 when $\text{pH}_{ss}=8$.