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

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A. Introduction

Current chemistry transport models (CTMs) generally apply instantaneous mixing of shipping emissions over the model grid cells. By instantly diluting the emissions, effects of non-linear, in-plume chemistry are neglected. This leads to overestimation of NO_x concentrations and ozone production over the oceans. In this study, we adapted a Gaussian plume dispersion model with chemistry (PARANOX, Meijer et al. [1997]), to explicitly simulate NO_x decay and net ozone production during the early stages of plume dispersion and implemented it using a look-up table (LUT) in a global CTM (GEOS-Chem).

B. Goal

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, which can be seen for the Indian Ocean in Figure 1. By comparing observed with accurately simulated NO_2 columns we can provide top-down constraints on NO_x shipping emissions inventories. By taking non-linear, in-plume chemistry into account, we can also improve ozone simulations over the oceans.

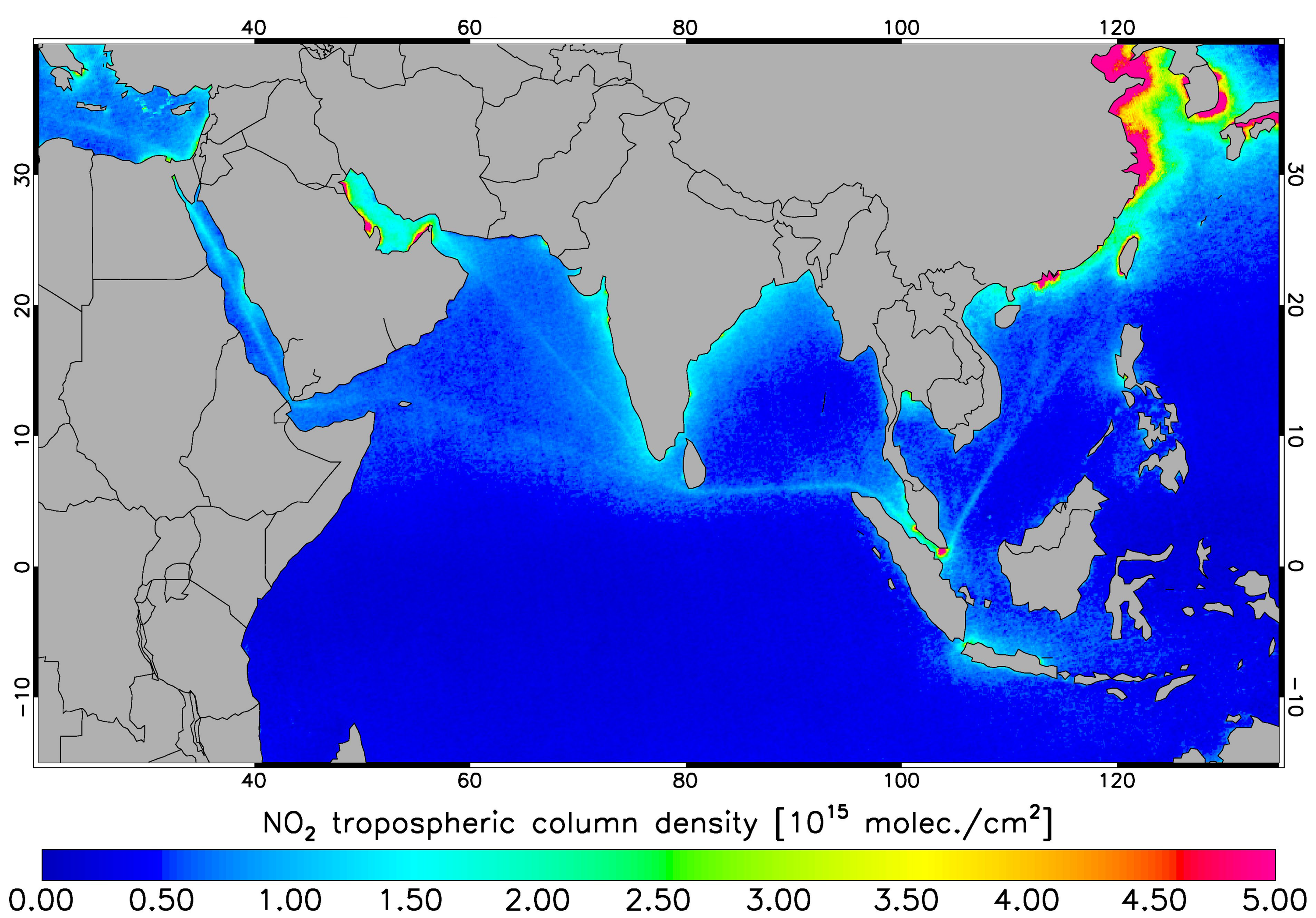

Figure 1: OMI tropospheric NO_2 columns averaged over March - April - May 2005-2006 on 0.1° by 0.1° grid cells, clearly showing 6 ship tracks. Land masses have been greyed-out. Only observations with an estimated cloud radiance fraction less than 0.5 have been used.

C. Approach

In order to account for the effects of in-plume chemistry in the global GEOS-Chem CTM, we ran PARANOX and constructed a look-up table (LUT). This LUT contains the fraction of NO_x remaining and the integrated net ozone production 5 hours after initial release and will be used to preprocess the emissions before release in GEOS-Chem. Figure 2 illustrates the concept of our approach.

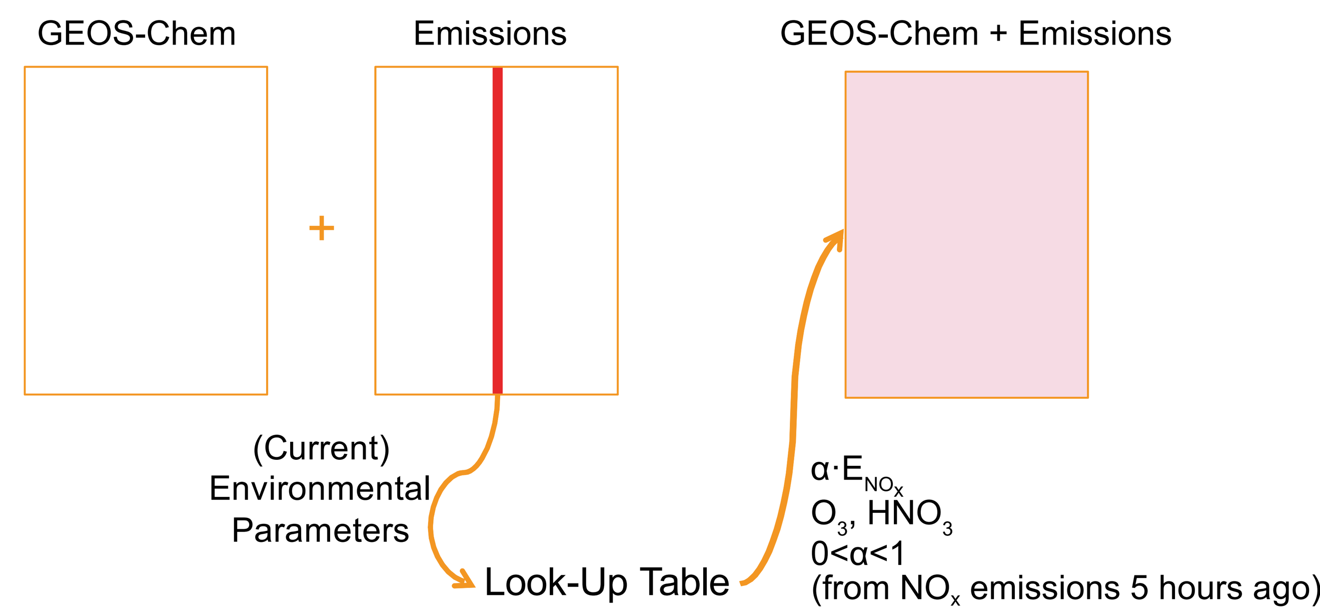

Figure 2: Concept of new approach of handling shipping NO_x emissions in GEOS-Chem.

After performing a sensitivity study, we found that the fraction of NO_x remaining and the integrated net ozone production are a function of 7 important environmental parameters in the marine boundary layer: temperature, O_3 concentration, NO_x concentration, the solar elevation angle at the time of initial and actual release, and photolysis rate constants for NO_2 and $O(^1D)$.

D. Results

Figure 3 shows that GEOS-Chem simulations with our new approach (SHIP) result in NO_x concentrations that agree best with observations from the PEM-West B campaign.

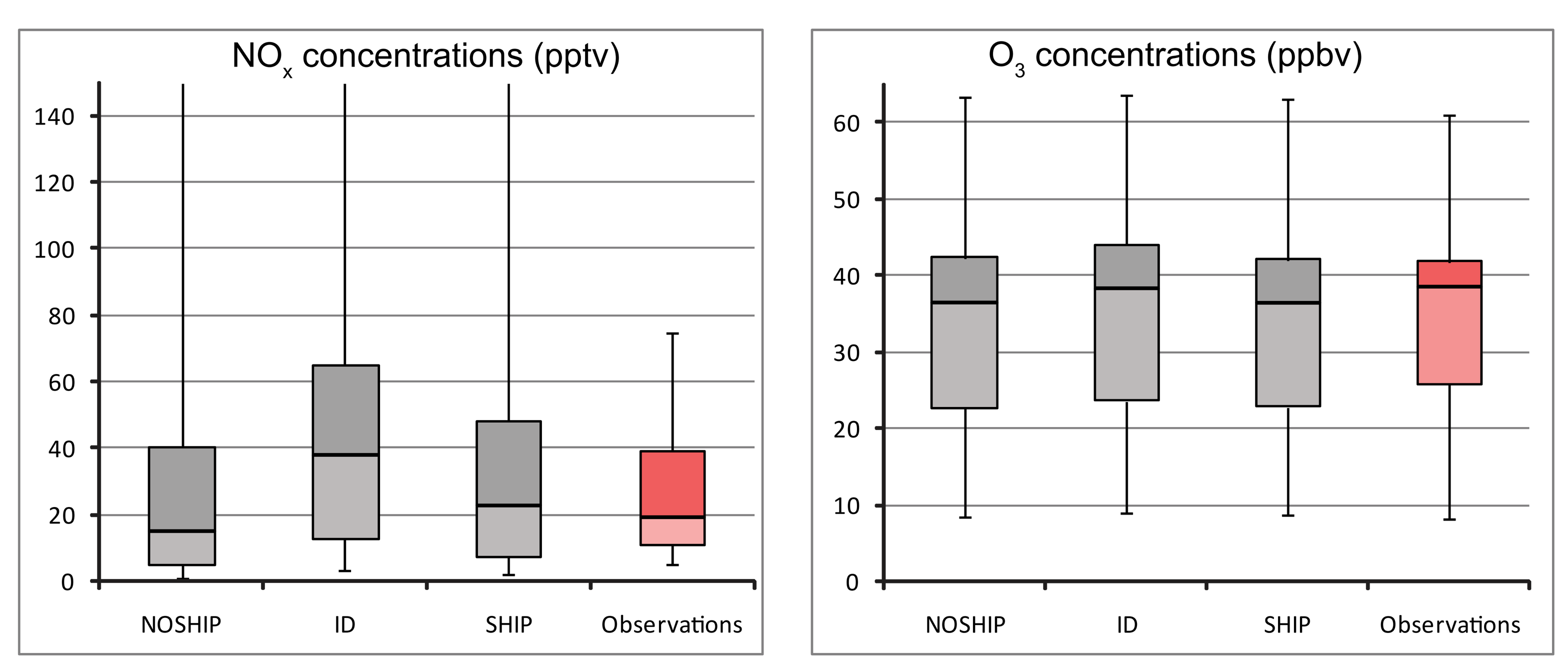

Figure 3: Comparison of O_3 and NO_x simulations and observations for the PEM-West B campaign (7 Februari till 14 March 1994). All simulations are for the year 2005, NOSHIP represents a simulation with no ship emissions, ID represeents instantly diluting ship NO_x emissions and SHIP represents simulations with our preprocessed emissions.

D. Results - continued

Figure 4 shows that in July, the simulation with our new approach (SHIP) results in lower NO_x concentrations over the ocean (about 60% less), compared to instantly diluting ship emissions (ID). In the relative difference plot, all frequently traveled ship tracks can be seen.

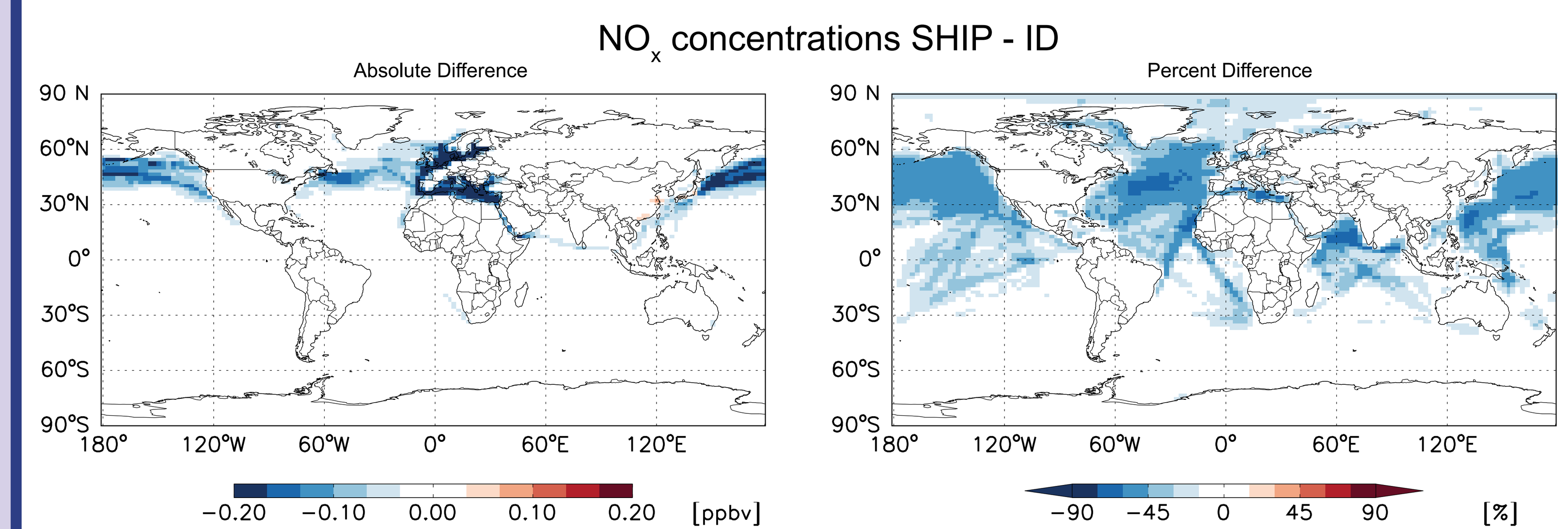

Figure 4: Absolute and relative difference in monthly mean global NO_x concentrations for the lowest model layer (0-0.3 km), simulated with GEOS-Chem for July 2005 for the instant diluting case (ID) and with our new approach (SHIP).

Figure 5 shows that, as a result of the lower NO_x concentrations in the ship tracks, the simulation with our SHIP approach results in lower O_3 concentrations over the ocean (up to 5 ppbv) in July, compared to instantly diluting ship emissions (ID). Differences over heavily polluted areas (e.g. North Sea or coastal China) are small.

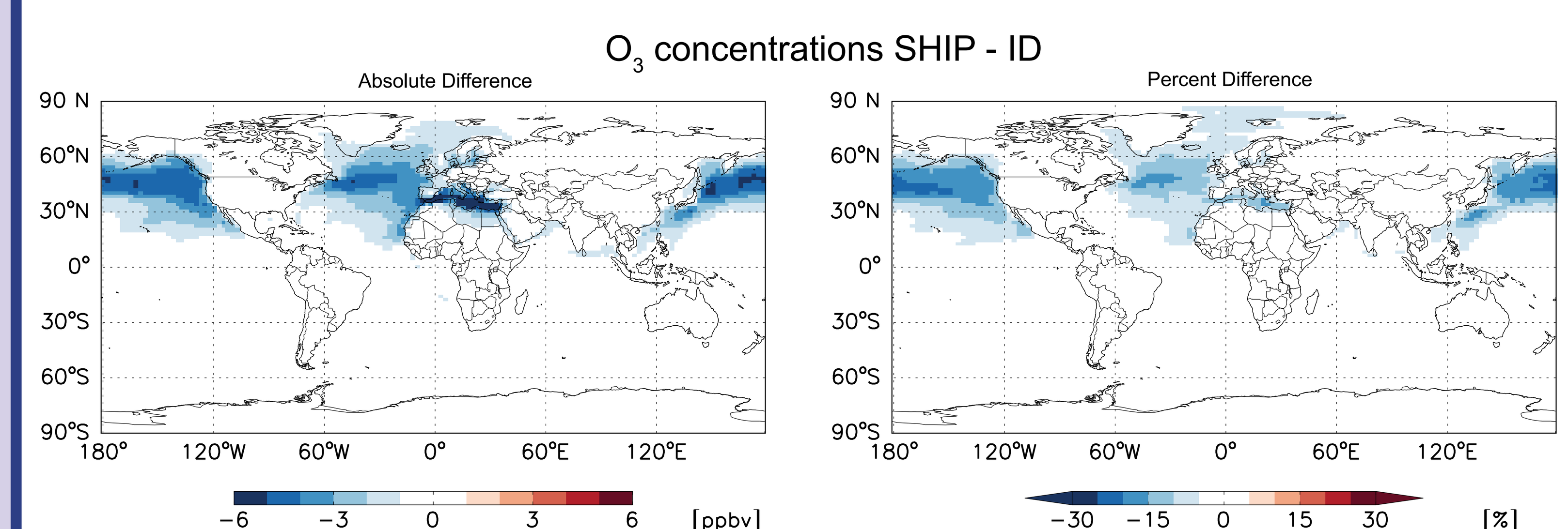

Figure 5: Absolute and relative difference in monthly mean global O_3 concentrations for the lowest model layer (0-0.3 km), simulated with GEOS-Chem for July 2005 for the instant diluting case (ID) and with our new approach (SHIP).

E. References and Acknowledgements

[Meijer et al., 1997] The effects of the conversion of nitrogen oxides in aircraft exhaust plumes in global models. Geophysical Research Letters, 24(23):3013-3016.

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