

Rising Asian ozone and implications for North American air quality

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1. What are the key challenges or questions for Earth System Science across the spectrum of basic research, applied research, applications, and/or operations in the coming decade?

The 2014 NASA workshop on Outstanding Questions in Atmospheric Composition (Final report: https://espo.nasa.gov/home/content/NASA_SMD_Workshop) identified several questions of critical importance for improving our understanding of present-day and future ozone air quality:

What are the major pollution sources in the developing world and how can we improve projections of future air quality and global composition?

How is tropospheric ozone changing globally and regionally, and what drives these long-term trends?

What controls background ozone concentrations in surface air?

These questions are particularly germane to western North America, a region impacted by the import of background ozone which is strongly influenced by ozone precursor emissions from South and east Asia. Background ozone refers to the ozone that would exist in the absence of any anthropogenic ozone precursor emissions from North America; a quantity that cannot be measured by instruments, but must instead be calculated by global scale atmospheric chemistry-transport models. Observed ozone that comes ashore along the North American west coast is referred to as baseline ozone and is the product of all upwind sources including North American emissions that have circled the globe. An influential modelling study by *Jacob et al.* [1999], written over 16 years ago, demonstrated that increasing ozone precursor emissions in Asia would have implications for western North America. They predicted that by the year 2010 increasing levels of background ozone transported from Asia across the North Pacific Ocean would offset some of the ozone reductions expected in the western USA from domestic ozone precursor emissions reductions.

In the decade following the publication of *Jacob et al.* [1999] routine baseline ozone observations along the US west coast were not available to confirm the predicted increasing impact of Asian ozone. Despite limited observations, *Cooper et al.* [2010] were finally able to compile enough observations from various platforms to demonstrate that baseline ozone had indeed increased in the free troposphere above western North America during springtime, and that the rate of increase was greater when the sampled air masses were transported directly from Asia. A recent update to the study (shown below in Figure 1) demonstrates that the positive trend has continued through 2014 at a rate comparable to the observed ozone increases at two rural high elevations sites near the US west coast. As predicted by *Jacob et al.* [1999] and confirmed by recent observational and modelling studies [*Lin et al.*, 2012; *Verstraeten*, 2015], the increase in ozone in the free troposphere and at the high elevation rural sites has occurred even though reductions in domestic ozone precursor emissions have clearly reduced the extreme urban ozone pollution episodes most detrimental to human health [*Cooper et al.*, 2014]. This phenomenon is consistent with observed ozone increases in east Asia [*IPCC*, 2013; *Cooper et al.*, 2014].

On October 1, 2015, the US EPA lowered the National Ambient Air Quality Standard for ozone from 75 ppbv to 70 ppbv. This lower standard poses ozone attainment challenges for high elevation regions of the western United States [Cooper *et al.*, 2015] due to the impact of strong and significant contributions of background ozone on US surface ozone mixing ratios [EPA, 2014ab]. Despite the compelling evidence for increasing background ozone there is no ozone monitoring network across the western United States that can report ozone observations at a temporal and spatial frequency high enough to reveal the daily quantity of ozone advected into the western USA from across the North Pacific Ocean. These observations are not even of a high enough temporal or spatial frequency to effectively evaluate the models used to estimate background ozone.

2. Why are these challenge/questions timely to address now especially with respect to readiness?

Ensemble chemistry-climate model simulations indicate that the tropospheric ozone burden will increase by 18% during the 21st century under the RCP8.5 business-as-usual global emissions scenario [Young *et al.*, 2013]. As shown in Figure 1 baseline ozone impacting western North America has increased since the 1990s, and model estimates indicate that present-day background ozone can account for 43% (Los Angeles) to 55% (Denver) of observed ozone in western US cities on high ozone days (> 60 ppbv) [EPA, 2014ab]. The US EPA is developing new policies to address the impact of background ozone on surface ozone monitors in the USA (<http://www3.epa.gov/ozonepollution/actions.html>) which may provide regulatory relief to ozone non-attainment regions. While these new policies have not yet been announced it is clear that the impact of background ozone cannot be adequately addressed if the quantity of background ozone is unknown or poorly estimated.

From both scientific and regulatory points of view, the lower ozone standard is motivating air quality control planners to seek more accurate and precise attribution of observed ozone to local, upwind and stratospheric sources of ozone to determine the degree to which domestic emissions must be reduced in order to attain that standard (http://www.csg.org/aapca_site/). Accurate quantification of background ozone under this new paradigm requires enhanced baseline ozone observations at a spatial density and temporal frequency adequate for evaluating and improving the models. Once the models can replicate baseline ozone, greater confidence can be placed on their estimates of background and locally produced ozone. Providing timely information on background ozone to air pollution control districts requires a prompt assessment of the observations and modelling required to accurately quantify the background ozone impacting the western United States.

3. Why are space-based observations fundamental to addressing these challenges/questions?

The impact of rising Asian emissions on western US surface ozone was predicted nearly 20 years ago but the successful and accurate quantification of these changes has been severely hampered by the scarcity of observations. The prospect of geostationary satellite retrievals of tropospheric ozone at high temporal and spatial resolution combined with in situ observations at high vertical resolution would finally reveal the magnitude and trends of baseline ozone in all seasons.

NASA's Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument is under development and is designed to operate on a geostationary satellite and provide high frequency retrievals of tropospheric O₃, NO₂, SO₂, H₂CO, C₂H₂O₂ and aerosols across the USA and eastern North Pacific Ocean. The temporal and spatial coverage of TEMPO will be unprecedented and the instrument's multi-spectral observations will provide sensitivity to ozone in the lowermost troposphere and will capture ozone's diurnal cycle. However, even if TEMPO is operational by 2019 it will be several years before the record is long enough to establish a trend. In the meantime observations through 2014 indicate that ozone has been increasing in the free troposphere above western North America for at least 20 years, and continuous high quality observations are required to determine if this trend continues.

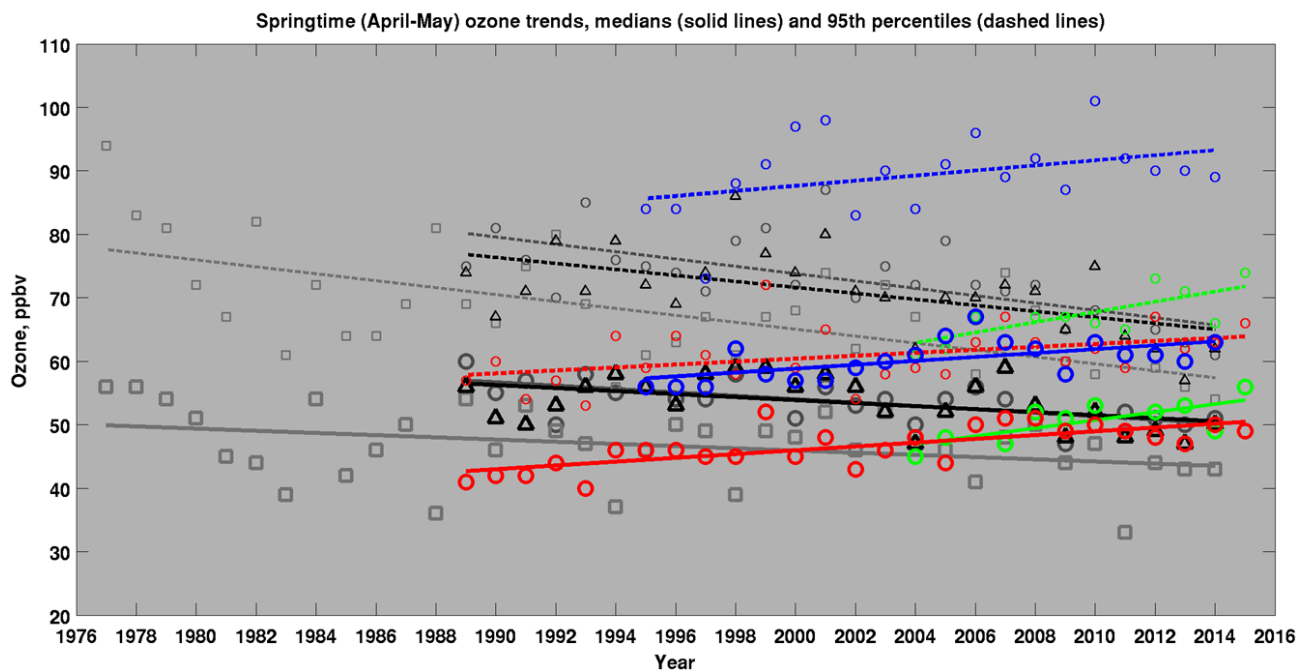
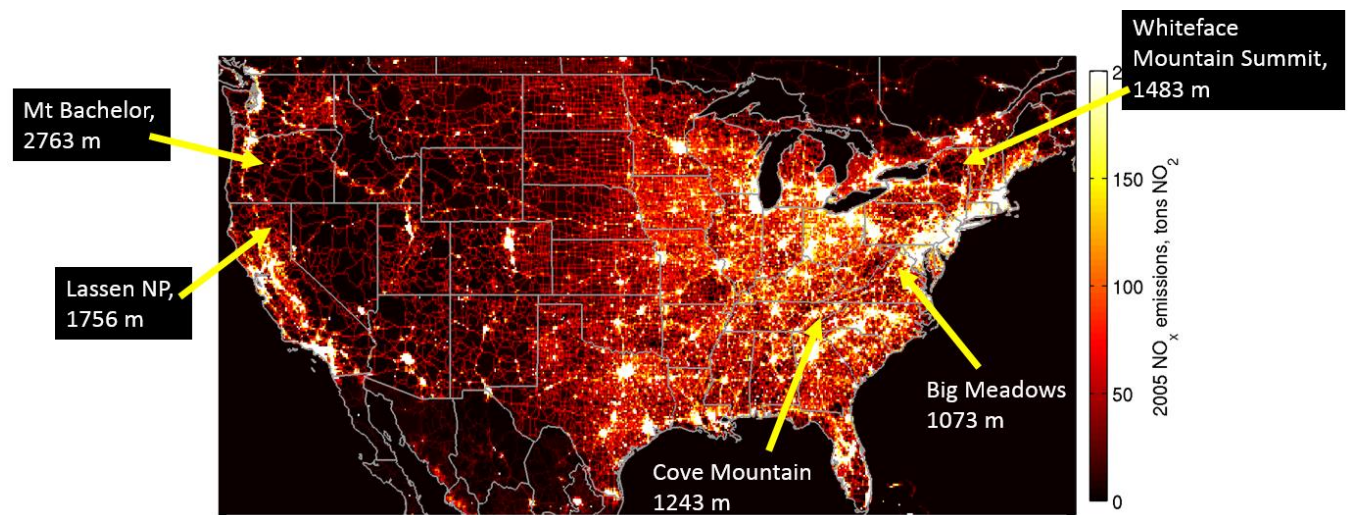
The free tropospheric ozone trend shown in Figure 1 depends on all available observations from any in situ observation platform above western North America. The frequency of these observations has diminished in recent years and continued data loss would make it impossible to continue the trend

analysis. Analogous to NASA's IceBridge Mission (http://www.nasa.gov/mission_pages/icebridge) that is using aircraft to bridge the present gap in the satellite record of polar ice observations, new in situ ozone observations can bridge the gap until TEMPO is fully operational. In a recent commentary in the journal *Science*, Cooper et al. [2015] describe several observational platforms that can provide high frequency observations of the baseline ozone flowing into western North America. Methods include routine vertical ozone profiles at multiple coastal and inland sites using balloon-borne ozonesondes, ground-based ozone lidars, or possibly commercial aircraft. Related options include augmenting the NASA/NOAA Tropospheric Ozone Lidar Network (TOLNet), the NOAA Global Greenhouse Gas Reference Network's aircraft program, or the European In-service Aircraft for a Global Observing System (IAGOS). New ozone and precursor monitoring sites at inland rural locations (especially high elevation) would be useful for gauging the descent of baseline ozone from the free troposphere into the boundary layer.

These additional observations would improve detection of inter-annual variability and long-term trends in baseline ozone. The observations would then be used to improve the coarse-scale global models needed for the routine estimation of background ozone and precursors that are subsequently down-scaled and input into the best regional air quality models covering the U.S. They would also allow improved monitoring of the changes in baseline ozone that have so far only been observed during spring, the only season with adequate observations for quantifying trends. Availability of year-round observations, especially in summer, would allow scientists to explore the possibility of ozone trends in seasons outside of spring. Importantly, these observations could be continued into the TEMPO era and utilized for instrument evaluation as well as providing useful benchmarks at higher vertical resolution.

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Light gray squares- Whiteface Mountain Summit, NY, 1483 m, daytime data. 50th% trend: -0.17 ± 0.16 ppbv, 95th% trend: -0.55 ± 0.21 ppbv
 Dark gray circles- Big Meadows, Shenandoah N.P., VA, 1073 m, daytime data. 50th% trend: -0.27 ± 0.13 ppbv, 95th% trend: -0.58 ± 0.27 ppbv
 Black triangles- Cove Mountain, TN, 1243 m, daytime data. 50th% trend: -0.24 ± 0.19 ppbv, 95th% trend: -0.47 ± 0.30 ppbv
 Red - Lassen Volcanic National Park, northern CA, 1756 m, daytime data. 50th% trend: 0.30 ± 0.12 ppbv, 95th% trend: 0.23 ± 0.21 ppbv
 Green - Mt Bachelor Observatory, OR, 2763 m, nighttime data. 50th% trend: 0.63 ± 0.41 ppbv, 95th% trend: 0.81 ± 0.50
 Blue - western N. America free troposphere, all available observations, 3.0 - 8.0 km. 50th% trend: 0.31 ± 0.21 ppbv, 95th% trend: 0.40 ± 0.48

Figure 1. Despite domestic emissions reductions, ozone continues to increase in the rural western USA. (Top) US EPA NEI 2008 NO_x emission inventory indicating major sources of anthropogenic ozone precursor emissions. Highlighted are five high-elevation rural ozone monitoring sites. (Bottom) Springtime ozone trends (April-May) of the 50th (solid lines) and 95th (dashed lines) ozone percentiles at

the five surface sites plus a 20-year record in the free troposphere above western North America. Ozone decreases in the eastern US are in response to domestic ozone precursor reductions. Even though emissions have decreased in the western US, ozone at sites strongly impacted by background ozone is increasing, likely due to the impact from South and east Asia.

Data courtesy of:

Whiteface Mountain Summit: J. Schwab, University of Albany

Big Meadows, Shenandoah National Park and Lassen Volcanic National Park: EPA CASTNET

Cove Mountain, Great Smoky Mountain National Park: US National Park Service

Mt Bachelor: Dan Jaffe, University of Washington

Free tropospheric observations: an update to Cooper et al., 2010.