

**American Petroleum Institute**

**September 25, 2015 10 AM**

**EO 12866 Meeting- EPA Ozone NAAQS Review**

**EPA should allow the current standard 75ppb to be fully implemented**

- There is no need to lower the current standard.
- We have not yet realized the benefits of fully implementing 75 ppb.
- As we'll discuss the science shows that 75 ppb is protective of human health and the environment.

**The Current Standard of 75 ppb is supported by science and is protective of public health**

EPA places greatest weight on controlled human exposure studies and much less on epidemiological studies

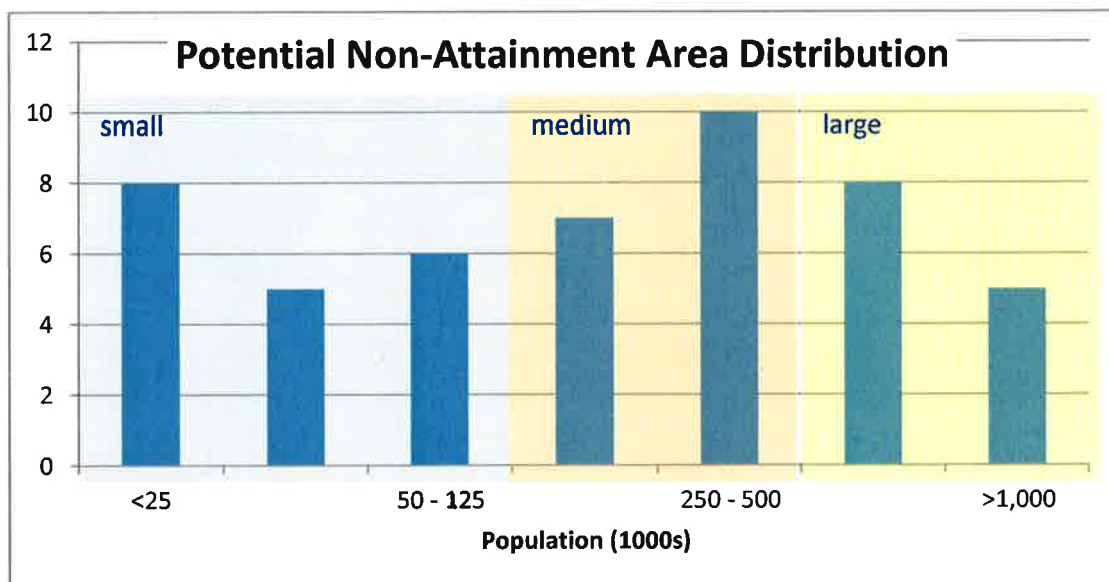
- EPA relies heavily on one clinical study conducted below the current standard -- Schelegle at 72 ppb ozone which showed a very small (<6%) mean, reversible decrease in lung function capacity
- The study also showed a mean increase in respiratory symptoms not correlated with individual lung function decrements as explained and discussed by Gradient last week
- The test was conducted for 6.6 hours with moderate exercise, 10 minute breaks every 50 minutes and was designed to simulate the worst exposure scenario (outdoor workers)
- It was conducted on young healthy adults, which studies support as being the most responsive age group. Interestingly, the best available scientific evidence does not suggest a greater response in children, people with asthma and older adults.
- In conclusion, the one study EPA largely relies on, was conducted on the most responsive population, using conditions that represent the worst case exposure.
- Even if one concluded the small effects observed at 72ppb are adverse, a standard set at 70 ppb would protect public health with an adequate margin of safety especially given exposure scenarios and uncertainties from the studies. The science does not support going lower.

**Lowering the Standard will intensify regulatory burden to states by needlessly adding large areas of nonattainment.**

- Analysis shows that the import of emissions from China has neutralized ~43% of the ozone decrease that would have otherwise occurred in the western US with US air pollution controls. (We will also leave them this article for the docket)  
[www.nature.com/ngeo/journal/vaop/ncurrent/full/ngeo2493.html](http://www.nature.com/ngeo/journal/vaop/ncurrent/full/ngeo2493.html)

- Background ozone will be presented fully in a meeting with you later today but in summary- EPA has not sufficiently addressed background ozone and exceptional events.
- As mentioned before, API's evaluation of the science highlights no justification to change the Ozone NAAQS. That being said, it has been reported in the trade press that members of the Administration are suggesting a final Ozone NAAQS of 68 ppb. In light of that information I want to take a couple of minutes to help you understand the impact of that suggested option.
  - First, using the 2012-2014 data, there are 134 monitored counties that could be designated nonattainment under a standard of 68 ppb as opposed to 70 ppb.
- The number is not the whole story, it is also important to know those non-attainment designations would be taking place in areas that have never been in nonattainment before. Thus those areas have no familiarity with non-attainment which could make administering the NAAQS compliance programs problematic for the states.

These additional counties span 29 states. The counties are either individual rural counties or counties with mostly small to medium size cities. (table)



- Grand Canyon National Park (AZ) and Mammoth Cave National Park (KY) could be non-attainment as well as the Blackwater National Wildlife Refuge on Maryland's eastern shore.
- Costs in these areas might largely be borne by light manufacturing and small businesses.

**Lowering the Standard may have direct and significant negative impacts to the Refinery System and Exploration and Production**

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# 2012-2014 OZONE DESIGN VALUES

2012-2014 CASTNET O3 Design Values  
September 23, 2015

# Rapid increases in tropospheric ozone production and export from China

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**Rapid population growth and industrialization have driven substantial increases in Asian ozone precursor emissions over the past decade<sup>1</sup>, with highly uncertain impacts on regional and global tropospheric ozone levels. According to ozonesonde measurements<sup>2,3</sup>, tropospheric ozone concentrations at two Asian sites have increased by 1 to 3% per year since 2000, an increase thought to contribute to positive trends in the ozone levels observed at North America's West Coast<sup>4,5</sup>. However, model estimates of the Asian contribution to North American ozone levels are not well-constrained by observations<sup>6,7</sup>. Here we interpret Aura satellite measurements of tropospheric concentrations of ozone and its precursor NO<sub>2</sub>, along with its largest natural source, stratospheric ozone, using the TM5 global chemistry-transport model. We show that tropospheric ozone concentrations over China have increased by about 7% between 2005 and 2010 in response to two factors: a rise in Chinese emissions by about 21% and increased downward transport of stratospheric ozone. Furthermore, we find that transport from China of ozone and its precursors has offset about 43% of the 0.42 DU reduction in free-tropospheric ozone over the western United States that was expected between 2005 and 2010 as a result of emissions reductions associated with federal, state and local air quality policies. We conclude that global efforts may be required to address regional air quality and climate change.**

Elevated tropospheric ozone (O<sub>3</sub>) concentrations have a direct adverse impact on human<sup>8</sup> and ecosystem health<sup>9</sup> at the surface, whereas in the free troposphere O<sub>3</sub> acts as a greenhouse gas<sup>8,10</sup> and drives the production of the hydroxyl radical, which controls the chemical lifetime of many atmospheric pollutants and reactive greenhouse gases. Previous studies have pointed not only to changing emissions of O<sub>3</sub> precursors<sup>6,7,11–14</sup>, but also to changes in both the net inflow of O<sub>3</sub> from the stratosphere<sup>14–16</sup> and large-scale transport patterns<sup>7,13–16</sup> as contributing to observed trends and variability in tropospheric O<sub>3</sub>. For many regions outside Asia, *in situ* measurements suggest that tropospheric O<sub>3</sub> has remained relatively constant in the 2000s following substantial increases in the 1980s–1990s, at least partially due to changes in the emissions of O<sub>3</sub> precursors, including nitrogen oxides (NO<sub>x</sub>) and hydrocarbons<sup>17</sup>. However, chemistry-transport models used for attribution of trends in Europe and North America tend to greatly underestimate the observed O<sub>3</sub> changes over the past several decades<sup>18</sup>. Furthermore, trends are generally derived from sparse surface and balloon measurements that are not necessarily representative of larger regions. This is particularly true for China, where measurements from only two ozonesonde stations are reported in the literature<sup>2,3</sup>.

Over the past decade, satellite instruments have provided valuable information on the global distribution of tropospheric O<sub>3</sub> and NO<sub>x</sub> concentrations and their evolution in time. Here we use O<sub>3</sub> retrievals from the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS) sensors and NO<sub>2</sub> from the Ozone Monitoring Instrument (OMI), all onboard NASA's Aura satellite (Methods). We investigate changes in TES tropospheric O<sub>3</sub> and OMI NO<sub>2</sub> over China and the western United States (US) between 2005 and 2010, and attribute the observed O<sub>3</sub> changes to emissions, transport and chemistry using the TM5 chemistry-transport model<sup>19</sup> (Methods), with both emissions and stratosphere-troposphere exchange constrained by the satellite measurements.

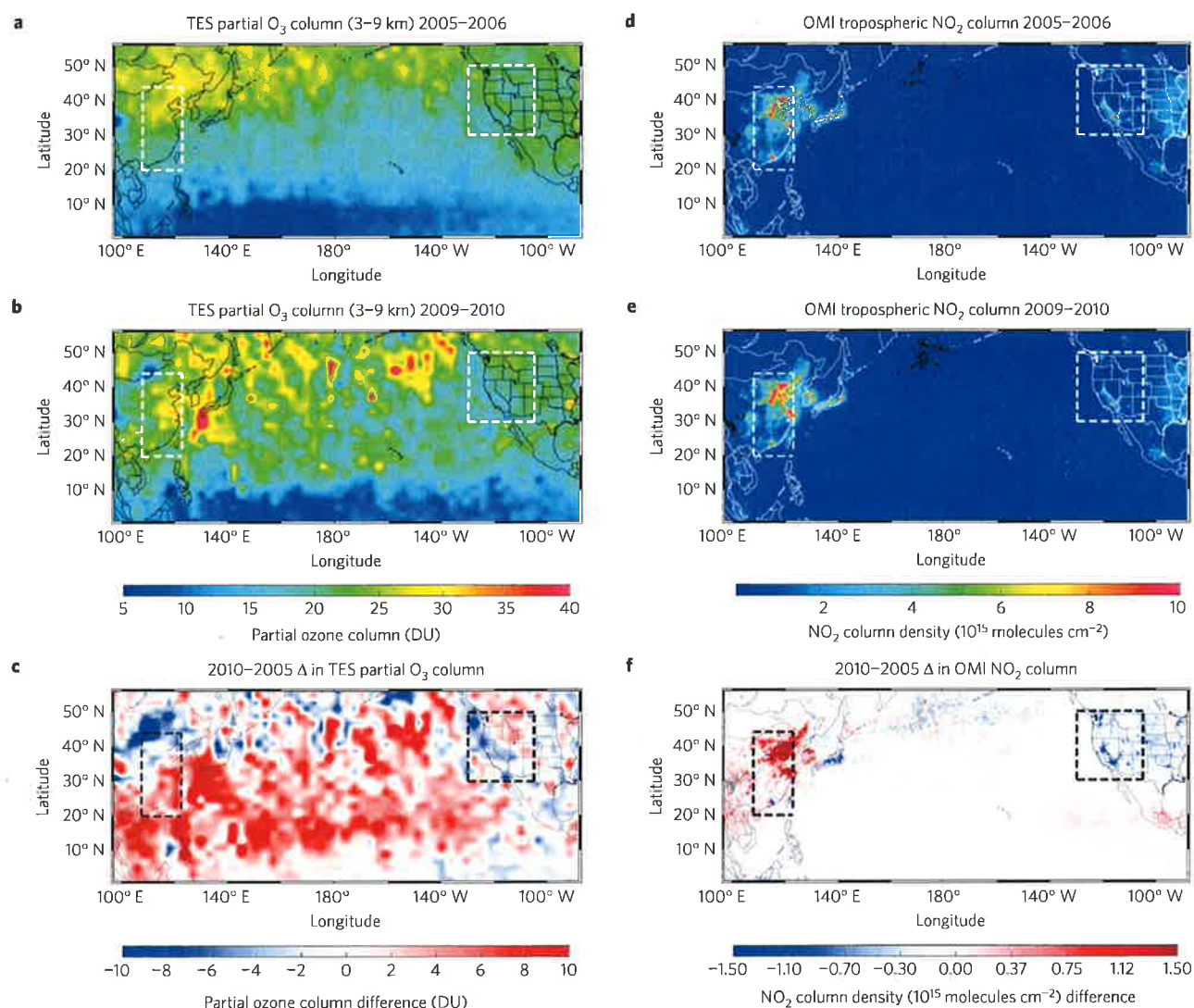
During summertime, when photochemical tropospheric O<sub>3</sub> production is most active, TES observes substantial increases of >10 DU in the partial O<sub>3</sub> column (3–9 km) over eastern China between 2005–2006 and 2009–2010 (Fig. 1a–c). At the same time, OMI observes NO<sub>2</sub> column increases of >1.5 × 10<sup>15</sup> molecules cm<sup>-2</sup> (Fig. 1d–f). Figure 1a–c also reveals a strong increase in O<sub>3</sub> off the coast of China (>6.0 DU), suggestive of O<sub>3</sub> production and transport aloft via the dominant westerlies<sup>12</sup>, and weaker increases over the eastern Pacific (~3.7 DU). OMI observes significant reductions in NO<sub>2</sub> (>0.3 × 10<sup>15</sup> molecules cm<sup>-2</sup>) over much of the western US, especially California and Washington (Fig. 1d–f). TES shows decreases in O<sub>3</sub> over the western US in the areas with the largest NO<sub>2</sub> decreases, but the region as a whole shows an O<sub>3</sub> increase of ~2.7 DU.

Monthly averaged O<sub>3</sub> values derived from TES measurements show the detailed temporal evolution of partial O<sub>3</sub> columns (3–9 km) for eastern China and the western US (Fig. 2a,e). These results are not strongly dependent on sampling (Supplementary Information 1). We interpret the observed satellite O<sub>3</sub> short-term trends with simulations from TM5 (Fig. 2a,e). To account for the rapid changes in NO<sub>x</sub> emissions that have occurred between 2005 and 2010 (as reflected in OMI NO<sub>2</sub> measurements in Fig. 1), we use monthly means of the OMI tropospheric NO<sub>2</sub> column record to derive improved estimates of global monthly NO<sub>x</sub> emissions for 2005–2010 (Methods and Supplementary Information 2). These top-down estimates<sup>20</sup>, in line with published trends (Methods), indicate a 21% increase in NO<sub>x</sub> over China from 25.1 to 30.3 Tg yr<sup>-1</sup> NO<sub>x</sub> for 2005–2010 (Supplementary Table 1). Over the western US, a 21% decrease is estimated (4.8 to 3.8 Tg yr<sup>-1</sup> NO<sub>x</sub>) following strict American emission controls for vehicles and power plants<sup>21</sup>.

In our attribution of the observed signals in tropospheric O<sub>3</sub>, we consider the effects of changes in anthropogenic NO<sub>x</sub> emissions, stratosphere-troposphere exchange of ozone (STF) and long-range transport. We focus on NO<sub>x</sub> emissions rather than other

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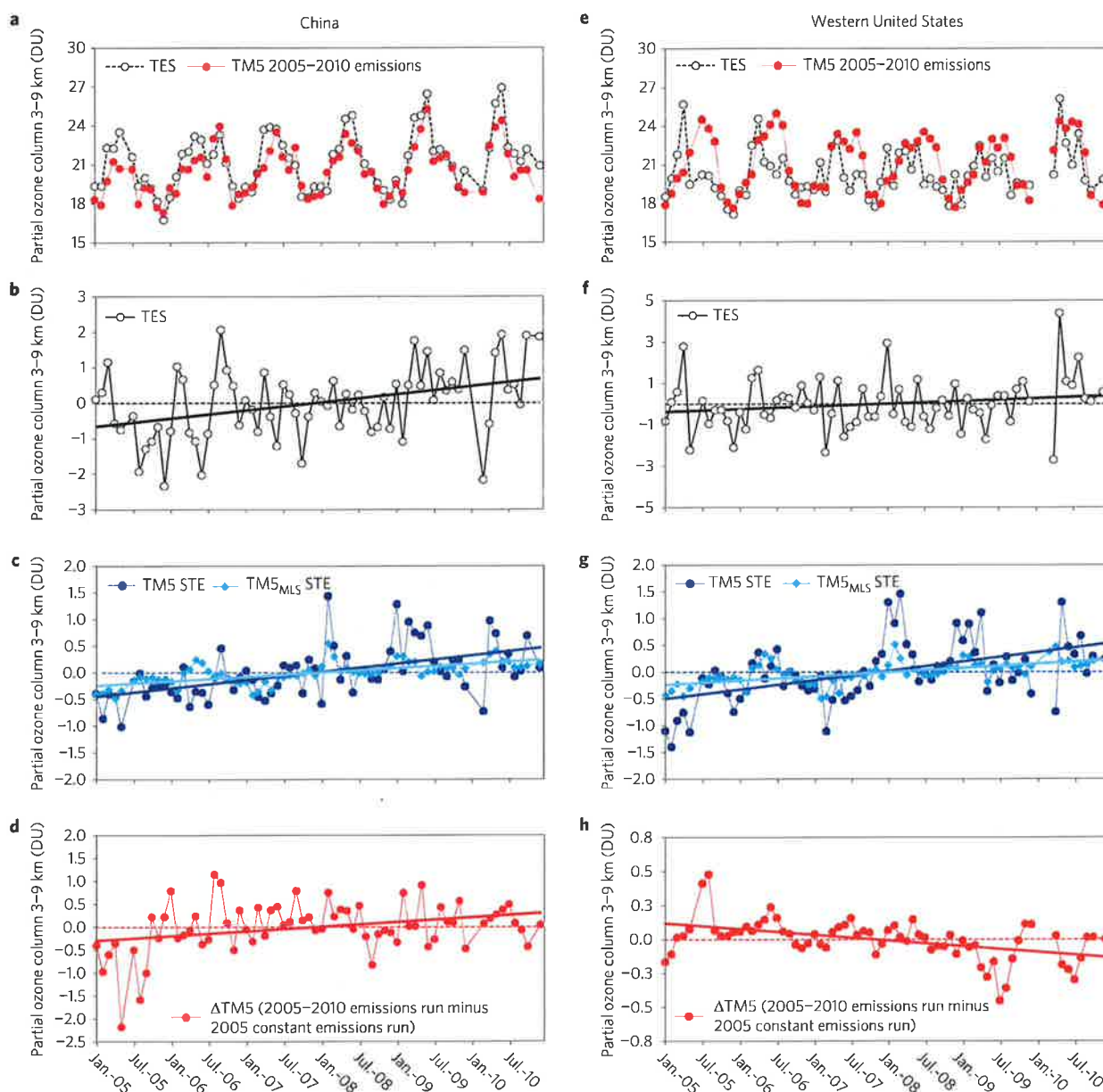
**Figure 1 | Summertime tropospheric  $O_3$  and  $NO_2$  distributions for 2005–2006 and 2009–2010 observed by TES and OMI.** **a**, Averaged TES partial  $O_3$  column (3–9 km) for May–August 2005–2006 (clear-sky conditions: cloud top pressure >750 hPa, effective optical depth <50). **b**, Same as **a** but for 2009–2010. **c**, Difference between 2010 and 2005 TES observations. **d–f**, Corresponding results from OMI tropospheric  $NO_2$  columns (mostly clear-sky situations: cloud radiance fraction <0.5). TES (OMI) observations have been aggregated over  $3^\circ \times 2^\circ$  ( $0.25^\circ \times 0.25^\circ$ ) longitude  $\times$  latitude grid cells. Dashed lines: Eastern China:  $105^\circ$ – $129^\circ$  E,  $18^\circ$ – $42^\circ$  N and western US:  $130^\circ$ – $105^\circ$  W,  $20^\circ$ – $50^\circ$  N (typically including  $\sim 70$  TES data points per month).

precursors because both regions are  $NO_x$ -limited throughout the photochemically active seasons, and top-down scaling suggests that lower tropospheric  $O_3$  concentrations are 5–15 times less sensitive to changes in anthropogenic volatile organic compounds (VOCs) and CO than to changes in  $NO_x$  (ref. 20; Supplementary Information 2). We use TM5 to evaluate the contributions of these different drivers to the observed 2005–2010  $O_3$  changes (Methods). TM5 with the OMI-based  $NO_x$  emissions generally captures the absolute magnitude and temporal evolution of the TES  $O_3$  observations over China (Fig. 2a). Over the western US, TM5 also reproduces the monthly observations, but with slightly larger deviations from TES during the summer months, which we attribute primarily to errors in modelling local influences rather than long-range transport (Supplementary Information 3).

Deseasonalized TES observations (Methods and Fig. 2b,f) show a large, statistically significant ( $P = 0.002$ , 95% confidence interval, two-tailed) increase in tropospheric  $O_3$  over eastern China of  $1.08\% \text{ yr}^{-1}$  ( $0.23 \text{ DU yr}^{-1}$ ) for the period 2005–2010, and a smaller, insignificant increase ( $P = 0.16$ ) over the western US ( $0.56\% \text{ yr}^{-1}$ ,

$0.13 \text{ DU yr}^{-1}$ ) (Table 1). Local soundings over Beijing<sup>3</sup> show an increase in  $O_3$  of  $\sim 5\% \text{ yr}^{-1}$  (2002–2011) at 3–9 km altitude; co-located TES and TM5 grid cells show an increase of  $\sim 3$  and  $\sim 4\% \text{ yr}^{-1}$ , respectively.

To investigate the STE contribution to the observed  $O_3$  short-term trends, we examine TM5 simulations with tagged stratospheric  $O_3$  (inflow driven by ECMWF cross-tropopause fluxes of satellite-constrained stratospheric  $O_3$  distributions<sup>22</sup>). These show a substantial positive short-term trend ( $P < 0.001$ ) in the stratospheric contribution, with STE accounting for an increase of  $\sim 0.80\% \text{ yr}^{-1}$  ( $0.78$ – $0.84$ ) ( $P < 0.001$ ) in the 3–9 km tropospheric  $O_3$  over both regions (Fig. 2c,g, Table 1 and Supplementary Information 4). MLS observations of the Northern Hemisphere mid-latitudes, however, suggest only a small positive trend in the stratospheric contribution to tropospheric  $O_3$  for the period 2005–2010, driven almost entirely by the 2009/2010 El Niño/Easterly shear quasi-biennial oscillation (ref. 16). The large model STE trend seems to arise from an unrealistically large trend in lower stratospheric  $O_3$  as compared to MLS

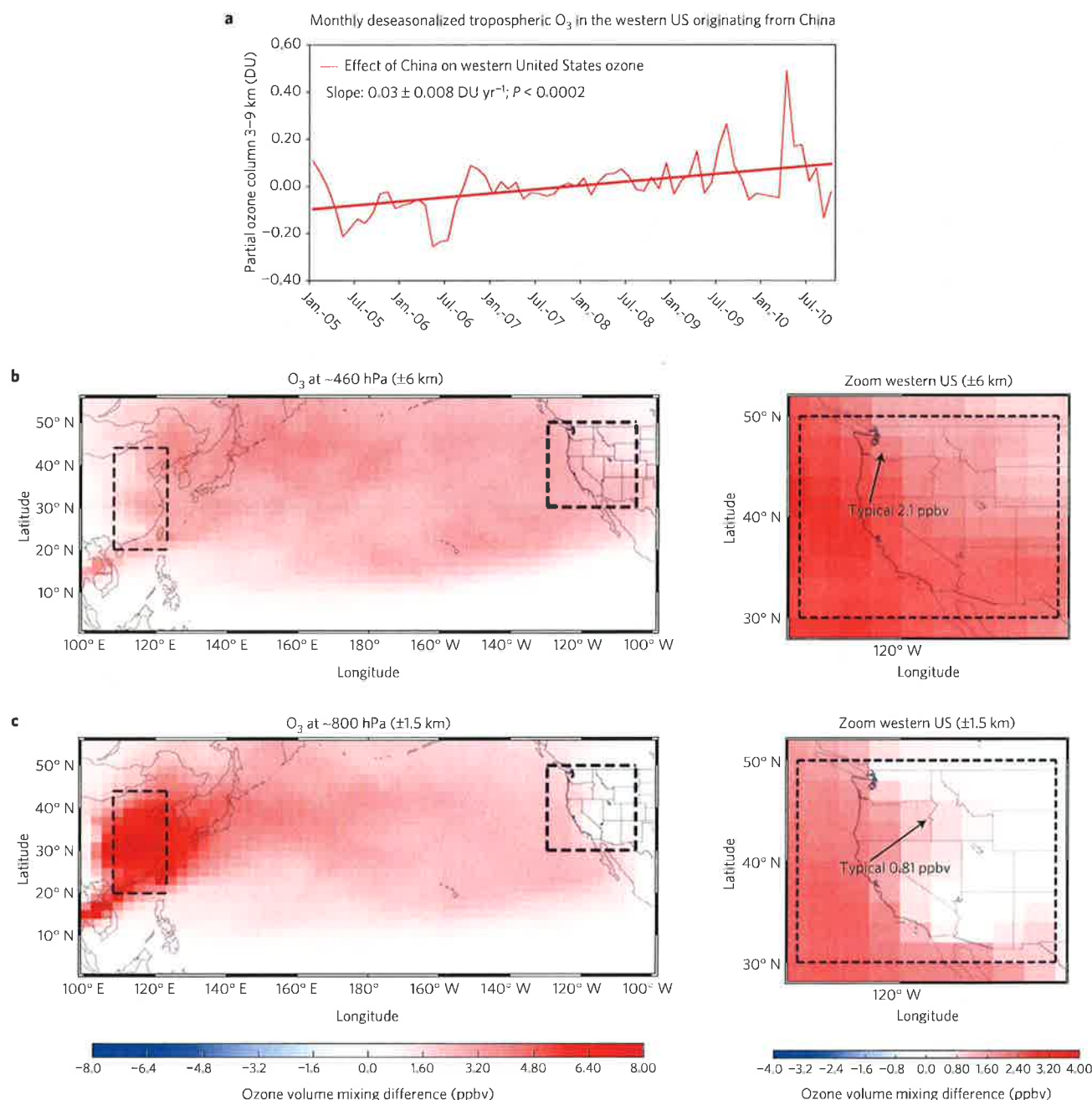


**Figure 2** | 2005–2010 observed and simulated changes in monthly tropospheric partial  $O_3$  columns over eastern China and the western US. **a–d**, Eastern China. **e–h**, Western US. **a, e**, TES 3–9 km  $O_3$  time series (dashed black curve) and TM5 simulations (dashed red) with OMI-inferred anthropogenic  $NO_x$  emissions. **b–d, f–h**, Short-term trends in deseasonalized  $O_3$  columns. **b, f**, TES deseasonalized time series (solid black line). **c, g**, 2005–2010 changes in the contribution of stratospheric  $O_3$  to the tropospheric column for TM5 (dark blue) and TM5<sub>MLS</sub> (bright blue). **d, h**, Effect of changing  $NO_x$  emissions (OMI-inferred 2005–2010  $NO_x$  emissions run minus the constant year 2005 emissions run).

(Supplementary Information 4). To better reflect the considerable uncertainties in STE, we provide a second estimate of the STE contribution to the tropospheric  $O_3$  changes by constraining TM5 lower stratospheric  $O_3$  concentrations using MLS measurements<sup>16</sup> (TM5<sub>MLS</sub>; Supplementary Information 4), which results in a smaller STE contribution of  $\sim 0.5\% \text{ yr}^{-1}$  (Table 1). With the MLS-corrected STE (Fig. 2c,g), the modelled  $O_3$  trend is reduced from  $1.33\% \text{ yr}^{-1}$  to  $1.03\% \text{ yr}^{-1}$  over China and from  $0.65\% \text{ yr}^{-1}$  to  $0.27\% \text{ yr}^{-1}$  over the western US (Table 1). Although TM5<sub>MLS</sub> trends are slightly lower than those from TM5, both agree with TES trends within their uncertainties.

To quantify the impacts of  $NO_x$  emissions changes on the partial  $O_3$  columns, we subtract the TM5 run with updated 2005–2010  $NO_x$  emissions from a simulation with constant 2005 emissions,

so that all processes driven by annually varying meteorology, including STE, cancel out. The 21% increase in  $NO_x$  emissions between 2005 and 2010 (inferred from monthly OMI observations) is responsible for a  $0.10 \text{ DU yr}^{-1}$  ( $0.58\% \text{ yr}^{-1}$ ) trend in tropospheric  $O_3$  over China (Fig. 2d and Table 1). Furthermore, the increases over eastern Asia are strongest in summertime (Supplementary Information 5), indicative of an important role for enhanced photochemical production. For the western US, 21% reductions in OMI-inferred  $NO_x$  emissions result in a clearcut decrease ( $-0.04 \text{ DU yr}^{-1}$ ) in modelled tropospheric  $O_3$  (Fig. 2h and Table 1). Note that the short-term trends are significantly different from zero despite the relative high standard errors (s.e.m.); the relative changes in the partial  $O_3$  columns for 2005–2010 from TM5 agree well with TES only if the  $NO_x$  emissions changes are considered



**Figure 3 | The effect of Chinese emissions and long-range transport on tropospheric  $O_3$  over the western US. **a**, Monthly deseasonalized difference between the global TM5 run with observationally constrained 2005–2010 anthropogenic  $NO_x$  emissions and the TM5 run with constant 2005  $NO_x$  emissions for China and observationally constrained emissions everywhere else. **b**, Difference in  $O_3$  in the free troposphere ( $\sim 460$  hPa) between the two TM5 runs for April–May 2010 sampled at the Aura overpass time. **c**, As for **b** but showing the lower troposphere ( $\sim 800$  hPa).**

(Supplementary Information 2). Over China, the  $NO_x$  emissions update results in an increase in  $O_3$  of  $\sim 0.6\% \text{ yr}^{-1}$  and thus explains roughly half of the TM5 short-term trend ( $1.03\text{--}1.33\% \text{ yr}^{-1}$ , Table 1). Over the western US, non-significant  $O_3$  increases were found ( $P = 0.159$  and  $0.120$  for TES and TM5, respectively), despite the significant  $NO_x$ -driven  $O_3$  decrease of  $0.2\% \text{ yr}^{-1}$ .

From the above attribution of  $O_3$  sources, we find that increased STE may have masked the effects of reductions in anthropogenic  $NO_x$  emissions in the US for 2005–2010. A critical question, however, is how the 2005–2010 observed Chinese emissions and  $O_3$  increases have influenced tropospheric  $O_3$  concentrations over the western US (refs 5,23). To obtain a quantitative estimate

of the impacts of Chinese emissions alone, we compare the global 2005–2010 emission update simulation against a simulation with constant (year 2005) emissions for China combined with OMI-constrained emissions for the rest of the world (Methods and Supplementary Information 2). In this way, the domestic  $NO_x$  emissions reductions in the US and the changes in the other parts of the world are identical in both runs, except for China. The  $O_3$  decrease over the western US associated with both local and Asian  $NO_x$  emissions changes is  $0.04 \text{ DU yr}^{-1}$  (Table 1), but the contribution of emissions and  $O_3$  transport from China to tropospheric  $O_3$  over the western US has grown by  $0.03 \text{ DU yr}^{-1}$  over 2005–2010 (Fig. 3a). The import from China has thus neutralized



**Table 1 | Attribution of absolute and relative changes in 2005–2010 partial O<sub>3</sub> column trends over China and western US.**

	China				Western United States			
	Absolute change ±s.e.m. (DU yr <sup>-1</sup> )		Relative change (% yr <sup>-1</sup> )		Absolute change ±s.e.m. (DU yr <sup>-1</sup> )		Relative change (% yr <sup>-1</sup> )	
	TM5	TM5 <sub>MLS</sub>	TM5	TM5 <sub>MLS</sub>	TM5	TM5 <sub>MLS</sub>	TM5	TM5 <sub>MLS</sub>
TES observed	0.23 ± 0.07		1.08		0.13 ± 0.09		0.56	
TM5 simulated	0.23 ± 0.07	0.18 ± 0.04	1.33	1.03	0.13 ± 0.16	0.04 ± 0.04	0.65	0.27
STE contribution	0.13 ± 0.03	0.08 ± 0.02	0.78	0.45	0.17 ± 0.04	0.08 ± 0.02	0.84	0.47
NO <sub>x</sub> contribution	0.10 ± 0.04		0.58		−0.04 ± 0.01		−0.2	

First row: TES-observed changes in 3–9 km O<sub>3</sub> column (DU yr<sup>-1</sup> and % yr<sup>-1</sup>). Second row: Simulated O<sub>3</sub> column changes with OMI-constrained 2005–2010 NO<sub>x</sub> emissions for both TM5 and the simulation with MLS-corrected STE (TM5<sub>MLS</sub>). Third row: STE contribution to the simulated O<sub>3</sub> changes for TM5 and TM5<sub>MLS</sub>. Fourth row: O<sub>3</sub> column changes attributable to changing (OMI-inferred) NO<sub>x</sub> emissions. Changes are statistically significantly different from zero ( $P < 0.05$ ), except for TES and TM5 trends over western US ( $P > 0.15$ ).

~43% of the 0.07 (= 0.03 + 0.04) DU yr<sup>-1</sup> O<sub>3</sub> decrease that would have occurred largely in response to air pollution controls in the western US, had Chinese emissions been constant. This is consistent with the spatial pattern of O<sub>3</sub> changes seen in Fig. 1c. O<sub>3</sub> over the western US decreased only in regions with very large NO<sub>x</sub> reductions; NO<sub>x</sub> reductions elsewhere (Fig. 1f) have been offset by a combination of STE and Chinese emissions.

The influence of Chinese pollution on the western US is largest during spring<sup>5</sup>, with 2010 (Fig. 3b,c) representing the maximum Chinese influence for the time series (Fig. 3a). The contribution to western US free-tropospheric O<sub>3</sub> is widespread (Fig. 3b) and larger than the contribution to near-surface O<sub>3</sub> (Fig. 3c). Our estimate of the western US response to Chinese emission changes is of the same order of magnitude as reported values for source–receptor relationships established from other models<sup>6,11,24</sup>, which have shown the importance of large-scale transport patterns and processes such as sustained O<sub>3</sub> production during trans-Pacific transport driven by decomposition of nitrogen reservoirs in descending air masses<sup>12,25</sup> in generating this response. The 2010 peak is probably related to the strong El Niño<sup>16</sup>, which favoured efficient transport towards the West Coast<sup>7,15</sup>.

The rapid increase in tropospheric O<sub>3</sub> (almost 7% over six years) from 2005 to 2010 detected by TES over China is evocative of much stronger changes in the lower troposphere (Fig. 3c). TM5 simulations reproduce the observed O<sub>3</sub> increases within their uncertainties (Supplementary Information 6 and Supplementary Table 2) when changes in NO<sub>x</sub> emissions and STE are considered. Our results indicate that policy-driven domestic emissions reductions decrease free-tropospheric O<sub>3</sub> over the western US, but the effect is offset by changes in STE combined with increasing long-range inflow of O<sub>3</sub> and its precursors from China. This Chinese export offsets ~43% of a nominal 0.07 DU yr<sup>-1</sup> domestic O<sub>3</sub> reduction, suggesting that air quality and regional climate change mitigation policies could eventually have limited impact if not considered in a global context, at least for free-tropospheric O<sub>3</sub> and its precursors<sup>10</sup>.

## Methods

Methods and any associated references are available in the online version of the paper.

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### Author contributions

W.W.V. performed the research, drafted the manuscript, prepared the figures and developed the analysis methods. J.L.N. provided analysis and interpretation of the MLS O<sub>3</sub> measurements and aided in drafting the manuscript. J.E.W. contributed to the development of TM5 and the nested anthropogenic emission estimates, and supported

the data interpretation. K.W.B. and J.R.W. provided analysis tools and interpretation. K.W.B., J.L.N. and J.R.W. are responsible for the TES experiment and instrument and project planning. K.F.B. is responsible for the Dutch OMI NO<sub>2</sub> data product, aided in drafting the manuscript and methods, and supported the development of the analysis methods and interpretation. All authors contributed to discussions of the results and preparation of the manuscript.

### Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at [www.nature.com/reprints](http://www.nature.com/reprints). Correspondence and requests for materials should be addressed to W.W.V.

### Competing financial interests

The authors declare no competing financial interests.

## Methods

**TES tropospheric O<sub>3</sub> data.** TES global survey standard products consist of 16 orbits of nadir vertical profiles with  $5 \times 8 \text{ km}^2$  horizontal resolution spaced  $1.6^\circ$  along the polar Sun-synchronous orbit track. In general, the TES vertical resolution for O<sub>3</sub> profiles is 6–7 km, corresponding to  $\sim 1$ –2 degrees of freedom (DOF) in the troposphere. Nonlinear spectral fitting of retrieval parameters is applied based on the optimal estimation technique for deriving TES O<sub>3</sub> from radiative transfer forward model simulations and observed radiances. A priori O<sub>3</sub> information is taken from monthly mean simulations from the MOZART model and averaged over  $10^\circ \times 60^\circ$  grids (latitude by longitude), and the averaging kernel (AK) is used to evaluate the vertical sensitivity of TES-retrieved O<sub>3</sub>. We have used V004 data (2005–2009) and V005 data (2010). Validation of these data has demonstrated that TES accurately captures spatial and temporal patterns in O<sub>3</sub> and, critical to this study, does not exhibit a trend in the retrieval bias<sup>26</sup>. References for TES O<sub>3</sub> retrievals and validation can be found in ref. 26. TES measurements continue, but have limited availability after 2010.

**OMI tropospheric NO<sub>2</sub> column data.** OMI tropospheric NO<sub>2</sub> columns are retrieved under cloud-free conditions (cloud fraction below 20%) at  $\sim 13 \text{ h } 30 \text{ min}$  local time (DOMINO v2; ref. 27) with a three-step approach: retrieval of NO<sub>2</sub> slant columns with Differential Optical Absorption Spectroscopy (405–465 nm wavelength range, spectral resolution of 0.5 nm), followed by assimilation of OMI NO<sub>2</sub> slant columns in the global chemistry–transport model TM4 for providing stratospheric background and a priori NO<sub>2</sub> profile shapes needed to calculate the tropospheric air mass factors. These are then used to convert the tropospheric slant columns into vertical columns. The OMI pixel size varies from  $24 \text{ km} \times 13 \text{ km}$  at nadir to  $140 \text{ km} \times 26 \text{ km}$  at the edges of the swath. Individual pixels were re-gridded to  $2^\circ$  latitude by  $3^\circ$  longitude, consistent with the TM5 grid. Pixels affected by the OMI row anomaly were excluded for the entire 2005–2010 period<sup>27</sup>. OMI tropospheric NO<sub>2</sub> vertical column densities show substantial sensitivity to boundary layer NO<sub>2</sub> levels and the DOMINO v2 product has been used and validated extensively<sup>28</sup>.

**MLS stratospheric O<sub>3</sub> data.** MLS measures microwave thermal emissions by scanning the Earth's atmospheric limb. The measurements have a horizontal resolution of  $\sim 200$ – $500 \text{ km}$  and are spaced  $\sim 165 \text{ km}$  apart along-track, for a total of  $\sim 3,500$  high-resolution ( $\sim 2$ – $3 \text{ km}$ ) vertical O<sub>3</sub> profiles and other species globally each day. Like TES, MLS retrievals are based on the optimal estimation technique. The single-profile precision for the 215–100 hPa O<sub>3</sub> measurements used here is  $\sim 0.04 \text{ ppmv}$ , and biases against ozonesonde measurements are  $<20\%$  (ref. 29). We show results for v2.2 MLS O<sub>3</sub> measurements, but find similar results with v3.3.

**TM5 (Tracer Model 5) chemistry–transport model.** TM5 (ref. 19) v3 simulates tracers at 34 vertical layers (surface–0.1 hPa) in a standard  $3^\circ \times 2^\circ$  (longitude/latitude) horizontal resolution using ERA-interim reanalysis meteorological fields. The chemistry scheme in TM5 is based on the Carbon-Bond Mechanism IV (CBM-IV). Stratospheric O<sub>3</sub> is constrained by assimilated total O<sub>3</sub> columns from a multi-sensor reanalysis data set<sup>22</sup>. The anthropogenic emission inventories were taken from the RETRO project, supplemented with those from the REAS (ref. 30) inventory for the Asian region. Biogenic emissions were adopted from climatologic values, either from GEIA (1990) or from the 12-year (1983–1995) average output of the ORCHIDEE model. Biomass burning emissions originated from the Global Fire Emissions Database version 2 (GFEDv2) 8-day inventories and lightning NO<sub>x</sub> emissions were parameterized. We have run the TM5 model for the period 2005–2010 with a spin-up period of one year (2004) using the relevant meteorology and available emission inventories.

**Top-down NO<sub>x</sub> emissions estimated from OMI NO<sub>2</sub>.** We extend the mass balance approach<sup>20</sup> to estimate top-down anthropogenic NO<sub>x</sub> emissions globally based on OMI NO<sub>2</sub> observations from 2005 to 2010. Surface NO<sub>x</sub> emissions are strongly related to satellite observations of tropospheric NO<sub>2</sub> columns owing to the short NO<sub>x</sub> lifetime combined with the high NO<sub>2</sub>/NO<sub>x</sub> ratio in the boundary layer. We first established the relationship between changes in surface NO<sub>x</sub> emissions and changes in tropospheric NO<sub>2</sub> columns in TM5 by computing the sensitivity of modelled tropospheric NO<sub>2</sub> columns to increases in surface NO<sub>x</sub> emissions. We then apply equation (1), effectively scaling the a priori emissions (from RETRO (ref. 31) supplemented with REAS (ref. 30) for the Asian region) with the relative trends observed by OMI, taking into account the response of NO<sub>2</sub> columns to changing NO<sub>x</sub> emissions (so-called  $\beta$ -factor). We further use a monthly scaling factor  $\gamma$  ( $\gamma = N_{\text{t,OMI}}/N_{\text{t,TM5}}$ , where  $N_{\text{t,OMI}}$  is the OMI NO<sub>2</sub> value for a specific month of year  $t$  and  $N_{\text{t,TM5}}$  is the TM5 NO<sub>2</sub> value for the same month and year) to match the TM5 tropospheric NO<sub>2</sub> columns to OMI observations. This also provides better agreement with anthropogenic NO<sub>x</sub> emissions reported in the literature<sup>22,33</sup> (discussed below) than straightforward application of the

approach in ref. 20. The monthly time-dependent top-down NO<sub>x</sub> emissions ( $E_t$ ) are calculated as:

$$E_t = E_a \left( 1 + \beta \frac{N_{\text{t,OMI}} - N_{\text{a,OMI}}}{N_{\text{a,OMI}}} \right) \gamma \quad (1)$$

where  $E_a$  represents the a priori NO<sub>x</sub> emissions (subscript  $a$  refers to a priori) in TM5 (from the 2005 nested RETRO/REAS inventories),  $\beta$  is the sensitivity, which relates an increase of 15% in surface emissions of NO<sub>x</sub> in TM5 to resulting changes in modelled tropospheric NO<sub>2</sub>,  $N_{\text{a,OMI}}$  is the OMI NO<sub>2</sub> for a specific month of 2005. We compute  $\beta$  on a monthly basis using the TM5 simulation for 2006. NO<sub>x</sub> emissions in TM5 are distributed over the days of the month and over the hours of the day following prescribed day-of-week and hour-of-day patterns. The monthly values are then used to calculate the top-down estimates ( $E_t$ ) for each month of year  $t$ . Absolute uncertainties in  $E_t$  are considerable (order 50%) because of errors in both TM5 and the OMI retrievals, but the top-down estimates capture the changes in NO<sub>x</sub> emissions associated with relevant variations in economic activity and environmental legislation in different parts of the world between 2005 and 2010. At the grid cell size of TM5, the  $\beta$ -factors describing the sensitivity of NO<sub>2</sub> columns to NO<sub>x</sub> emissions are largely driven by the seasonal cycle in photochemistry, suggesting a small role for nonlinear chemistry in modelling and explaining O<sub>3</sub> changes over time. The time series of tropospheric NO<sub>2</sub> columns observed by OMI over China and the western US used for updating the NO<sub>x</sub> emissions in TM5 are shown in Supplementary Fig. 1 (more details in Supplementary Information 2). The yearly OMI-inferred NO<sub>x</sub> emissions for China and the western US are given in Supplementary Table 1 (see Supplementary Information 2). We find a 21% increase in NO<sub>x</sub> emissions over China and a 21% reduction over the western US, comparable to reported estimates<sup>32</sup> for 2005–2010 (27% growth in Chinese NO<sub>x</sub> emissions and 23% decrease over the United States) and estimates<sup>30</sup> based on SCIAMACHY (2006–2009 increase in anthropogenic emissions of 19% over East Asia and decrease of 6% over North America), given differences in time period and area. The anthropogenic NO<sub>x</sub> emissions and their evolution are of the same order of magnitude as in ref. 33 for the given regions.

**Comparing TES tropospheric O<sub>3</sub> with TM5.** For comparisons with TES, the TM5 O<sub>3</sub> profiles from simulations with constant 2005 emissions and with 2005–2010 updated emissions were sampled along the AURA orbit track at the observation times and interpolated onto the TES pressure grid. Using partial column averages, no substantial difference is observed, whether or not the vertical sensitivity of each TES retrieval<sup>26</sup> is considered.

**TM5 simulations.** Attribution of the observed tropospheric O<sub>3</sub> values to various sources is based on three different TM5 model runs. A first (base) run for the period 2005–2010 consists of TM5 simulations using a fixed anthropogenic emission scenario based on 2005 values of the REAS inventory, resulting in tropospheric O<sub>3</sub> changes due to meteorology, long-range O<sub>3</sub> transport and STE. The second run evaluates the effect of changing anthropogenic NO<sub>x</sub> emissions on tropospheric O<sub>3</sub> by updating the emissions based on OMI NO<sub>2</sub> observations for the entire 2005–2010 period. By comparing the first and second runs the effect of the changes in NO<sub>x</sub> emissions on O<sub>3</sub> production can be assessed because the changes due to STE and meteorology cancel out. Tagged O<sub>3</sub> (O<sub>3,t</sub>) TM5 simulations allow us to quantify the contribution of STE, tracing the O<sub>3</sub> originating from the stratosphere to the troposphere, where it is subject to loss processes. A final run was set up to evaluate the influence of increased NO<sub>x</sub> emissions and O<sub>3</sub> production in China on the United States by using the OMI-based update of anthropogenic emissions worldwide, except for China, where year 2005 NO<sub>x</sub> emissions were used. Subtracting this run from the world-wide updated second run allows us to isolate the effect of growing Chinese emissions on the United States with respect to O<sub>3</sub> pollution.

**Time series analysis.** Deseasonalized monthly time series (both from satellite data and model output) are computed by subtracting from each monthly value the six-year average for that specific month.

**MLS-corrected TM5 stratosphere–troposphere O<sub>3</sub> exchange.** We constrain TM5 STE in a two-step linear regression (see also Supplementary Information 4). First we establish a relationship between the model's lower stratospheric O<sub>3</sub> (at 200–250 hPa) and the monthly averaged contribution of TM5 STE O<sub>3</sub> to the 3–9 km partial column for  $30$ – $50^\circ \text{ N}$  (see Supplementary Fig. 5). We calculate a corrected zonal mean STE contribution using 215 hPa MLS O<sub>3</sub> measurements for this same latitude band. We then diagnose the relationship between TM5 STE over China and the Western US and the zonal mean TM5 STE to derive corrected regional STE trends. Giving 50% weight to MLS retrievals, we calculate a lower but still positive contribution of STE to the TM5 short-term O<sub>3</sub> for both China and the western US, as reported in Table 1.

**Data sources.** All Aura satellite measurements are publicly available. TES O<sub>3</sub> data are available at <http://tes.jpl.nasa.gov>; OMI NO<sub>2</sub> data are available at <http://www.temis.nl/airpollution/no2.html>; MLS O<sub>3</sub> data are available at <https://mls.jpl.nasa.gov>.

**Code availability.** The TM5 model code used to generate the simulations analysed here is available at <http://tm5.sourceforge.net>.

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