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Supplement of

The climate impact of ship NO_x emissions: an improved estimate accounting for plume chemistry

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Supplementary Information for "The climate impact of ship NO_x emissions: an improved estimate accounting for plume chemistry"

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Gaussian plume model

Vinken et al. (2011) provide a full description of the Gaussian plume model that underpins the look-up tables used in the global CTM. We provide only a short summary here. The plume model contains 10 concentric rings representing a cross section of the plume perpendicular to the wind direction. Plume expansion, entrainment of environmental air, and mixing between rings are controlled by wind speed using standard dispersion relations for marine conditions (Hanna et al., 1985; Song et al., 2003). The chemical solver contains 98 reactions between 43 O_3-NO_x-HO_x-hydrocarbon species, including photolysis and heterogeneous uptake of N_2O_5 on aerosols. The final composition of the exhaust plume after 5 hours of aging is tabulated as a function of 8 environmental variables and this look-up table is used in the global CTM to calculate effective emissions from ships. Figure S1 compares the Gaussian plume model to field observations of an aging ship plume. Figure S2 shows how the 8 environmental variables influence the plume chemistry.

Radiative forcing from long-lived O_3 and CH_4

Here, we derive the *a* term introduced in Eq. (4). Holmes et al. (2011) analyzed factors controlling long-lived RF from tropospheric O_3 and CH_4. Combining Eqs. (3) and (4) from that earlier work, the long-lived RF from these species is related by

$$F_{\text{long-O3}} = F_{\text{CH4}}\,(dF/d[CH_4])^{-1}\,(d[O_3]/d[CH_4])\,(dF/d[O_3]), \qquad \text{(S1)}$$

where $d[O_3]/d[CH_4] = 3.5 \pm 1.0$ DU (O_3) ppm(CH_4)$^{-1}$ is the change in tropospheric O_3 responding to a change in global-mean CH_4; $dF/d[O_3] = 36 \pm 8$ mW m^{-2} DU(O_3)$^{-1}$ is the radiative forcing due to changes in tropospheric O_3; and other terms are defined in Sect. 2.2 of the main text. Values of each factor are based on a literature survey of multiple CTMs and radiative transfer models (Holmes et al., 2011). If we define

$$a = (dF/d[CH_4])^{-1}\,(d[O_3]/d[CH_4])\,(dF/d[O_3])$$

and propagate the best estimates and uncertainties in all factors, then $a = 0.34 \pm 0.13$.

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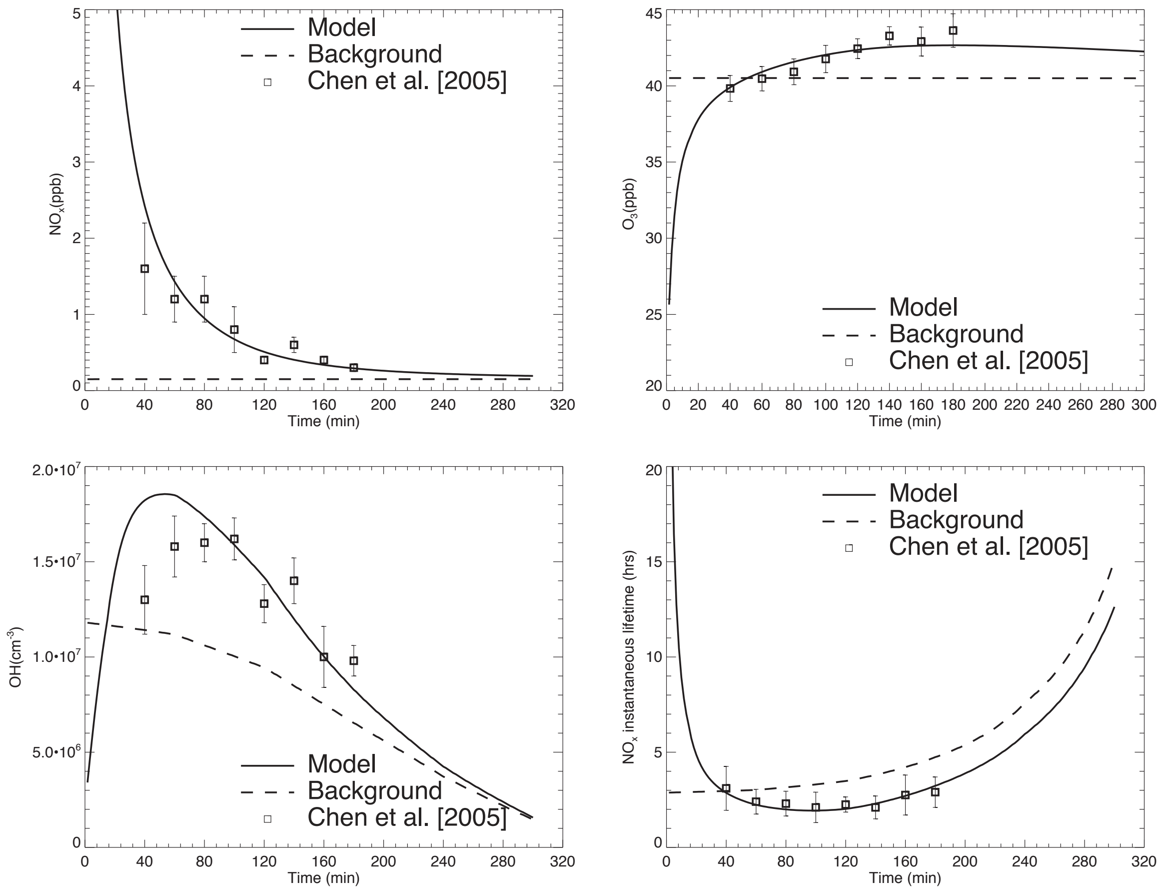

Figure S1. Observed and simulated NO_x (ppb), O_3 (ppb), OH (molec cm^{-3}), and the NO_x instantaneous lifetime (h) within a ship plume released at 12:00 h local time. Model and data are averaged across the cross section of the plume. Observations were made on 8 May 2002 during the ITCT 2002 aircraft campaign (Chen et al., 2005). Vertical bars indicate 1-σ range. The model is configured as described by Vinken et al. (2011) to match observed environmental conditions.

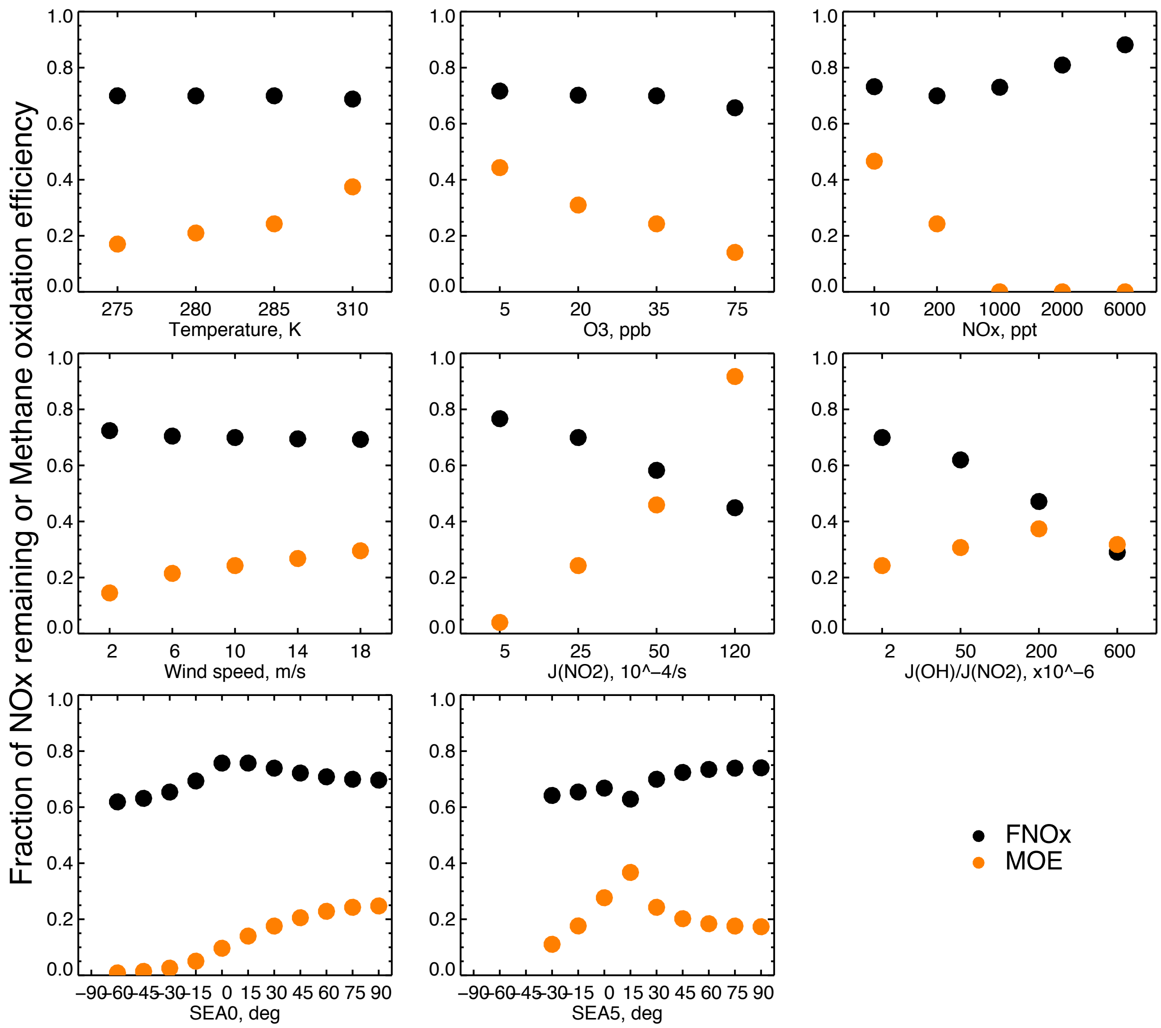

Figure S2a. Effect of environmental variables on ship plume composition and chemistry after 5 h of plume aging. FNOx is the fraction of emitted NOx that remains as NOx after 5 h. MOE is the methane oxidation efficiency (mol/mol). Each panel shows the effect of changing one variable while others are held constant at conditions similar to the ITCT 2002 field experiment (Chen et al., 2005): temperature is 285 K; background O_3 is 35 ppb; background NOx is 200 ppt; wind speed is 8 m s^{-1}; $J(NO_2)$ is 2.5×10^{-3} s^{-1}; $J(OH)/J(NO_2)$ is 2×10^{-6}; solar elevation angle at emission time (SEA0) is 75°; and solar elevation angle after 5 hours (SEA5) is 30°. $J(NO_2)$ is the photolysis rate of NO_2 and J(OH) is the photolysis rate of O_3 times the yield of OH from subsequent reaction with H_2O. Note that some abscissae are not linearly distributed.

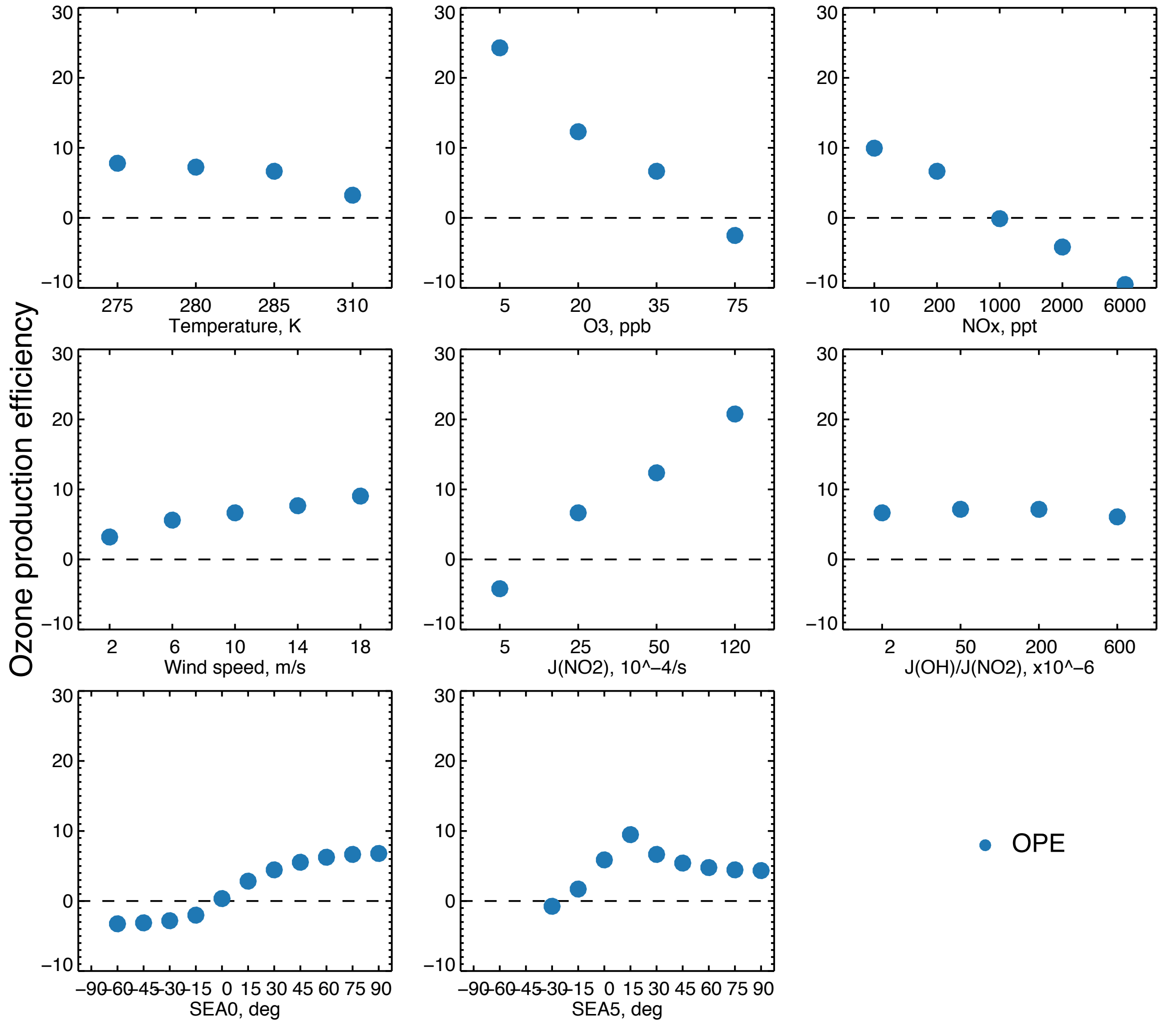

Figure S2b. Same as Fig. S2a, except showing ozone production efficiency (OPE, mol/mol).