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# Coordinated profiling of stratospheric intrusions and transported pollution by the Tropospheric Ozone Lidar Network (TOLNet) and NASA Alpha Jet experiment (AJAX): Observations and comparison to HYSPLIT, RAQMS, and FLEXPART

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## ABSTRACT

Ground-based lidars and ozonesondes belonging to the NASA-supported Tropospheric Ozone Lidar Network (TOLNet) are used in conjunction with the NASA Alpha Jet Atmospheric eXperiment (AJAX) to investigate the transport of stratospheric ozone and entrained pollution into the lower troposphere above the United States on May 24–25, 2013. TOLNet and AJAX measurements made in California, Nevada, and Alabama are compared to tropospheric ozone retrievals from the Atmospheric Infrared Sounder (AIRS), to back trajectories from the NOAA Air Resources Laboratory (ARL) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model, and to analyses from the NOAA/NESDIS Real-time Air Quality Modeling System (RAQMS) and FLEXPART particle dispersion model. The measurements and model analyses show much deeper descent of ozone-rich upper tropospheric/lower stratospheric air above the Desert Southwest than above the Southeast, and comparisons to surface measurements from regulatory monitors reporting to the U.S. EPA Air Quality System (AQS) suggest that there was a much greater surface impact in the Southwest including exceedances of the 2008 National Ambient Air Quality Standard (NAAQS) of 0.075 ppm in both Southern California and Nevada. Our analysis demonstrates the potential benefits to be gained by supplementing the existing surface ozone network with coordinated upper air observations by TOLNet.

## 1. Introduction

It is well established that even moderate concentrations of ozone ($O_3$) can harm human health (EPA, 2013) and impair plant growth and productivity (EPA, 2013; Lefohn et al., 1988). Ozone was accordingly designated a criteria air pollutant by the 1970 U.S. Clean Air Act (CAA), and the U.S. Environmental Protection Agency (EPA) established National Ambient Air Quality Standards (NAAQS) to protect human health and welfare from its adverse effects. The CAA requires that the NAAQS for ozone and other criteria pollutants be periodically reviewed and adjusted, if necessary, to provide an adequate margin of safety for the public. The most recent such review led to the lowering of the ozone NAAQS from the value of 0.075 parts-per-million (ppm) for the 3-yr average of the 4th highest maximum daily 8-h average (MDA8) concentration set in 2008 to a value of 0.070 ppm in October of 2015 (U.S. Environmental Protection Agency, 2015).

Ozone is a secondary pollutant formed through photochemical reactions between nitrogen oxides (NOx) and volatile organic compounds (VOCs), and efforts to control ambient concentrations have sought to regulate anthropogenic emissions of these precursors. Ozone also has

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significant background concentrations, however, that lie outside local control and complicate regulatory efforts. Background ozone is derived from NOx and VOCs emitted from distant anthropogenic sources, as well as from natural sources, such as soils, vegetation, lightning, wildfires, and from naturally formed ozone transported downward from the lower stratosphere (EPA, 2014).

Most stratosphere-to-troposphere transport (STT) at midlatitudes occurs through the formation of tropopause folds (Danielsen, 1968), tongues of upper tropospheric and lower stratospheric (UT/LS) air (Wernli and Davies, 1997) extruded downward into the free troposphere beneath the jet stream circulating around extratropical cyclones. Tropopause folds can occur year-round, but are most frequent during fall, winter, and spring in the Northern Hemisphere (Elbern et al., 1998) and probably form during the life cycle of most midlatitude cyclones (Johnson and Viezee, 1981). The majority are dissipated in the middle and upper troposphere by turbulence (Shapiro, 1980), convection (Cho et al., 1999; Langford and Reid, 1998), or breaking gravity waves (Langford et al., 1996), or are irreversibly stretched into streamer-like structures (Appenzeller and Davies, 1992) that slowly become part of the tropospheric background (Bithell et al., 2000). Those that reach the lower troposphere are usually highly diluted (Trickl et al., 2014), but on rare occasions, deep tropopause folds can cause large spikes in surface ozone (Attmannspacher and Hartmannsgruber, 1973; Davies and Schuepbach, 1994; Haagenson et al., 1981; Lamb, 1977; Stohl et al., 2000; Viezee et al., 1983). A growing number of studies (Hess et al., 2015; James et al., 2003; Skerlak et al., 2014; Wernli and Bourqui, 2002) have shown that the West Coast of North America is one of the preferred locations for deep tropopause folds (Bourqui and Trepanier, 2010), which can also entrain wildfire plumes (Brioude et al., 2007) or anthropogenic pollution transported across the Pacific Ocean from Asia during their descent (Cooper et al., 2004; Lin et al., 2012) and carry these contaminants toward the surface.

Deep STT episodes have been implicated in exceedances of the ozone NAAQS in the western U.S. (Kaldunski et al., 2017; Langford et al., 2009, 2015), and Section 319b of the U.S. Clean Air Act establishes a mechanism known as the Exceptional Events Rule (EER) to exclude those exceedances caused by stratospheric intrusions, wildfires (Jaffe et al., 2013), or other natural events from regulatory consideration (U.S. Environmental Protection Agency, 2007; 2016). Likewise, Section 179B provides a mechanism to exclude exceedances caused by intercontinental transport of pollution. It has proven challenging for state, tribal, and local regulatory agencies to implement these rules, however, in large part because of the difficulty in quantifying or even identifying the impact of STT and long-range transport (LRT) on surface ozone using only regulatory surface monitors, and only one ozone exceptional events report has been accepted by the EPA to date (Kaldunski et al., 2017; State of Wyoming, 2013). One reason for this difficulty is the sparsity of ground-based ozone monitors in the western U.S. (Gustin et al., 2015) where the impact of STT on surface ozone appears to be greatest (Lefohn et al., 2014, 2011, 2012; Lin et al., 2012). Another is the fact that stratospheric intrusions and associated pollution transport events do not always produce easily flagged spikes in surface $O_3$, but instead may cause the concentrations to increase slowly as the descending air is entrained into the mixed layer (Kunz and Speth, 1997; Langford et al., 2017; Viezee et al., 1983). Even these “unexceptional” events can cause exceedances of the 2015 NAAQS if they add 10 to 20 parts-per-billion by volume (ppbv) to the typical $O_3$ concentrations of 50–60 ppbv found in the southwestern U.S. during late spring (Fiore et al., 2014).

The most serious obstacle preventing state and local regulatory agencies from distinguishing the influence of STT and long-range transport on surface ozone is a lack of knowledge about the ozone concentrations above the surface (Cooper et al., 2015). Ozonesondes are routinely launched only about once per week from a small number of locations in the U.S. (Newchurch et al., 2003) and ozone retrievals using current space-based sensors have limited sensitivity and vertical resolution in the lower troposphere (Duncan et al., 2014; Zoogman et al., 2011). These observational deficiencies also make it difficult to evaluate the performance of satellite retrievals and the regional and global chemical transport models (Fiore et al., 2014; Lin et al., 2012; Zhang et al., 2011, 2014) that could potentially be used to quantify the impacts of STT on surface ozone for EER purposes.

The dearth of vertical ozone profiling in North America motivated the creation of the NASA-sponsored Tropospheric Ozone Lidar Network (TOLNet) to coordinate the activities of existing ground-based tropospheric ozone lidars in the U.S. and Canada. TOLNet includes fixed and transportable lidar systems that operate both routinely and on a campaign basis. Much of the data is posted on the TOLNet website (http://www-air.larc.nasa.gov/missions/TOLNet/index.html) and other measurements can be made available to state, tribal, and local regulatory agencies on request. TOLNet measurements can potentially be used to supplement routine surface monitoring, to investigate the synoptic-scale behavior of tropospheric ozone, and to help assess the impact of stratospheric intrusions and transported ozone on surface air quality. TOLNet measurements will also be used to help validate the high temporal and spatial resolution data obtained from the upcoming GEO-CAPE (Fishman et al., 2012) and TEMPO (Zoogman et al., 2014) satellite missions.

In this paper, we use TOLNet measurements made in California, Nevada, and Alabama to characterize the descent of two stratospheric intrusions that developed above the contiguous U.S. on May 24–25, 2013. The lidar measurements in California and Nevada were coordinated with an AJAX research flight, and the Nevada and Alabama measurements were part of larger efforts in support of the Las Vegas Ozone Study (LVOS) (Langford et al., 2015, 2017) and Southeast Nexus (SENEX) (Kuang et al., 2017; Warneke et al., 2016) field campaigns. Both intrusions were forecast by the NOAA National Environmental Satellite, Data, and Information System (NESDIS) Real-time Air Quality Modeling System (RAQMS) assimilation/forecast model (Pierce et al., 2003, 2007) and FLEXPART particle dispersion models (Brioude et al., 2013; Stohl et al., 2005) used in support of these campaigns. We compare the lidar and aircraft measurements to TOLNet ozonesonde profiles and to retrievals from the Atmospheric Infrared Sounder (AIRS) (Martin, 2008; Zoogman et al., 2011) aboard the NASA *Aqua* satellite, as well as back trajectories from the NOAA Air Resources Laboratory (ARL) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Rolph et al., 2017; Stein et al., 2015) and the RAQMS and FLEXPART analyses. Finally, we use surface measurements from regulatory monitors reporting to the U.S. EPA Air Quality System (AQS) to show how the stratospheric intrusions sampled by the coordinated TOLNet lidar and AJAX aircraft observations impacted surface air quality on May 23–25, 2013.

## 2. Tropospheric Ozone Lidar Network (TOLNet)

The Tropospheric Ozone Lidar Network or TOLNet was organized in 2010 under the auspices of the NASA Earth Sciences Division. The network loosely coordinates the activities of existing ground-based tropospheric ozone lidar systems in the U.S. and Canada and provides recommendations for the development of future systems. Three of the six lidar systems currently belonging to the network are operated by NASA through the Jet Propulsion Laboratory (JPL) Table Mountain Facility (TMF) in California (McDermid et al., 2002), the Langley Research Center (LaRC) in Virginia (Sullivan et al., 2015a), and the Goddard Space Flight Center (GSFC) in Maryland (Sullivan et al., 2015b). The remaining three are operated by the NOAA Earth System Research Laboratory (ESRL) (Alvarez et al., 2011), the University of Alabama at Huntsville (UAH) (Kuang et al., 2013), and Environment and Climate Change Canada (ECCC) (Strawbridge et al., 2017).

All of the TOLNet lidars use the differential absorption lidar (DIAL) technique (Proffitt and Langford, 1997) to measure ozone, but with hardware configurations and operating characteristics determined by

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