Changes in US background ozone associated with the 2011 turnaround in Chinese NOx emissions

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Abstract
Assessing the effectiveness of surface ozone air quality regulations is complicated by non-local sources and inhibited by sparse in situ observational networks. Here, we harness satellite measurements of ozone and its precursors over Asia to identify the Asian contribution to background ozone over the United States (US) from 2006–2016 using a state-of-the-art chemical data assimilation system. Our results indicate that Chinese NOx emissions reached an apex in 2011, increasing daily average ozone concentrations over Los Angeles by up to 3.8 ppb at 750 hPa and 0.7 ppb at the surface in 2012 relative to 2006, with somewhat larger impacts in the high altitude regions of the Western US (assuming constant VOC emissions). Decreasing Chinese NOx emissions after 2011 contributed to a reduction of similar magnitude in Western US background ozone from 2012–2016. We also assess the impact of direct assimilation of satellite-based tropospheric ozone profile measurements over Asia and find that it substantially modifies global ozone, including US surface concentrations. These results suggest that measurements of both ozone and its precursors over Asia play a substantial role in evaluating not only local but also global air quality. The simultaneous assimilation of ozone and its precursors proffers a powerful way to constrain the vertical profile of ozone and improve understanding of ozone variations. In the future, a more extensive satellite observing system has great potential to better constrain both local pollution and background ozone globally when employed in conjunction with chemical data assimilation.

1. Introduction
Ozone is an important air pollutant and essential for understanding atmospheric chemistry. It influences the oxidation capacity of the atmosphere and tropospheric chemistry more broadly. Furthermore, short- and long-term exposure to ambient ozone and its precursors adversely affects human health [1]. Tropospheric ozone is formed by photochemical oxidation of CO and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NOx = NO + NO2). Because of its relatively long lifetime, the variability of ozone is controlled by both local and non-local sources. The ability to distinguish local and remote influences on ozone is important for air quality regulation and for assessing the co-benefits of those regulations for climate mitigation [2, 3].

Rapid ozone changes associated with remote sources have been identified using various measurements and chemical transport models [4–8]. Previous modeling studies have found that Asian sources contribute ~5 ppb to surface ozone in the Western United States (US) during springtime, with occasional enhancements as large as 15 ppb at rural sites [9, 10]. The term ‘background ozone’ refers to ozone attributed to non-local sources along with non-anthropogenic contributions from lightning, wildfires, and the stratosphere [11–13]. The US Environmental Protection Agency (EPA) defines the US background ozone as ozone that would exist in the absence of anthropogenic emissions of ozone precursors from within the US. US background ozone concentrations are highest in the Western US, and can limit states’ ability to meet the federal ozone standard in some locations [13]. Section 179B of the US Clean Air Act provides relief for locations that would meet the
standard if not for transport of ozone from international sources, but applying for this relief requires quantifying
the influence of such transport.

Surface measurements provide long-term records of surface ozone, but have limited geographic coverage
especially in developing countries [14]. Ozonesonde observations provide vertical profiles of ozone, which are
essential for capturing long-range transport associated with non-local sources in the free troposphere [15].
Nevertheless, ozonesondes do not provide a full global picture of tropospheric ozone variations. When
combined with chemical transport models (CTMs), ozone precursor emissions play an essential role in
understanding changes in local and non-local contributions to ozone [3]. However, current emission
inventories suffer from large uncertainty and substantial latency, limiting our understanding of recent
changes [16].

Satellite measurements have provided an unprecedented global picture of ozone and its precursors.
Tropospheric ozone profiles have been retrieved from the Tropospheric Emission Spectrometer (TES), which
flew on the NASA Aura satellite [17], and the Infrared Atmospheric Sounding Interferometer (IASI) [18].
Recently developed multi-spectral retrievals, for instance from a combination of radiances from TES and the
Ozone Monitoring Instrument (OMI) (TES/OMI) [19, 20], the Atmospheric Infrared Sounder (AIRS) and OMI
(AIRS/OMI) [21] and IASI and the Global Ozone Monitoring Experiment-2 (IASI/GOME-2) [22] provide
global records of tropospheric ozone with increased degrees-of-freedom (DoFs) in the troposphere and
enhanced surface sensitivity relative to measurements from individual instruments. Meanwhile, satellite
measurements of ozone precursors have been shown to substantially improve emission estimates at various
spatiotemporal scales [16, 23–25], which in turn aids understanding of the mechanisms driving formation of
local and non-local air pollutants [26]. However, it can be challenging to combine information from multiple
satellites due to their differences in overpass time, spatial representativeness, vertical sensitivity, and uncertainty.
Chemical data assimilation provides physically and chemically consistent composition fields by combining
information from multiple satellites with a model [27, 28]; the results provide improved understanding of the
relationship between precursor emissions and ozone. In addition, data assimilation can help quantify the
relative value of any existing set of measurements through observing system experiments (OSEs) [26, 29] and of
future observations through Observing System Simulation Experiments (OSSEs) [30].

In this study, we quantify changes in the Asian contribution to US background ozone from 2006–2016,
maximally leveraging satellite observations of ozone and its precursors through data assimilation in a relatively
simple setting to clearly demonstrate the impact of changing NOx emissions on ozone. We assess the impacts of
the observations on modeled surface and tropospheric ozone over both Asia and the US, including evaluation of
changes in the number of exceedances of surface ozone air quality standards associated with their assimilation.
Satellite NO2 measurements indicate that Chinese anthropogenic NOx emissions experienced a rapid increase
before 2012 and then a decrease afterwards [24, 31]. This provides us a unique opportunity to evaluate the
linearity of the ozone response to changing NOx emissions. We also discuss the potential value of current and
future satellite observations over Asia for the evaluation of local and non-local air pollutant sources when used in
conjunction with chemical data assimilation.

2. Methodology

2.1. Multi-constituent chemical data assimilation

The chemical data assimilation technique used here is based on a local ensemble transform Kalman filter
(LETKF) approach [32] that uses an ensemble forecast to estimate the background error covariance matrix
[27, 33]. This approach has been implemented within four different CTMs to construct a Multi-mOdel Multi-
cOstituent Chemical data assimilation (MOMO-Chem) framework [27]. This framework directly accounts for
model errors in transport and chemistry and quantifies uncertainties in the data assimilation analysis due to
model errors. Parts of the MOMO-Chem system have been used to produce the Tropospheric Chemistry
Reanalysis version 1 (TCR-1) and version 2 (TCR-2) products [33, 34].

In this study, we used the GEOS-Chem model within the MOMO-Chem system to evaluate NOx emission
changes and their influence on ozone. GEOS-Chem has been widely used to quantify US background ozone
[35, 36]. The GEOS-Chem model is driven by assimilated meteorological data from the NASA Global Modeling
and Assimilation Office (GMAO) Modern-Era Retrospective analysis for Research and Applications, Version 2
(MERRA-2) [37]. The adjoint model version 35 [38], which corresponds to version 9 of the forward model, with a
horizontal resolution of 2° × 2.5° and 47 vertical levels extending from the surface to 0.1 hPa, was used here as
a forward forecast model, without adjoint calculations. The a priori emission data were obtained from the
Emission Database for Global Atmospheric Research (EDGAR) version 3 inventory [39] for anthropogenic
emissions and from the Global Fire Emissions Database (GFED) version 2 inventory [40] for biomass burning
emissions. Emission data for North America were replaced with the 2008 National Emissions Inventory (NEI).
Nevertheless, the use of the state-of-the-art assimilation, based on emission changes. Data assimilation corrects biases in bottom-up emission inventories and improves understanding of long-term changes. Section 3.1. Decadal changes in NOx emissions and ozone

3. Results

3.1. Decadal changes in NOx emissions and ozone

Data assimilation corrects biases in bottom-up emission inventories and improves understanding of long-term emission changes [16, 24]. As shown in figure 1, the estimated Chinese emissions (108°E–123°E, 20°N–44°N) increased from 2006 to 2012 by about 30% and decreased about 20% from 2013 to 2015, with smaller decreases during 2015 and 2016. Interestingly, there are substantial spatial differences in the estimated positive and negative trends. These variations are attributable to the competing influences of economic development and air quality controls [31], as commonly suggested by previous studies [44–46]. For the Western US (105°W–130°W, 30°N–50°N), the estimated emissions show a rapid reduction of 21% from 2006 to 2010, a slight increase for 2011–2013, and a slow reduction from 2013 through 2016. These emission changes at regional and country scales are broadly consistent with our previous estimates using TCR-1 [24, 47] and TCR-2 [33]. In contrast, even most recent regional inventories, such as an updated NEI inventory over the US and MEIC and CAPPS, still show distinctly different emission patterns from top-down estimates [47, 48], which would provide different ozone changes. The uncertainty of these emissions is estimated to be 21% for East China and 17% for the US based on the multi-model spread of the posteriori emissions using MOMO-Chem [27]. In spite of large differences among the inventories, the choice of prior emissions provided only a limited influence on the posteriori emissions (not shown). This confirms that the OMI measurements provide sufficient constraint on NOx emissions.
The ozone analysis from GEOS-Chem has already been validated against independent observations, such as those from ozonesondes, within the MOMO-Chem framework [27]. This evaluation confirmed that data assimilation substantially reduces model errors globally. Here we show an evaluation of the GEOS-Chem ozone data assimilation analysis used in this paper against TES L2 tropospheric ozone retrievals [17] over China and the western US for 2005–2010, when Chinese NOx emissions increased rapidly. The TES retrievals have been extensively validated against ozonesonde observations [49, 50] and provide unprecedented information on global ozone changes for this early time period, including for regions without any ozonesonde observations such as China. Previous work found that the TES retrievals show a large, significant increase in tropospheric ozone at 3–9 km over eastern China (1.08%/year) for the period 2005–2010, and a smaller, insignificant increase over the western US (0.56%/year) [8]. This same work used the TM5 model to show that increases in ozone from China offset about half of the reductions in mid-tropospheric ozone over the Western US associated with US air quality regulations during this period [8]. Changes in surface ozone, however, were not quantified.

As shown in figure 2, the model control run without any data assimilation generally underestimates ozone in the free troposphere over China (108 °E–123 °E, 20 °N–44 °N) by up to 10 ppb, except during spring. The data assimilation removes most of the model negative bias, with a mean bias reduction of 37% (from −3.1 ppb to −1.9 ppb) and an increase in temporal correlation from 0.88 to 0.96. The interannual changes are also well reproduced by the reanalysis, which shows increasing trends from 2005 to 2010 of +0.33 ppb/year (p > 0.05 from the Mann-Kendall test, p-values greater than a significance level of 0.05 fail to reject the null hypothesis of no trends), consistent within the uncertainties with the +0.44 ppb/year increase (p > 0.05) in the TES observations. The model control simulation, in contrast, has only a +0.11 ppb/year increase (p > 0.05). Over the western US (105 °W–130 °W, 30 °N–50 °N), the model reproduces the observed variations in the free troposphere relatively well. However, it underestimates the springtime peak by up to 6 ppb in some years. The data assimilation reduces these model negative biases and decreases the root mean square error (RMSE) from 4.5 ppb to 2.6 ppb.

Free tropospheric ozone variations are largely controlled by NOx emissions, including the recent rapid changes during the COVID-19 pandemic [51–53], and by variations in transport of ozone from the stratosphere to the troposphere [8, 54]. Since tropospheric ozone observations were not assimilated, the improved agreement with independent TES measurements demonstrates that the assimilation provides a reasonable representation of NOx emissions and of stratospheric ozone. Nevertheless, there are many other factors affecting ozone.
variations, including precursor emissions other than NOx. While we focus on NOx in this paper, these influences need to be explored to fully address the causal mechanisms of observed ozone changes.

3.2. Impacts of Chinese NOx emission variations on ozone

3.2.1. Spatial distributions

To measure the impact of changing NOx emissions on ozone for each year during 2006-2016, two sets of model simulations were conducted using the optimized emissions obtained from the data assimilation runs. In baseline simulations, the optimized emissions for each year were used. In sensitivity calculations, the emissions at 2006 levels were used for 2007-2011, and the emissions at 2011 levels were used for 2012-2016. Differences between the baseline and sensitivity model simulations illustrate global ozone anomalies due to changing emissions. While we have assimilation results for 2005 (c.f. Figure 2), we start with 2006 in this analysis in order to examine the same number of years for both increasing and decreasing emissions (5 years each).

First, we investigated the impacts of global NOx emissions changes. Variations in global NOx emissions relative to 2006 led to differences in monthly mean ozone concentrations of up to 18 ppb at the surface and up to 20 ppb in the lower troposphere (750 hPa), with mostly negative values during 2007-2009 and mixed positive and negative values in 2010 and 2011 (figure S1 available online at stacks.iop.org/ERC/4/045003/mmedia).

Large surface ozone anomalies were found over polluted areas. At 750 hPa, the anomalies were propagated across the northern extratropics, including remote areas, through atmospheric transport. These changes in ozone reflect changes in both natural and anthropogenic sources of NOx across the globe, including biomass burning emissions, and confirm the substantial impact of NOx emissions on global background ozone levels. In addition, year-to-year ozone variations can also be driven by changes in meteorological or climate conditions [55], such as wind, temperature and humidity. They control ozone production efficiency and transport patterns, including inflows from the stratosphere [54].

The remainder of our analysis focuses on the influence of Chinese NOx emission changes on ozone. The turnover in Chinese emissions during the satellite period provides a unique opportunity to evaluate the linearity of the ozone response to both increases and decreases in NOx. As shown in figures 3 and 4, the increase in Chinese NOx emissions from 2006-2011 had substantial influence on ozone across the NH extratropics, including the Arctic region. The ozone response varies strongly with region and season. In winter over highly polluted areas, ozone near the surface was decreased due to the NOx titration effect under high NOx conditions (figure not shown). This was most obvious over northern China. During other seasons and in the free troposphere, there was a broad increase in ozone due to increased Chinese NOx emissions. In boreal spring (figure 3), the ozone anomaly was strongly propagated into the Pacific and North America via westerly winds at NH mid latitudes. The cross-Pacific transport was stronger in spring than in other seasons (not shown). The increase in Chinese NOx emissions increased monthly mean ozone at 750 hPa by up to 1.8 ppb in East Asia and up to 0.4 ppb over the Western US in 2011 relative to 2006. Meanwhile, the impacts of Chinese emission
variations and differences in meteorology reveal strong year-to-year changes in ozone, such as weaker anomalies over the Pacific and North America in 2009 than in 2008 and 2010. The ozone anomaly over Canada and the Arctic region is largest in 2010, with up to a 0.4 ppb increase.

Similarly, the reduction in Chinese NOx emissions from 2012 onward decreases ozone, with maximum reductions in 2015 over many regions, e.g., up to a 0.7 ppb reduction over the Northern Pacific and 0.4 ppb

Figure 3. Monthly mean ozone anomaly due to Chinese NOx emission changes since 2006 during 2007-2011 (left panels) and since 2011 during 2012-2016 (right panels) in April at 750 hPa.

Figure 4. Same as in figure 3, but at the surface.
reduction over the Western US at 750 hPa. The monthly mean changes in ozone are typically \(+0.2\text{–}0.5\text{ ppb}\) and \(-0.15\text{–}0.25\text{ ppb}\), respectively, over these regions for the 2013–2017 period. The interannual variability of the simulated ozone concentration driven by meteorology is typically 1–3 ppb over Northern Pacific and 0.5–15.5 ppb over the Western US (not shown), which is larger than the evaluated influence of Chinese emissions. Nevertheless, the systematic ozone anomaly seen throughout the 2013–2017 period provides evidence of a robust influence of the Chinese NOx emissions. An exception is northern China, where ozone near the surface increases, again reflecting NOx titration effect. The general spatial patterns at 750 hPa, with the largest anomalies extending from southern China to the mid-latitude Pacific Ocean and North America, are common for the positive anomalies during 2007–2011 and for the negative anomalies during 2012–2016. These results suggest that variations in NOx emissions in Asia affect background ozone levels and explain part of the observed variability over many remote regions and the Western US. Peroxyacetyl nitrate (PAN) is a long-lived reservoir species for NOx and can be transported long distances before decomposing. Figure S2 shows large anomalies in PAN in the free troposphere associated with changes in Chinese NOx emissions, highlighting the role of long-range transport of ozone precursors in remote impacts of international NOx emission changes on US background ozone.

Again, because changes in emissions other than NOx, such as those of VOCs and CO, were not considered, the actual ozone responses to Chinese anthropogenic emission changes could be broader and more complex [56]. Satellite measurements of formaldehyde (CH\(_2\)O) from OMI and GOME-2 have been used to evaluate VOC emission changes in previous studies, focusing on biogenic isoprene emissions [57–59]. The VOC emission analysis can be extended using more recent satellite CH\(_2\)O products, such as from TROPOMI [60] and OMPS, including anthropogenic VOC sources [61]. Nevertheless, evaluation of anthropogenic VOC emissions remains largely uncertain for many regions of the world [16]. Using model sensitivity calculations, changes in VOC emissions by 20% led to local monthly mean ozone concentration changes by 3 ppb over eastern China at the surface [52]. Meanwhile, MOPITT CO measurements have been used to infer \(-2\%\) per year decrease in primary CO sources in East Asia from 2005–2016, mainly attributing to source reductions in China [62]. These changes in VOC and CO emissions would amplify the impact of Chinese emission influences on the US background ozone. Further studies would be required to evaluate decadal changes in global anthropogenic VOC emissions and its impacts on ozone.

### 3.2.2. Impacts on Western US background ozone

Surface ozone over the US west coast is heavily influenced by long-range transport from China [8, 63]. Our model simulations constrained by satellite measurements show that the ozone changes over Asia associated with Chinese NOx emissions are propagated across the Pacific to the Western US. While the magnitude of the ozone anomalies decay significantly during transport, they are nevertheless non-negligible in the US at daily scales. As seen in figure 5, daily surface ozone anomalies over Los Angeles associated with Chinese NOx emissions are about 3.8 ppb higher in 2012 than in 2006 at 750 hPa and 0.7 ppb higher at the surface, with strong day-to-day and seasonal variations. The ozone anomaly is largest from spring through early summer at both levels. The seasonality is driven by changes in transport and ozone production efficiency [64]. In total, over the Western US, the number of exceedances of the US national ozone standard (70 ppb for daily maximum 8-hour concentrations: MDA8) increased by about 6% from 2006 to 2012 due to the Chinese NOx emission changes (figure S3). Because we stored 6-hourly mean ozone concentration outputs only and used them for this analysis, actual exceedance cases due to the Chinese NOx emission changes could be biased in some locations. Using 2-hourly model output, we confirmed that the use of 6-hourly averages, instead of MDA8, tend to slightly underestimate daytime maximum ozone concentrations over the Western US and overestimate it over central and eastern US, by up to 1 ppb in monthly means and 1.5 ppb in daily concentrations over polluted areas with high ozone levels. Differences in daily concentrations are as large as 5 ppb over remote areas like the oceans. A more accurate evaluation of the impact on the US standard needs to be obtained using our results as boundary conditions to a regional model that better represents the detailed spatiotemporal evolution of ozone at the relevant scales, as similarly demonstrated by previous studies [63, 65].

We also conducted model simulations in which we fully removed anthropogenic NOx emissions in China. The surface ozone anomaly due to all Chinese anthropogenic NOx emissions is about 7 ppb at 750 hPa and 4 ppb at the surface over Los Angeles (figure 5). Chinese anthropogenic NOx emissions are associated with approximately a 97% increase in the number of exceedances of the US national ozone standard (figure S2). This demonstrates the important contributions of human activity in remote countries to local air quality and the resulting human health impacts, not only in the Western US, but in any location downwind of significant ozone sources. In the western US, the remote influence is greatest at high elevation locations, consistent with previous findings [13]. The Chinese emission impact was also large over Mexico (up to 4 ppb), which increased the occurrence of the exceedances of the Mexican national ozone standard (also 70 ppb) by 6 days.
Meanwhile, the large difference in the ozone response per unit NOx emission in the two simulations (about 30% emission reductions in the upper left plot in figure 5 and 100% emission reductions in the upper right) and in the number of Western US ozone exceedances (6% versus 97%) confirms the non-linearity of ozone response, the degree of which also differs between the surface and free troposphere. This nonlinearity suggests that realistically-achievable decreases in remote emissions using current technologies will have at most a modest effect on local ozone. Anthropogenic emissions from countries other than China, such as India and Southeast Asian countries, generally had smaller influences on US background ozone than China (results not shown). Nevertheless, we find that the rapid increase in Indian emissions from 2006 to 2016 substantially increased background ozone levels over remote regions at low latitudes (within ±30°). Biomass burning emissions from tropical regions also had substantial influences on background ozone levels and the tropospheric ozone burden [66]. Better understanding of these complex effects on background ozone over the US and other countries is needed to further improve air quality control strategies.

We emphasize that the ozone response to changing emissions can be strongly model dependent. Model sensitivities are the primary drivers of differences in chemistry-climate estimates of quantities such as ozone radiative forcing [67, 68]. Our previous work has shown that the sensitivity of ozone to NOx emissions varies by a factor of 2 among the four different chemical transport models in the MOMO-Chem system, revealing fundamental differences in the representation of fast chemical and dynamical processes [27]. Thus 200% can be regarded as the possible uncertainty range in the estimated ozone response we show here. In another study assessing background ozone from multiple models and other approaches, the uncertainty in total US background ozone was estimated to be about 10 ppb for seasonal mean values and higher for individual days [13]. Future work will focus on better quantifying model-based uncertainties in US background ozone estimates. In particular, the use of a suite of modeling systems in a consistent manner would provide a more systematic assessment, as suggested by previous study [13]. Meanwhile, the ozone response showed large spatial variations. Thus, monitoring detailed spatial patterns of both ozone and its precursors regionally or globally may provide important information in understanding background ozone changes.

3.3. Impact of assimilating ozone observations over Asia

Our previous studies have shown that the simultaneous assimilation of ozone and its precursors is the most powerful way to constrain the vertical distribution of ozone for the entire troposphere within East Asia [26] and

Figure 5. Ozone anomaly (left) due to Chinese NOx emission changes since 2006 and (right) due to all Chinese NOx emissions in 2012. The upper panels show time series over Los Angeles at the surface (black) and 750 hPa (red). The lower panels show spatial distributions of maximum ozone anomaly in 2012 associated with Chinese emissions over the US at the surface in ppb.
for the globe [23]. Using the regional (RE for East Asia: 20°N-50°N, 80°E-130°E with denser spatial sampling) multi-spectral AIRS/OMI tropospheric ozone profile retrievals [21] generated for the KORUS-AQ campaign studies [26, 69], we demonstrate the impact of dense satellite measurements of tropospheric ozone profiles over Asia on ozone within the broader Northern Hemisphere during April-May 2016. The purpose here is to explore the value of such measurements in isolation using OSEs rather than to provide a best estimate of the impact of Asian ozone on remote locations. While utilizing the optimized NOx emissions and stratospheric ozone constrained by satellite measurements (c.f., sections 3.1 and 3.2), our OSEs are designed to provide insight into the ability of satellite measurements of ozone over Asia to inform global atmospheric composition in a realistic data assimilation setting. Assimilating multiple satellite tropospheric ozone measurements simultaneously from

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**Figure 6.** Spatial distributions of (a) data assimilation ozone increments by AIRS/OMI ozone data assimilation at 500 hPa at the initial data assimilation step on April 1st 2016, (b) ozone anomaly, as estimated from differences between the data assimilation run and model control run, on the 40th day after the beginning of data assimilation, and (c) maximum ozone anomaly during April-May 2016 at each grid point at 500 hPa and (d) at the surface in ppb.
the current satellite observing system would provide a more quantitative assessment of the value of individual measurements. Nevertheless, the presence of systematic biases among measurements [70] complicates such an evaluation, and thus this was not considered in our stand alone OSEs. In the future, similar observational information on the vertical distribution of tropospheric ozone could be obtained from geostationary satellites such as the Korean Geostationary Environment Monitoring Spectrometer (GEMS) if combined with a high spatial sampling, high accuracy thermal IR instrument in geostationary or even polar orbit (for once per day profiles at the overpass time).

As shown in figure 6, large negative data assimilation increments in free tropospheric ozone appear over Asia early on in the AIRS/OMI data assimilation cycle. The magnitude and structure of the data assimilation increments reflect complex factors such as the model-observation differences (departure), averaging kernel profile, retrieval uncertainty, and sampling pattern of the assimilated measurements. The positive model bias relative to the AIRS/OMI data, suggested by the negative data assimilation increments (figure 6), is opposite to the negative model bias relative to TES seen in figure 2. This reflects the large impacts of optimizing NOx emissions and stratospheric ozone concentrations in the OSEs, together with errors in the model initial condition used in the OSEs, especially for the first few assimilation cycles. In addition, any systematic biases between TES and AIRS/OMI will be reflected as disparate model biases. We focus here on the spatial propagation of the information provided by the assimilation of the AIRS/OMI measurements.

The increments are mainly limited to the free troposphere because of the maximum vertical sensitivity of AIRS/OMI to this region. The ozone anomalies, however, are carried into the lower troposphere through vertical atmospheric transport, decaying as they near the surface. After a few days, the adjustments made over East Asia begin to spread into a larger area. After 40 days, we find large negative ozone anomalies across the Northern Hemisphere, with up to 13 ppb reductions in the continental US, relative to the control model simulation without any assimilation. The maximum ozone anomaly in the Western US occurs during April-May 2016 and is about 20 ppbv at 500 hPa and 3 ppb at the surface. Over Los Angeles, it reaches 9 ppb at 500 hPa but is greatly reduced at 750 hPa and at the surface (figure S4). These results demonstrate the value of direct satellite-based ozone measurements over Asia for informing the global distribution of ozone given model biases associated with chemical production of ozone and difficulty in reproducing free tropospheric ozone over Asia [71].

However, the changes in background ozone levels associated with ozone assimilation are strongly dependent on model performance, including errors in emissions of many precursors and the impacts of stratospheric ozone. Ozone assimilation would have a smaller impact in a simulation that better represented 2016 conditions of the complex atmospheric system. Meanwhile, more comprehensive OSEs, for instance, using a combination of other satellite measurements of ozone, such as those from current polar orbiting satellites, as well as surface observations of ozone and its precursors, would provide further insights into the relative value of the Asian measurements in the current observing system. This type of OSE needs to be done in a separate study.

4. Conclusion

We investigated changes in the Asian contribution to US background ozone from 2006-2016, maximally leveraging satellite observations of ozone and its precursors over Asia. Distinguishing local and remote influences is important for air quality regulation, including relief from such regulation for pollution from international and other non-controllable sources, and also for mitigating potential climate and human health impacts [72]. The use of global NOx emission changes for 2006-2016 estimated from satellite data assimilation has a strong impact on the evolution of regional and global ozone over this period. Chinese anthropogenic NOx emissions constrained by satellite measurements exhibit an increase of nearly 30% from 2006 to 2011 and then a slightly smaller decrease from 2012 to 2016. This turnover in emissions during the satellite period provides an unique opportunity to evaluate the linearity of the ozone response, and we find that this response is of similar magnitude for both increases and decreases in NOx given the degree of interannual variability driven by meteorology and other factors. Model simulations using the optimized emissions show Western US surface ozone changes of less than 1 ppb in response to the 30% changes in Chinese NOx emissions, while the surface ozone anomaly due to all Chinese anthropogenic NOx emissions is as high as 4 ppb. Although the changes in Chinese emissions are associated with a not-insignificant 6% change in the number of Western US exceedances of the US daily ozone standard, the relatively small response of remote surface ozone to large changes in NOx highlights the challenge of reducing background ozone through realistically-achievable NOx controls in long-distance source regions.

Our results, including the large changes in global ozone associated with direct assimilation of ozone profiles over Asia, suggest that measurements of both ozone and its precursors over Asia play substantial roles in evaluation of local and global air quality. The simultaneous assimilation of ozone and its precursors is a powerful
way to constrain the vertical profile of ozone and improve understanding of ozone variations. Thus global chemical data assimilation has the potential to benefit various applications; for instance, it can provide improved boundary conditions for regional air quality forecasts. Newly available satellite data provides detailed spatial and temporal patterns for various species. Multispectral retrievals from the NASA Tropospheric Ozone and its Precursors from Earth System Sounding (TROPESS) system, including Cross-track Infrared Sounder (CrIS)/TROPOspheric Monitoring Instrument (TROPOMI), AIRS/OMI, and TES/OMI, along with IASI/GOME-2 provide ozone profiles with improved vertical sensitivity to the lower troposphere, whereas geostationary satellite measurements from GEMS, Tropospheric Emissions: Monitoring of Pollution (TEMPO), and Sentinel-4 provide hourly observations of NO₂ and other important air quality species at high spatial resolution. These observations have great potential to constrain both local pollution and global background ozone when used in conjunction with chemical data assimilation.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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