



Short communication

Ozone production by corona discharges during a convective event in DISCOVER-AQ Houston



Alexander Kotsakis^a, Gary A. Morris^b, Barry Lefer^c, Wonbae Jeon^a, Anirban Roy^a, Ken Minschwaner^d, Anne M. Thompson^e, Yunsoo Choi^{a,*}

^a Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, USA

^b St. Edward's University, School of Natural Sciences, Austin, TX, USA

^c National Aeronautics and Space Administration Headquarters, Washington DC, USA³

^d New Mexico Tech, Department of Physics, Socorro, NM, USA

^e National Aeronautics and Space Administration, Goddard Space Flight Center, Greenbelt, MD, USA

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ABSTRACT

An ozonesonde launched near electrically active convection in Houston, TX on 5 September 2013 during the NASA DISCOVER-AQ project measured a large enhancement of ozone throughout the troposphere. A separate ozonesonde was launched from Smith Point, TX (~58 km southeast of the Houston site) at approximately the same time as the launch from Houston and did not measure that enhancement. Furthermore, ozone profiles for the descent of both sondes agreed well with the ascending Smith Point profile, suggesting a highly localized event in both space and time in which an anomalously large enhancement of 70–100 ppbv appeared in the ascending Houston ozonesonde data. Compared to literature values, such an enhancement appears to be the largest observed to date. Potential sources of the localized ozone enhancement such as entrainment of urban or biomass burning emissions, downward transport from the stratosphere, photochemical production from lightning NO_x, and direct ozone production from corona discharges were investigated using model simulations. We conclude that the most likely explanation for the large ozone enhancement is direct ozone production by corona discharges. Integrating the enhancement seen in the Houston ozone profile and using the number of electrical discharges detected by the NLDN (or HLMA), we estimate a production of 2.48×10^{28} molecules of ozone per flash which falls within the range of previously recorded values (9.89×10^{26} – 9.82×10^{28} molecules of ozone per flash). Since there is currently no parameterization for the direct production of ozone from corona discharges we propose the implementation of an equation into a chemical transport model. Ultimately, additional work is needed to further understand the occurrence and impact of corona discharges on tropospheric chemistry on short and long timescales.

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1. Introduction

Ozone plays an important role in tropospheric chemistry and driving global climate change. The well-known sources of tropospheric ozone are through photochemistry and stratosphere-troposphere exchange (STE). The sources for photochemical ozone formation are reactions of nitrogen oxides with volatile organic compounds (VOCs) in the presence of ultraviolet radiation. Anthropogenic and biogenic sources constitute the majority of NO_x

emissions, with lightning accounting for 10–20% of the global NO_x (Lee et al., 1997). Photochemical production of ozone from lightning NO_x has been well documented in numerous field and remote sensing studies (Lelieveld and Crutzen, 1994; Martin et al., 2007). The amount of ozone produced by lightning NO_x has been estimated to be 1.5 times that transported from the stratosphere (Price et al., 1997).

While ozone is photochemically produced from electrically active storms by lightning NO_x, it can also be produced directly by corona discharges. Compared to high-energy discharges such as cloud-to-ground and cloud-to-cloud lightning, corona discharges occur at lower energies, thus occurring before the conduction breakdowns that lead to lightning strikes. Corona discharges

* Corresponding author.

E-mail address: ychoi23@central.uh.edu (Y. Choi).

generally will occur on the streamer tips of charge draining regions. Streamers are what connect the areas of opposite charge to each other prior to a lightning discharge. Shlanta and Moore (1972) observed ozone concentrations that were 2.2 and 2.6 times larger than pre-storm surface concentrations just beneath a thundercloud and at an altitude of 6 km respectively. Other observations have shown minimal net production of ozone from corona discharges around thunderstorms due to the significantly larger amounts of NO_x that can also be produced by higher energy discharges (Sisterson and Liaw, 1990). Ozonesonde, aircraft, and ground-based observations around thunderstorms have shown ozone enhancements ranging from 12 to 30 ppbv that was attributed to production by corona discharge (Bozem et al., 2014; Minschwaner et al., 2008; Winterrath et al., 1999).

This study investigates an exceptionally large ozone enhancement observed from an ozonesonde launched near an electrically active convective cell during the National Aeronautics and Space Administration's (NASA) Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) field campaign in Houston on 5 September 2013. To accurately identify the large source of ozone in this study we analyze data from ozonesondes, radar, a lightning mapping array and model simulation output. The findings of this study help in further understanding a source of ozone around convection that has been scarcely observed.

2. Data and methods

2.1. Measurements

Electrochemical concentration type (ECC) (Komhyr, 1986; Komhyr et al., 1995) En-Sci 2Z-V7 ozonesondes (Droplet Measurement Technologies, Boulder, CO) were launched at least once a day from two sites in the Houston, TX area as part of NASA's DISCOVER-AQ field campaign, which took place during September 2013. The primary operating principle of ECC ozonesondes involves an anode and cathode chamber, which contain different concentrations of potassium iodide solution. When ozone enters the cathode chamber, an iodide-iodine redox reaction occurs creating a current, which can be used to derive an ozone concentration. All of the ozonesondes in this study used 0.5% buffered potassium iodide cathode solution as recommended by Smit et al. (2007). With a typical rise rate of ~5 m per second and measurement response time of 20–30 s, the effective vertical resolution of the ozone measurement is 100–150 m. Two launch sites were maintained during DISCOVER-AQ Houston. The first launch site was located at the University of Houston-Main Campus (29.72° N, 95.34° W), an urban site ~3 km south-southeast of downtown Houston and ~8 km west-southwest of the highly industrialized Houston Ship Channel. The second launch site was located at Smith Point, TX (29.55° N, 94.78° W), a rural marine site located on the east side of Galveston Bay. On the afternoon of 5 September 2013, an ozonesonde was launched from Smith Point (SP), TX at 13:20 local time (18:20 UTC) and another ozonesonde was launched from the University of Houston (UH) at 13:47 local time (18:47 UTC). The University of Houston ozonesonde took approximately 57 min to ascend and 45 min to descend and the Smith Point ozonesonde took approximately 113 min to ascend and 27 min to descend. The difference in flight time between the two sites is due to UH launching the ozonesonde with a 350 gram balloon and SP launching with a 600 gram balloon. The larger 600 gram balloon can fly approximately 5 km higher than the 350 gram balloon based on the manufacturer listed burst height specifications, so total flight time with a 350 (600) gram will be shorter (longer). These launch sites are separated by 57.8 km (36 miles), and the launches occurred

within 27 min of each other. Each ozonesonde was coupled with an iMet-1-RSB radiosonde (International Met Systems, Grand Rapids, MI), which measured parameters such as temperature, relative humidity, and pressure. A global positioning system (GPS) on the radiosonde provided latitude, longitude, altitude, and wind speed and wind direction derived from the GPS data.

2.2. Lightning and radar data

Data from the Houston Lightning Mapping Array (HLMA) (Cullen et al., 2008) provided horizontal and vertical distribution of lightning discharges. Additional raw lightning data from the U.S. National Lightning Detection Network™ (NLDN) (Cummins et al., 1998) provided data for our ozone calculation. Both the HLMA and NLDN measure cloud-to-ground and cloud-to-cloud lightning discharge by measuring the Very High Frequency (VHF) impulse from the electrical breakdown and lightning propagation processes (Cullen et al., 2008). Base radar reflectivity and enhanced echo top data from the Houston National Weather Service (KHGX) WSR-88D Radar was used for tracking the movement, size, height, and strength of convection.

2.3. Model set up

This study employed the United States Environmental Protection Agency (USEPA)'s Community Multi-scale Air Quality (CMAQ) model V5.0.2 (Byun and Schere, 2006). Previous studies have employed the model extensively for simulating and investigating air quality issues around southeastern Texas (Czader et al., 2015; Li et al., 2016). Meteorological inputs were developed using the Weather Research and Forecasting (WRF) model V3.7 (Skamarock and Klemp, 2008). The model domain was set up over the contiguous United States covering 470×310 horizontal grid cells. Based on this domain setup, CMAQ and WRF featured a 12 km horizontal grid and a vertical grid extending to 20 km. The USEPA's National Emission Inventory (NEI) for 2011 was used for anthropogenic emissions; these were prepared for modeling using the Sparse Matrix Kernel Emissions Modeling System (SMOKE) V3.6 (Houyoux et al., 2000). Biogenic emissions were estimated using BEIS3 (Biogenic Emission Inventory System version 3) (Pierce et al., 2002). Biomass burning (BB) emissions were taken from the National Center for Atmospheric Research (NCAR)'s (FINN V1.5) Fire Inventory (Wiedinmyer et al., 2011). The inventory was not provided for CMAQ chemical mechanism, and thus the FINN MOZART-4 mechanism was mapped into CB05/AERO6. The model used motor vehicle emissions inputs from the USEPA's Motor Vehicle Emissions Simulator (MOVES) model. The chemical boundary conditions of the domain were taken from the GEOS-Chem output (v10).

3. Results and discussion

The comparison of the observed and modeled vertical ozone profiles for the base (without biomass burning), biomass burning added, and lightning NO_x cases are plotted in Fig. 1. The UH and SP ascent profiles show significant differences in ozone concentrations with the largest differences occurring in the free troposphere (Fig. 1c). The SP ascent profile shows ozone remaining below 100 ppbv throughout the troposphere, while UH ascent profile shows ozone concentrations consistently over 100 ppbv above 4.5 km and over 150 ppbv above 6 km. The Houston profile record (2004-present) does not contain another example with such a large enhancement in the troposphere over such a depth and compared to literature values; such an enhancement appears to be the largest observed to date. While we expected that the boundary layer ozone concentrations between the two sites would be different given

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