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A Call for an Aloft Air Quality Monitoring Network: Need, Feasibility, and Potential Value

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Abstract

Changing precursor emission patterns in conjunction with stringent health protective air quality standards, necessitate accurate quantification of non-local contributions to ozone pollution at a location due to atmospheric transport, that by nature predominantly occurs aloft nocturnally. Concerted efforts to characterize ozone aloft on a continuous basis to quantify its contribution to ground-level concentrations however are lacking. Applying our classical understanding of air pollution dynamics to analyze variations in widespread surface-level ozone measurements, in conjunction with process-based interpretation from a comprehensive air pollution modeling system and detailed backward-sensitivity calculations that quantitatively link surface-level and aloft pollution, we show that accurate quantification of the amount of ozone in the air entrained from aloft every morning as the atmospheric boundary layer grows is the key missing component for characterizing background pollution at a location, and propose a cost-effective continuous aloft ozone measurement strategy to address critical scientific gaps in current air quality management. Continuous aloft air pollution measurements can cost-effectively be achieved through leveraging advances in sensor technology and proliferation of tall telecommunications masts. Resultant improvements in ozone distribution characterization at 400–500m altitude are estimated to be 3–4 times more effective in characterizing the surface-level daily maximum 8-hour average ozone ($DM8O_3$) than improvements from surface measurements since they directly quantify the amount of pollution imported to a location, and furnish key-missing information on processes and sources regulating background ozone and its modulation of ground-level concentrations. Since >80% of the $DM8O_3$ sensitivity to tropospheric ozone is potentially captured through measurements between 200–1200m altitude (a possible design goal for future remote sensing instrumentation), their assimilation will dramatically improve air quality forecast and health advisories.

Graphical Abstract

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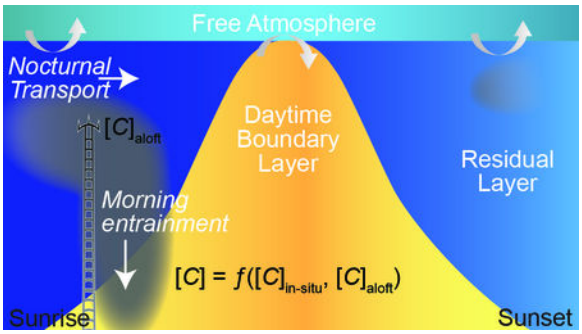

Introduction

Information on the state of the atmosphere inferred from ground-based air quality monitoring networks has played a central role in characterizing and improving air quality in many parts of the world. For instance, the Air Quality System (AQS) *(1)* which contains ambient concentration measurements of criteria air pollutants, collected by EPA, state, local, and tribal air quality agencies, has helped to identify areas where current air quality is unacceptable as well as to prevent deterioration in areas where air is relatively free of contamination. Trends in ground-level ozone (O_3) design values (i.e., the three-year average of the annual 4th highest daily maximum 8-hour average O_3 mixing ratio) show significant reductions resulting from implementation of control measures and technological advances that have reduced emissions of NO_x and volatile organic compounds (VOCs) precursor species across the U.S. *(2)*. However, as air quality standards have tightened from a 1-hour averaged concentration to an 8-hour averaged concentration with a lower threshold value, more rural and suburban regions have fallen into non-attainment *(3)*, emphasizing the need to quantify the contributions of long-range air pollutant transport to local air quality.

At most land-based locations, ground-level O_3 exhibits a characteristic, well-recognized diurnal cycle *(4)* with a typical mid-afternoon maximum and a night-time minimum that arises from diurnal variations in activity patterns of precursor emission sources, the atmospheric boundary layer, and photo-chemistry in the atmosphere. Surface deposition, titration by NO emissions, and the lack of chemical production in a shallow inversion layer which decouples O_3 at the ground from upper levels, lead to the night-time suppression of ground-level O_3. The break-up of the nocturnal inversion, entrainment of O_3-rich air from above and onset of O_3 production via photochemical reactions involving NO_x and VOC precursor species, result in increase in O_3 levels in the morning hours leading to afternoon peak values that then decline as the sun sets and chemical production declines.

Influence of Aloft Air Pollution on Surface Concentrations

The classical view of diurnal O_3 variations can alternatively be examined as a rate of change (or tendency) in the ground-level O_3, and comprehensive atmospheric chemistry-transport models can be instrumented to provide a quantitative view of the relative importance of the various atmospheric dynamical and chemical process in shaping this diurnal evolution of O_3 mixing ratios. As an example, Figure 1 presents representative summer-time diurnal

variations in median process tendencies regulating ground-level O_3 for the contiguous U.S., simulated by the Community Multiscale Air Quality (CMAQ) modeling system *(5)*. While O_3 mixing ratios typically peak around mid-day to early afternoon, the rate of increase in ground-level O_3 is highest in the morning hours and peaks around 9:00 am local time; similar variations are also seen in ground-level O_3 measurements (Figure 1, inset A). Also shown in Figure 1 are the model estimates for rate of change of O_3 due to the dominant atmospheric processes. Turbulent mixing and chemical production contribute to the accumulation of O_3 at the surface, while dry deposition is the dominant sink. The net vertical O_3 flux at the surface is dictated by the turbulent mixing responding to the evolution of the planetary boundary layer and dry deposition at the earth's surface. This vertical flux is positive in the morning, and is the dominant contributor to the net increasing O_3 tendency during the morning period, indicative of the prominent contribution of O_3 trapped in the nocturnal residual layer aloft to daytime ground-level O_3. The magnitude of the net O_3 tendency during the morning period varies spatially and is smaller at locations influenced by fresh NO traffic emissions that titrate O_3 as it is entrained from aloft (Figure 1, inset B).

In its simplest form, ground-level O_3 mixing ratios at any given location can thus be considered to be comprised of an *in-situ* component determined from local source/sink balance, and a local *background* component representing non-local O_3 that was transported to that location. Intrinsic to the concept of background concentrations is a space and time-scale so that it can be expected to vary both spatially and temporally *(6)*. In actuality the background O_3 at any location is comprised of O_3 photo-chemically produced upwind as well as a natural component that originated in the stratosphere. If background concentration for a geographic region is defined as the concentration attributable to sources (natural or anthropogenic) outside of the region, then the hemispheric O_3 background influences the regional background which in turn influences a local background. Thus, understanding the origin of ground-level O_3 at a location, requires not only an accurate description of its local sources and sinks but also an accurate attribution of the background over the different space and time scales. Though air masses are advected to a location through the course of a day, the significantly larger contribution of O_3 in the air entrained from above during the morning hours as the nocturnal inversion breaks up, can be considered to be the starting point (or background) on which the in-situ component builds to shape that day's peak O_3.

It can thus be argued that accurate characterization of this nocturnal aloft O_3 is key for quantifying the impacts of regional to continental scale transport of O_3 on ground-level O_3 mixing ratios. The recognition of such a relationship between ozone storage in the residual layer overnight and evolution of near-surface O_3 on the following day is not new. Neu et al. *(7)*using a model based on transilient turbulence theory and initialized using vertical profiles from tethered balloon soundings, estimated that 50–70% of the maximum near-surface O_3 levels were mixed down from the previous night's residual layer. Using a simple one-dimensional model, Zhang and Rao *(8)* showed that entrainment of O_3 rich air from aloft contributed to ground-level O_3 buildup. Through complementary analysis of continuous vertical measurements at a tall tower and results from a comprehensive atmospheric model, Aneja et al. *(9)* illustrated strong correlations between nocturnal O_3 in the residual layer aloft and ground-level maximum O_3 the following afternoon. In spite of this well-acknowledged vertical coupling of ground-level daytime O_3 with its nocturnal component in the aloft

residual layer, and the need to quantify it on a routine basis for areas not attaining the National Ambient Air Quality Standard (NAAQS), little coordinated effort to date has been devoted towards measuring and characterizing regional nocturnal O_3 distributions aloft *(10)*

Changing emission patterns across the globe *(11)* are now necessitating an improved characterization of long-range air pollution transport, which by nature occurs aloft. Based on Figure 1, if we assume that the rate of change of ground-level O_3 during the morning hours (8:00–9:00 am) is indicative of the influence of the aloft O_3 reservoir on ground-level O_3, then long-term trends in this morning-time O_3 tendency could be suggestive of changes in this reservoir influenced in turn by changes in local, regional, and hemispheric scale O_3 distributions. Figure 2 presents this inferred change in seasonal aloft-O_3 entrainment rates over the 1990–2010 period using hourly measurements at monitors from the Clean Air Status Trends Network (CASTNET; https://www.epa.gov/castnet). The directionality of these observed trends (Figures 2a and b) is generally consistent with previous analysis of trends of surface-level O_3 distributions *(11-13)*, with decreases at most locations in the eastern U.S. indicative of reductions in tropospheric O_3 from emission reductions, but a contrasting increasing trend at many monitoring sites in the western U.S. attributed to increasing O_3 in air masses entering the western boundaries of the U.S. Figure 2 also suggests that the spatial distribution of the O_3 reservoir in the residual layer has changed and will continue to change in response to changes in local and global emissions and any climate-driven changes in stratosphere-troposphere exchange. Also shown in Figures 2c and 2d are the corresponding changes in seasonal aloft O_3 entrainment rates inferred from multi-decadal simulations with the CMAQ model *(14)*. Comparison of model and observed trends in these entrainment rates show discrepancies both in magnitude and direction at several locations especially in the western U.S. Availability of continuous aloft O_3 measurements would help better assess the ability of current models in representing the aloft O_3 reservoir and constrain modeled entrainment rates, thereby improving confidence in model estimates of spatial and temporal variations in background O_3 pollution.

As local precursor emissions (and consequently the in-situ chemical O_3 production) change, the relative importance of the *local background* to NAAQS attainment increases. Consequently, measurements that can help assess changes in this component and enable robust evaluation of the abilities of regional and global scale model in representing it are needed to build confidence in the resulting inferences on source attribution. Satellite measurements from instruments such as the Tropospheric Emission Spectrometer (TES) are now providing new perspectives on large scale tropospheric O_3 distributions *(15)* but the relatively coarse vertical resolution (~6–7 km) is inadequate to quantify O_3 in the nocturnal residual layer. Aircraft *(16)* and balloon based measurements *(17)* have provided useful information on the vertical O_3 structure from the surface to the stratosphere but are resource intensive and impractical for establishing a long-term record of O_3 in the residual layer over regional scales. Ground based ozone lidars can provide high temporal resolution of the vertical structure of O_3 in the troposphere *(18)*, but are expensive to deploy continuously to cover a region such as the contiguous U.S.

A Cost-effective Aloft Air Pollution Monitoring Strategy for Now

Past efforts at instrumenting tall towers with O_3 monitors have yielded valuable insights on the diurnal variations in vertical O_3 distributions. Resources associated with installation and maintenance of long sampling lines have most likely been a deterrent to the long-term operation of these limited efforts. Nevertheless, analysis of limited measurements from such platforms *(9)* have provided compelling indication of the dependence of maximum ground-level O_3 on nocturnal residual layer amounts, and consequently the potential value of in-situ continuous aloft measurement of the residual layer O_3. Additional observations from emerging remote sensing platforms, higher frequency ozonesonde launches, expansion of a ground-based lidar network, and commercial aircrafts will provide valuable information on the large-scale processes dictating baseline O_3 *(19)* and in describing specific episodic cases, but may still not provide adequate constraints to accurately represent and forecast the day to day variability in ground-level O_3. Continuous O_3 measurements at an altitude of 400–500m above ground level will help directly link the variability in ground level O_3 at a location with the transported (or local background) component since a direct measurement of the amount of O_3 entrained to the surface will be available. An appropriately spaced network of such measurements could then provide the vital missing information on regional-scale nocturnal transport aloft that links surface measurements with the larger scale free-troposphere column information, as well as a key measurement to evaluate and improve the source attribution models.

Advances in air pollution monitoring technologies have recently yielded compact and low-power instrumentation that accurately measure ambient ozone. Technological advances are also now making available powerful microprocessors with low power draw and high on-board memory storage, low power cellular modems, increased cellular communications coverage, and lower cost solar panels to power such an instrument package. Together, these technology shifts are already generating a significant change in the spatial and temporal availability of surface air monitoring data *(20)*. Compact air monitoring systems, with self-contained power and wireless data communications, can allow for nearly autonomous monitoring in remote areas, such as tall towers, and circumvent the need to run long power cables or sampling lines that hindered previous efforts. Jiao et al. *(21)* demonstrated that a solar-powered UV absorbance-based compact ozone monitor was able to provide accurate readings, despite frequent power cycling and operation in an environment without controlled temperature or humidity. Similar instrumentation deployed on the top of tall building in Hong Kong was also shown to provide accurate data even after being subjected to frequent power cycling, subtropical conditions, and several major weather events *(22)*, suggesting the feasibility of an instrument package that could provide reliable measurements over a long-term in an autonomous manner.

The proliferation of tall telecommunication and broadcasting masts and towers across the world provide an existing set of platforms to possibly host such compact, autonomous, high accuracy O_3 (and in future other species) sensors. Figure 3 presents the summer-average e-folding distance *(23-24)* for night time O_3 distributions (representative of spatial scales over which O_3 mixing ratios are correlated) at the ground and an altitude of 400–500m based on O_3 distributions simulated by CMAQ. Expectedly, the e-folding distances increase with

altitude, with values of 200 km or greater at altitude of 400–500m AGL. This implies that a potential monitoring network grid of 21 × 13 (east-west x north-south) with a separation of about 200 km could provide a complete view of the spatial distribution of nocturnal O_3 in the residual layer aloft over the contiguous U.S. This possible continuous record of O_3 measurements aloft would for the first time provide a comprehensive view of nocturnal O_3 transport aloft and furnish key missing information to evolve our understanding of the processes and sources regulating background O_3, it's spatial and temporal variability, and most importantly it's modulation of ground-level O_3 and consequent impacts on human and ecological health.

Potential Benefits of Improved Aloft Characterization for Managing Air Quality

The expected impacts of accurately quantifying aloft O_3 for improved surface-level daily maximum 8-hour average O_3 ($DM8O_3$) characterization is demonstrated through detailed calculations with the CMAQ-Adjoint model *(25)* (see supplementary material: CMAQ-Adjoint). Note that since the adjoint calculations are computationally intensive, for demonstration purposes we limit the domain size to the southeastern U.S. Figure 4 illustrates the backward (adjoint) sensitivity of surface $DM8O_3$ > 70ppb (current O_3 NAAQS) to a possible 1 ppb change in characterization of O_3 at different altitudes. These simulations for the 1–14 July, 2011 period are representative of typical current summertime conditions over the southeastern U.S. As is evident from comparison of Figures 4a and 4b, $DM8O_3$ is 3–4 times more sensitive (and over larger spatial regions) to O_3 distributions at 400–500m than that at the surface, implying that measurements that help better characterize O_3 aloft would yield far greater benefit than enhancements to the surface networks. Removal of O_3 by dry deposition and titration by fresh NO_x emissions (Figure 1) result in shorter O_3 lifetime at the surface and thus the smaller sensitivity. As local NO_x emissions decline, the O_3 transported nocturnally in the residual layer to a region and entrained to the surface, becomes a more significant contributor to the $DM8O_3$. Figure 4c presents the cumulative $DM8O_3$ sensitivity over the study domain and period to O_3 at different altitudes of the model troposphere and suggests that about 80% of the $DM8O_3$ sensitivity could be captured by improved characterization of O_3 distributions between 200–1200m. Thus, future remote sensing instruments that have greatest sensitivity within this altitude range would yield maximum benefits in improving surface-level O_3 (and possibly other criteria pollutants) air quality characterization. As compact, low power draw, and reliable NO_x sensors become available additional $DM8O_3$ improvements can also be realized through improvements in aloft NO_x characterization (Figure S2).

The potential benefits of deploying an aloft air quality monitoring network based on the relatively simple idea outlined above can be tested immediately. A relatively good density of tall masts (height > 400m) used by broadcasting companies already exists in the U.S. (especially the eastern U.S., see Figure 3) and other parts of the world. With an estimated cost of ~$5,000–6,000 in materials for a rugged monitoring system that includes a UV absorbance-based O_3 instrument (~$3500), and supporting equipment (~$1500) including a microprocessor, solar panel and battery power, celluar modem for data communications, and

weather-proof enclosure, the deployment of a few hundred monitoring systems on existing masts could be achieved with a relatively modest (and low compared to other conventional vertical profiling techniques) capital investment. Public-private partnerships between Federal, State and private broadcasting organizations could be pursued to help with the deployment and maintenance of this aloft network. These continuous O_3 measurements could be assimilated into comprehensive atmospheric models in real-time to develop reliable next-day air quality forecast to guide public health warning and promote restrictive actions (e.g., agricultural burn decisions). Data from the network could also guide episodic deployment of lidars in networks such as TOLNet (http://www-air.larc.nasa.gov/missions/TOLNet) to capture the detailed three-dimensional O_3 structure and transport over large regions. Aloft measurements of air pollution in conjunction with measurements of diurnal evolution of mixing heights *(26)* planned at several Photochemical Assessment Monitoring Stations (PAMS) would help constrain and improve model representation of vertical pollutant transport and consequently surface-level background pollution. These data can also be used to guide constrained observing system simulation experiments (OSSE) to further refine the aloft network configuration.

Compared to other factors influencing environmental quality (land, water, built environment, sociodemographic), human exposure to air pollution exhibits the strongest association with all-cause and cause-specific (heart disease, cancer) mortality *(27)*. While significant improvements in air quality have occurred in the U.S., studies suggest that still more than half of the population is exposed to unhealthy levels of particle and ozone pollution *(28)*, point to adverse health effects from to exposure to O_3 and $PM_{2.5}$ concentrations below current NAAQS *(29)*, and recommend air quality standards lower than the current *(30)*. Implementation of the current NAAQS and future revisions would greatly benefit from improvements in our understanding of background pollution levels. Measurements of air pollution in the residual layer will provide key missing information on regional air pollution transport that is prominent at night, regulates the local background pollution, influences subsequent day peak values, and helps connect the variability in ground-level pollution with larger scale pollution features that descend from the free troposphere. Technological advances in air sensor development coupled with the availability of a platform in the form of tall telecommunications and broadcasting masts, provide a timely and unique opportunity for deployment of a cost effective aloft air quality monitoring network whose value can be immediately tested. Continuous measurements of air pollution aloft from such a network hold potential to provide vital information for quantifying and accounting for background air pollution when designing and implementing air quality standards.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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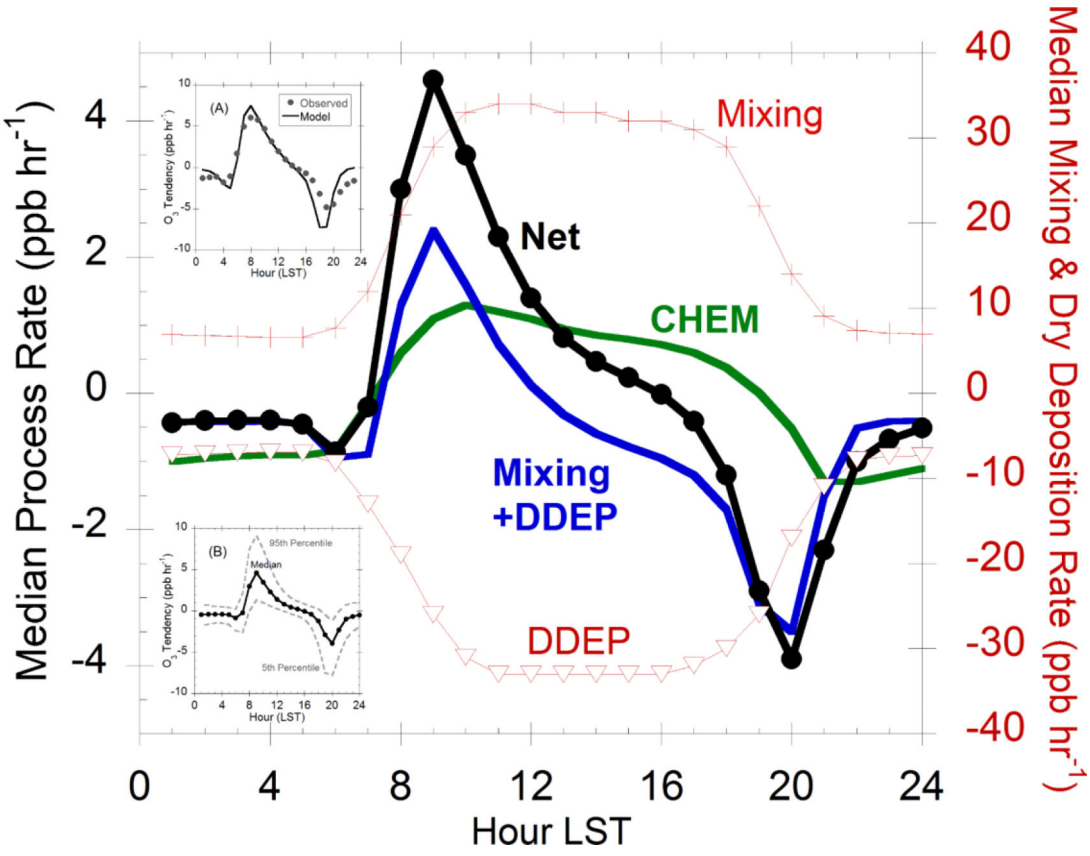

Figure 1:

Diurnal variation of CMAQ estimated O_3 process tendencies regulating variations in ground-level O_3. Values shown are median values (of July 2010 monthly average diurnal variations) across all Continental U.S. land cells. CHEM represents the net rate of change due to atmospheric chemical production and loss, DDEP is the rate of change due to dry deposition to the earth's surface, Mixing represents the rate of change due to turbulent mixing, and Net is the sum of tendencies from all modeled processes regulating O_3. Note that the values for the Mixing and DDEP terms are shown on the y-axis on the right. Inset A: Comparison of model and observed average (for July 2010 and across all AQS sites in the Continental U.S) diurnal surface O_3 tendency. Inset B: Distribution of model O_3 tendency across all land cell across the Continental U.S. The lower percentile curve represents locations influenced by fresh NO_x emissions as is evident by the large negative tendency associated with the evening traffic rush hours. Traffic related NO_x emissions in the morning rush hours result in titration of O_3 entrained from aloft and the consequent smaller magnitude of O_3 tendency at these locations.

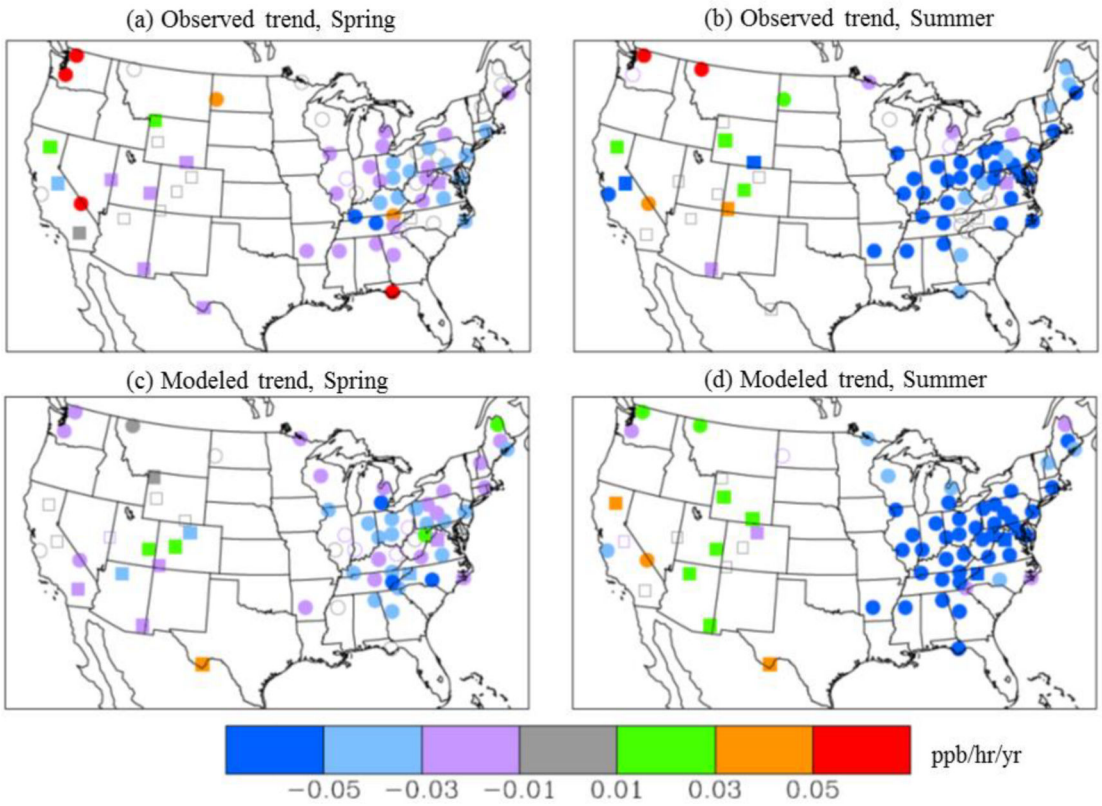

Figure 2:

Inferred changes in seasonal entrainment rates of aloft O_3 to the surface over the past two decades. Trends in observed average 8–9 am ground-level O_3 tendency in units of *ppb* hr^{-1} yr^{-1} during 1990–2010 at CASTNET monitoring locations are shown for (a) Spring (March-May) and (b) Summer (June-August). (c and d) same as (a and b) but based on model simulations. The hourly tendency at each monitor is estimated as the difference in measured O_3 at an hour and at the previous hour. Seasonal-averages of the estimated tendencies at 8:00am and 9:00 am (times when the net O_3 tendency peak) are computed for each year and the trends for the 21-year period are shown. Squares indicate high-elevation sites (altitude > 1000m ASL), while circles represent sites with altitude < 1000m ASL. Filled symbols represent locations where the estimated trends are significant at the 95% level, while open symbols are locations where they are not. These trends are only computed at CASTNET monitors that reported data for at least 11 of the 21-years. *It should be noted that both the magnitude and directionality of the trend at individual locations can change if a different subset of years within the 21-year period or a different data completeness-criteria is chosen.*

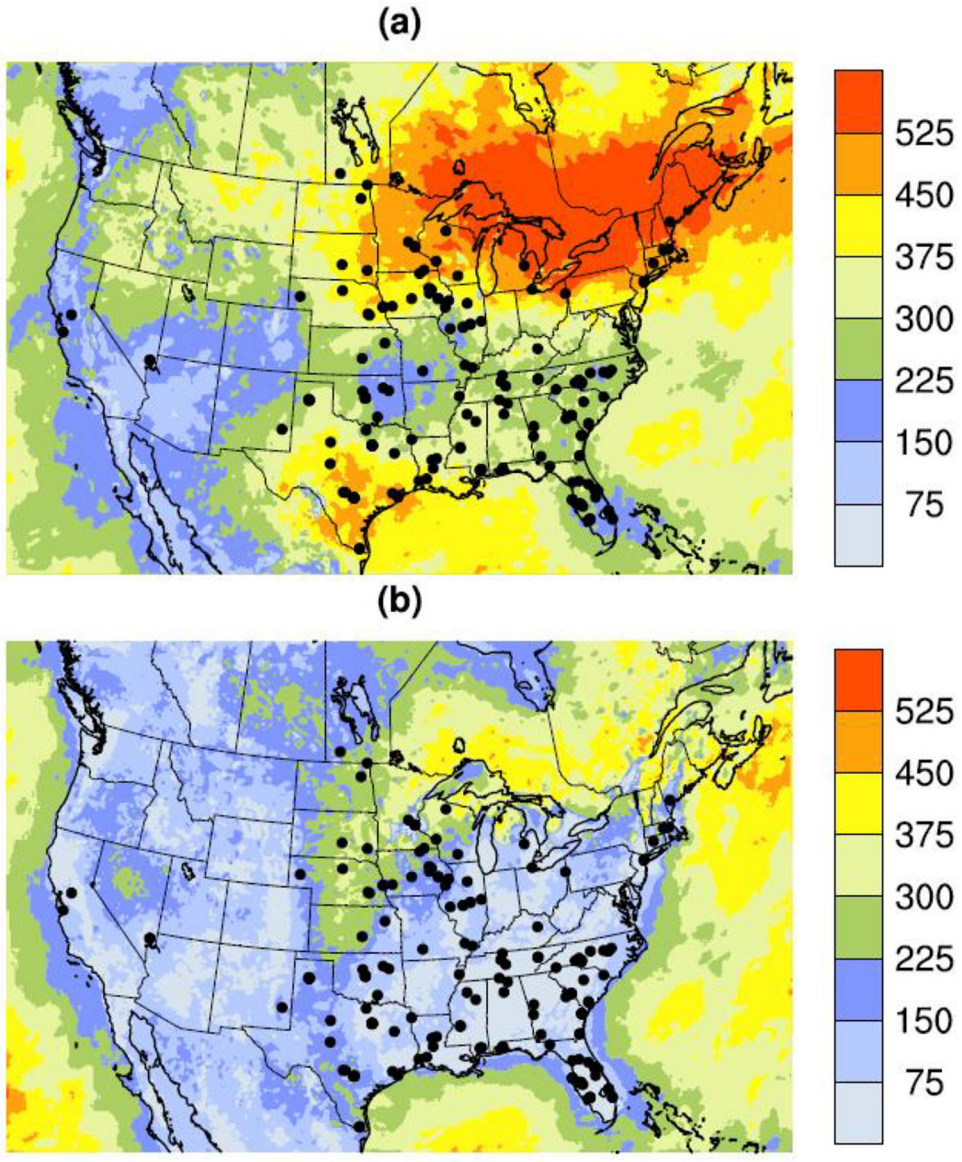

Figure 3:

May-September 2010 average e-folding distances for nocturnal O_3 (at 0700 UTC) for altitudes (a) between 400–500m AGL and (b) 10m AGL inferred from CMAQ O_3 simulations. The dots indicate locations of tall towers (>400 m) based on the list of tallest structures between 400–500m height available at: https://en.wikipedia.org/wiki/List_of_tallest_structures_%E2%80%93_400_to_500_metres. To estimate the e-folding distance, for a given model altitude, we first calculate the correlation between the synoptic component of the 0700 UTC O_3 time series at a grid point with that at each of the other grid points. For each location we then examine the relationship between this correlation and the distance between the points (Note that the relationship between correlation and distance generally follows an exponential decline). The e-folding distance at a location is the distance at which correlations in O_3 at this location with that at any remote location drops below 0.37. The e-folding distance is thus an indicator of spatial correlation in the O_3 field.

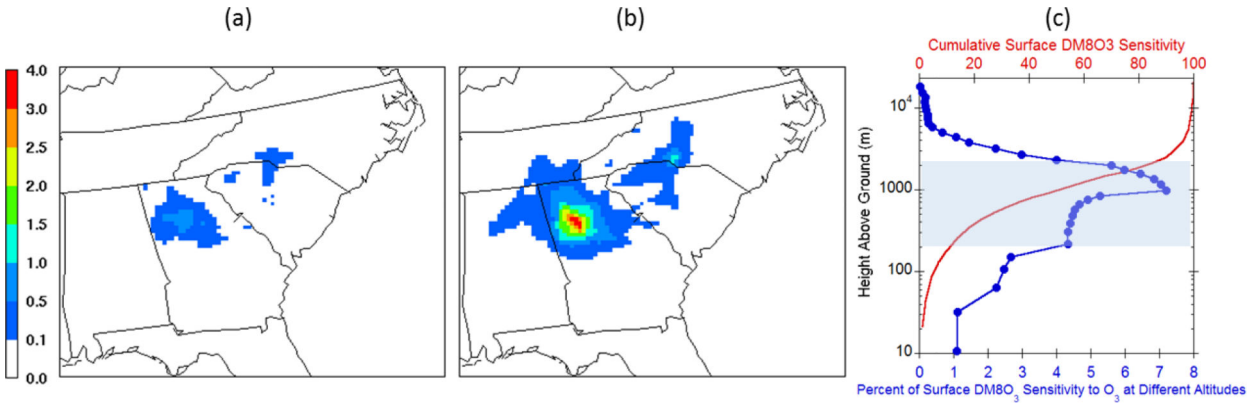

Figure 4:

Adjoint sensitivity of surface-level daily maximum 8-hour average O_3 >70ppb ($DM8O_3$) across the southeastern U.S. during 1–14 July, 2011. Estimated potential change in simulated $DM8O_3$ (in ppb) due to (a) a 1 ppb change in representation of O_3 at the surface (0–20m) and (b) a 1 ppb change in representation of O_3 at 400–500m altitude. (c) Sensitivity of $DM8O_3$ across the southeastern U.S. to O_3 at different altitudes expressed as percent of total sensitivity integrated through the model depth (in blue). Also shown in red is the cumulative sensitivity of $DM8O_3$ as function of height. Measurements characterizing O_3 distribution at 200–1200 m altitude (shaded region) would yield greatest benefit in improving $DM8O_3$ predictions via data assimilation.