



Impact of northern hemisphere mid-latitude anthropogenic sulfur dioxide emissions on local and remote tropospheric oxidants

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Abstract. The unintended consequences of reductions in regional anthropogenic sulfur dioxide (SO₂) emissions implemented to protect human health are poorly understood. SO₂ decreases began in the 1970s in the US and Europe and are expected to continue into the future, while recent emissions decreases in China are also projected to continue. In addition to the well documented climate effects (warming) from reducing aerosols, tropospheric oxidation is impacted via aerosol modification of photolysis rates and radical sinks. Impacts on the hydroxyl radical and other trace constituents directly affect climate and air quality metrics such as surface ozone levels. We use the Geophysical Fluid Dynamics Laboratory Atmospheric Model version 3 nudged towards National Centers for Environmental Prediction (NCEP) reanalysis wind velocities to estimate the impact of SO₂ emissions from the United States, Europe, and China by differencing a control simulation with an otherwise identical simulation in which 2015 anthropogenic SO₂ emissions are set to zero over one of the regions. Springtime sulfate aerosol changes occur both locally to the emission region and also throughout the Northern Hemispheric troposphere, including remote oceanic regions and the Arctic. The presence of sulfate aerosol strongly reduces hydroxyl (OH) and hydroperoxy (HO₂) radicals by up to 10% year-round throughout most of the troposphere north of 30 °N latitude, the latter of which is directly removed via heterogeneous chemistry on aerosol surfaces, including sulfate, in the model. Regional SO₂ emissions significantly increase nitrogen oxides (NO_x) by about 5-8% throughout most of the free troposphere in the Northern hemisphere by increasing the NO_x lifetime as the heterogeneous sink on sulfate aerosol declines. Despite the NO_x increases, tropospheric ozone decreases at northern mid-latitudes by 1-4% zonally averaged and by up to 5 ppbv in surface air over China, as its response is dominated by the larger decreases (up to 2x) in HO₂ and OH. Since 2015 anthropogenic SO₂ emissions in China exceed those in the US or Europe, the oxidative response is greatest for the China perturbation simulation. Chemical effects of aerosols on oxidation (reactive uptake) dominate over radiative effects (photolysis rates), the latter of which are only statistically significant locally for the large perturbation over China. We find that the emissions decrease in China, which has yet to be fully realized, will have the largest impact on oxidants and related species in the Northern Hemisphere free troposphere compared to changes in Europe or the USA. Our results bolster previous calls for a multipollutant strategy for air pollution mitigation, to avoid the unintended consequence of aerosol removal leading to surface ozone increases that offset or



mask surface ozone gains achieved by regulation of other pollutants, especially in countries where current usage of high-sulfur emitting fuels may be phased out in the future.

35 1 Introduction

Understanding and constraining tropospheric oxidants such as the hydroxyl radical (OH) remains a key challenge of direct relevance to understanding the oxidizing power of the atmosphere, radiative forcing, and surface air quality. Despite the critical role in atmospheric chemistry, OH abundances differ widely among chemistry-climate and chemical transport models (Stevenson et al., 2020; Zhao et al., 2019). In addition, global, annual mean OH response to historical anthropogenic emission
40 changes between the preindustrial and the present-day ranged from a 12.7% decrease to a 14.6% increase across 17 global models (Naik et al., 2013), with similar discrepancies across simulations of future composition and climate (Voulgarakis et al., 2013). These differences between model estimates of OH suggest major knowledge gaps in our understanding of the drivers of OH. One potential driver of tropospheric oxidant changes that has not received sufficient study is aerosols, which can uptake radical species (chemical effect) and scatter or absorb incoming solar radiation (radiative effect), thereby impacting OH and
45 other important chemical species (Jacob, 2000; Wild et al., 2000).

Anthropogenic emissions of sulfur dioxide (SO₂), a precursor to sulfate aerosol, have significantly decreased in the United States and Europe for the last several decades and are projected to continue to decline (Riahi et al., 2011; Vuuren et al., 2011; Westervelt et al., 2015). In China, emissions of anthropogenic aerosols began to decline in about 2013 after increasing for decades (Fontes et al., 2017; Li et al., 2017; Samset et al., 2018). Previous research has indicated that these past and
50 forthcoming emission changes have the potential to influence the tropospheric oxidation capacity on both a regional and global basis (Dentener and Crutzen, 1993; Dickerson et al., 1997; Martin et al., 2003). The aerosol decreases in China were associated with subsequent increases in summertime surface ozone (O₃) in China, attributed to a reduction in the sink of radical species such as the hydroperoxyl radical (HO₂) that promote O₃ production (Li et al., 2019b). Using a model and observations, the authors found that a 40% decrease in fine particulate matter (PM_{2.5}) in China between 2013 and 2017 led to an increasing
55 ozone trend of up to 3 ppb per year in eastern China and was found to be a more important factor than NO_x emissions reductions over the same time period (Li et al., 2019b, 2019a). These findings confirm earlier modelling work and point to an important role for aerosol impacts on tropospheric oxidation with implications for surface O₃ concentrations, especially over China (Li et al., 2018; Lou et al., 2014).

On a global scale, few studies have addressed the impact of aerosols on tropospheric oxidants. Often, aerosol impacts
60 are assumed to be negligible in constraining present and future OH concentrations (Voulgarakis et al., 2013). Primary production of OH depends on the amount of water vapor and O(¹D) present (formed via O₃ photolysis), and is the dominant pathway of OH formation in most locations except for high latitudes (Spivakovsky et al., 2000). Secondary production includes reactions of HO₂ or RO₂ (organic peroxy) radicals generated from oxidation of volatile organic compounds (VOCs) or carbon



monoxide (CO) with nitrogen oxide (NO) which regenerates OH. Concentrations of these atmospheric constituents and certain meteorological factors such as absolute humidity, temperature, and ultraviolet radiation are thought to predominantly control OH abundance (Spivakovsky et al., 2000). However, by differencing a Goddard Earth Observing System Chemistry Transport Model (GEOS-Chem) control simulation of late 1990s atmospheric composition with a sensitivity simulation in which the offline global aerosols are excluded, Martin et al. (2003) find that the presence of all aerosols decreases OH by 9% globally and 5-35% in the northern hemisphere boundary layer. The authors also find 15-45 ppbv decreases in boundary layer O₃ over India in March associated with the presence of all aerosols compared to all aerosols removed. In a similar global study, (Tie et al., 2005) use the Model for Ozone and Related Chemical Tracers version 2 (MOZART-2) to show that the net effect of all aerosols (natural and anthropogenic) reduces HO_x (defined as OH + HO₂) and O₃ by 30% and 20%, respectively, improving on past methodology (e.g., Martin et al., 2003) by calculating aerosol abundances interactively. Past studies only considered global distributions of aerosols and often focused on natural aerosols such as dust or sea salt (Bian and Zender, 2003; Liao et al., 2003). The impact of rapidly changing spatially heterogeneous anthropogenic aerosol abundances on tropospheric OH and O₃ is thus an open question.

We expand on past studies by considering individual regions (China, Europe, and the US) of sulfate aerosol decrease via SO₂ emissions reductions, and quantify the local and remote impacts of changing aerosol emissions on atmospheric HO_x, NO_x, and O₃ concentrations on a seasonal basis within a chemistry-climate model nudged to observed meteorology. We focus on anthropogenic SO₂ emissions which have decreased most dramatically in many regions compared to anthropogenically-sourced carbonaceous aerosols or natural aerosols such as dust and sea salt. We seek mechanistic understanding on the interactions between aerosols, oxidants and radical species, and photolysis rates over different regions and in different seasons. We consider two main pathways through which aerosols can affect oxidation: modification of photolysis rates via extinction of incoming solar radiation (radiative effect) and heterogeneous uptake of radical species onto aerosol surfaces (chemical effect). Finally, we consider the impact of anthropogenic SO₂ emissions reductions on boreal summertime surface O₃ concentrations in China, Europe, and the US.

2 Model and simulations

We use the National Oceanic and Atmospheric Administration Geophysical Fluid Dynamics Laboratory Atmospheric Model version 3 (GFDL-AM3), which is the atmosphere-only component of the GFDL coupled climate model, CM3 (Donner et al., 2011). The model has been rigorously evaluated against observations in previous work (Donner et al., 2011; Naik et al., 2013). The model has 48 vertical layers from the surface up to about 0.01 hPa and a six-face cubed-sphere grid with 48 cells along each edge (C48), which is regridded to a 2 degree latitude by 2.5 degree longitude Cartesian grid. Emissions of anthropogenic trace gases and aerosols for year 2015 emissions are from the Representative Concentration Pathway 8.5 (RCP8.5) scenario (Riahi et al., 2011). The tropospheric chemical mechanism for aerosols and gas-phase species follows the work of Horowitz et al. (2003) and Horowitz et al. (2007) with updates to photolysis, radical uptake by aerosols, and convective



wet scavenging of aerosols. The Fast-JX module (Bian et al., 2003; Wild et al., 2000) calculates the impact of online aerosols and clouds on photolysis rates and actinic fluxes, implemented into GFDL-AM3 according to Mao et al. (2013b). Heterogeneous uptake of radical species is simulated according to Mao et al. (2013b) and Mao et al. (2013a) using a first order reactive uptake rate constant k (Equation 1):

$$k = - \left(\frac{r_e}{D_g} + \frac{4}{\gamma v} \right)^{-1} A \quad (\text{Equation 1})$$

where r_e is the aerosol effective radius (m), D_g is the gas-phase molecular diffusion coefficient, v is the mean molecular speed of the gas, and A is the aerosol surface area per unit volume of air. Here we set the heterogeneous reactive uptake coefficient (γ) of HO₂ to 0.2 instead of the value of 1.0 in Mao et al. (2013a). Though estimates of γ are uncertain, recent literature suggests such high values of 1.0 are not supported by observations and that the parameter is likely closer to 0.2 (Abbatt et al., 2012; Li et al., 2019a, 2019b; Taketani et al., 2012). Taketani et al. (2012) recommends a middle γ value of 0.24 based on measurements at two high altitude sites in China. Reactive uptake coefficients for all other reactions including N₂O₅, NO₃, and NO₂ are shown in Table 1, taken from Jacob (2000). We allow uptake of HO₂, N₂O₅, NO₃, and NO₂ onto all aerosol types, including sulfate, black carbon, organic carbon, sea salt, and dust using the same coefficients for each composition. We also include updates to convective wet scavenging of aerosols in the form of finer vertical discretization of convective updraft plumes, resulting in improvements in aerosol budgets (Paulot et al., 2015). Horizontal wind velocities are nudged using a pressure-dependent technique towards reanalysis values from the National Centers for Environmental Prediction Global Forecast System (NCEP GFS; (Lin et al., 2012)). Further model description and model evaluation against observations can be found in Donner et al. (2011), Naik et al. (2013a), and (Rasmussen et al., 2012).

We conduct a two-year (2014-2015) nudged control simulation in which emissions of aerosols and their precursors follow RCP8.5 and contrast it with three perturbations: one in which all anthropogenic SO₂ emissions are set to zero over the United States (30°N - 50°N, 70°W - 125°W), all anthropogenic SO₂ emissions are set to zero over Europe (35°N - 70°N, 15°W - 55°E), and all anthropogenic SO₂ emissions are set to zero over China (15°N - 50°N, 95°E - 130°E). SO₂ is oxidized by the hydroxyl radical in the gas phase and by ozone and hydrogen peroxide in clouds to form sulfate aerosol, which is a dominant component of total aerosol in GFDL-AM3 (Westervelt et al., 2015, 2017). We separately subtract each regional SO₂ perturbation simulation from the control simulation, thereby isolating the impact of SO₂ emissions (and subsequent sulfate formation) on tropospheric oxidants and related species. We test for statistical significance using a Student's t-test on seasonal mean responses with the null hypothesis being that the difference between the control and the perturbation simulation is zero. Only the full year of 2015 is used for analysis. SO₂ perturbations from our simulations are 10.8, 12.4, and 16.2 Tg SO₂ y⁻¹ for US, Europe, and China, respectively.

The global annual mean OH for the 2015 control simulation is 7.0×10^5 molecules cm⁻³, which is within the range of the 14 Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) for year 2000 and 14 Chemistry Climate Model Initiative (CCMI) models (Voulgarakis et al., 2013b; Zhao et al., 2019) for years 2000-2010. The global annual



tropospheric burden of O₃ in the 2015 control simulation is 356 Tg, which compares well to the year 2000 O₃ burden mean across the ACCMIP models of 337 ± 23 Tg (Young et al., 2013).

130 3 Results

Figure 1 shows the percent increase in seasonal (March-April-May, MAM) sulfate concentrations at the surface (right column) and at altitude (left column) for the presence of all anthropogenic US SO₂ (first row), all European SO₂ (second row), and all Chinese SO₂ (third row) based on year 2015 anthropogenic emissions. Additional seasons are shown in the supplemental (Figs. S1-3). The zeroing of 2015 emissions in each location results in the largest perturbation in China, where emissions are highest, followed by Europe and the US. Sulfate increases are largest closest to the source region, but all three regional simulations show statistically significant remote impacts both horizontally and vertically in the atmosphere, as evidenced by the spatial and zonal plots in Fig. 1. The US, Europe, and China perturbations all significantly increase sulfate throughout the troposphere up to 200 hPa and higher towards the North Pole, with the largest increases of up to 30-40% resulting from a zeroing of China SO₂. Transport to the Arctic is a common feature in all three perturbations, and is consistent with previous studies on aerosol transport to the Arctic (Shindell et al., 2008; Stohl, 2006). The US perturbation impacts sulfate concentrations significantly at the surface and at altitude over the North Atlantic Ocean, while the China perturbation exerts a heavy influence over the Pacific reaching all of the way to the Western US. European SO₂ emissions have widespread influence on the Northern Hemisphere, but especially in the Arctic and the Mediterranean and northern Africa. In all cases, sulfate changes are nearly entirely confined to the Northern Hemisphere.

We analyze the impact of sulfate changes on oxidation, starting in Fig. 2 with OH (left column) and HO₂ (right column) for each of the three regional perturbations (rows of Fig. 2). Sulfate aerosol surfaces directly uptake HO₂ radicals as described in Sect. 2, resulting in significant decreases of HO₂ and OH (via their rapid cycling). For each perturbation, decreases in both OH and HO₂ occur throughout most of the Northern Hemisphere up to about 200 hPa vertically during the boreal spring (MAM). The largest decreases in OH and HO₂ occur in spring for each of the perturbations, followed by winter (December-January-February, DJF), autumn (September-October-November, SON), and summer (June-July-August, JJA). These additional seasons are plotted in Figs. S4-S6. In MAM, SO₂ emissions over the US decrease OH and HO₂ by about 5% within the US planetary boundary layer. In the mid-troposphere (400 – 600 hPa), OH decreases are 5% or greater and are located spatially above the Arctic. For the Europe SO₂ and China SO₂ cases during MAM, the Arctic middle troposphere OH decreases are larger in percent change (>10%) than the local changes near the surface (~8%). The presence of 2015 China SO₂ emissions also decreases OH and HO₂ by about 10% over the north Pacific Ocean middle troposphere (about 400-600 hPa). We conclude that regional SO₂ emissions may have stronger impacts remotely than locally, and OH may be relatively more sensitive to aerosol changes in the Arctic and remote oceans at higher altitudes where its production is more limited.

In Fig. 3 we plot spring (MAM) changes in NO_x (defined as NO + NO₂) concentrations in response to anthropogenic SO₂ emissions in the US, Europe, and China. While HO₂ and OH strongly decreased in response to SO₂ emissions, NO_x



160 significantly increases throughout most of the Northern Hemisphere. In the model, aerosols can take up NO_2 directly but with
a very low reaction probability (0.0001, Table 1), such that little uptake actually occurs and is easily offset by feedbacks onto
other chemical reactions involving NO_x . Instead, reduction in the sinks of NO_x via OH (nitric acid formation) during the day
and uptake of NO_x reservoir species at night dominates the response to SO_2 emission changes, increasing NO_x in the model as
OH decreases. At night, NO_x is removed by reaction with the nitrate radical (NO_3), which forms dinitrogen pentoxide (N_2O_5)
165 (Chang et al., 2011; Jacob, 2000). Sulfate aerosols are effective at removing both NO_3 and N_2O_5 via reactive uptake (reaction
probability of 0.1), slowing down these night time NO_x sinks and thus increasing NO_x abundance. This hindering of NO_x sinks
is most effective during MAM and DJF in the Northern Hemisphere mid-troposphere (Fig. 3a-c for MAM, additional seasons
shown in Figs. S7-S9). Mid-tropospheric northern hemisphere NO_x increases reach about 7-8% in response to Chinese SO_2
emissions specifically, with smaller effects for both US and Europe SO_2 perturbations. NO_x at the surface increases slightly
170 less at about 5-7% depending on the regional emissions perturbation, though these changes still are statistically significant.
Several previous studies have used smaller reactive uptake coefficients for N_2O_5 than the value of 0.1 used here (Evans and
Jacob, 2005; Holmes et al., 2019; Macintyre and Evans, 2010; McDuffie et al., 2019) based on more recent laboratory
experiments, but only find impacts on mean tropospheric O_3 burden of 2-4%. Using a box modelling approach, McDuffie et
al. (2019) find a median γ for N_2O_5 , of 0.076, reasonably close to our assumed value of 0.1. Using a smaller γ would lead to
175 less N_2O_5 uptake by aerosols, a smaller decrease in the NO_x sinks, and therefore a smaller increase in NO_x concentration via
the N_2O_5 uptake pathway.

In most of the Northern Hemisphere troposphere, O_3 decreases in MAM by up to 4% in response to US, European,
or Chinese SO_2 emissions increases in the model (Fig. 4), mostly coinciding with regions of large HO_x decreases (Fig. 2),
despite the increase in NO_x (Fig. 3). The O_3 increases in the upper troposphere are mostly not significant. We examine model
180 diagnostics of gross ozone production (the sum of HO_2+NO and all RO_2+NO reaction pathways) and O_3 loss (which includes
reaction of O_3 with HO_x and with alkenes, plus O_3 photolysis followed by $\text{O}^1\text{D}+\text{H}_2\text{O}$) to interpret further the O_3 decrease.
While both O_3 production (P_{O_3}) and loss (L_{O_3}) rates decline (Fig. S10 and Fig. S11), production decreases more strongly than
loss, lowering O_3 concentrations. We confirm that transport of O_3 from other latitudes is unlikely to contribute much to the
modelled O_3 response as the change in advective or convective tendency in O_3 (Fig. S12) is far smaller than the chemical
185 production and loss terms (Fig. S10). The O_3 production and loss rates decrease most strongly in the lower troposphere over
the source regions (Fig. S10 and Fig. S11) while the O_3 decreases (Fig. 4) propagate more widely through the free troposphere,
indicating reduced export from these source regions. Additional seasons for O_3 change are shown in Figs. S13-S15.

We find here that the decline in HO_x and its impact on P_{O_3} outweighs the aerosol-induced increases in NO_x and
decreases in O_3 - HO_x sinks, even during summer in all three source regions. We show the response of summertime surface 8-
190 hour maximum daily average (MDA) O_3 to increasing anthropogenic SO_2 emissions in the US, Europe, and China in Fig. 5.
Increasing sulfate aerosol increases the sink of HO_2 radicals and thus slows down O_3 production (Fig. S11), resulting in surface
 O_3 concentration decreases. Sulfate aerosol can also reduce NO_2 and O_3 photolysis rates. The combined effect of sulfate aerosol
on changes in photolysis rates and heterogeneous chemistry is a statistically significant decrease of about 5 ppbv over most of



195 eastern China, Korea, and Japan when Chinese SO₂ emissions are introduced, a decrease of about 3 ppbv over the eastern US
for US SO₂ emissions, and a decrease of about 3 ppbv over Eurasia for the Europe SO₂ perturbation. Decreases in surface
MDA8 O₃ are mostly confined to near the source region. Changes in similar magnitude have been reported over China using
both a chemistry-transport model and observations (Li et al., 2019). Large sulfate decreases have occurred since the 1970s in
both Europe and the US. The SO₂ perturbation in our study (zero-out 2015 level emissions) is 10.8, 12.4, and 16.2 Tg SO₂ y⁻¹
in the US, Europe, and China, respectively. These results imply that the sulfate decreases from clean air regulations and
200 technologies have had the unintended consequence of driving O₃ up by a few ppb during the summertime in the US and Europe.
NO_x emissions have also decreased dramatically over roughly the same time period and have likely more than offset any O₃
increase from decreasing sulfate. However, the full potential of possible O₃ improvement via NO_x and anthropogenic volatile
organic carbon (VOC) decreases may have been partially masked by sulfate decreases. These findings highlight the importance
of a multi-pollutant strategy for effective clean air regulation.

205 Finally, in Fig. 6 we plot the relative change in MAM O₃→O(¹D) and NO₂ photolysis rates, denoted j_{O^1D} and j_{NO_2} in
response to SO₂ emissions in each region. Photolysis of both species is slightly influenced by changing SO₂ emissions,
especially over China in response to China SO₂ emissions, where decreases in both photolysis rates are about 7%. For each of
the perturbations, especially the US and Europe cases, changes in photolysis rates rarely rise above the noise, which is likely
caused by meteorological factors such as slight changes in cloud cover. We conclude that while radiative effects via photolysis
210 are non-negligible, they are significantly less important than chemical effects for aerosol impacts on oxidation, consistent with
previous findings (Li et al., 2019).

4 Summary and conclusions

Using the updated GFDL-AM3 nudged chemistry-climate model with online aerosol heterogeneous chemistry and
interactions with radiation, we estimate the impact of northern hemisphere mid-latitude regional anthropogenic SO₂ emissions
215 on tropospheric OH, HO₂, O₃, and NO_x. Regional SO₂ emissions perturbations lead to significant changes to sulfate aerosol in
far-reaching regions of the world, particularly in the Arctic and the mid and upper troposphere. OH and HO₂ decrease
throughout the northern hemisphere mid-troposphere by up to 10%, which in turn increase NO_x concentrations by at least 5%.
NO_x is not efficiently removed by heterogeneous reactions on aerosols, while species that contribute to NO_x sinks such as OH
(via HO₂ uptake), N₂O₅, and NO₃ are efficiently removed, slowing down the NO_x sink and increasing NO_x concentrations.
220 However, any influence of NO_x increases on tropospheric O₃ are overwhelmed by HO₂ decreases, and the resulting decrease
in O₃ production offsets decreases in O₃ sinks, resulting in up to 4% decrease in O₃ in the free troposphere and at the surface.
Aerosols impact oxidation primarily through heterogeneous reactive uptake pathways over photolysis pathways.

Surface ozone decreases by 3 to 5 ppbv in response to the introduction of regional SO₂ emissions. If SO₂ emissions
decline in developing regions of the world such as South Asia and sub-Saharan Africa, a goal attained through air quality
225 improvements to protect human health, there could be an unintended increase in surface O₃ concentrations. Decreasing surface



O₃ in these regions will require a multipollutant approach in which NO_x and VOCs are simultaneously decreased with aerosols in order to offset the effect of decrease in aerosols and their precursors. While SO₂ and NO_x emissions decreases coincided to some extent in the US, end-of-pipe technologies at power plants allow for control of SO₂ and NO_x individually, and other sources of fine particulate matter (PM_{2.5}) such as waste burning and vehicle emissions will have a similar effect on ozone as sulfate aerosols. PM_{2.5} and SO₂ have decreased dramatically in recent years in the US and Europe, such that O₃ improvements may have been partially masked by the aerosol impact. SO₂ perturbations from our simulations are 10.8, 12.4, and 16.2 Tg y⁻¹ for US, Europe, and China, respectively, which result in a 3 to 5 ppbv surface ozone response over China, where SO₂ emission are the largest in 2015.

Future work is needed to improve estimates of reactive uptake of HO₂ and other radical species by aerosols, as great uncertainty still exists surrounding this parameter as well as the dependence of aerosol composition on reactive uptake parameters (George et al., 2013). We focus here on anthropogenic aerosols as they are changing rapidly and expected to continue to change. Previous work finds a large influence of Saharan dust aerosols on oxidation (Tie et al., 2005). Regions of biomass burning such as Africa and South America are also potential contributors to aerosol-driven oxidation changes. In order to avoid “trading one problem for another” in areas of the world that are experiencing both rapid emissions changes and high exposures to air pollutants, we must better understand the impact of aerosols on atmospheric photochemistry.

Author Contributions

DMW wrote the manuscript, created all figures, and conducted all simulations. AMF and DMW originally conceived the project. CBB assisted with model setup and analysis of output. GC developed the model for use at LDEO. All authors contributed to editing the manuscript.

Code Availability

The code for GFDL-AM3 is available here: <https://www.gfdl.noaa.gov/am3/>.

Data Availability

Data is available here: https://figshare.com/articles/dataset/Concentration_data_for_aerosol_impact_on_oxidants/13331066 (Westervelt, 2020)

Competing Interests

The authors declare no competing interests.

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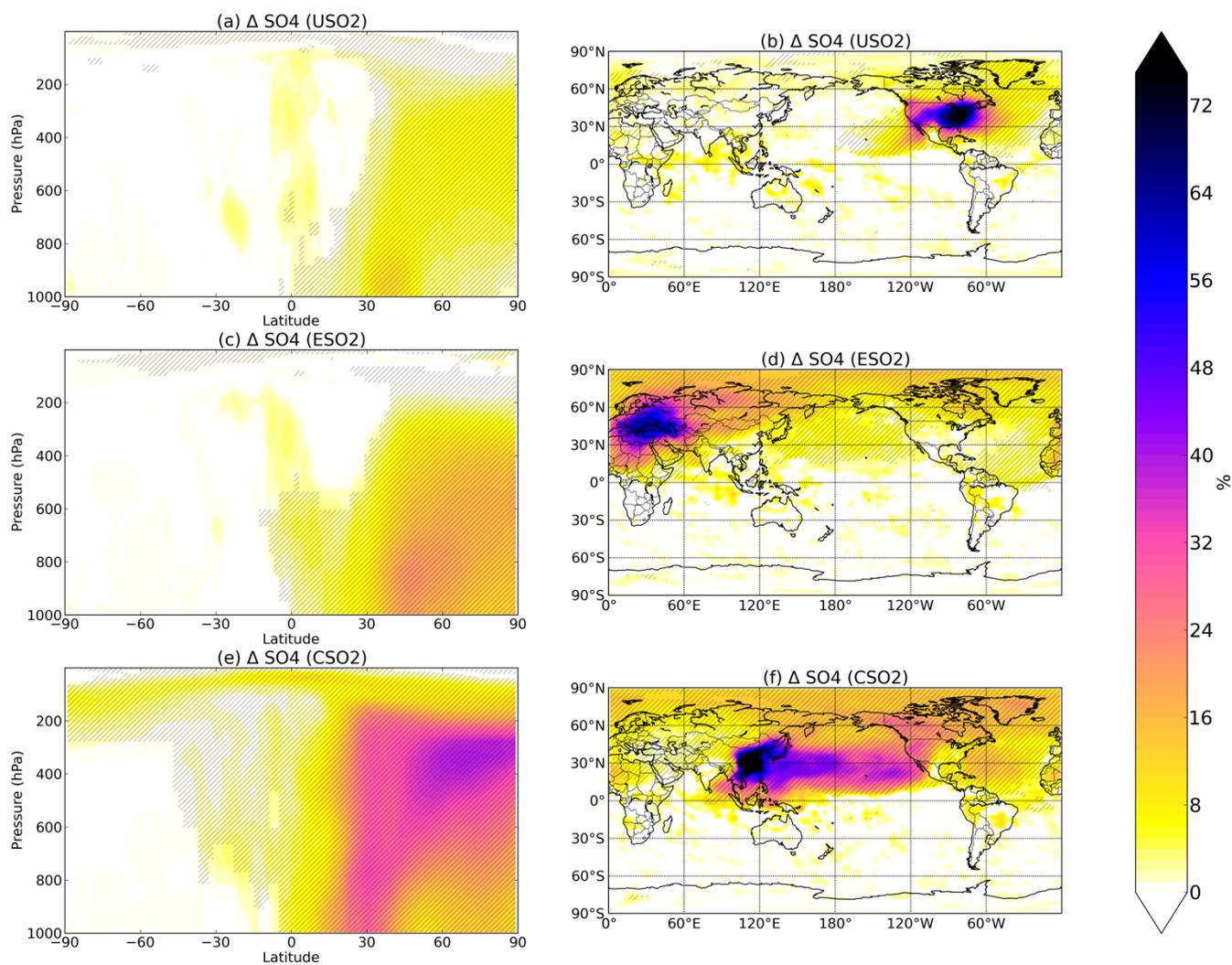
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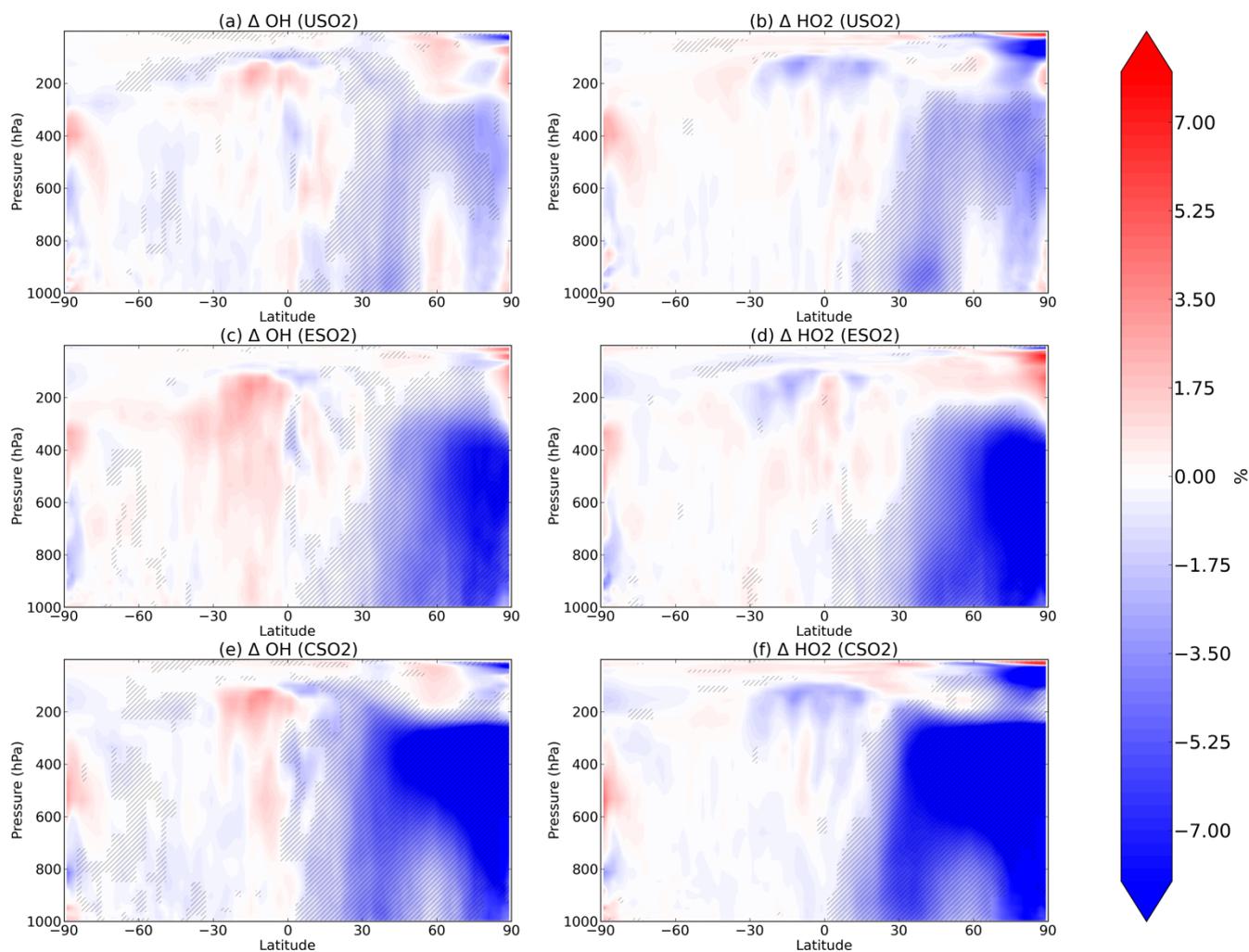
Table 1: Heterogeneous reactive uptake coefficients for several reactions in GFDL-AM3

Reaction	Uptake coefficient (γ)
$\text{HO}_2 \rightarrow \text{H}_2\text{O}_2 \text{ or } \text{H}_2\text{O}$	0.2
$\text{N}_2\text{O}_5 \rightarrow 2.0 \text{ HNO}_3$	0.1
$\text{NO}_3 \rightarrow 1.0 \text{ HNO}_3$	0.1
$\text{NO}_2 \rightarrow 0.5 \text{ HNO}_3$	0.0001

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420 **Figure 1: Boreal springtime (MAM) mean percent change in sulfate concentration between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a,b) US, (c,d) Europe, and (e,f) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.**



425 **Figure 2: Boreal springtime (MAM) mean percent change in OH (left column) and HO₂ (right column) between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a,b) US, (c,d) Europe, and (e,f) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.**

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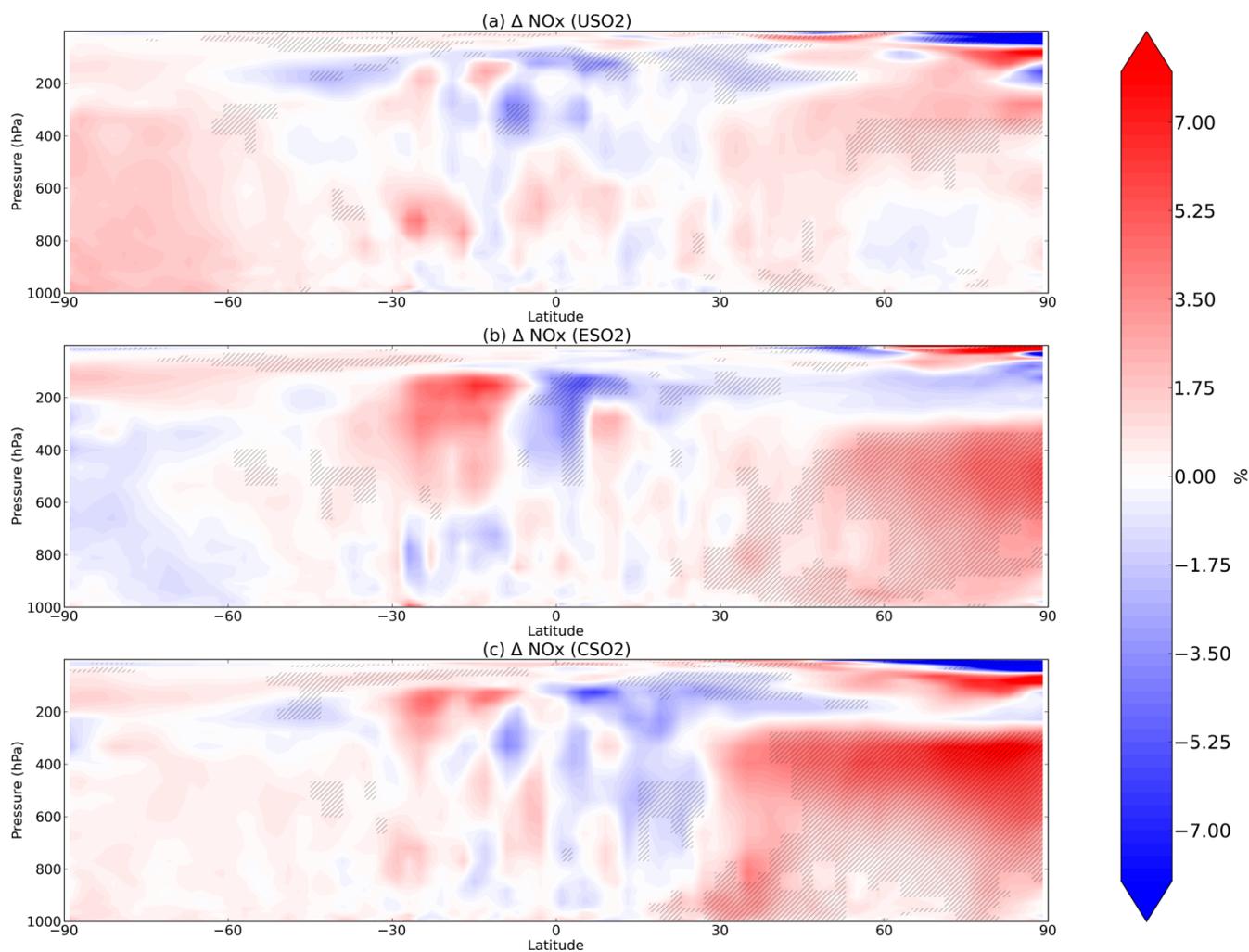


Figure 3: Boreal springtime (MAM) mean percent change in NO_x between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a) US, (b) Europe, and (c) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.

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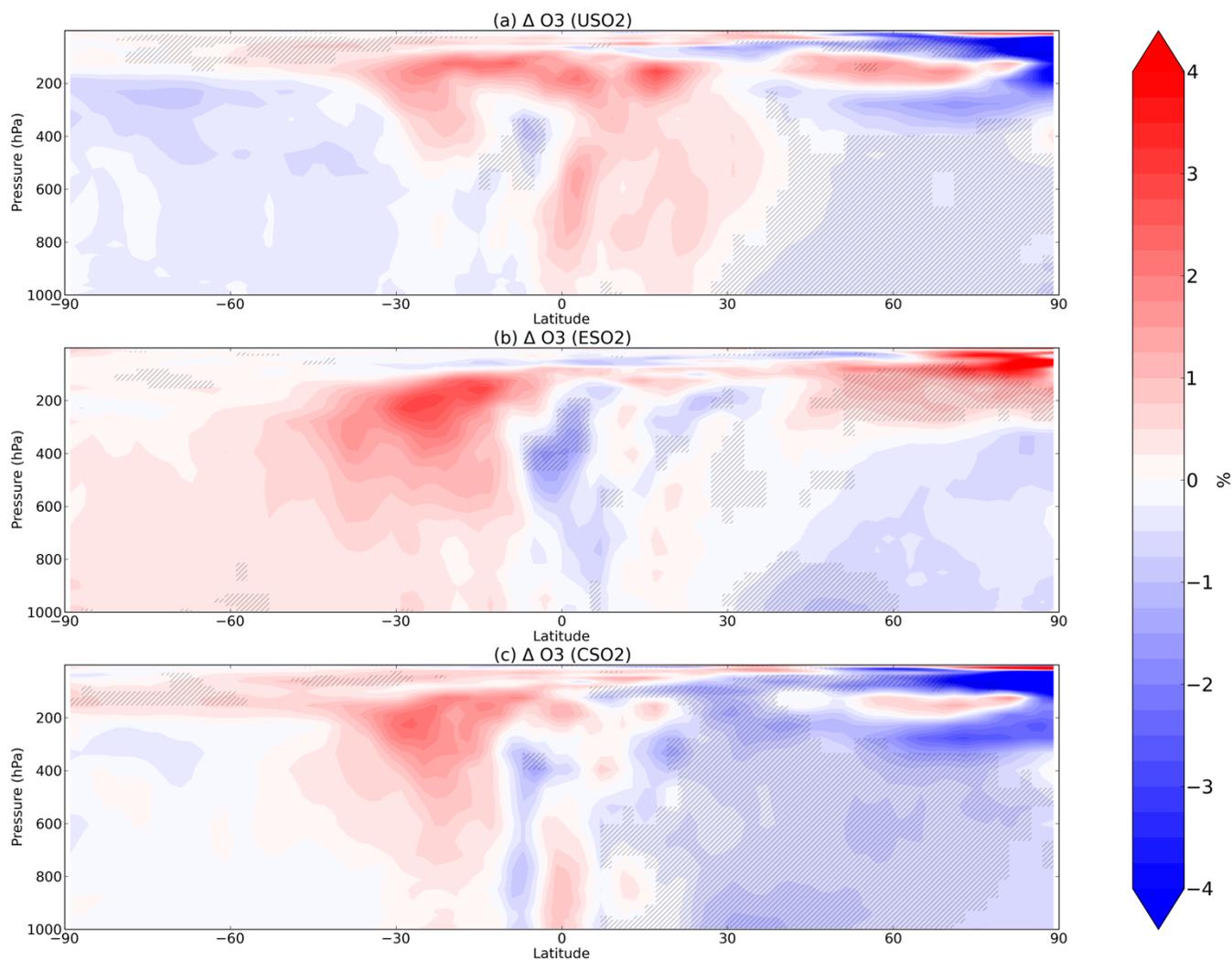
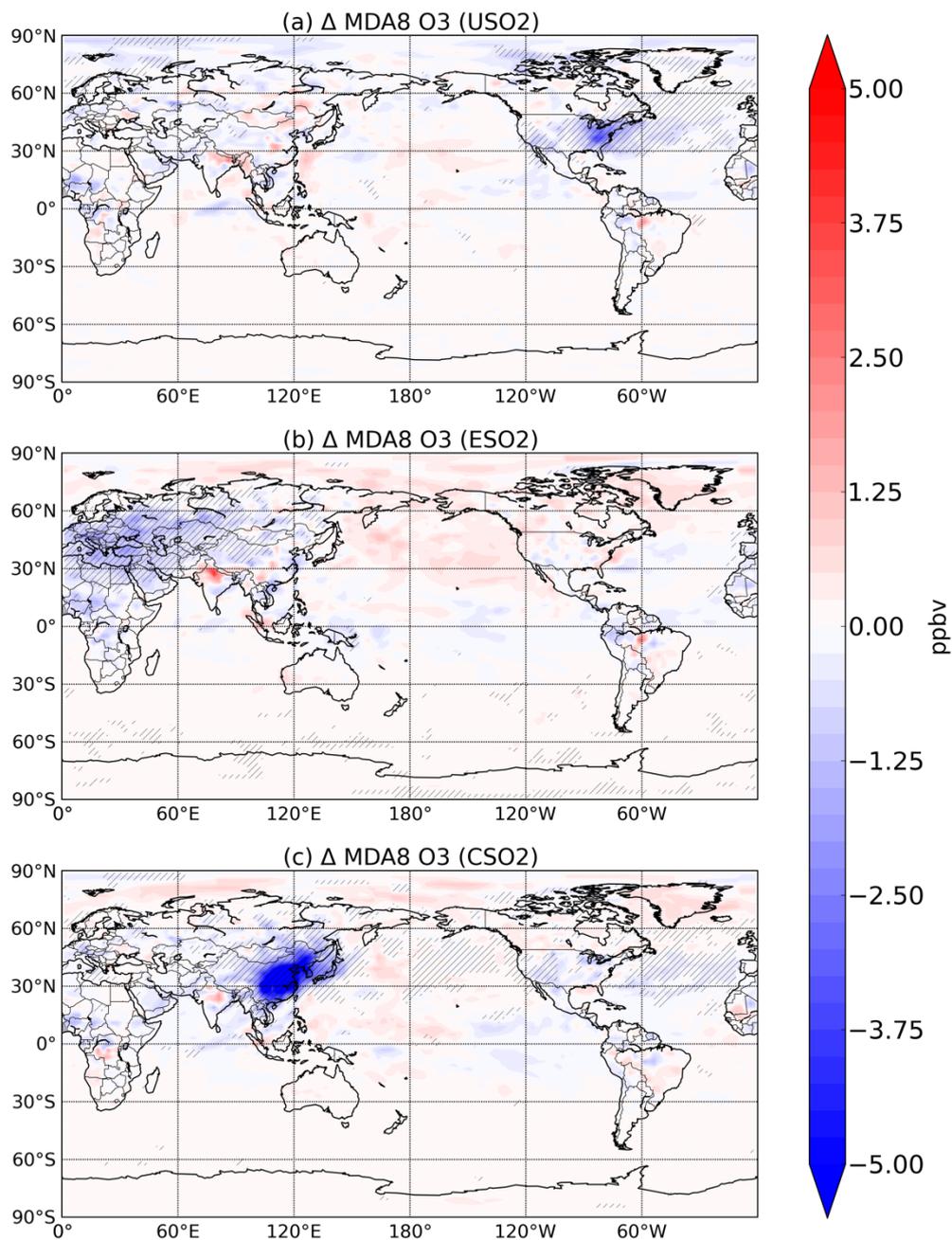


Figure 4: Boreal springtime (MAM) mean percent change in O₃ between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a) US, (b) Europe, and (c) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.

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460 **Figure 5: Summertime (JJA) surface O₃ change (in ppbv) between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a) US, (b) Europe, and (c) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.**

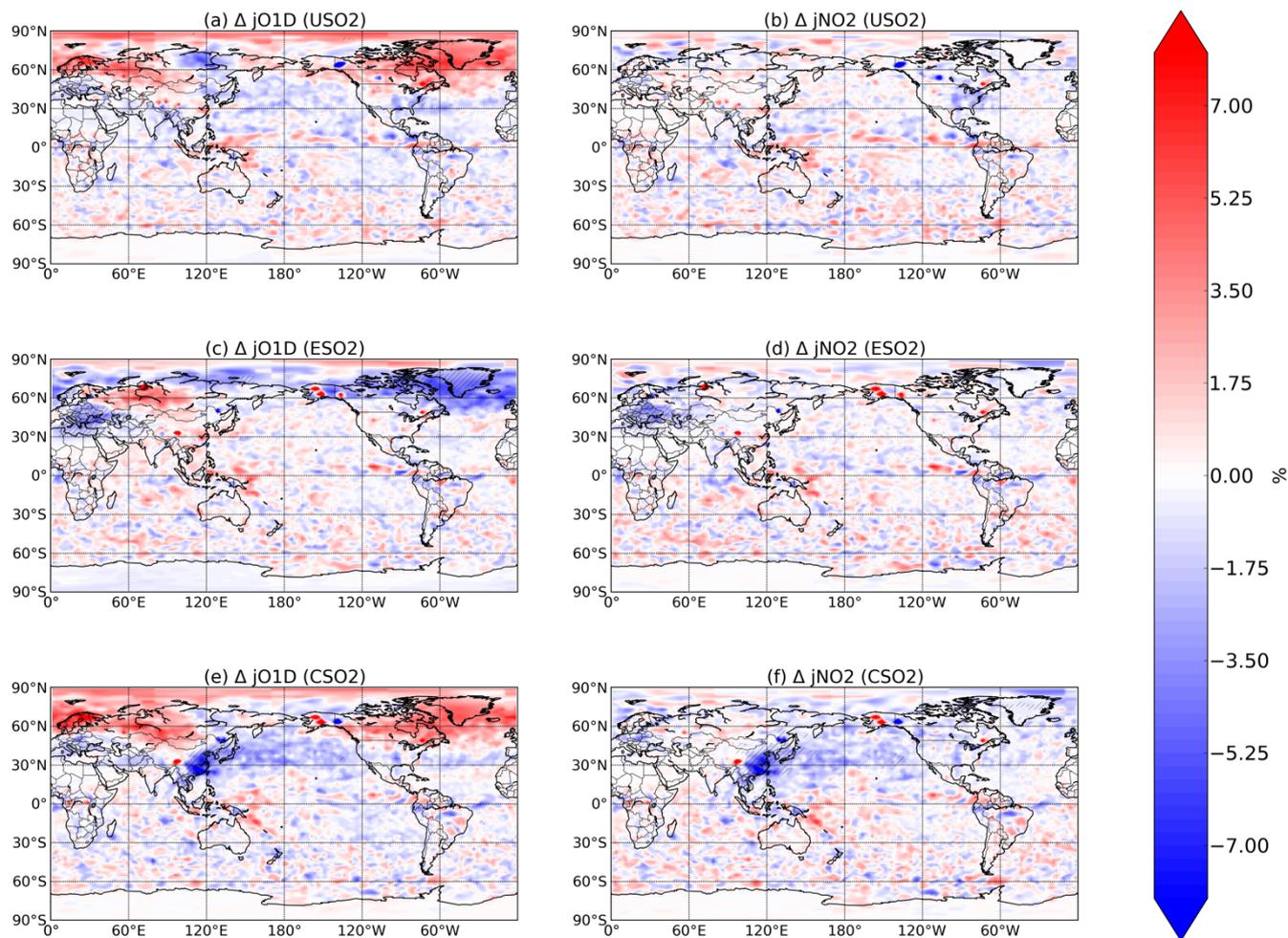


Figure 6: Boreal springtime (MAM) mean percent change in photolysis rates (j_{O1D} , left column and j_{NO2} , right column) between a control simulation and a perturbation simulation in which anthropogenic SO₂ emissions are removed over a certain region: (a,b) US, (c,d) Europe, and (e,f) China. Hatching denotes statistical significance according to a Student's t-test at the 95% confidence level.

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