Investigating the influence of long-range transport on surface O₃ in Nevada, USA, using observations from multiple measurement platforms

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Abstract

The current United States (US) National Ambient Air Quality Standard (NAAQS) for O₃ (75 ppb) is expected to be revised to between 60 and 70 ppb. As the NAAQS becomes more stringent, characterizing the extent of O₃ and precursors transported into the US is increasingly important. Given the high elevation, complex terrain, and location in the Intermountain West, the State of Nevada is ideally situated to intercept air transported into the US. Until recently, measurements of O₃ and associated pollutants were limited to areas in and around the cities of Las Vegas and Reno. In 2011, the Nevada Rural Ozone Initiative began and through this project 13 surface monitoring sites were established. Also in 2011, the NASA Ames Alpha Jet Atmospheric eXperiment (AJAX) began making routine aircraft measurements of O₃ and other greenhouse gases in Nevada. The availability of aircraft and surface measurements in a relatively rural, remote setting in the Intermountain West presented a unique opportunity to investigate sources contributing to the O₃ observed in Nevada. Our analyses indicate that stratosphere to troposphere transport, long-range transport of Asian pollution, and regional emissions from urban areas and wildfires influence surface observations. The complexity of sources identified here along with the fact that O₃ frequently approaches the threshold being considered for a revised NAAQS indicate that interstate and international cooperation will be necessary to achieve compliance with a more stringent regulatory standard. Further, on a seasonal basis we found no significant difference between daily 1-h maximum O₃ at surface sites, which ranged in elevation from 888 to 2307 m, and aircraft measurements of O₃ <2500 m which suggests that similar processes influence daytime O₃ across rural Nevada and indicates that column measurements from Railroad Valley, NV are useful in understanding these processes.

Key words

Asian pollution, stratosphere to troposphere transport, back trajectory

44 1. Introduction

45 Exposure to elevated ozone (O₃) can be detrimental to human (cf. Chen et al., 2007) and environmental
46 health (cf. Musselman et al., 2006). As a result, National Ambient Air Quality Standards (NAAQS) for ambient O₃
47 have been set by the United States (US) Environmental Protection Agency (EPA). The EPA is expected to revise
48 the current standard of 75 ppb (8-h averaging period) (EPA, 2010). Comments are currently being accepting on
49 an O₃ NAAQS between 60 and 70 ppb (<https://federalregister.gov/a/2014-28674>).

50 The Nevada Rural Ozone Initiative (NVROI) was established in 2011 in response to concerns that Great
51 Basin National Park (GBNP), which is located in a remote, rural area of eastern Nevada near the border of Utah,
52 would not be in compliance with a more stringent O₃ NAAQS (Fine et al., 2014; Gustin et al., 2015).
53 Anthropogenic sources of O₃ precursors are limited in Nevada (cf. Cooper et al., 2012; Van Curen and Gustin,
54 this issue). Past measurement campaigns suggest that entrainment of air from the free troposphere influences
55 surface measurements in Nevada (Weiss-Penzias et al., 2009; Huang and Gustin, 2012; Gustin et al., 2013; Fine
56 et al., 2014). Modeling studies have indicated that long-range transport (LRT) of Asian pollution and
57 stratosphere to troposphere transport (STT) influence surface measurements in Nevada (Lefohn et al., 2012; Lin
58 et al., 2012a; Lin et al., 2012b). Entrainment of O₃ and precursors from the free troposphere can occur through
59 a number of mechanisms including subsidence behind cold fronts, tropopause folding, and growth of the
60 daytime boundary layer (Brown-Steiner and Hess, 2011; Lin et al., 2012b; Gustin et al., 2015).

61 Past work has shown that field measurements of meteorological parameters, traces gases, and
62 particulate matter are useful for identification of pollution sources (cf. Ambrose et al., 2011; Hart et al., 2006;
63 Hastie et al., 1996; Huang et al., 2013; Van Curen, 2003; Yates et al., 2013). Vertical profiles of the trace gases in
64 the atmosphere are particularly useful for understanding transport mechanisms and identification of layers of
65 pollution; however, vertical profiles derived from sonde and in-situ aircraft measurements are spatially and
66 temporally limited (Cooper et al., 2011). Trinidad Head (THD), located on the coast of northern California, is the
67 only location with a long-term record of vertical profiles of O₃. Given the rural coastal setting, measurements at

THD are likely representative of baseline O_3 , that is the O_3 entering the western US derived from non-domestic sources (Dentener et al., 2011; McDonald-Buller et al., 2011); however, Cooper et al. (2011) using measurements from a network of ozone sondes launched from sites spanning the coast of California noted that western US baseline O_3 varies with latitude. In addition to sonde measurements, aircraft platforms have been used to measure vertical profiles of trace gases in the atmosphere. In the western US, aircraft measurements tend to be limited to short-term intensive campaigns; however, the NASA Ames Alpha Jet Atmospheric eXperiment (AJAX) has been conducting routine airborne measurements of trace gases over California and Nevada since 2011 (Yates et al., 2013). Approximately once per month, there are flights to Railroad Valley, NV (RRVA) to measure vertical profiles of O_3 , methane (CH_4), carbon dioxide (CO_2), and water vapor in the atmosphere. The routine AJAX vertical profile measurements at RRVA coupled with the extensive surface O_3 monitoring network, recently established by the NVROI (Fine et al., 2014; Gustin et al., 2015), provide a unique opportunity to investigate the sources of O_3 aloft and the extent to which they influence boundary layer measurements in rural Nevada using in situ measurements. Further, measurements in this high elevation, rural area with low emissions provide an ideal means to identify remote contributions to the western US.

The objectives of this study were to (1) understand seasonal variability in baseline O_3 in rural Nevada based on vertical profiles measured by instruments on board the Alpha Jet; (2) investigate how baseline O_3 derived from vertical profiles in RRVA compares to baseline O_3 derived from THD profiles and NVROI surface measurements; and (3) characterize sources that influenced surface O_3 in rural Nevada using 4 specific case studies. Our hypothesis for this work was that by understanding the vertical distribution of O_3 throughout the troposphere, we can then better understand the sources of O_3 intercepted at surface sites in this region.

2. Methods

Vertical profiles

The analyses presented here are based on measurements collected from January 2012 to July 2013. Vertical atmospheric profiles of O_3 , CH_4 , CO_2 , and water vapor were collected as part of the AJAX project by

instruments on board the Alpha Jet research aircraft. Details regarding these measurements are described in Yates et al. (2013) and Tadić et al. (2014). Seasonal vertical flight profiles were constructed using data from 23 flights from this period. Following established conventions, seasons were classified as winter (December to February), spring (March to May), summer (June to August), and fall (September to November). To enable consistent comparison of O₃ between sites, data points were averaged in 500 m intervals throughout the vertical column (i.e. 1-1.5km, 1.5-2.0, etc.). However, individual point measurements (i.e. not interval averaged) are presented in the individual case studies. Measurements collected between >1 and 8.5 km altitude were considered in the seasonal comparisons. We also considered vertical profiles of O₃ from sonde measurements at THD and Hilo, Hawaii (HILO) which are operated by the Global Monitoring Division (GMD) of the Earth System Research Laboratory (ESRL) at NOAA (<http://www.esrl.noaa.gov/gmd/dv/data/>). We present vertical profile data in units altitude (i.e. units above sea level – asl) when making direct comparisons between two different locations whereas, data are presented in units above ground level (agl) when we discuss trajectories and profiles over a single location. In all figures and associated discussion, the units considered are noted. We felt that both metrics were useful particularly because the complex terrain in Nevada influences air movement and subsequently the extent of entrainment.

Surface measurements

We considered hourly O₃ data measured at 11 ground sites within the NVROI network (Table 1) for the period of January 2012 to July 2013. Specific site characteristics and measurements at these surface sites are described in Gustin et al. (2015) and Fine et al. (2014). Seasonal mean daily 1-h maximum values were derived by averaging the daily 1-h maximum for all days within a particular season. Sites with <50% of data available for a particular season were not included in seasonal summary plots or corresponding statistical analyses, but were discussed in the case studies if measurements were available. Ancillary trace gas measurements, specifically of CO, NO, and NO_x, from Paradise Valley (PAVA; el. 1385 m) and GBNP (el. 2060 m) were considered when available. Details regarding these particular measurements can be found in Gustin et al. (2015) and Miller et al.

(this issue). We also used the chemical composition of particulate matter collected by the National Park Service at GBNP reported by the IMPROVE network (Data downloaded from: views.cira.colostate.edu).

Trajectory Analyses

Backward trajectories were calculated using the PC version of the HYSPLIT4 model available from <http://ready.arl.noaa.gov/HYSPLIT.php>. Technical details for the model can be found in Draxler and Hess (1997 and 1998). Global Data Assimilation System (GDAS) archives were used as the input meteorology. An ensemble of trajectories were initiated at 2000 UTC (1200 PST) from 9 starting locations in a 0.5° x 0.5° grid centered on the RRVA ground site and at starting heights coincident with laminae of interest for each particular case study. The duration of backward trajectories was 240 hours (10 days). We also used the web based version of HYSPLIT to construct individual trajectories from sites other than RRVA to support interpretation of the data (Draxler and Rolph, 2014; Rolph, 2014).

Case Studies

Four case studies are presented here. We selected one flight day from each season for in-depth analyses. We based the selection on the availability of regional surface site and flight data.

Statistical Analyses

Statistical analyses were conducted using Microsoft Excel 2010, Minitab 16, and ESRI ArcGIS software. Results are considered significant at the 95% confidence level ($\alpha = 0.05$). Paired t-tests were used to test if there were significant differences between two sets of data, specifically, the seasonal flight profiles as well as flight profiles from the boundary layer and surface measurements. We calculated the median of O₃ data at 500 m intervals within the flight profiles measured for each season. Because of the fine temporal resolution of the flight measurements (e.g. 2 seconds for O₃ measurements), we assumed that the data used to calculate the median 500 m interval values followed a normal distribution. We compared the median values throughout the vertical profiles for each season to identify the extent of seasonal variability in O₃. Data at surface sites across

139 Nevada were collected on a continuous basis. Generally, a diel pattern was evident in O₃ data measured at
140 surface sites throughout rural Nevada. Peak O₃ concentrations occurred during midday as a result of
141 establishment of a well-mixed boundary layer, and minimum O₃ concentrations occurred at night as a result of
142 surface deposition processes and collapse of the daytime boundary layer (cf. Gustin et al., 2015). The
143 magnitude of the diel pattern varied site to site as a result of differences in terrain features. To avoid
144 introducing bias resulting from the diel patterns at each site, we calculated the seasonal mean daily 1-h
145 maximum O₃ concentration for each site. These mean values from the surface sites were compared to median
146 O₃ data calculated from O₃ measured during flights over RRVA when aircraft were <2500 m (approximate height
147 of the boundary layer during flight measurements) in an effort to determine if the distribution of O₃ measured
148 by aircraft in the boundary layer at RRVA were representative of surface measurement across rural Nevada.

149 **Results and Discussion**

150 **O₃ at grounds sites in rural Nevada**

151 Here, in addition to the maximum daily 8-h average (MDA8), we used the daily 1-h maximum O₃ value,
152 because it was less likely to be biased by nocturnal surface depositional processes and represents the maximum
153 for each day, thus is not influenced by averaging. Further, use of the 1-h metric allowed us to highlight the fact
154 that relatively high peak O₃ is measured across rural Nevada, which may be obscured by other metrics including
155 MDA8 which is used for assessment of compliance with the NAAQS.

156 During the time period considered here, seasonal mean MDA8 was greatest in the summer (47 to 60
157 ppb; 8 sites) followed by the spring (43 to 56 ppb; 9 sites), fall (38 to 48 ppb; 8 sites), and winter (32 to 45 ppb; 8
158 sites) (data not shown). If we consider the current NAAQS of 75 ppb, the MDA8 measured at these rural sites
159 averaged 63 to 80%, 57 to 75%, 51 to 64%, and 32 to 45% of the NAAQS for the summer, spring, fall, and winter,
160 respectively. If we consider the lowest end of the range in the EPA proposal for the revised NAAQS (60 ppb),
161 then the average MDA8 at the rural Nevada sites could range from 78 to 100%, 72 to 93%, 63 to 80%, and 53 to
162 75% of the proposed NAAQS in the summer, spring, fall, and winter, respectively. It is important to note that

163 we reported average MDA8 across particular seasons in the above discussion and assuming that MDA8 values
164 follow a fairly normalized distribution around the mean at these sites, then the data indicate that O₃ routinely
165 approaches a value that could lead to exceedance of the current standard and suggests that compliance with a
166 more stringent standard will be challenging.

167 Consistent with the MDA8 metric, mean daily maximum 1-h O₃ was greatest in the summer (50 to 62
168 ppb; 8 sites) and spring (45 to 59 ppb; 9 sites) and lowest in the winter (34 to 47 ppb; 9 sites) (Table 2). Using
169 regression analyses, we found a moderate to strong positive linear relationship between site altitude and
170 seasonal mean daily 1-h maximum O₃. The strongest correlation was in the summer ($r^2 = 0.95$; $p = 0.00$)
171 followed by the fall ($r^2 = 0.79$; $p = 0.00$), winter ($r^2 = 0.77$; $p = 0.00$), and spring ($r^2 = 0.67$; $p = 0.01$) (Table 3).
172 High elevation sites more readily intercept pollution transported in the free troposphere and the results of the
173 regression analyses indicate an increase of 13 ppb in the daily maximum 1-h O₃ for every km in altitude occurred
174 in the summer, and that the rate was 7 to 10 ppb/km in the other seasons that emphasizes the importance of
175 the high elevation setting of Nevada. The summer months throughout Nevada are characterized by increasingly
176 arid conditions and rapid surface heating that promote the development of a very deep boundary layer (cf.
177 Whiteman, 2000; Gustin et al., 2015). The strong relationship between site altitude and maximum 1-h O₃
178 indicate that higher elevation sites are more directly exposed to the free troposphere, and experience less
179 dilution with “cleaner” boundary layer air and deposition of O₃ to surfaces.

180 **Vertical profiles of O₃ at Railroad Valley, NV**

181 During the time period considered, there were 23 flights over Railroad Valley with 11 in the summer, 5
182 in the spring and winter, and 2 in the fall. Flights were targeted for clear sky conditions over RRVA, which could
183 have biased the patterns reported. Median O₃ in the vertical profile (calculated in 500 m intervals ranging from
184 1.5 to 8.5 km altitude) ranged from 53 to 80 ppb, 56 to 71 ppb, 41 to 50 ppb, and 36 to 56 ppb in the summer,
185 spring, winter, and fall, respectively (Figure 1). Using a paired t-test (as a function of altitude), we found no
186 significant difference between median O₃ profiles in the spring and the summer ($p = 0.61$). Median O₃

187 throughout the profile was an average of 16 ppb lower in the winter, relative to the spring and summer. Below
188 2.5 km altitude (1 km agl at RRVA), median O₃ averaged 58, 57, 43, and 49 ppb in the summer, spring, winter,
189 and fall, respectively. Using a paired t-test, we found no significant difference between median O₃ in the vertical
190 profile <2.5 km altitude and mean daily 1-h maximum O₃ at ground sites in rural Nevada (p = 0.23, 0.09, and 0.84
191 in the winter, spring, and summer respectively; Due to the small sample size for the flight data, a statistical
192 comparison could not be made for the fall season). These results suggest that, at least on a seasonal time-scale,
193 the processes influencing daytime O₃ are similar across sites in rural NV and that measurements at RRVA can be
194 effectively scaled up to understand these processes.

195 Because data were available, we compared the seasonal profiles of O₃ at RRVA with those measured by
196 sondes launched from Trinidad Head, CA (THD) (Figure 2). To avoid bias introduced by the local maritime
197 climate and the marine boundary layer at THD, we did not consider measurements made < 2 km altitude in the
198 statistical comparison. However, some bias may have been introduced to the comparison because, although the
199 seasons are the same, the particular days of the flights and the sonde launches were different. Using a paired t-
200 test to compare median seasonal values at 500 m intervals throughout the vertical column, we found no
201 significant difference between vertical profiles of median O₃ above RRVA and THD in the summer (p-value =
202 0.81). In the winter, median O₃ was an average of 5.5 ppb (95% Confidence Interval = 3.1, 7.8 ppb; p-value =
203 0.00) higher throughout the vertical profile at THD whereas median O₃ was an average of 2.4 ppb (95%
204 Confidence Interval = 0.3, 4.6 ppb; p-value = 0.03) higher at RRVA than at THD in the spring. The consistency
205 between profiles at THD and RRVA in summer is coincident with limited activity along the Pacific storm track. In
206 the winter, the difference between the THD and RRVA profiles increases with increasing altitude which is
207 suggestive of divergent synoptic scale conditions. The trajectory of the polar jet stream likely influences the
208 composition of air aloft. The winter months considered in this analysis were among the driest on record in the
209 western US (cf. <http://www.ncdc.noaa.gov/sotc/national/2012/2> and
210 <http://www.ncdc.noaa.gov/sotc/national/2013/2>), activity along the Pacific storm track was limited, and the jet
211 stream was positioned farther north of the Great Basin and the northern Sierra Nevada Mountains than is

212 typical. The meander of the jet stream could have led to a situation where air intercepted at THD moved north
213 along the path of the jet stream rather than west. Thus, air intercepted at RRVA during the winter months was
214 along the margins of the jet stream and had a different composition than that at THD. Due to the anomalous
215 winter conditions, this may be a unique bias for the time period considered. In contrast, the difference between
216 the spring profiles is most pronounced below 3 km altitude which would suggest a regional enhancement at
217 RRVA that is not present at THD or, more likely, greater top-down entrainment of air aloft at RRVA. The arid
218 climate and lack of vegetation at RRVA promote rapid surface heating and subsequently more rapid boundary
219 layer growth relative to THD, which is characterized by a maritime climate.

220 In the next section, we investigate in detail the link between coincident measurements of O_3 and other
221 parameters throughout rural Nevada to understand sources influencing measured O_3 observed at surface sites
222 with specific seasonal case studies (Figure Supplementary Information (SI) 1; Table SI 1).

223 **Spring Case Study: 9 April 2012**

224 The flight day was characterized by warm, dry surface conditions and clear skies across Nevada with the
225 exception of thin clouds in the far northwest sector. In the days leading up the flight, an area of low pressure
226 took up residence off the coast of northern California and was evident in National Weather Service (NWS)
227 surface analyses plots in the upper and lower atmosphere for most of the preceding week and in the days
228 following the flight to RRVA. Farther south, an area of high pressure was located offshore, southwest of
229 southern California and extended to the Hawaiian Islands. Modeled back trajectories (Figure SI 2) and NWS
230 forecasts indicate that these two (surface) pressure centers were persistent and effectively recirculated air
231 offshore for several days.

232 The vertical profile of O_3 from the aircraft measurements at RRVA ranged from 52 to 76 ppb with
233 laminae of elevated O_3 present between 2.6 and 3.6 km above ground level (agl) and above 5 km agl (Figure SI
234 1). Relative to the rest of the vertical column, CO_2 and CH_4 were depleted in the laminae with elevated O_3
235 (Figure 3). Trajectory calculations indicate that air throughout the vertical column descended from high altitude

236 (>5 km agl) to RRVA (Figure 4). Laminae intercepted in RRVA at 4.5 and 6.0 km agl followed a trajectory across
237 the northern Pacific Ocean before approaching RRVA from a southwesterly direction. Air arriving at this
238 elevation maintained a fairly constant altitude and was unlikely to have influenced surface observations in
239 Nevada. Peak O₃ concentrations of 75 to 76 ppb were measured at 2.8 and 3.4 km agl, respectively. Back
240 trajectory calculations indicated that air at 3 km agl approached RRVA from the south and traveling from the
241 central Pacific Ocean near the Hawaiian Islands. It is interesting to note that the vertical profile of O₃ from
242 sonde measurements at Hilo, HI on 4 April 2012 (5 days prior the RRVA flight) shows laminae with elevated O₃
243 present at 4.4 (72 ppb) and 5.1 km asl (72 ppb), which is markedly similar to the elevated laminae encountered
244 at RRVA (Figure SI 3). Given that trajectory calculations indicate that air descended as it approached RRVA from
245 the south, the elevated laminae observed at Hilo at 4.4 and 5.1 km asl could be derived from the same source as
246 that observed at RRVA at 4.2 and 4.8 km asl (2.8 and 3.4 km agl, respectively). Back trajectories initiated from
247 Hilo on the day of the sonde launch indicate that air descended from high altitude (>6.5 km asl) and was
248 recirculated above the Hawaiian Islands for several days (Figure SI 2). Further, sonde measurements from THD
249 on April 10 (1 day after the RRVA flight) indicate a laminae of enhanced O₃ present at 4.9 (78 ppb) and 5.8 km
250 (76 ppb) asl, which is similar in altitude and concentration to laminae intercepted at RRVA and HILO in the
251 preceding days (Figure SI 3). Back trajectories initiated from THD were similar to those for RRVA (Figure SI 2).
252 Below 2.6 km agl at RRVA, O₃ was relatively homogenous (60 ppb) suggesting that O₃ was well-mixed near the
253 surface throughout the region. No significant difference ($p = 0.30$) was found between aircraft measurements $\leq$
254 2.5 km agl and maximum 1-h O₃ measurements from the 5 active surface sites in rural Nevada that ranged from
255 52 to 65 ppb further indicating that daytime O₃ was consistent and well-mixed throughout the region.

256 Trajectory analyses, meteorological conditions, and the similarities in O₃ aloft measured at 3 different
257 sites throughout the eastern Pacific region indicate that meso-scale conditions effectively transported a widely
258 dispersed plume of O₃ across the Pacific. This is consistent with typical spring plumes of Asian pollution
259 described by Brown-Steiner and Hess (2011). Given the consistent high altitude transport and geographic origin
260 indicated in the back trajectory calculations, a mixture of Asian pollution and stratospheric O₃ likely contributed

261 to the observed O₃ in the laminae. Relative humidity was persistently low at all surface sites throughout the
262 day, with minimum relative humidity ranging 52 to 87% below the hourly monthly average at all 5 rural ground
263 sites (Figure SI 4). The elevated O₃ coincident with low humidity is indicative of subsidence of dry air from aloft
264 which could include O₃ derived from a stratospheric intrusion or LRT of pollution. To distinguish between LRT or
265 stratospheric intrusion or lofted regional pollution, we considered additional parameters measured at PAVA and
266 GBNP. A marker of local emissions, NO, was markedly low at GBNP and PAVA (< 2 ppb) (Figure SI 5). In several
267 past studies, CO has been used as an indicator of LRT (cf. Ambrose et al., 2011; Lin et al., 2012b). At PAVA and
268 GBNP, the daily average of CO was slightly below the monthly mean (80 versus 84 ppb and 112 versus 138 ppb
269 at GBNP and PAVA, respectively); however, we found a weak to moderate linear correlation between O₃ and CO
270 for the day ($r^2 = 0.12$; $p = 0.10$ and 0.28 ; $p = 0.00$ at GBNP and PAVA, respectively) (Figure SI 5). At the GBNP
271 IMPROVE site, fine particulate made up the majority (58%) of the total particulate. The fine aerosol composition
272 was dominated by sulfate (SO₄ + NH₄SO₄ = 47% of mass) (Figure 5), which is characteristic of long-range
273 transport from Asia (Brock et al., 2004; Dunlea et al., 2009; Lin et al., 2012b).

274 Relative to GBNP, PAVA was in closer proximity to the location of the offshore low pressure center
275 driving the cyclonic flow. Back trajectory calculations indicate that air intercepted at PAVA took a more rapid,
276 direct trans-Pacific route than air arriving at GBNP which circulated around the central Pacific (near the Hawaiian
277 Islands) prior to descending to GBNP (Figure SI 6). The gaseous composition was disrupted as outflow
278 encountered the mountainous terrain of Nevada; however, the particulate signature was retained (even if mass
279 was not) by tropospheric air intercepted at GBNP. CO concentration and sulfate mass were somewhat lower
280 than that of Asian pollution layers reported by other studies which indicated that dilution with stratospheric air
281 occurred for this particular event (Brock et al., 2004; Lin et al., 2012b). This is consistent with the conclusion
282 from other studies indicating that stratospheric air intermixes with Asian pollution in the free troposphere (cf.
283 Cooper et al., 2004; Cooper et al., 2006; Lin et al., 2012b; Trickl et al., 2014). It is interesting to note that the
284 chemical composition of the aerosol sample at GBNP was similar to that of the average monthly composition
285 which indicates that pollution derived from LRT was routinely intercepted at GBNP in April 2012 (Figure 5).

286 **Summer Case Study: 17 July 2012**

287 An area of surface high pressure was stationed off the coast of the western US. Aloft, a closed low was
288 located off the coast of northern California and established a strong vorticity gradient and conditions conducive
289 to STT. Above 4 km agl, O₃ averaged 26 ppb ranging from 16 to 43 ppb. A lamina with elevated O₃ was present
290 between 3 and 4 km agl. In this lamina, O₃ ranged from 72 to 91 ppb and was coincident with depleted water
291 vapor and CO₂, and enhanced CH₄ (relative to the rest of the column) (Figure 3). CH₄ and CO₂ are derived
292 predominantly from anthropogenic sources; however, the magnitudes and daily cycles of individual source areas
293 and types are not well resolved (Hartmann et al., 2013; Wecht et al., 2014). CH₄ in particular has a number of
294 sources that have not been well quantified including marine emissions and offshore methane hydrate extraction
295 which could influence air masses within the marine boundary layer indicating that CH₄ may be a somewhat
296 ambiguous tracer until sources are better resolved. CO₂ has a well-defined tropospheric seasonal cycle and no
297 known stratospheric sinks (Sawa et al., 2008); as such, Yates et al. (2013) noted that during the summer, CO₂ in
298 the stratosphere is expected to be less than that in the troposphere and, thus, can serve as an unconventional
299 tracer of STT.

300 The vertical profile of trace gases at RRVA indicated that the air was well-mixed up to 3 km agl and O₃
301 was 60 ppb (Figure 3), whereas CO₂ concentrations were higher than that measured in the lamina and slightly
302 less than that measured above. Because RRVA is a playa devoid of vegetation, local (soil) CO₂ efflux would be
303 driven by increased moisture and temperature. In the days prior to the flight, the playa received several inches
304 of precipitation. Past work has shown that moisture increases CO₂ efflux from a wide variety of soils (cf. Smith
305 et al., 2003; Harms and Grimm, 2012); however, the mixing depth was >2 km agl at the time of flight and it is
306 unlikely that local, episodic soil emissions alone would have driven concentrations observed throughout the
307 entire boundary layer in RRVA. Given that the boundary layer CO₂ concentration is similar to that measured
308 aloft; the CO₂ in the boundary layer in this instance is likely representative of global tropospheric background.
309 Entrainment of air from the lamina would have reduced CO₂ and enhanced O₃ in the boundary layer at RRVA and

310 transitional concentrations of these two gases were observed just below the lamina indicating that entrainment
311 was in progress (Figure 3). The coincident peak in O_3 and minima in CO_2 is consistent with O_3 in the lamina being
312 derived from STT at this time of the year (cf. Yates et al., 2013).

313 Maximum 1-h O_3 concentrations at sites in rural Nevada ranged from 54 to 68 ppb and were higher at
314 sites in the western part of the state compared to the eastern part of the state, regardless of site elevation
315 (Figure SI 7). We found a strong linear correlation between 1-h O_3 and relative humidity which further supports
316 the conclusion that entrainment of air aloft, which is characterized by low humidity, influenced boundary layer
317 measurements in rural Nevada (Figure SI 8). Back trajectory calculations indicate that air intercepted
318 throughout the vertical column at RRVA was derived from high altitude (> 5 km agl) (Figure 6). Air intercepted in
319 the boundary layer at RRVA descended from high altitude off the coast of northern California (in the vicinity of
320 the closed low aloft) and then traveled within the marine boundary layer off the coast of California and within
321 the boundary layer of southern California (circa San Diego county) and southern Nevada (circa Las Vegas) before
322 arriving in RRVA (Figure 6). Given this trajectory, we would expect that there was some regional influence on
323 CO_2 concentrations with uptake in active growing regions of California coupled with (and likely offset by)
324 emissions from urban areas of San Diego and Las Vegas. Back trajectories initiated at 6 km agl, that indicate
325 that air was lofted from the continental boundary layer of east Asia, followed a rapid trans-Pacific route and
326 descended off the coast of northern California, then traveled at high altitude (> 5 km agl) offshore along the
327 California coast before moving across southern California and into Nevada (with no boundary layer interactions).
328 Back trajectories initiated at 3.5 km agl (mid-point of lamina with peak O_3) followed a similar route as those
329 intercepted at 6 km agl except that interaction with the eastern Asia boundary layer was less evident, and the
330 route directly intercepted the closed low off the coast of California (Figure 6). The trajectories and associated
331 trace gases suggest that air in the upper part of the column was derived from a combination of Asian LRT and
332 stratospheric O_3 ; air in the lamina with peak O_3 was primarily derived from the lower stratosphere; and air in the
333 well-mixed boundary layer was derived from Asian LRT, stratospheric O_3 , and regional urban emissions. A
334 lamina of elevated O_3 was also present in the vertical profile of O_3 measured by a sonde launched from THD on

335 the day following the RRVA flight (Figure SI 9) which suggests that a reservoir of elevated O_3 persisted in the area
336 offshore of northern California. For the sake of comparison, the O_3 measured from the sonde launch at Hilo did
337 not indicate this same enhancement (Figure SI 9).

338 Surface measurements of CO, NO, and NO_x were lower than average for the month at PAVA (Figure SI 5;
339 measurements were not available at GBNP). Interestingly, the regression slopes between CO, NO, NO_x and O_3
340 at PAVA were negative. We would expect CO and O_3 to be enhanced during a LRT event; however, there was a
341 negative relationship at PAVA indicating a source of O_3 without associated precursors such as STT. The west >
342 east gradient in O_3 suggests that the offshore dynamics had a greater influence on sites in closer proximity. In
343 addition, back trajectories initiated from PAVA and GBNP indicate that air intercepted at sites on the eastern
344 side of the state intermingled with boundary layer pollution in upwind urban areas (San Diego, Las Vegas)
345 (Figure SI 10), suggesting that dilution with urban boundary layer air decreased ambient O_3 . The composition of
346 particulate samples from the IMPROVE site at GBNP showed a strong sulfate component as well as fine soil and
347 organics (Figure 5). Back trajectories indicate significant residence time in the marine boundary layer near
348 southern California ports. The sulfate component was likely derived from shipping emissions rather than LRT
349 and the fine soil and organic component also could have been derived from regional sources.

350 **Case Study 3: 21 September 2012**

351 More than a dozen large wildfires were burning in the Pacific Northwest with significant smoke output
352 on the day of the flight to RRVA (NOAA NCDC, 2012; Figure SI 11). Skies were clear and a ridge of surface high
353 pressure was present across the state, which is conducive to the accumulation of pollution in the boundary
354 layer. Below 3 km agl, O_3 in RRVA was well-mixed at 55 ppb with the exception of the first few hundred meters
355 above ground level where it was below 50 ppb (Figure 3). Water vapor was elevated relative to the upper
356 portion of the vertical column indicating boundary layer transport rather than subsidence from the free
357 troposphere. Back trajectory calculations indicate that air in the well-mixed lower half of the column at RRVA
358 followed a clockwise circuitous route around the northern sector of Nevada before approaching RRVA from the

359 south whereas air in the upper half of the column was derived from high altitude and the marine boundary layer
360 (Figure 7). Maximum 1-h O_3 concentrations at sites in rural Nevada ranged from 51 to 58 ppb and were higher
361 in the northeast half relative to the southwest half of the state (Figure SI 7). This pattern in maximum 1-h O_3
362 concentrations is consistent with wildfire emissions exerting a stronger influence on air in the northeast half of
363 the state due to the closer proximity to active wildfires in the Pacific Northwest (Figure SI 11). Measurements of
364 CO were significantly higher at the surface site in PAVA relative to GBNP (mean of 335 and 167 ppb,
365 respectively) whereas NO_x was similar (mean = 2.5 ppb) (Figure SI 5). Chemical analyses of PM samples
366 collected at the IMPROVE sites in Nevada (Jarbridge and GBNP) had elevated potassium (Figure SI 11), an
367 indicator of wildfire emissions (cf. Bein et al., 2008), on the day of the flight to RRVA suggesting that the plume
368 from the Pacific Northwest wildfires was indeed intercepted at surface sites throughout Nevada. It is important
369 to note that while measurements indicate that the gas and particulate composition of air parcels intercepted at
370 surface sites in Nevada were influenced by wildfire emissions, the O_3 concentration of the boundary layer was
371 only 11 ppb greater than air aloft which was not influenced by wildfire emissions to the same extent as the
372 boundary layer air (Figure 7).

373 **Case Study 4: 19 December 2012**

374 An area of high pressure was centered over central Nevada on the day of the flight and an area of low
375 pressure was stationed offshore to the northwest of Washington state adjacent to a cold front overtaking a
376 warm front indicating the presence of a mid-latitude cyclone. STT is often promoted by these conditions (cf. Lin
377 et al., 2012a; 2012b). At RRVA, O_3 ranged from 34 to 66 ppb in the vertical column and average of 42 ppb in the
378 boundary layer (Figure 3). The vertical profile was characterized by multiple, filamentous layers with variable O_3
379 concentrations. CH_4 and CO_2 were also variable; however, concentrations of these two trace gases were
380 significantly higher throughout the column than the other two cases studies when carbon measurements were
381 available suggesting that anthropogenic emissions had a greater influence on the composition of air in the
382 column than the two prior case studies (Figure 3). Decreased continental uptake of CO_2 would also be lower

383 during this time in the Northern Hemisphere, because this time is not within the active growing season. Back
384 trajectory analyses indicate that air intercepted throughout the vertical column at RRVA was rapidly transported
385 from continental Asia along the path of the jet stream (Figure 8). Air intercepted in the RRVA boundary layer
386 spent several days within the marine boundary layer of the northern Pacific Ocean. Air intercepted at higher
387 altitudes in RRVA was transported at primarily high altitude (> 5 km agl) before descending to RRVA (Figure 8).
388 The back trajectory calculations and the enhanced CH_4 and CO_2 suggest Asian LRT contributed to measured O_3 ;
389 however, the dynamics present off the coast in the vicinity of the descending air mass likely contributed
390 stratospheric O_3 as well. At surface sites in rural NV, maximum 1-h O_3 ranged from 34 to 46 ppb (Figure SI 7).
391 Measured O_3 concentrations were significantly higher at the high elevation sites (> 2 km asl) than at the low
392 elevation sites (Figure SI 7). At PAVA, CO was slightly lower than the monthly mean (daily mean = 121 v. 132
393 ppb); however, we detected a moderate, linear correlation between O_3 and CO ($r^2 = 0.45$; $p = 0.00$) (Figure SI 5).
394 At GBNP, CO was elevated above the monthly mean (daily mean = 157 v. 130 ppb) and PM composition
395 measured at the IMPROVE site was dominated by organic carbon, elemental carbon, and sulfate (Figure 6). The
396 fact that O_3 was higher at the high elevation sites is consistent with a free tropospheric source of O_3 . Due to low
397 temperatures and the limited surface interactions, the free troposphere can accumulate O_3 and O_3 precursors
398 derived from multiple sources including the stratosphere, long-range transport, and regional emissions. It is
399 interesting to note that there was a significant, negative correlation between O_3 at PAVA and GBNP ($r = -0.80$; p
400 $= 0.00$) which we interpret to indicate that with growth of the boundary layer, entrainment of boundary layer air
401 reduced O_3 at GBNP (situated at high elevation and largely exposed to the free troposphere) whereas at PAVA
402 entrainment of free tropospheric air during growth of the boundary layer enhanced O_3 . The elevated CO_2 and
403 CH_4 in the vertical profile as well the elevated CO and composition of PM at GBNP are consistent with an
404 anthropogenic source area influencing the composition of air intercepted at these rural NV sites. Back trajectory
405 calculations indicate Asia as a likely source area; however, the high altitude transport pattern and presence of a
406 mid-latitude cyclone provide conditions in which stratospheric O_3 could have been intermingled with O_3 derived
407 from Asian LRT. Recent work by Trickl et al. (2014) found that CO is rarely depleted in stratospheric intrusion

layers. The authors conclude that this was due to stratospheric air mixing rather slowly with tropospheric air near the thermal tropopause in the absence of significant wind shear or convection.

Conclusions

The measurement of O_3 at an extensive network of rural sites coupled with routine aircraft observations of O_3 and trace gases in the vertical column in RRVA presented a unique opportunity to investigate sources of O_3 intercepted at sites in rural Nevada. Surface and vertical profile measurements were highest in the summer and spring. On a seasonal basis, we found no significant difference between daily 1-h maximum O_3 at surface sites, which ranged in elevation from 888 to 2307 m, and aircraft measurements of O_3 <2500 m. This finding suggests that similar processes influence daytime O_3 across rural Nevada and indicate that column measurements from RRVA, derived from aircraft campaigns or potentially satellite measurements (if vertical resolution can be improved) are useful in understanding these processes. Using surface, sonde, and aircraft measurements we presented analyses of 4 case studies to characterize sources influencing O_3 observed in rural Nevada. These analyses indicated that STT, Asian LRT, and regional emissions from urban areas and wildfires influenced surface observations. Back trajectory analyses coupled with trace gas measurements and speciated particulate measurements indicate that air masses influenced by STT were intermingled with pollution derived from Asian LRT. Regional emissions were more prominent in measurements made during the summer and fall, whereas evidence of Asian LRT and STT intercepted by surface sites occurred in the winter, spring, and summer. The complexity of sources identified in these analyses and the fact that O_3 throughout rural NV frequently approaches the threshold being considered for a revised NAAQS emphasizes that interstate and international cooperation will be necessary to design effective compliance strategies for this region. Fine resolution chemical transport modeling is necessary to determine the quantitative contribution from regional emissions, Asian LRT, and STT. The surface and airborne measurements presented here will be useful in validating chemical transport models.

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Figure 1: Vertical profiles of O₃ from aircraft measurements over Railroad Valley, NV. The bold lines represent median O₃ and the thin lines represent the 25th and 75th percentile O₃ measured during the season at each altitude. The number of aircraft profiles in each season during the time period considered is noted in parentheses next to the season. Because measurements from only 2 fall aircraft flights were available, data were averaged in 500 m intervals and plotted here for both fall flights (denoted with bold lines – bottom right hand figure). The dots represent seasonal mean daily 1-h maximum O₃ at ground sites located at a range of altitudes in rural Nevada. The vertical dotted lines at 60 and 70 ppb O₃ are for reference.

Figure 2: Seasonal midday vertical profiles of O₃ at Railroad Valley, NV (RRVA) and Trinidad Head, CA (THD). The bold line represents the median value and the thin lines represent the 25th and 75th percentile of the distribution. The number of flights (RRVA) or sonde launches (THD) is indicated in parentheses next to the site code in the legend. Because measurements from only 2 fall aircraft flights were available, data were averaged in 500 m intervals and plotted here for both fall flights (denoted with bold lines – bottom right hand figure). The vertical dotted line at 70 ppb is for reference.

Figure 3: Midday vertical profiles of O_3 , water vapor, carbon dioxide (CO_2), and methane (CH_4) at Railroad Valley on (from top to bottom) 09 April 2012, 17 July 2012, 21 September 2012, and 19 December 2012 from aircraft measurements.

Figure 4: Ensembles of 10-day back trajectories initiated from Railroad Valley, NV (RRVA) on 09 April 2012. The starting heights for the ensembles are indicated in the top left-hand corner of each plot.

Figure 5: Chemical composition of IMPROVE particulate samples from GBNP on select days. Measured fine mass ($\mu\text{g}/\text{m}^3$) is in parentheses in the figure legends. A description of each parameter is given in Table SI 2.

Figure 6: Ensembles of 10-day back trajectories initiated from Railroad Valley, NV (RRVA) on 17 July 2012. The starting heights for the ensembles are indicated in the top left-hand corner of each plot.

Figure 7: Ensembles of 10-day back trajectories initiated from Railroad Valley, NV (RRVA) on 21 September 2012. The starting heights for the ensembles are indicated in the top left-hand corner of each plot.

Figure 8: Ensembles of 10-day back trajectories initiated from Railroad Valley, NV (RRVA) on 19 December 2012. The starting heights for the ensembles are indicated in the top left-hand corner of each plot.

Site Name	Code	Elevation (m)	Latitude (N)	Longitude (W)	Date Established (M/YYYY)	Percent of days with O ₃ data available			
						Winter	Spring	Summer	Fall
Angel Peak	AGPK	2705	36.3182	115.5749	6/2013	0	0	5	0
Warm Springs Summit	WSSU	2307	38.1844	116.4250	6/2012	60	52	70	100
Echo Peak	ECHO	2286	37.2128	116.3236	6/2012	60	51	100	73
Berlin Ichthyosaur State Park	BISP	2082	38.8700	117.6033	7/2011	100	100	75	86
Great Basin National Park	GBNP	2060	39.0050	114.2161	7/2011	99	97	100	92
White River Valley	WRVA	1728	38.9163	115.0686	7/2011	100	65	60	100
South Fork State Recreation Area	SFRA	1602	40.6767	115.7453	7/2011	94	70	100	100
Cathedral Gorge State Park	CGSP	1513	37.8100	114.4097	7/2011	93	85	100	91
Railroad Valley	RRVA	1438	38.5044	115.6935	11/2012	48	49	40	32
Paradise Valley	PAVA	1385	41.5031	117.4981	11/2011	93	84	99	100
Pahrump	PRMP	888	36.3489	116.0317	5/2013	0	0	27	0

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627 **Table 1:** Description of rural Nevada O₃ monitoring sites and availability of data for the time period considered
628 in the analysis.

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	AGPK	WSSU	ECHO	BISP	GBNP	WRVA	SFRA	CGSP	RRVA	PAVA	PRMP
Winter		43	47	44	46	41	40	40	40	34	
Spring		58	58	56	59	52	52	53	54	45	
Summer	69*	62	62	57	61	55	53	51	53*	50	59*
Fall		47	50	48	48	43	43	44	38*	42	
Site Altitude (m)	2705	2307	2286	2082	2060	1728	1602	1513	1438	1385	888

632 **Less than 50% data availability*

633 **Table 2:** Seasonal mean daily maximum 1-h O₃ (ppb) at rural Nevada surface sites.

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	r^2	p-value	intercept	slope
Winter	0.77	0.00	24	9.4
Spring	0.67	0.01	37	9.5
Summer	0.95	0.00	32	13.3
Fall	0.79	0.00	33	6.9

Table 3: Results of linear regression analyses of site altitude (km) and seasonal mean daily 1-h maximum O₃.