RESEARCH ARTICLE

Contrasting behavior of slow and fast photoreactive gases during the August 21, 2017, solar eclipse

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The total solar eclipse on August 21, 2017, provided a rare opportunity to observe and test our understanding of atmospheric dynamics and photochemical dependency on solar irradiance. Here, we utilize observations from the continuous monitoring of both slow and fast photochemically reacting trace gases near Boulder, Colorado, for evaluating the unique dynamic and photochemical forcings on the eclipse day. The monitoring station saw a 93% solar obstruction during the peak of the eclipse. Eclipse day data are contrasted with the full month’s record from this site. The loss of irradiance caused cooling of the surface air by ~3°C, and weakened convective and turbulent mixing. This resulted in a buildup of slow photoreactive gases (methane, short-chain non-methane hydrocarbons), as well as total nitrogen oxides (the sum of nitric oxide (NO) + nitrogen dioxide (NO$_2$)) in the surface layer. In contrast, ozone (O$_3$) declined by ~15 ppb during the first phase of the eclipse compared to median August diurnal mixing ratios. Similar O$_3$ signatures were observed at a series of network stations along the Northern Colorado Front Range. With the loss of irradiance, the initial ratio of NO/(NO + NO$_2$) of ~0.2 dropped steadily, bottoming out at <0.01, but rebounded to approximately two times above August median levels for this time to the end of the eclipse. Above average O$_3$ enhancements were seen in the afternoon hours following the eclipse at this and a series of other nearby surface O$_3$ monitoring sites. The contrasting behavior of these slow and fast photoreactive gases, and comparison with other published eclipse data, allow characterizing these responses as more typical for an urban/polluted environment.

Keywords: Solar eclipse; Atmospheric monitoring; Ozone; Nitrogen oxides; Photochemistry

Introduction

Atmospheric oxidation processes, convective mixing, and diurnal boundary layer growth are primarily driven by solar irradiance. The August 21, 2017, total solar eclipse crossed the United States along a west to east swatch (Figure 1) during a time of year with seasonally intense solar irradiance. This provided a rare opportunity to observe the response of atmospheric dynamics and photochemical reactions to a large sudden decline in solar irradiance during mid-day.

The response of photochemical oxidants in the atmospheric column and in surface layer concentrations due to the rapid change in solar irradiance during a solar eclipse has been studied on a number of occasions (e.g. Bojkov, 1968; Gerasopoulos et al., 2008). Research has addressed both micro and boundary layer meteorological influences as well as atmospheric trace gas behavior. The gradual loss of solar irradiance has been shown to result in cooling at the surface, and reduced convective and turbulent mixing in the surface layer (Founda et al., 2007; Tzanis et al., 2008; Hanna et al., 2015). Reductions in atmospheric concentrations have been reported for photochemically produced species, such as the instantaneously formed OH radical (Abram et al., 2000; Fabian et al., 2001). Changes have also been seen for surface ozone (O$_3$) that is formed on minutes to hours time scales (Eastman and Stedman, 1980; Chudzynski et al., 2001; Chung et al., 2010). Interestingly, even clustering processes leading to new particulate production have been seen to be suppressed during a partial eclipse (Jokinen et al., 2017). Responses have not just been noted at the surface, but even deeper into the atmosphere, such as for zonal winds, aerosol optical depth, and in the O$_3$ and NO$_x$ columns (Stranz, 1961; Bojkov, 1968; Dani and Devara, 2002; Adams et al., 2010; Dutta et al., 2011; Manchanda et al., 2012), though it has been cautioned that column measurements during a solar eclipse may be biased from the large relative increase in diffusive (over direct) solar irradiance (Zerefos et al., 2000).

We report continuous observation of meteorological conditions and photochemically reactive gases, i.e. of nitrogen oxides (nitrogen oxide (NO) + nitrogen dioxide (NO$_2$) = NO$_x$), and O$_3$ contrasted by the behavior of photochemically slow reacting species. For the latter, we utilize observations of methane and of the non-methane volatile organic compound (VOC) ethane. While ethane,
with a summertime mid-northern latitude mean atmospheric lifetime of ~1 month, is considered an \( \text{O}_3 \)-forming photoreactive compound, its reaction is slow on the scale of the changes seen during the few hours of the eclipse. Therefore, its atmospheric behavior is expected to be overwhelmingly driven by dynamic rather than photochemical effects during the eclipse. Data were obtained during the partial eclipse at the Boulder Reservoir, a monitoring site on the outskirts of the City of Boulder, Colorado. At this location, the eclipse lasted close to three hours, with a maximum solar obstruction of 93%. With a cloud-free sky during the morning hours, near ideal conditions existed for studying meteorological and photochemical responses to a rapid change in solar irradiance in a real world outside environment. We particularly focus on \( \text{NO}_x \) and \( \text{O}_3 \), which play an intricate role in photochemical oxidation, and indirectly in production of the hydroxyl radical, which both determine the oxidation and removal of most atmospheric trace gases and pollution. Turner et al. (2018) present observations of energy fluxes and changes in turbulent mixing during the August 2017 eclipse over north-central Oklahoma. To the best of our knowledge, our findings reported here are the most comprehensive data records of chemical species during a solar eclipse event in the U.S. to date.

**Methods**

Observations were conducted at a Colorado Department of Public Health and Environment (CDPHE) monitoring site at the Boulder Reservoir (40.0704 N, 105.2198 W, 1604 m asl) (Figure 1). The monitoring at this site is part of CDPHE’s statewide \( \text{O}_3 \) air quality network. In addition, atmospheric methane, VOCs, and \( \text{NO}_2 \) are monitored by the Institute of Arctic and Alpine Research (INSTAAR) through a contract from Boulder County Public Health (BCPH) for assessing atmospheric impacts from oil and natural gas development in the north to southeast upwind sectors of the site. Instrumentation is housed inside a shelter, with air being pulled to instruments from inlets mounted on a tower at 5 m (\( \text{NO}_x \), \( \text{CH}_4 \), VOCs) and 7 m (\( \text{O}_3 \)) above ground level. Data for methane, VOCs, \( \text{NO}_x \), \( \text{O}_3 \), and wind monitoring are posted in near-real time online (CDPHE, 2018; INSTAAR, 2018).

**Meteorological variables:** Ambient air temperature was measured at the Boulder Reservoir station by CDPHE with a Model 41382VC R.M. Young Company (Traverse City, MI, USA) relative humidity/temperature probe inside a model 41003 multi-plate radiation shield. Wind speed and wind direction were monitored with an RM Young Company Model 05305-5 propeller wind anemometer positioned on a tower at 5 m above ground level. Solar radiation data were monitored with a Davis Vantage Pro2 Weather station on the roof of the University of Colorado Duane Physics building at 40.008 N, 105.268 W, 1660 m asl, approximately 8.0 km southwest of the Boulder Reservoir site (Figure 1).

**Ozone:** Ozone was monitored by CDPHE using a Teledyne API Model 400E UV absorption spectrometer. Calibration and quality control follow the EPA 40 CFR Appendix D to Part 50 for \( \text{O}_3 \) analysis by UV absorption. Data were recorded at 1-minute time resolution, compiled from instantaneous readings every 6 seconds.

**Nitrogen oxides:** A Thermo Fisher Scientific (Franklin, MA, USA) Model 42TL chemiluminescence analyzer with a molybdenum oxide (MoO) catalyst heated to 325°C (Model 42C-TL) was used. The instrument has two channels. The first channel measures \( \text{NO} \) via \( \text{NO} + \text{O}_3 \) chemiluminescence. The second channel measures \( \text{NO}_2 \) by redirecting air through the MoO converter, which causes \( \text{NO}_2 \) – and other oxidized nitrogen compounds – to convert to \( \text{NO} \). The \( \text{NO}_2 \) mixing ratio is then determined by

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**Figure 1:** Top Right: Path of August 21, 2017, total solar eclipse, with the observation area indicated by the red square. Left: Study area, with the monitoring site at the Boulder Reservoir and the location of the solar radiation measurements on the University of Colorado, Boulder campus. (Map on left from https://www.google.com/maps/; map to the right from https://eclipse.gsfc.nasa.gov/SEgoogle/SEgoogle2001/SE2017Aug21Tgoogle.html). Eclipse map/figure/table/predictions courtesy of Fred Espenak, NASA/Goddard Space Flight Center, from eclipse.gsfc.nasa.gov. DOI: https://doi.org/10.1525/elementa.322.f1
subtracting NO, measured in the first channel. There are several interferences in this NO₂ measurement scheme (Steinbacher et al., 2007). The error in the NO₂ measurement increases with increasing amounts of interfering gases such as nitrous acid (HONO), peroxyacetyl nitrates (PAN), and alkyl nitrates that can contribute to the NO₂ mode signal. However, in urban environments, NO₂ typically constitutes the largest fraction of oxidized nitrogen compounds (Spicer, 1982; Steinbacher et al., 2007); hence, NO₂ mixing ratios obtained with the TEI 42C-TL represent a reasonable estimate if the site is mostly influenced by anthropogenic sources. A recent intercomparison of NO₂ measurement techniques showed that NO₂ measured with MoO converter instruments yielded values that differed from instruments using other techniques by 2–3% for measurements at a semi-rural site in Germany (Gilge et al., 2013). The instrument was calibrated monthly using a 5.07 ppm NO standard (Scott Marin, Riverside, CA, prepared June 2016) that was dynamically diluted using ultra zero air.

**Methane:** Methane was monitored with loop injection-gas chromatography (GC) and flame ionization detection (FID). The sample flow was dried with a Nafion dryer. A 3-ml temperature-controlled injection loop was used. The flow through the injection loop was stopped a few seconds before injection to allow equilibration with ambient pressure. Methane separation was achieved by isothermal GC with a packed 3.2/2.1 mm o.d./i.d. × 1.8 m length Porapack Q (Grace Discovery Sciences, Columbia, MD) column. Sample injections were made every 16 minutes, alternating with one working standard injection for every sample. The working standard was cross-calibrated against a primary methane standard obtained from and cross-referenced against the NOAA Global Monitoring Division, Boulder, CO, methane calibration scale.

**VOC monitoring:** The GC system is a further development of the system described by Tanner et al. (2006). The inlet/GC column conditions are tailored towards the quantitative analysis of C₂–C₇ VOCs; quantified compounds are ethane, acetylene, propane, iso-butane, n-butane, iso-pentane, n-pentane, benzene, toluene, ethylbenzene, and the three xylene isomers. Together, these quantified species constitute approximately 85–90% of the FID peak area within the C₂–C₇ retention time range. Air was pulled from the tower inlet, and aliquots of the sample stream were first passed through a Pelletier-cooled cryogenic water trap to dry the sample to a dew point of ~45°C. VOCs were then concentrated on a Pelletier-cooled (−40°C) multi-stage adsorbent trap. Analysis was accomplished by thermal desorption and injection onto an Al₂O₃/KCl PLOT column (Agilent, 0.53 mm × 50 m, 15 µm film) for cryogen-free separation on an SRI Model 310 GC with FID. With the 500 ml sample volume, the instrument achieves 10–20 parts per trillion (pmol mol⁻¹; ppt) detection limits. At 100 pmol mol⁻¹ mole fraction, analytical accuracy and precision errors are typically lower than 5%. Blank runs, with VOC-free zero air, and calibration runs with a primary standard were conducted every four days. The primary standard (National Physics Laboratory, Teddington, U.K.) is traceable to the World Meteorological Organization (WMO) Global Atmospheric Watch (GAW) scale for VOCs.

**Results and Discussion**

**Data presentations**

**Diurnal cycles for all August 2017 days, excluding Aug. 21, for NO, NO₂, O₃, methane, and ethane are available in Supplemental Materials (SM) Figure 1. Atmospheric mole fractions are highly variable with substantial day-to-day differences depending on emissions and atmospheric transport and dispersion, with the variability generally increasing with decreasing lifetime of the atmospheric species (Pollmann et al., 2016). For easier comparison, in Figure 2, we demonstrate the unique features seen in NO, NO₂, O₃, CH₄, and ethane by contrasting data from the August 21 eclipse day with the median diurnal cycle of the other 30 (non-eclipse) days during August 2017 (see SM_Figure 2 for the corresponding NO₂ data).

**Mean August diurnal cycles**

**Slow photoreactive gases:** Median methane (Figure 2d) exhibits a regular sinusoidal diurnal cycle, with increasing mixing ratios from late afternoon to ~6 hrs Mountain Standard Time (MST; MST = UTC − 7 hrs), and steadily decreasing mixing ratios during the day. We are not aware of methane sources that exhibit a diurnal cycle, nor would the rather slow methane oxidation by OH cause a notable diurnal signal. Therefore, we consider the pattern in these data a reflection of synoptic transport and boundary layer dynamics, where methane emissions from sources within the nighttime transport distance are trapped near the surface because of weak vertical transport, causing a gradual increase in mixing ratios. Declining methane starting shortly after sunrise is probably due to the onset of convective turbulence, causing dilution of the methane-enriched surface layer with lower methane mixing ratio air from aloft. This region also experiences regular diurnal upslope/downslope flow patterns. During nighttime, west/southwesterly flow brings in cleaner air. This transitions to upslope northeast/easterly flow during the day that typically is associated with more polluted air originating from urban, industrial, and oil and gas development areas to the east of the site (Johnson and Toth, 1982; Evans and Helmig, 2017).

The ethane (Figure 2d) August median cycle is less regular than for other gases, likely due to lower time resolution/fewer measurements going into the calculation. Ethane levels are highest during night and drop steadily during the daytime hours, likely for the same reasons as discussed above for methane.

**Reactive gases:** The August average diurnal cycles reflect the different dependency of these gases on their sources, photochemical production, and dispersion. NO, being an emission from combustion and a product of photochemical dissociation of NO₂, exhibits low mole fractions during the night (Figure 2a). The large increase seen between 5–8 MST coincides with the morning maximum of hourly traffic counts on the nearby (~700 m) Diagonal Highway (SM_Figure 3), one of the main traffic arteries into Boulder from the northeast, and is likely due to increased emissions from the onset of morning commuting traffic. Secondly, this morning increase likely reflects the onset of photochemical production of NO from the on average ~5 ppb NO₃ reservoir that persists through the night (SM_Figure 2).

**Mean August diurnal cycles**
The drop in NO and NO\(_2\) (Figure 2b) from ~8 MST throughout the early afternoon is likely due to the growth of the boundary layer, where dilution of the boundary layer with lower-NO\(_x\) containing air from aloft supersedes the rate of emissions from the surface. A very different behavior is seen for O\(_3\) (Figure 2c). After a steady drop during the night, O\(_3\) increases rapidly during morning hours, peaking at ~15–18 MST in the afternoon. This behavior reflects a steady 6–7 ppb hr\(^{-1}\) morning-to-early afternoon increase in O\(_3\). This increase likely has two contributing components, i.e. 1. mixing of higher O\(_3\)-containing air into the O\(_3\)-depleted surface layer with the onset of convective mixing in the morning, and 2. local O\(_3\) photochemical production. After ~18 MST, O\(_3\) production becomes weaker than O\(_3\) sinks (i.e. surface deposition, chemical destruction), which then causes the declining O\(_3\) mixing ratios. Boulder County is within the Northern Colorado Front Range (NCFR) O\(_3\) National Ambient Air Quality Standard (NAAQS) non-attainment area, and as such, a region of elevated O\(_3\). Elevated O\(_3\) occurrences have been shown to be largely due to regional O\(_3\) production in the NCFR (Cheadle et al., 2017; Evans and Helmig, 2017; Pfister et al., 2017).

**Behavior on the eclipse day**

Meteorological conditions: August 21 started out as a sunny day. The solar irradiance graph (Figure 3a) shows an unimpeded diurnal radiation increase in the morning, with radiation reaching 670 W m\(^{-2}\) by 9:25 MST (eclipse start was at 9:22 MST). This measurement record, indicating a cloud-free sky, is confirmed by photographs taken during the morning. Immediately after the onset of the eclipse, irradiance dropped steadily, bottoming out at 45 W m\(^{-2}\) at 10:50 MST (eclipse maximum was at 10:46 MST). After the peak of the eclipse, irradiance increased again, at about the same absolute rate as during the morning hour decline. After reaching a maximum value of 925 W m\(^{-2}\) at 11:55 MST, there was a sudden drop, most likely from clouds covering the sun. This drop in irradiance occurred 19 minutes before the eclipse ended. Since the radiation monitoring site is ~8 km southwest of the Boulder Reservoir, there is no certainty that the timing and degree of solar obstruction by clouds was exactly the same at the two sites. The sky/cloud conditions inferred from the radiation record were confirmed by citizen’s reports and inspection of photographs that were taken during the eclipse period within the Boulder area.

Non-reactive gases: Methane is monitored at ~10 times higher time resolution than VOCs. The higher number of data allow for a more robust contrasting of diurnal cycles than for the VOCs. On August 21, methane showed a nighttime maximum (Figure 2d), and then declining morning hour mixing ratios. The methane decline halted earlier than for the median monthly data and methane began increasing again after reaching its minimum at 10 MST. A notable feature is a ~100 ppb spike that lasted ~45 min right around noon at the end of the eclipse.
Reactive gases: Both NO and NO$_2$ show enhanced concentrations in the morning, with peak levels at ~7 MST (Figure 2a, b). These August 21 values exceed those of the average day by a factor of 3–4, and for both gases are the highest values observed on that day. These peaks can likely be attributed to two causes: 1. Again, the timing coincides with the morning traffic peak on Highway 119 (SM_Figure 3). 2. Winds during the August 21 morning hours were relatively weak, turning from south to the north/northeast (Figure 3d), which first placed the City of Boulder and then increasingly HW 119 and the City of Longmont upwind of the site. During the eclipse window, first, NO and NO$_2$ showed decreasing mole fractions. This behavior reversed shortly after the peak of the eclipse with a notable increase of both nitrogen gases. This increase is unusual for both species compared to the average diurnal cycle, however, it is similar to NO$_x$ observations during a solar eclipse in Southern India, where NO$_2$ and NO$_x$ roughly doubled shortly after the peak of the eclipse (Nishanth et al., 2011a; Nishanth et al., 2014). This NO$_x$ increase is likely driven by a combination of reduced vertical mixing/dilution of surface emissions and an increased NO$_x$ lifetime from a reduced oxidation rate by the diminishing concentrations of the primary NO$_x$ sink, i.e. [OH], during the eclipse.

Ozone started out at average August mixing ratios during the early part of the day (Figure 2c). Subsequently, the $O_3$ time series deviated markedly from the steady increase seen in the average diurnal $O_3$ cycle. First, there is a notable dip in $O_3$ between 6:30–7:00 MST. This coincides with the morning peak seen in NO, which suggests that this loss in $O_3$ is likely caused by reaction with NO during the the morning spike in the NO mole fraction. The typical morning increase in $O_3$ came to a halt right at the onset of the eclipse, at an $O_3$ mole fraction of ~55 ppb. Ozone then remained at a relatively stable mole fraction until the peak of the eclipse. It then decreased rapidly by some 15 ppb, indicating that the $O_3$ loss rate was higher than $O_3$ production during this phase of the eclipse. The morning drop in $O_3$ is highly unusual and deviates from the average diurnal cycle by well over the 3–σ (99%) significance level. In Figure 4, the rate of $O_3$ change was calculated in 1-min steps for 45 min time windows for each August 2017 day from ~3 hrs prior to ~3 hrs after the eclipse hour (7–14 MST; with slope values plotted at the center of the time interval). The range of the 3–σ standard deviation of all August (except eclipse day) values is indicated by the grey shaded area. The rate of the $O_3$ change (drop) during the peak of the eclipse reached a minimum slope value of ~300 ppt min$^{-1}$; this rate is more than 400 ppt min$^{-1}$ lower than the median value during that time of day and well outside the 3–σ shaded area that depicts the variability in the $O_3$ change during all other August 2017 days.

This dip coincides with the enhancement in NO and NO$_2$, suggesting that $O_3$ loss during the eclipse was to some degree caused by reaction with NO. Shortly after the peak of the eclipse, $O_3$ showed rapid recovery, increasing by ~15 ppb within ~1 hour. The behavior seen for these reactive gases is notably different than the observations for the photochemically much slower reacting methane and ethane.
Figure 5a shows the evolution of the NO/NO$_x$ ratio during the eclipse day. After peaking at ~6:30–7:30 MST at a value of ~0.35, NO/NO$_x$ dropped steadily, bottoming out at a value of 0.14, just nine minutes after the peak of the eclipse. This drop in the NO/NO$_x$ ratio is similar to that observed during an eclipse near Munich, Germany (Fabian et al., 2001), but to significantly lower values than the NO/NO$_x$ ratios of 0.2–0.3 that were observed during an eclipse in Seoul, South Korea during a similar time of day window (Kwak et al., 2011), and during an early afternoon eclipse in Southern India (Sharma et al., 2010; Nishanth et al., 2011a). During the recovery phase of the eclipse, NO/NO$_x$ increased rapidly and reached a maximum value that was approximately two times the August median noontime value within an hour after the peak of the eclipse. Figure 5b shows the NO/NO$_x$ ratio as a function of solar irradiance, with data color-coded by different periods during the eclipse day. NO/NO$_x$ shows a steady increase with solar irradiance, albeit with notable scatter in the data. Some of this scatter may be an artifact stemming from the distance between the irradiance and NO$_x$ measurement sites. A notable feature in these data is that, except during the morning rush hour NO traffic peak, NO/NO$_x$ during the occurrence of the eclipse does not deviate significantly from the period of increasing irradiance in the early morning hours, indicating that the NO$_x$ partitioning during the eclipse mostly resembles conditions during other times with corresponding levels of solar irradiance.

There was a shift in wind direction from south to north during the first half of the eclipse, and then gradually from north to northeast during the second half (Figure 3d). In the following, we further explore dependency of these gas observations on changes in radiation and convective mixing caused by the eclipse as well as air flow to the site.

Factors driving the trace gas behavior on the eclipse day

Unique features seen in the chemical eclipse data can be traced to surface/boundary layer dynamics and radiative changes exerted by the loss of solar irradiance. Atmospheric composition changes during the eclipse are closely linked to meteorological changes exerted by the eclipse-driven drop in solar irradiance. The drop in temperature (respectively the stalling of the typical morning temperature increase) observed at the Boulder Reservoir (Figure 3b) was on the order of 3°C, which is at the upper end of the range of cooling observed during other eclipses (Founda et al., 2007; Mauder et al., 2007; Chung et al., 2010; Subrahmanyan et al., 2011; Girach et al., 2012; Hanna et al., 2015). The drop in solar irradiance and surface heating has also been noted to result in increased stabilization of the surface layer, a drop in surface winds, weaker turbulent and convective mixing, and suppressed mixed boundary layer growth (Founda et al., 2007; Mauder et al., 2007; Tzanis et al., 2008; Nymphas et al., 2009; Subrahmanyan et al., 2011; Hanna et al., 2015; Kastendeuch et al., 2016). At a site in Oklahoma during the 21 August 2017 eclipse, turbulent fluxes of heat and momentum responded quickly to the decline in solar irradiance and surface heating has also been noted to result in increased stabilization of the surface layer, a drop in surface winds, weaker turbulent and convective mixing, and suppressed mixed boundary layer growth (Founda et al., 2007; Mauder et al., 2007; Tzanis et al., 2008; Nymphas et al., 2009; Subrahmanyan et al., 2011; Hanna et al., 2015; Kastendeuch et al., 2016). At a site in Oklahoma during the 21 August 2017 eclipse, turbulent fluxes of heat and momentum responded quickly to the decline in solar irradiance.

Figure 4: Mean rate of ozone change over 45 minutes time windows calculated in 1-min time steps for August 2017 (excluding the eclipse day) in orange, with the 3–σ standard deviation range shaded in grey. Results for the eclipse day are shown as the blue circle data. DOI: https://doi.org/10.1525/elementa.322.f4

Figure 5: (a) NO/NO$_x$ ratio and ozone prior, during, and after the eclipse. Medians were calculated from all data for the month, binned in 15-minute intervals. Error bars show the 1–σ variability of the 30 days of data (excluding August 21) for each bin. (b) NO/NO$_x$ on the eclipse day, with data color coded by different time periods. DOI: https://doi.org/10.1525/elementa.322.f5
solar radiation, resulting in a large decrease of turbulent mixing in the boundary layer and in stable atmospheric conditions, similar to a nighttime stable boundary layer, near the surface. The temperature decrease was most pronounced in the lowest 200 m of the atmosphere (Turner et al., 2018). Winds declined during the eclipse phase at the Boulder Reservoir (Figure 3c), but the site’s August 21 diurnal record is too noisy to clearly tie this dip to the timing of the eclipse occurrence. Unfortunately, there were no direct turbulence measurement data available from the site. Nonetheless, the temperature record, features in the wind data, and inferred reduction in convective mixing suggest overall reduced dilution of surface emissions during the eclipse period. This can likely serve as an explanation for the increases seen in NO$_3$ during, and in methane and ethane shortly after the eclipse (Figure 2). The delay in these mixing ratio changes to the timing of the eclipse is an indication of the influence of micro and macro-meteorological conditions versus the direct photochemical forcing. This effect is most clearly evident in the temperature record (Figure 3b). The temperature minimum at the Boulder Reservoir was recorded ~20 min after the peak of the eclipse. Delays of ~15 min of the temperature minimum (Founda et al., 2007) and ~30 min of atmospheric turbulent mixing (Foken et al., 2001) after the timing of peak of eclipses have been noted previously. A delay of 10–15 min was found for near-surface temperature and wind speed at three sites in Oklahoma during the August 21, 2017, eclipse (Turner et al., 2018). This time shift can be explained by the inertia of the surface heating and resulting turbulence to the incident radiative forcing.

As seen in the mean August diurnal O$_3$ cycle, on average there is significant daytime photochemical O$_3$ production at the Boulder Reservoir site. From 6–12 MST during August, O$_3$ on average increases at a rate of 6–7 ppb h$^{-1}$; the O$_3$ growth rate was even steeper at ~10 ppb h$^{-1}$ from 7–9 MST during the eclipse day. Because of the frequency of elevated summer O$_3$, the NCFR has been designated as a ‘Moderate’ non-attainment area for the O$_3$ US National Ambient Air Quality Standard (Bien and Helmig, 2018). Elevated O$_3$ occurrences are mostly due to 1. regional O$_3$ production from diverse regional NO$_x$ sources and elevated VOCs, with fugitive natural gas (and to a lesser extent oil) drilling/distribution emissions contributing significantly to the O$_3$ production chemistry (Gilman et al., 2013; Swarthout et al., 2013; McDuffie et al., 2016) (i.e. the annual mean ethane mixing ratio at the Boulder Reservoir in 2017–2018 was higher than in 27 (out of 28) large American cities in 1999–2004; SM Figure 4), and 2. by upslope (towards the west) transport of high O$_3$ containing air masses from the northeastern to southeastern sectors, where oil and natural gas (O&NG) operations constitute a major VOC emissions source (Cheadle et al., 2017; Evans and Helmig, 2017). The behavior seen in these data is typical for a receptor site. Ozone remains relatively high during the night, indicating that local sources of NO are too weak during the evening and might to titrate the O$_3$ efficiently.

Given that O$_3$ is exclusively produced by photochemistry, the decline in solar irradiance would be expected to cause a slowdown or halt in the O$_3$ growth. Remarkably, not just a halt in production, but a dramatic reduction of ~15 ppb in O$_3$ occurred. The O$_3$ drop seen in our data is on the upper end of surface O$_3$ changes that have been reported in other studies, i.e. drops of ~15 ppb in Illinois (Eastman and Stedman, 1980), ~10 ppb during a 97% solar eclipse in England (Abram et al., 2000), 10–25 ppb during a 84% solar eclipse in 2010 at sites in India (Nishanth et al., 2011a; Nishanth et al., 2011b; Girach et al., 2012), and 0–20 ppb in Seoul, Korea (Kwak et al., 2011). These responses show that in these environments O$_3$ sinks become dominant processes in determining the O$_3$ behavior. A probable explanation is that O$_3$ production ceased from the reduction in the NO$_2$ photolysis rate (j(NO$_2$)). There were no direct j(NO$_2$) measurements available for the site or nearby. Visible irradiance dropped to ~7% (Figure 3a). Due to the spectral dependence of limb darkening there is a proportionally more pronounced decrease of radiation at shorter wavelengths (Kazadzis et al., 2007; Suzuki and Yamamoto, 2013), with the loss in UV-B irradiance at 310 nm being as much as 60% more than for a hypothetical sun (Koepke et al., 2001). This implies a probable larger drop for the UV wavelength window of j(NO$_2$) (than to the 7% seen in the visible wavelength data). This would compare well with reductions to less than 5% in direct j(NO$_2$) measurements that have been recorded during other eclipses with similar solar obstruction (Fabian et al., 2001; Nishanth et al., 2011b).

Primary sinks of O$_3$ near the surface are 1. photolysis, 2. reaction with OH and HO$_2$, radicals, 3. deposition to the surface, and 4. chemical reaction with NO and other reactive gases. Ozone photolysis and reaction with OH/NO$_2$ are relatively weak O$_3$ sinks, causing a combined ~1–2 ppb diurnal O$_3$ loss in the clean atmosphere (Monks et al., 2000). Under the reduced irradiance conditions during the eclipse, O$_3$ loss by photolysis is expected to diminish to insignificant levels. Similarly, given the strong dependence of OH production on irradiance and the short OH lifetime, atmospheric OH during the eclipse is expected to drop steeply (i.e. see Burnett and Burnett, 1985; Burnett et al., 1988; Abram et al., 2000; Fabian et al., 2001) with reports on the decline of atmospheric OH during prior eclipses), and O$_3$ loss from reaction with OH is expected to be negligible. Further, O$_3$ surface deposition is expected to weaken because of the reduced convective mixing, which lowers the surface uptake rate. This leaves chemical losses, with NO and unsaturated organic compounds being possible reactants, as the most likely destruction path for the drop in O$_3$. Other previous work has found that the degree of O$_3$ loss during the eclipse is also a function of the O$_3$/NO$_2$ ratio. Comparing data from polluted and clean sites, Wu et al. (2011) found larger O$_3$ loss rates and higher NO$_2$ enhancements in polluted areas. Similar conclusions were derived in a comparison of eclipse data from four sites in Greece. At two relatively unpolluted sites there was no clear signal in O$_3$, NO, and NO$_2$, while at two urban sites, decreases in O$_3$ and NO, and an increase in NO$_2$ (Zanis et al., 2007) were observed. Evidently, under eclipse conditions, in environments with higher NO$_2$ emission, there is a higher rate of NO$_2$ buildup from the loss of turbulent mixing, increase in NO$_2$ from the NO + O$_3$ --> NO$_2$ reaction, and conversely a higher rate of O$_3$ loss. Because of these dependencies, atmospheric composition...
changes in these reactive gases during an eclipse are more evident in urban/polluted areas, whereas they may be masked by transport effects in rural environments (Zanis et al., 2001). In light of this previous literature, the Boulder Reservoir data clearly show the more ‘urban’, i.e. higher NO\textsubscript{x} environment behavior, despite the site being located in the outskirts of the city.

A striking feature is that during the end phase of the eclipse, the NO/NO\textsubscript{x} ratio rapidly increased to levels that were higher than the monthly median. Further, there is also a notable steady O\textsubscript{3} increase during the afternoon, with resulting O\textsubscript{3}~10 ppb above the monthly median. This ‘rebound’ in these photochemically dependent variables is surprising and hasn’t been reported in prior literature cited above. In Figure 6 we analyze O\textsubscript{3} records from five other nearby CDPHE-operated sites (within ~80 km, see SM_Figure 5 for locations) to examine if the O\textsubscript{3} dynamics seen at the Boulder Reservoir were a mere local phenomenon. The O\textsubscript{3} drop during the eclipse peak phase is clearly apparent at all of these sites, with drops in surface O\textsubscript{3} of similar magnitude (10–20 ppb) as for the Boulder Reservoir. There is a slight indication that the dip in O\textsubscript{3} is more pronounced at the further northern sites, which would be expected due to the larger degree of the solar irradiance drop with closer proximity of these sites to the center of the eclipse swath (Figure 1). The nearby Boulder Reservoir, Rocky Flats, and NREL sites all show an above average O\textsubscript{3} increase occurring during the mid-afternoon.

![Figure 6](https://doi.org/10.1525/elementa.322.f6)
hours, which labels this behavior as a regional effect along that section of the Colorado Front Range. With our available information it is not possible to assess how much of these differences may be due to the gradient in the loss of solar irradiance (also from variable cloud cover at these sites) versus spatial heterogeneity. Transport patterns during the eclipse phase will also need to be considered. For instance, eclipse data from a network in Seoul, South Korea, have shown larger effects downwind of the eclipse swath (Kwak et al., 2011). Hourly back trajectories from the Boulder Reservoir (SM_Figure 6) show that the general air transport flow was from the northwest during the morning (8 MST), and then rotated counterclockwise over the south before settling on consistent northeasterly flow from 11–16 MST, consistent with the inferred transport from the site wind direction measurements. The trajectories place the northeastern sector, including Weld County upwind and as the source region for air masses measured at the Boulder Reservoir during the afternoon hours. This region has been identified as a source of elevated $O_3$, with the $O_3$ production being fueled to a significant degree by O&NG emissions (McDuffie et al., 2016; Pfister et al., 2017). Therefore, it appears probable that the enhancements seen in the afternoon hours are a reflection of the particular synoptic transport conditions that brought in air from this previously identified $O_3$ production region (CDPHE, 2008; Cheadle et al., 2017; Evans and Helmig, 2017). This conclusion is confirmed by the diurnal cycles of the O&NG tracer ethane and the pentane isomeric ratio. The isomeric ratio of i/n-pentane has been shown to be a selective indicator for O&NG influence, as pentanes display lower values in O&NG emissions (0.9–1.0) compared to urban air (>2) (Gilman et al., 2013; Thompson et al., 2014). The enhancements (compared to the median monthly diurnal cycle) in ethane, and the drop in the i/n-pentane values (SM_Figure 7) seen in the afternoon, support associating the afternoon conditions to transport from the north to east upwind O&NG regions. There is also a possibility of higher than average NO emissions and $O_3$ production as a result of the high southbound traffic on roads leading into the state as a record number of people were returning back to Colorado from viewing the total eclipse in the states to the north. This hypothesis, however, is not supported by the diurnal NO record at the Boulder Reservoir (SM_Figure 2) that shows mostly average diurnal NO$_x$ mixing ratios during the afternoon.

**Summary**

Concurrent measurements of reactive and non-reactive gases, meteorological conditions, and cloud-free conditions during most of the eclipse offered a rare opportunity for the study of radiative effects on photochemistry and boundary layer dynamics in a natural outdoor setting. The loss of irradiance caused cooling of the surface air by $-3^\circ$C, and a buildup of non-reactive gases (methane, VOCs) as well as nitrogen oxides (NO, NO$_x$), are indications of weakened convective and turbulent mixing and nearby emission into the surface layer of these gases. In contrast, O$_3$ declined by $-15$ ppb compared to median August diurnal mixing ratios during the $\sim$2 hours of the first phase of the eclipse. With the loss of irradiance, the initial ratio of NO/(NO + NO$_x$) of $-0.2$ dropped steadily, bottoming out at $<$0.01, but rebounded to $\sim$twice above average levels towards the end of the eclipse. Another striking and unusual feature was the “rebound” (overshooting) of surface $O_3$ towards the end and in the hours after the eclipse. Similar $O_3$ enhancements were observed at a series of network stations along the Northern Colorado Front Range. Wind and trajectory analyses indicate that the $O_3$ enhancements seen post-eclipse are likely due to the transport from $O_3$ producing source regions northeast of the site. The relatively large response in photochemical species seen during the eclipse at the Boulder Reservoir are characteristic for an urban environment with dominant daytime $O_3$ production.

**Data Accessibility Statement**

The most recent ozone data are available from CDPHE (https://www.colorado.gov/pacific/cdphe) online. Ozone records from prior years are available at the US Environmental Protection Agency (EPA) Air Quality System (AQS) archive https://aqsdr1.epa.gov/aqsweb/aqstmp/aqsdmp/airstmp/data/download_files.html.

The methane and ethane data are posted graphically at http://instaar.colorado.edu/arl/boulder_reservoir.html#monitoring. Numerical data can be requested from the corresponding author.

**Supplemental files**

The supplemental files for this article can be found as follows:

- **SM_Figure 1a, b.** (a) Diurnal cycle of 1-min NO data for all days of August 2017 (except August 21), with median diurnal cycle overlaid in blue. (b) Same as (a) for NO$_x$. DOI: https://doi.org/10.1525/elementa.322.s1
- **SM_Figure 1c, d.** (c) Diurnal 1-hour NO data for all days of August 2017 (except August 21), with median diurnal cycle overlaid in blue. (d) Same as (c) for methane, with methane measurements performed on a 16-minute interval. DOI: https://doi.org/10.1525/elementa.322.s1
- **SM_Figure 1e.** Diurnal cycle of 2-hour ethane measurements for all days of August 2017 (except August 21), with the August median diurnal ethane cycle overlaid in blue. DOI: https://doi.org/10.1525/elementa.322.s1
- **SM_Figure 2.** Diurnal cycle of NO$_x$ on the eclipse day August 21 (filled circles) in comparison with the median diurnal cycle for August 2017 (except August 21). The time window of the eclipse is indicated by the vertical dotted lines (eclipse start and end), and the center of the eclipse is indicated by the solid line. DOI: https://doi.org/10.1525/elementa.322.s1
- **SM_Figure 3.** Averaged diurnal northbound, southbound, and total hourly traffic count on Highway 119, for August 23–24, 2016. DOI: https://doi.org/10.1525/elementa.322.s1
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Competing interests

Representatives of CDPHE were contacted for permission to use the agency’s O\(_3\) data in this publication. While they approved this request, this approval does not imply endorsement of any of the analyses and/or conclusions that are presented in this work. An Elementa EIC is an author on this publication. The anonymous peer-review of the manuscript was handled by the Elementa Sustainable Engineering Domain EIC, without any influence of the EIC-author on the peer-review, and with preserving the anonymity of the reviewers.

Author contributions

• DH: Project management, data quality control, data analyses, manuscript preparation.
• BB: Data processing, data quality control, graphical data analyses.
• JH: Instrument development, calibration, maintenance.

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