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Supplement of

N_2O_5 uptake onto saline mineral dust: a potential missing source of tropospheric $ClNO_2$ in inland China

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Text S1. Parameterization for N_2O_5 uptake on aqueous aerosols in GEOS-Chem

$ClNO_2$ can form from heterogeneous reaction of N_2O_5 with Cl^- on aqueous aerosols:

$$\mathrm{N_2O_5} + \varphi\mathrm{Cl^-} + (1-\varphi)\mathrm{H_2O} \rightarrow \varphi\mathrm{ClNO_2} + (2-\varphi)\mathrm{NO_3^-} + 2(1-\varphi)\mathrm{H^+} \quad \text{(R1)}$$

The rate reaction (R1) is determined by a reactive uptake coefficient, $\gamma(N_2O_5)$, representing the probability that a gas-phase N_2O_5 molecule impacting the aerosol surface and to react in the bulk. For aqueous aerosols, the model assumes that reaction (R1) happens on internally mixed sulfate, ammonium, nitrate, sea salt and organic aerosols, and account for the effect of organic coating as:

$$\frac{1}{\gamma_{\mathrm{N_2O_5}}} = \frac{1}{\gamma_{\mathrm{core}}} + \frac{1}{\gamma_{coat}} \qquad (1)$$

where γ_{core} represents the reactive uptake mechanism of Bertram and Thornton (2009), and γ_{coat} represents the retardation from organic coating. Calculation of γ_{coat} is based on Riemer et al. (2009) with the relative humidity (RH) dependence of coating properties from Gaston et al. (2014). This parameterization has been described in detail by McDuffie et al. (2018a):

$$\gamma_{\mathrm{N_2O_5}} = \frac{4V}{cS_a} H_{aq} k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3[\mathrm{H_2O}]}{k_{2b}[\mathrm{NO_3^-}]}\right) + 1 + \left(\frac{k_4[\mathrm{Cl^-}]}{k_{2b}[\mathrm{NO_3^-}]}\right)}\right) \qquad (2)$$

$$k'_{2f} = \beta\left(1 - e^{-\delta[\mathrm{H_2O}]}\right) \qquad (3)$$

$$\gamma_{\mathrm{coat}} = \frac{4RTH_{org}D_{org}R_c}{clR_p} \qquad (4)$$

where R is the ideal gas constant, c is the gas-phase thermal velocity of N_2O_5, V and S_a are particle volume and surface area density, H_{aq} is Henry's law constant for N_2O_5 in water; β is equal to 1.15×10^6 s^{-1}, δ is equal to 0.13 M^{-1}, k_3/k_{2b} is equal to 0.06, k_4/k_{2b} is equal to 29; $H_{org}D_{org} = H_{aq}D_{aq}$ where D_{aq} is N_2O_5 liquid diffusion coefficient, and ε is a scaling coefficient which increases linearly with the increase of RH; R_c and R_p are radius of inorganic core and the whole particle with organic coating, and l is the thickness of organic coating which is calculated using the volume ratio of organic and inorganic aerosols:

$$l = R_p(1 - \alpha^{\frac{1}{3}}) \qquad (5)$$

$$\alpha = \frac{1}{1+\frac{V_{organic}}{V_{inorganic}}} \qquad (6)$$

The production yield of $ClNO_2$ in reaction (R1), φ, is calculated using the mechanism of Bertram and Thornton (2009) with a scaling factor of 0.25 following the suggestion by McDuffie et al. (2018b):

$$\varphi = 0.25\left(\frac{k_2[\mathrm{H_2O}]}{k_3[\mathrm{Cl^-}]} + 1\right)^{-1} \qquad (7)$$

where k_3/k_2 is equal to 450, as given by Roberts et al. (2009).

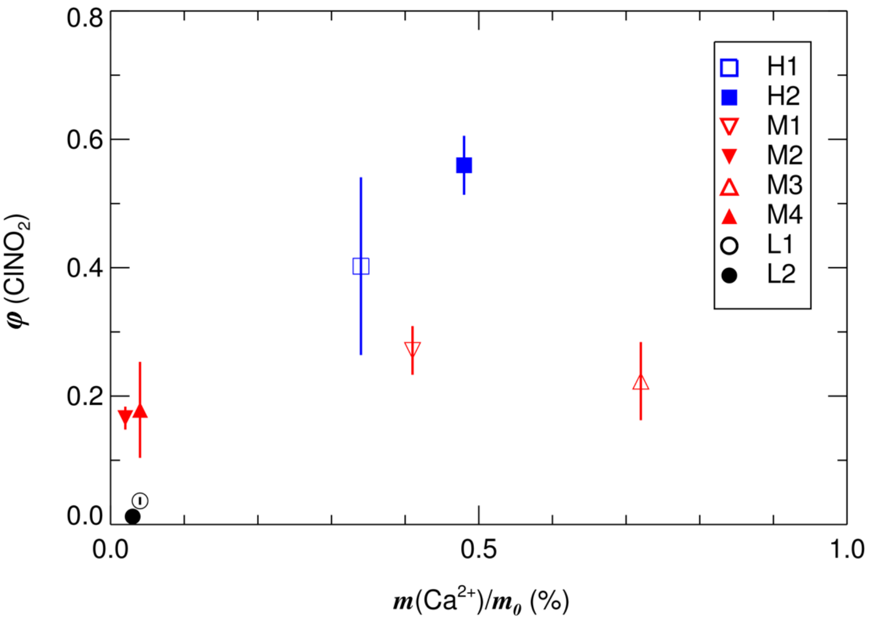

Figure S1. Measured $ClNO_2$ yields at 18% RH versus mass fractions of soluble Ca^{2+} for the eight saline mineral dust samples.

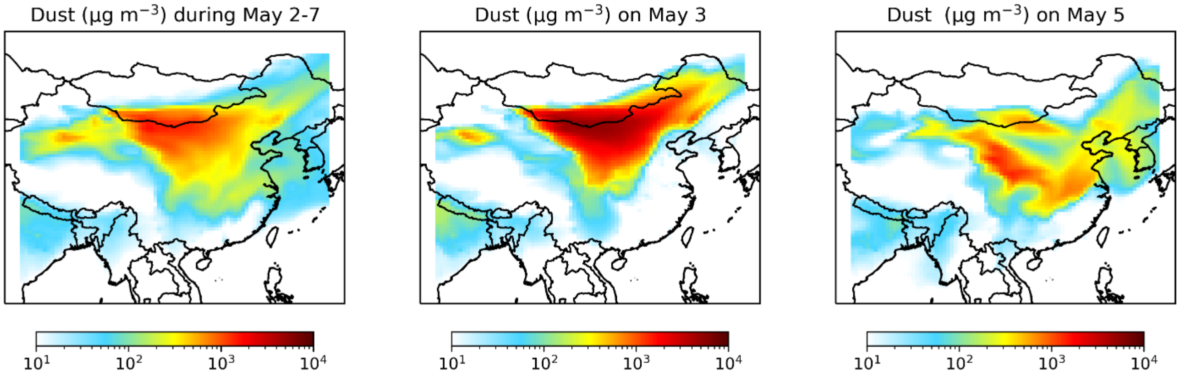

Figure S2. Modeled mean dust concentrations in the surface air over China during 2-7 May (left), on 3 May (middle) and on 5 May (right).

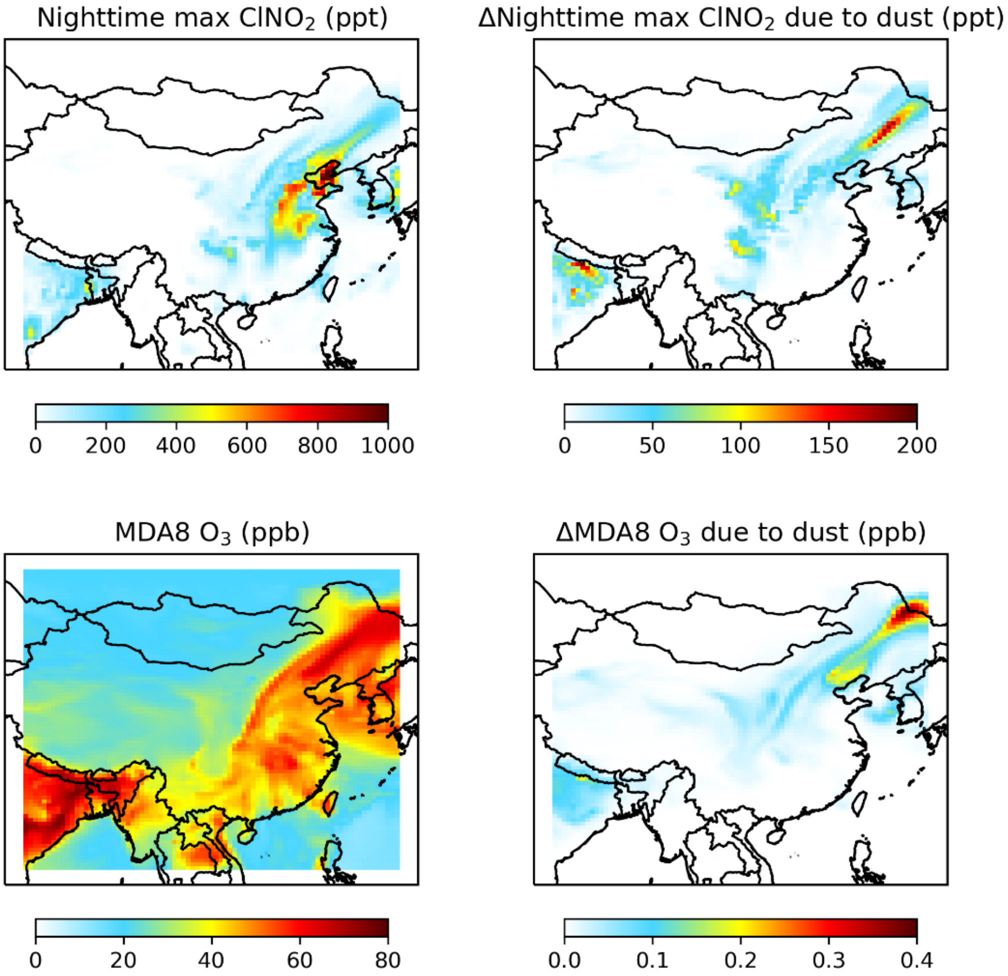

Figure S3. Modeled weekly mean mixing ratios of nighttime maximum $ClNO_2$ (upper panels) and maximum daily 8-h average (MDA8) ozone (bottom panels) in surface air over China on 3 May 2017. The left panels show simulated mixing ratios in our standard case in which $\varphi(ClNO_2)$ is assumed to be 0 for N_2O_5 uptake onto dust aerosol. The right panels show impacts of $ClNO_2$ formation due to N_2O_5 uptake onto dust, calculated as the difference between the standard case and the case in which $\varphi(ClNO_2)$ is assumed to be 0.1 for N_2O_5 uptake onto dust.

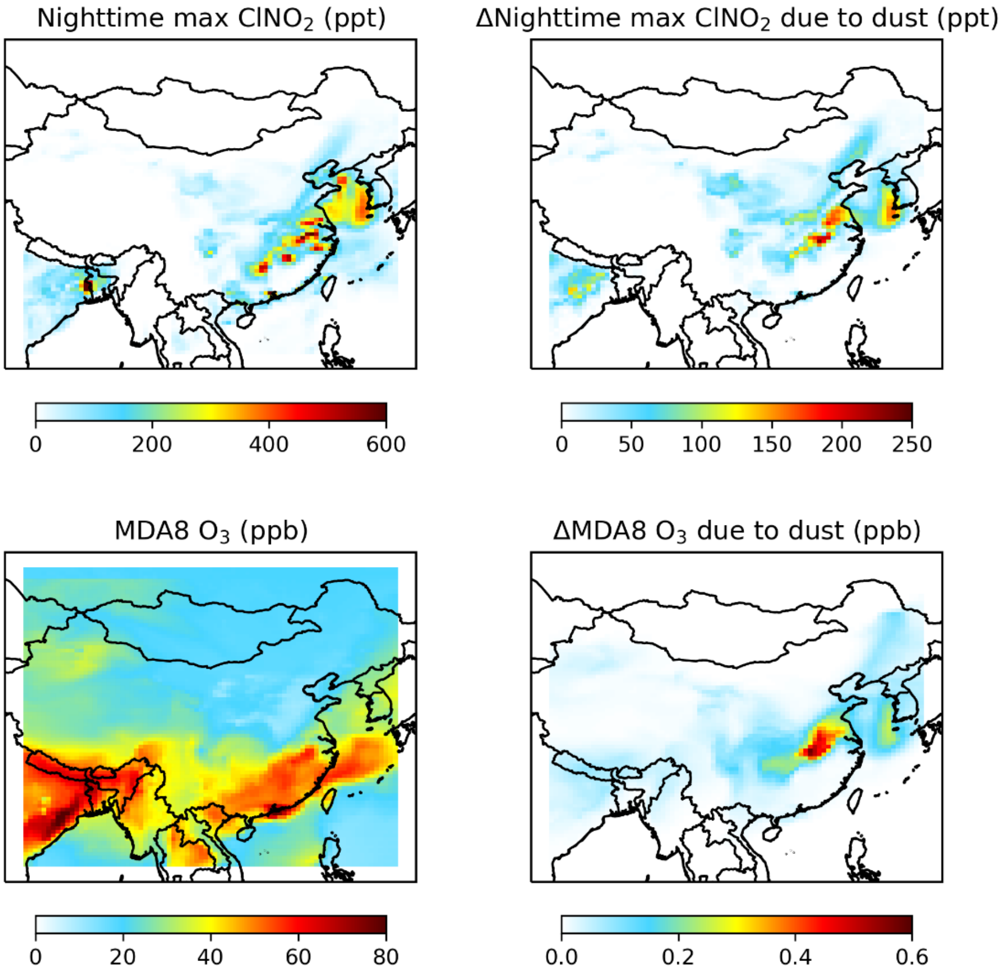

Figure S4. Same as Figure S3 but for 5 May 2017.

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