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Accounting for non-linear chemistry of shipping plumes in the GEOS-Chem global chemistry transport model

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Current chemistry transport models (CTMs) generally apply instantaneous mixing of shipping emissions over the model grid cells, thereby neglecting the effects of non-linear, in-plume chemistry, and overestimating NO_x concentrations and subsequent ozone production over the oceans. In this study, we adapted a Gaussian plume dispersion model with chemistry, to explicitly simulate NO_x decay and ozone production during the early stages of plume dispersion. By taking non-linear, in-plume chemistry into account, we can improve ozone simulations over the oceans, but our main goal is to achieve a meaningful comparison between simulated NO_x concentrations and observed tropospheric NO_2 columns from satellite sensors over a number of distinct shipping lanes. In order to account for the effects of in plume chemistry in the global GEOS-Chem CTM, we parameterized the dispersion model by constructing a look-up table (LUT) with the fraction of NO_x remaining and the integrated ozone production 5 hours after initial release and implemented it. The fraction of NO_x remaining and the integrated ozone production are a function of 7 important environmental parameters in the marine boundary layer: temperature, ozone concentration, CO concentration, the solar elevation angle at the time of initial and actual release, and photolysis rate constants for NO_2 and $O(^1D)$. To our knowledge, this is the first time that the effects of non-linear, in-plume chemistry are taken into account in a global CTM. We will show that our improved model simulates NO_x concentrations over the Pacific Ocean that agree best with observational data from the Pacific Exploratory Mission West-B (PEMWB). Simulations with the popular instant dilution approach result in NO_x concentrations that are a factor 2 too high. The improved model leads to summertime O_3 concentrations that are 2-3 ppbv lower than with the instant dilution approach over areas with intense ship traffic, but in winter the differences are small. We found that ship emissions are responsible for up to 90% of surface NO_x concentrations over the North Atlantic. We will show first comparisons of satellite measured NO_2 columns with NO_2 columns modelled with the new approach, a first step towards providing constraints on shipping NO_x emissions from space.