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Development and applications of newly-developed photochemical/dynamic ship-plume model

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A photochemical/dynamic ship-plume model, which can consider the ship-plume dynamics and ship-plume chemistry, simultaneously, was developed to gain a better understanding of atmospheric impact of ship emissions. The model performance was then evaluated by a comparison with the observation data measured on a NOAA WP-3D flight during the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) airborne field campaign. The simulation conditions and parameters, such as meteorological conditions, emission rates, and background gas and particulate species concentrations, were obtained directly and/or inferred indirectly from the ITCT 2K2 observation data. The model-predicted concentrations showed good agreement with the observed concentrations of five ambient species (NO_x, NO_y, ozone, HNO_3, and H_2SO_4) at the eight plume transects by the WP-3D flight with strong correlations around the 1:1 line ($0.64 \leq R \leq 0.85$). In addition, a set of tests were carried out to approximate the magnitude of the reaction probability of HNO_3 onto sea-salt particles in the model-observation comparison framework. These results suggest that the reaction probability of HNO_3 onto sea-salt particles may be in the order of 0.05-0.1. The equivalent NO_x lifetime throughout the "entire plume" was also estimated from photochemical/dynamic ship-plume modeling. The NO_x lifetimes estimated throughout the entire ship plume ranged from 2.64 hrs to 3.76 hrs under stable to neutral stability conditions. The short NO_x lifetime over the entire ship plume clearly shows that the ship-plume chemistry shortens the NO_x lifetime considerably. Moreover, numerical analysis was carried out to exam the detailed sources (and/or source contributions) of the elevated levels of HCHO in the ship corridors. From the multiple simulations, CH_4 oxidation by elevated levels of OH radicals is found to be mainly responsible for the elevated levels of HCHO in the ship corridors. More than $\sim$90% of HCHO within the entire ship plumes is produced by this atmospheric chemical process, except for very near the ship stacks where the main source of the elevated HCHO levels would be primary HCHO from ships (due to deactivation of CH_4 oxidation from the depletion of OH radicals). Therefore, the ship-plume chemistry model should be used to model the changes in ship-plume chemical compositions and better evaluate the atmospheric impact of ocean-going ship emissions.