Detecting and Attributing Episodic High Background Ozone Events

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A summary of recent work by AQAST members that combines satellite products, in situ measurements, and models to detect and attribute observed episodic high-ozone events to three specific background sources: wildfires, stratospheric intrusions, and Asian pollution.

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The present formulation of the National Ambient Air Quality Standard (NAAQS) for ozone (O_3) considers the fourth highest daily maximum 8-hour average concentration to determine attainment status within specific areas. In order to achieve compliance with the 8-hour O_3 NAAQS, the three-year average of this statistic must not exceed the designated level, currently 75 parts per billion (ppb). The U.S. Clean Air Act Section 319 (b)(3)(B), however, recognizes that events can episodically exceed thresholds for O_3 and other NAAQS due to influences beyond the control of domestic air agencies. Such "exceptional events" can be exempted from counting toward regulatory decisions, such as non-attainment determinations, if air agencies can demonstrate that specific components of background O_3 led to the observed exceedance (see sidebar, "What Are Background Ozone and Exceptional Events?" on page 25). The origin of individual high O_3 events thus needs to be understood and contributions from individual sources quantified. This attribution to specific sources becomes increasingly important as the O_3 NAAQS is tightened (i.e., the threshold lowered), as has been proposed on the basis of health evidence.[1]

Ozone source attribution is confounded by a lack of coincident measurements of related species (e.g., water vapor, carbon monoxide) at sufficient spatial resolution that could enable an unambiguous attribution from observations. Many background events, however, are associated with broad, synoptic-scale features evident from satellite products and resolved in models. Following are examples from recent work by AQAST members that combines satellite products, in situ measurements, and models to detect and attribute observed episodic high O_3 events to three specific background sources: wildfires, stratospheric intrusions, and Asian pollution. While the ground-based and space-based measurements are usually provided on a daily basis, the in situ aircraft and sonde measurements in the examples below are generally not routinely available.

Wildfires

In early July 2002, lightning sparked several fires in central Quebec that consumed approximately 2.5×10^9 m^2 of forest. A massive smoke plume, visible from space (see Figure 1) was swept by the meteorological conditions to the heavily populated areas of the U.S. East Coast.[2] Coincident with the arrival of the smoke was an air pollution event in which ambient monitors in Maryland exceeded the O_3 NAAQS. Was the fire to blame?

Fires generate aerosols and O_3 precursor gases such as carbon monoxide (CO) and volatile organic compounds (VOCs), but not necessarily nitrogen oxides (NO_x) needed alongside the VOCs to produce O_3, and thus, do not always lead to substantial O_3 production.[3] Measurements from aircraft (see Figure 2) revealed a peak ozone mixing ratio over Maryland of nearly 160 ppb, more than twice the current 75-ppb ozone NAAQS threshold, coinciding with enhanced fire effluents (aerosols and CO), but not U.S. anthropogenic emissions (e.g., sulfur dioxide).

These in situ measurements, combined with numerical simulation and lidar records, demonstrated that the O_3 pollution layer generated by the fires mixed down to the surface, leading to the observed ground-level O_3 above the NAAQS. A similar approach can be used to identify high fine particulate matter ($PM_{2.5}$) events resulting from wildfires. On the basis of this integrated analysis of in situ measurements, satellite products, and model data, the Maryland Department of the Environment

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Figure 1. MODIS (Moderate Resolution Imaging Spectrometer) visible image (July 7, 2002) of the smoke plume that was advected from Quebec (under the influence of a low pressure system over the Maritime provinces) and fanned out over the U.S. East Coast. Red dots indicate the locations of active fires.[2a]

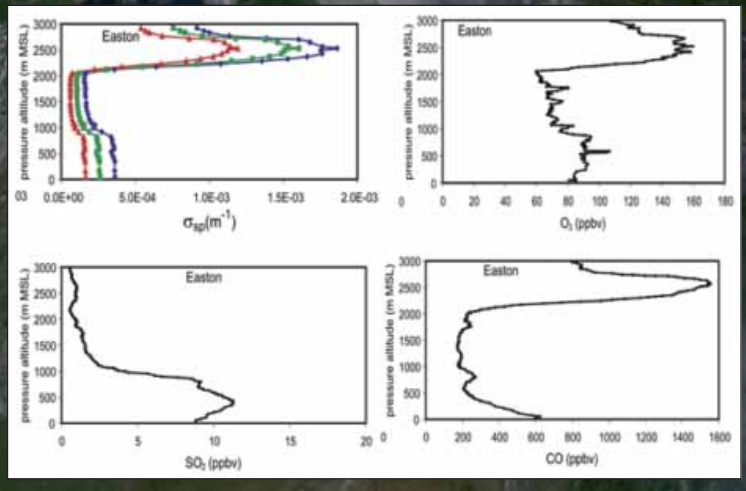

Figure 2. Measurements of pollutant abundances by altitude on July 8, 2002, over Easton, Maryland by University of Maryland and NASA investigators from a small research aircraft flown through the smoke plume and its surroundings. Shown are aerosol scattering, a proxy for $PM_{2.5}$ concentration, at three different wavelengths: 450 nm (blue), 550 nm (green), and 700 nm (red) (upper left); O_3 (upper right); sulfur dioxide (SO_2), indicative of regional anthropogenic pollution, especially coal combustion (lower left); and carbon monoxide (CO; bottom right). The smoke plume from the forest fires in Quebec (Figure 1) is evident at ~2.5 km altitude with enhanced aerosol, O_3, and CO but not SO_2. For details see Ref[2a] and Ref[2b].

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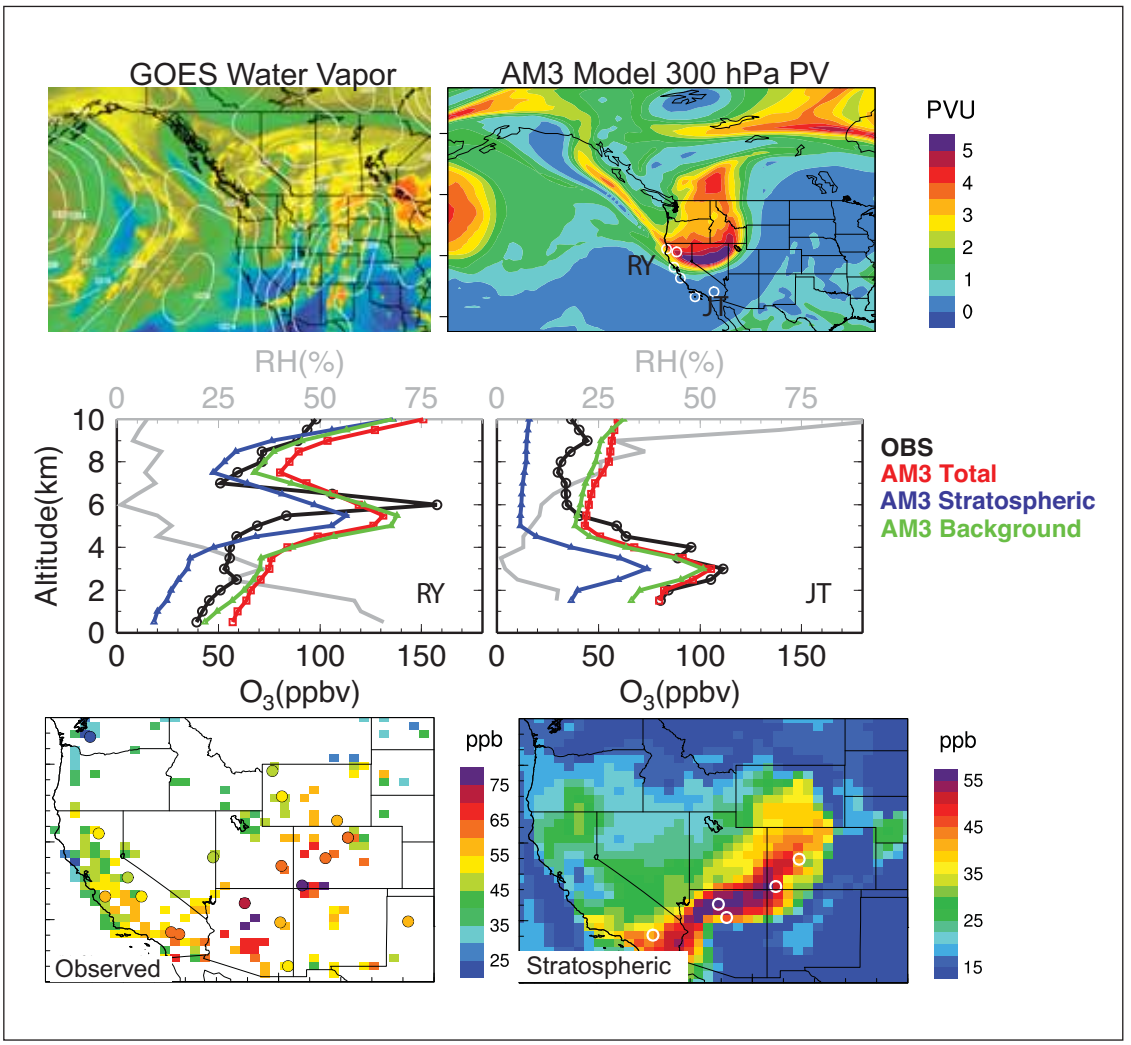

Figure 3. Example of integrating satellite, in situ, and model data to demonstrate the role of a stratospheric intrusion in enhancing surface O_3 over the western United States.

The GOES water vapor images show dry air in the mid-to-upper troposphere (blue colors in upper left; contours show sea level pressure) on May 28, 2010, coincident with a tongue of enhanced potential vorticity (PV), a dynamical marker for stratospheric air, simulated by the AM3 model (upper right). O_3 measured by balloon sondes on May 28, 2010 (black line in middle panels), launched at the locations indicated in the upper right panel, show an enhanced O_3 layer at ~6km in northern California (Point Reyes; RY) and another in the lower troposphere (~3 km) in southern California (Joshua Tree; JT) associated with dry air (grey line, relative humidity). The AM3 model (red line) attributes this high O_3 layer to North American Background (NAB) O_3 (green line), predominantly due to stratospheric influence (blue line; using approach described in Ref[4]). Ground-based monitors record a surface O_3 enhancement on May 29, 2010, extending southwest-northeast over Arizona and Colorado (bottom left) in the same region where the AM3 model simulates elevated stratospheric O_3 levels (bottom right). Adapted from Ref[4].

requested, and the U.S. Environmental Protection Agency (EPA) granted, "exceptional event" status during this episode for both O_3 and $PM_{2.5}$.

Stratospheric Intrusions

Thirteen stratospheric intrusions were found to contribute to raising western U.S. surface O_3 levels during April through June of 2010 (see Table 1).[4] Figure 3 shows one of these events, diagnosed using the GFDL AM3 model[5] and a suite of observations. On May 28, 2010, space-based observations of upper tropospheric water vapor show dry air where the GFDL AM3 model simulates high potential vorticity (PV; a marker for stratospheric air). The model and balloon sonde measurements show layers with enhanced O_3 descending in altitude southward along the coast of California. These enhancements are attributed by the model to stratospheric influence, consistent with measured dry air (low relative humidity).

A model simulation of North American Background (NAB) O_3 (see sidebar) indicates that the observed enhancements are not mainly associated with domestic O_3 pollution. The following day (May 29, 2010), the model indicates a surface influence from stratospheric O_3 coincident with observed regional surface O_3 enhancements. In light of events such as those demonstrated in Figure 3, EPA recently formed a stratospheric O_3 intrusion (SI) working group consisting of federal government scientists and air quality managers, with input from state and local agencies and academia. One of their foci is to provide satellite-based support for SI forecasting and related exceptional event analysis.

The Infusing satellite Data into Environmental Applications – International (IDEA-I; http://cimss.ssec.wisc.edu/imapp/ideai_v1.0.shtml) software package is being adapted to provide satellite based

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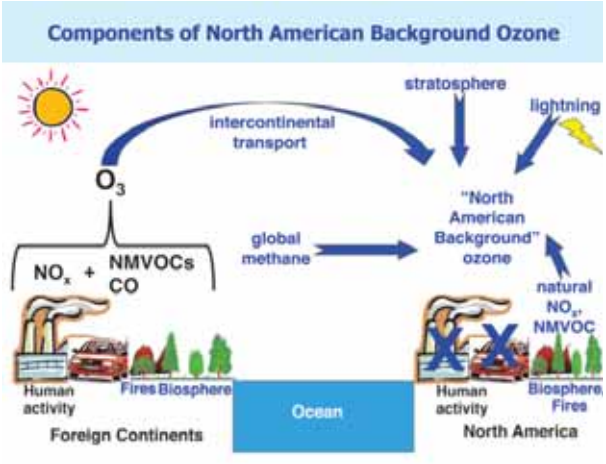
SI forecasting capabilities. IDEA-I is an open source, globally configurable version of Infusing satellite Data into Environmental air quality Applications (IDEA)[6] that provides a satellite-based aerosol forecasting, visualization, and data synthesis tool. An expansion of IDEA-I includes SI trajectory forecasts initialized using real-time, space-based measurements of O_3, temperature, and water vapor to identify signatures of stratospheric air (dry air with high O_3) within the troposphere. IDEA-I SI forecasts use measurements from several instruments aboard satellites: the Atmospheric Infrared Sounder (AIRS), the Infrared Atmospheric Sounding Interferometer (IASI), and the Cross-track Infrared Sounder (CrIS; http://cimss.ssec.wisc.edu/cspp/ uwhsrtv_edr_v1.2.shtml). When potential SI profiles are identified (based on three criteria: pressures below 500 hPa, O_3 above 80 ppb and dew point depressions of more than -15 °C), forward trajectories are initialized and used to predict

What Are Background Ozone and Exceptional Events?

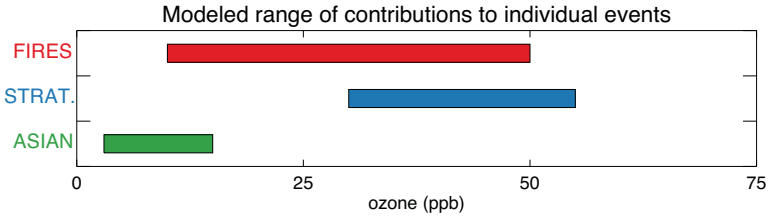
North American Background (NAB) O_3 is the concentration of O_3 in surface air that would occur in the absence of North American anthropogenic emissions.[11] NAB O_3 thus includes O_3 produced from natural nitrogen oxides (NO_x) and non-methane volatile organic compound (NMVOC) precursors, from foreign anthropogenic precursors, from global methane (including U.S. methane emissions which are not regulated), and transported from the stratosphere (see schematic). By definition, estimating background relies on models.

Ozone concentrations measured at remote sites, termed "baseline O_3" by a recent National Research Council report,[12] provide important constraints on model NAB estimates. NAB O_3 levels vary with altitude, season, and synoptic conditions, with the highest levels generally occurring over the high-altitude western United States during spring.[13] The figure below shows model-based ranges for maximum episodic contributions from fires,[14] stratospheric intrusions,[4] and Asian pollution[10,15] to high O_3 events in U.S. surface air. These model estimates motivate a need for accurate observational constraints to attribute unambiguously observed high O_3 events to sources other than North American anthropogenic emissions.

Such attribution could qualify the event for "exceptional event" status under the U.S. Clean Air Act. An exceptional event is a NAAQS exceedance that would not have occurred but for the influence of sources beyond the control of U.S. air agencies, and can be exempted from the dataset used for regulatory determinations, such as classifying a region as non-attainment.



Sources of North American Background (NAB) O_3 (blue labels and arrows). The blue Xs over North American human activities leading to O_3 precursor emissions indicate that those sources are not included in the definition of NAB O_3.



Range of contributions from components of North American Background (NAB) O_3 estimated with regional or global atmospheric chemistry models to individual high O_3 episodes in surface air. Shown are estimates for wildfires (FIRES),[14] stratospheric intrusions (STRAT.),[4] and Asian pollution (ASIAN).[10, 15] These components add to local and regional pollution, and can contribute to total surface ozone exceeding the current 75 ppb threshold for the U.S. O_3 NAAQS.

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Table 1. Stratospheric ozone intrusion events over the western United States from April–June 2010, adapted from Reference 4.[4] The event shown in Figure 3 is highlighted in bold.

Event Date(s)	Major surface impact regions	Peak daily maximum 8-hr average O_3	Number of exceedances at ground sites	Type of observations used to detect the intrusion
Apr. 7	Colorado, New Mexico	71	-	satellite
Apr. 9	Wyoming	75	1	satellite
Apr. 12–15	Four Corners Region	86	13	satellite
Apr. 21–23	Colorado, New Mexico	72	-	satellite
Apr. 28–29	Colorado, Wyoming	69	-	satellite
May 11–13	Arizona, New Mexico, W. Texas	74	-	satellite, lidar, ozonesondes,
May 18–21	Wyoming	74	-	satellite, ozonesondes
May 22–24	Colorado, New Mexico	79	4	satellite, lidar, ozonesondes
May 27–29	**Arizona, California, Colorado**	**82**	**5**	**satellite, ozonesondes**
Jun. 7–8	Idaho, Utah, Wyoming	76	3	satellite, ozonesondes
Jun. 9–14	California, Utah, Spread in Southwest	73	-	satellite, ozonesondes
Jun. 16–17	Colorado	67	-	satellite, ozonesondes
Jun. 22–23	Colorado	77	1	satellite

whether these stratospheric air masses are likely to impact the surface over the next 48 hours.

Figure 4 illustrates the IDEA-I SI trajectory forecasts along with model simulations and in situ measurements. These images provide corroborating evidence to demonstrate that a deep stratospheric intrusion led to a surface O_3 event above the NAAQS threshold. Figure 4a shows an IDEA-I SI forecast indicating downward transport of high O_3 air over northeastern Wyoming and Montana on June 6, 2012, when the Thunder Basin monitor in northeastern Wyoming reached 100 ppb at local noon. Enhanced O_3 (above 80 ppb) and PV (above 1.5 PVU) over central California on June 5, 2012, from the Real-time Air Quality Modeling System (RAQMS)[7] indicates stratospheric air extending down into the troposphere along the northern flank of a relatively strong (45 ms^{-1}) jet at 120°W (see Figure 4b). Elevated O_3 levels were also measured by the NASA Ames AJAX project as the aircraft[8] intersected the analyzed SI event (see Figure 4c). This analysis is included in a submitted exceptional event demonstration package (by Wyoming Department of Environmental Quality/ Air Quality Division to EPA; http://deq.state.wy.us/aqd/ExceptionalEvents/June_6_2012ThunderBasin/June_6_2012_SI_Package.pdf).

Asian Pollution

A comprehensive review of Asian pollution impacts on North American surface O_3 was recently published.[9] Figure 5 demonstrates a new approach to identifying days with the highest potential for importing Asian pollution into western United States surface air.[10] Daily CO columns from the AIRS instrument aboard the Aqua satellite are correlated with a time series of Asian O_3 pollution estimated with the GFDL AM3 model, sampled at Grand Canyon National Park, Arizona, and lagged by two days. The region over the eastern North Pacific Ocean with higher correlations implies that the AIRS CO columns are generally enhanced there two days before the model simulates Asian pollution reaching Grand Canyon National Park. While the correlations are not sufficiently strong to use directly as a predictor, they imply there is useful information in the space-based observations. The region identified here could be used to initialize the IDEA-I model (as for the stratospheric intrusion application) and forecast Asian pollution plumes reaching U.S. surface air.

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Figure 4. Example analysis used to document a stratospheric intrusion that led to a high-ozone event in excess of the U.S. NAAQS threshold and included in an exceptional event demonstration package.

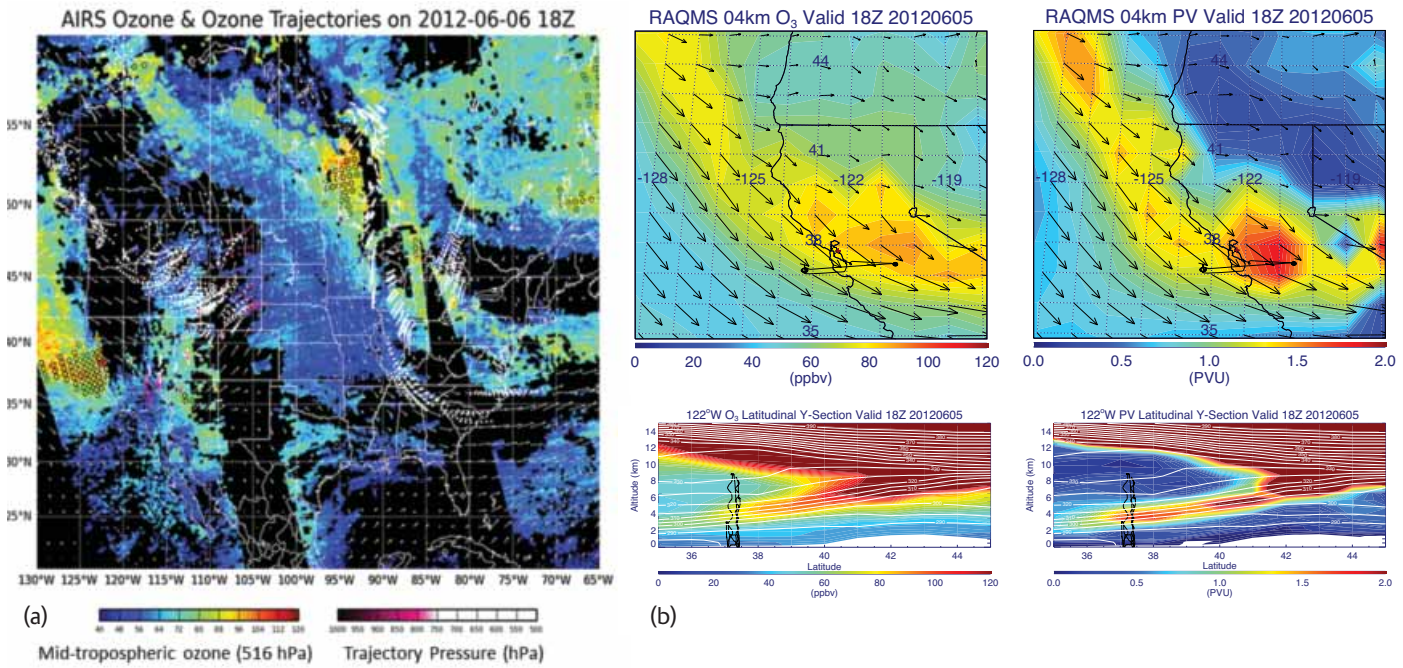

Figure 4a. IDEA-I SI forecast (magenta and white trajectories over Wyoming, Idaho, and Montana) valid at 18Z (11:00 a.m. MST) on June 6, 2012, initialized with mid-tropospheric O_3 measured by the Atmospheric Infrared Sounder (AIRS) instrument aboard the Aqua satellite on June 4, 2012. The IDEA-I SI trajectories are initialized in regions of high O_3 (open circles) off the coast of California and show the downward transport (magenta indicates high trajectory pressures near the surface) of the high O_3 air over northeastern Wyoming and Montana. The initial mid-tropospheric O_3 (516 hPa) observed with the AIRS instrument is also shown (blue-to-red color scale) along with mid-tropospheric winds (500 hPa; white arrows) from the NOAA Global Forecasting System (GFS), which are used to predict the trajectory movement. Note that the AIRS data indicate high O_3 over Canada (north of Minnesota) and trajectories were initialized there, but they never made it into the boundary layer (white trajectories over Tennessee and South Carolina).

Figure 4b. Maps at 4 km altitude (top) with wind vectors (black arrows) and cross sections at 120° W (bottom) of O_3 (ppb, left) and potential vorticity (PV; PVU, right) on June 5, 2012, at 1800 UTC from the Real-time Air Quality Modeling System (RAQMS) analyses.[7] RAQMS stratospheric O_3 analyses are constrained through assimilation of near-real-time (NRT) O_3 profiles from satellite instruments: the Microwave Limb Sounder (MLS)[16] above 50 hPa and NRT cloud cleared total column O_3 from the Ozone Monitoring Instrument (OMI),[17] both aboard the Aura satellite. Also shown is the AJAX flight track (black; measurements shown in Figure 4c). Note: the tropopause fold indicated by the tongue of relatively strong PV and high O_3 extending from the lower stratosphere into the mid-troposphere (lower panels). Reproduced from Ref[8].

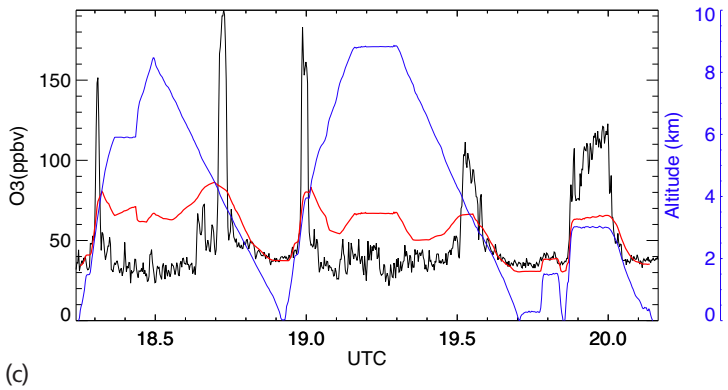

Figure 4c. Time series of observed in situ (black) and RAQMS analyzed (red; see Figure 4b) O_3 (ppb) along part of the Alpha Jet Atmospheric eXperiment (AJAX) Flight 47 track on June 5 2012. The altitude (km) of the aircraft is shown in blue. Reproduced from Ref[8].

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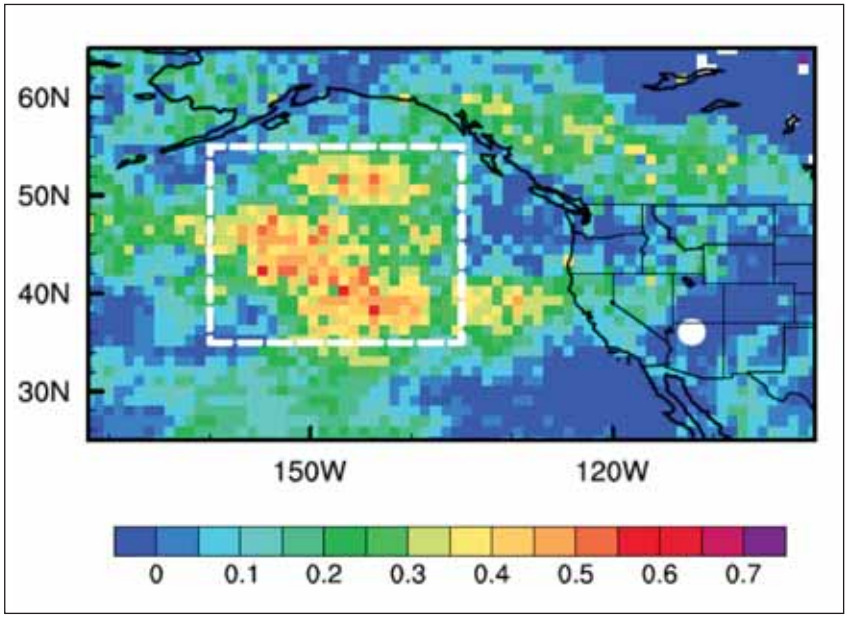

Figure 5. Correlation coefficients (r) at each 1°x1° grid box of May–June 2010 daily CO columns measured by the AIRS instrument on the Aqua satellite and model (AM3) estimated Asian anthropogenic enhancements to daily maximum 8-hour average O_3 at Grand Canyon National Park (white filled circle), Arizona, with a two-day time lag. The rectangle indicates the region where AIRS CO can be used to derive a space-spaced indicator of Asian influence on surface O_3 in the western U.S. Asian O_3 pollution is estimated as the difference between a standard AM3 model simulation and one with Asian anthropogenic emissions set to zero. Adapted from Ref[10].

Summary

The examples above from NASA AQAST members combine in situ and remotely sensed data with numerical simulations to attribute O_3 events measured at ground monitoring sites to three specific sources of background O_3: wildfires, stratospheric intrusions, and Asian pollution. Air agencies can use these integrated analyses to support "exceptional event" claims for situations where multiple lines of evidence indicate that the event would not have occurred but for the influence of sources outside of their control. NASA AQAST is working to make these tools and analysis techniques publicly available. **em**

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