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ABSTRACT Measuring plant health is a key aspect in maximizing crop outputs. One often overlooked risk to crop fields is damage caused by stomatal ozone uptake; measuring this uptake is an important tool in understanding crop losses. Traditional methods for measuring plant ozone uptake are prohibitively expensive and rely on equipment that cannot easily be moved. Here, we propose high-altitude weather ballooning as a cost-effective alternative for measuring ozone uptake on a regional (~10 km) spatial scale. Ozonesonde data was obtained with weather balloons launched from the National Oceanic and Atmospheric Administration research station in Boulder, Colorado. This data was then compared to back trajectory data to determine if wind blowing from over cropfields from the east had a significant increase in ozone concentration with altitude, indicating uptake. While initial results seemed promising, the results were compromised by the complex meteorology, terrain and landuse near Boulder, including the large urban area near Denver to the southeast and the mountain region to the west. The complexity of the local area confounded any possible relationships in the data. However, promising initial results and ozone concentration patterns indicate the potential of this new method, if performed in a more suitable area.

INTRODUCTION

As the human population continues to grow, crop production will need to keep up to feed the increasing number of people. Unfortunately, it has become more and more difficult to find new land for farming, and in the face of impending climate changes, new methods must be found to increase crop productivity on current fields. Air pollution, particularly from fossil fuel combustion, damages crops. Reducing and controlling air pollution can be a reliable way to increase crop yields without having to expand fields to new lands.

Ozone, an important pollutant produced by power production and transportation sources, has been shown to damage plant health significantly. This damage results in decreased efficiency of photosynthetic processes, resulting in a decrease in carbon assimilation and premature leaf senescence. Studies by Avnery et. al (2011) estimated that crop yields were reduced by 15%

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in the year 2000 and predicted that crop damages in 2030 could cost from 11 to 35 billion USD due to air pollution. Hong et. al (2020) also found in that most crops grown in California showed a negative correlation between crop yield and ambient ozone, with some crops, such as table grapes, showing roughly 20% reduced crop yields due to increased ambient ozone. Additionally, damages due to ozone reduce water use efficiency, increasing demand for crop irrigation (Peng et. al, 2020). This may result in droughts having more severe effects on crop production and yields.

Ozone can cause significant damage to plants, reducing crop yields. Finding accurate, cost-effective methods to measure ozone uptake provides critical information that can be used to develop strategies to minimize ozone uptake. Additionally, stomata are thought to close during droughts, which would impact ozone uptake (Averney et. al, 2011a & 2011b). Direct measurements of ozone flux could confirm if stomata do close during droughts, as the ozone uptake via plants is directly due to open stomata. This would increase the understanding of the interactions between drought, ozone uptake, and plant damage. Measuring ozone concentrations directly has long been simplified using satellite technology, such as the Total Ozone Mapping Spectrometer launched by NASA in 1996 (NASA, 2016), measuring ozone uptake by plants (flux) has historically been much more difficult. Despite this difficulty, measuring flux is more important since ozone flux is directly related to plant damage and crop loss. Primary methods of measurement include aircraft measurements (Desjardins et. al, 2016), models such as the Ozone Deposition Model developed by Bassin et. al (2004), or surface ozone flux monitoring towers. However, aircraft measurements are expensive, models are heavily dependent on often specific factors in the modeled ecosystem. Bassin et. al found that the Ozone Deposition Model, while effective for wheat under wet conditions, was less accurate during drier conditions (Bassin et. al, 20). and surface measurement towers, while effective, are both expensive and spatially sparse since they are fixed in one location.

This research seeks to fill the gap in ozone flux measurement by proposing High-Altitude Balloons (HABs) as a suitable platform for measuring landscape-scale ozone flux into crops. Previous research by Bouche et al. (2016) has found HABs to be suitable for measuring carbon dioxide flux from agriculture, and this research aims to emulate this success for ozone. HABs would provide an inexpensive and easy method of measuring ozone flux at low altitudes on an intermediate scale of 10s of km. If successful, this study would provide a method for researchers to measure and hence model ozone flux, which can inform crop science studies and drive policy and farming decisions in the future. Increasing our understanding of how plants uptake ozone could provide information on if ozone uptake via plants results in significant enough damage to warrant changes in farming practices, such as crop selections, as some crops may be more sensitive to ozone than others. Additionally, this may drive policy to limit ozone emissions due to potential crop losses

RESEARCH PLAN PRIOR TO COVID-19

Our research was planned to be conducted in person but was disrupted by the pandemic. We were intending to launch HABs near Pontiac, IL (40.88 N, -88.63 W), with sensors for altitude, GPS, ozone concentration, and carbon dioxide concentration. This data would then be used to calculate the gas flux between the surface and the atmosphere (Equation 1).

$$F_s = k * \frac{dC}{dZ} \quad (1)$$

Where F_s is surface flux, k is the atmospheric eddy diffusivity for ozone, and dC/dZ is the vertical gradient in concentration of the gas. This surface flux does not include ozone flux caused by near-surface air chemistry. The vertical gradients of ozone and carbon dioxide would have been measured by the HAB experiment. The atmospheric diffusivity is a property of the atmosphere that changes depending on atmospheric conditions. To determine k, the flux would have been calculated for carbon dioxide, using methods provided in Bouche et. al (2016). This flux, along with the vertical gradient in carbon dioxide gathered from the lower part of the flight data, could then be plugged into the equation to solve for the atmospheric diffusivity.

This value for k could then be plugged into the equation with the measured dC/dZ for ozone and the ozone surface flux could be calculated. While the k value is partially dependent on the gas species it is measured for, this would provide a reasonable approximation of surface flux. This technique is similar to the traditional Flux-Gradient technique, which uses the eddy diffusivity of specific heat as an estimate for the eddy diffusivity of ozone (Clifton et. al, 2020).

REMOTE RESEARCH MODIFICATIONS AND REVISED RESEARCH PLAN

Due to the onset of COVID-19 which prevented us from launching balloons, our research had to be done remotely using open data. We searched for available ozone profile data near crop fields, and found Boulder, Colorado to be the best location of data, however Boulder only had crop fields to the east. Additionally, the research station in Boulder was primarily used for measuring stratospheric ozone. We would need to limit the height of the profile data to the troposphere. Boulder is not directly located on top of crop fields. In order to measure crop uptake, we related the Ozonesonde data measured with wind trajectories calculated through HYSPLIT, a model run through NOAA to calculate wind trajectories. The location, Boulder, CO, as well as the time of the ozonesonde launch were inputted into HYSPLIT, which then calculated the wind trajectory at the time of the ozonesonde launch. Boulder is located to the east of the great plains; we hypothesized that on days when the wind blew from the east, and therefore from over crop fields, the ozone concentration in the lower troposphere would be significantly lower, indicating ozone uptake from the crop fields (Figure 1). We used the ozone profile data from Boulder, combined with calculated wind trajectories through HYSPLIT. We then did further analysis using archived weather station data near Boulder to discern if temperature had any impact on ozone uptake. If the HAB setup can detect uptake of surface ozone, the concentration of ozone at the surface should be lower than the concentration of ozone slightly above the surface. That is, dC/dZ should be greater than zero. Our study, focusing on ozone uptake at the surface, was interested in the change in ozone concentration with height at the surface.

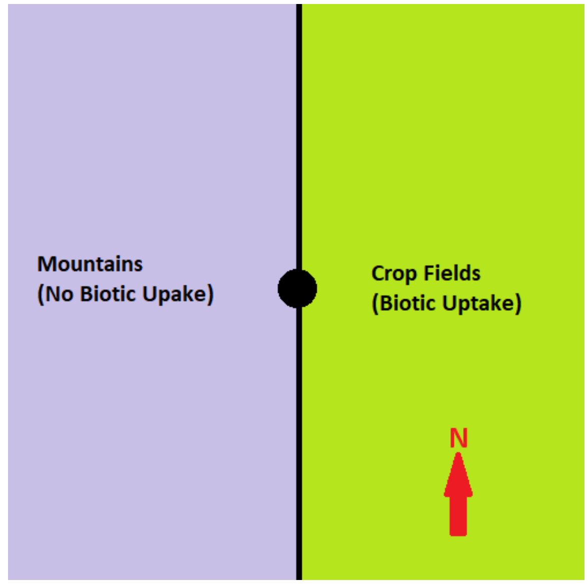

Figure 1: A diagram visualizing our wind trajectory hypothesis. Boulder is located in the center, represented by the black dot. The mountain terrain to the West corresponds to no biotic ozone uptake. From the east, the wind blows over crop fields, representing biotic ozone uptake. Thus, when the wind blows from the east, the ozone concentrations at the surface should be lower.

METHODS

OZONESONDE DATA

The Global Monitoring Laboratory (GML) managed by the Earth System Research Laboratories, a division of the National Oceanic and Atmospheric Administration (NOAA), measures atmospheric ozone profiles out of Boulder, Colorado. To do this, the GML launches weekly HABs with Ozonesondes and radiosondes. Radiosondes measure temperature, pressure, and humidity. An Ozonesonde is a small device that pumps air into a cell containing potassium iodide. The potassium iodide reacts with ozone in the air and produces electricity. The Ozonesonde records the ozone concentration measured every 100 meters during the ascent. The Ozonesonde records data from ground level to between 25 and 30 km above sea level. The recorded dataset is then provided online (Bryan et. al, 2018). The dataset includes altitude, pressure, humidity, temperature, and ozone quantity in pressure (absolute) units, concentration units, and Dobson units. Our experiment was primarily focused on surface level uptake and used relative volumetric concentration for the ozone concentration data (ppmv).

OZONESONDE CALCULATIONS

The Ozonesonde data provides ozone concentration in ppmv; however, our study was focused on measuring surface uptake from plants. To calculate this uptake, it is necessary to look at the change in ozone concentration with height at the surface level: dC/dZ. Our study, focusing on ozone uptake at the surface, was interested in the change in ozone concentration at the surface. Each Ozonesonde profile, the distribution of ozone concentration with height in the atmosphere, which according to Eqn. 1 requires that ozone concentration increase with increasing altitude. To calculate this from the Ozonesonde data, data points, starting from the ground level, were put into a set, following the relationship

$$N_1 > N_{i-1} \qquad 2$$

Where Ni is the current data point in the Ozonesonde data, and Ni-1 is the previous data point. Starting from ground level, every data point was checked with this equation. If a data point obeyed the relationship, it was added to the list; when a data point did not follow the equation, the process ended with the current set of data points. A linear regression was then performed on this set of data points to calculate a linear equation, C=mZ + constant, where Z is the altitude, and C is the concentration of ozone, and m is dC/dZ. The slope is proportional to the uptake of ozone by the surface (Eqn. 1); a larger m value corresponds to more ozone uptake at the surface. If the data for any particular day had an i value of greater than or equal to five, the day was considered “optimal” (Figure 2). If a day had an i value between two and four, the day was considered “sub-optimal” (Figure 3). If a day had an i value of one, there was no slope calculated, as a linear regression cannot be calculated on one point (Figure 4). Ozone was either constant with height or decreased with height in those situations.

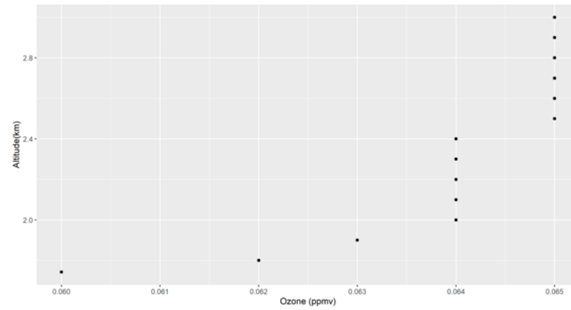

Figure 2: An example of an optimal Ozonesonde graph. The steady increase in ozone concentration with height up until 2.2 km indicates possible surface uptake by plants.

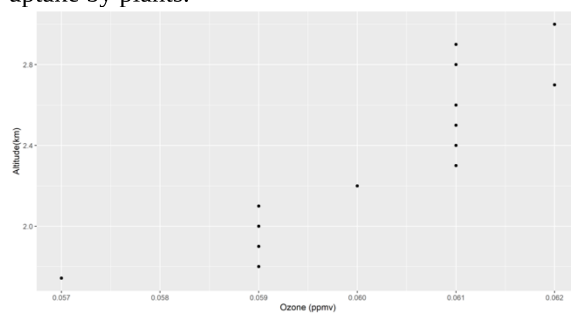

Figure 3: An example of a suboptimal Ozonesonde graph. The lack of a steady increase at low altitudes indicates a lack of ozone uptake at the surface.

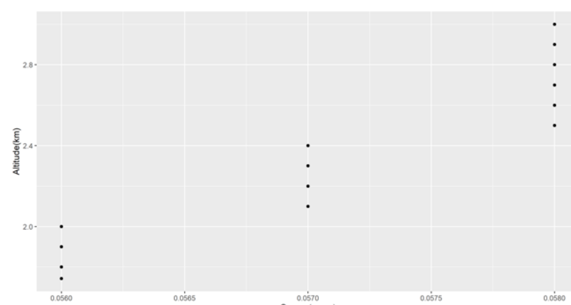

Figure 4: A “No Slope” Ozonesonde graph. The constant ozone concentration at the surface means that a positive slope was not present and there was no surface deposition.

WIND DATA

Air parcel source direction was obtained by using HYSPLIT to calculate backwards wind trajectories starting from the coordinates of the Ozonesonde launch site. HYSPLIT is a model that calculates air parcel movement such as forward and backward wind trajectories, smoke forecasting, and pollution dispersion (Stein et. al, 2015 & Rolph et. al, 2017). The HYSPLIT model works through a combination of Eulerean and Lagrangian methods for calculation. HYSPLIT first takes Eulerean atmospheric motion data from a separate system, the Global Data Assimilation System (GDAS). GDAS gathers atmospheric data from the numerous surface weather stations and radiosondes deployed across the continental United States and assimilates the data with an atmospheric transport model. The atmospheric motion data is transformed into an XYZ grid of the atmosphere, with the meteorological and dynamical state of the atmosphere defined at each point on the grid. This grid is the basis for HYSPLIT's calculations. Our research then used wind trajectories going backwards in time. HYSPLIT uses a Lagrangian approach for calculating the trajectories from its Eulerean grid. The desired ending location, starting time, and trajectory duration are input into HYSPLIT. The model then begins at the ending destination and calculates the trajectory of the wind blowing to the end destination, going backwards in time. The model produces the history of the wind trajectory backwards with a time step of one hour, for a number of steps until the total trajectory duration is reached. For our experiment, the trajectory duration was 6 hours, so each wind calculation had six steps located 1 hour apart. The calculated positions are outputted as a KMZ file, a file type commonly associated with Google Maps. These trajectories were calculated for every day that had Ozonesonde profiles, for the same location and time of day as the launch. The KMZ data was then analyzed by RStudio to find the air parcel source direction. For each profile, the coordinates of Boulder, CO, and the six-hour back-trajectory location were used to calculate the source angle. These angles were binned into four groups (0 – 90 °, etc): NE, SE, SW, NW.

WEATHER DATA

The National Centers for Environmental Information (NCEI), a newly merged group of environmental data centers, is the primary data measuring system used by NOAA. NCEI manages surface weather stations throughout the United States, measuring meteorological variables, such as temperature, dew point, humidity, atmospheric pressure, and more. This data is available at a variety of timescales and we employed the daily summaries. The closest station to the launch site is located at the Denver International Airport. The data included minimum, maximum, and average daily temperature and was matched to Ozonesonde profile launch dates.

STATISTICAL TESTS

Statistical tests were performed using R version 4.0.2 (R Core Team, 2020) in R-Studio version 1.3.1093 (RStudio Team, 2020). Several packages were used for data importation and analysis, including dunn.test (Dinno, 2017), ggplot2 (Wickham, 2016), readr (Wickham, 2018), stringr (Wickham, 2019) and sf (Pebesma, 2018). Daily max temperature, ozone slope, ozone value, and wind angle values were all tested for normality with Shapiro-Wilk tests. Kruskal-Wallace tests were run between daily max temperature and wind angle bins, and ozone slope and wind angle bins. A spearman's ranked correlation test was run between ozone slope and daily max temperature. Kruskal-Wallace tests were run between the wind angle bins on optimal days and the temperature on optimal days, and the wind angle bins on optimal days and the ozone slope on optimal days.

RESULTS

	Optimal	**Sub optimal**	**No Slope**	**Total**
Northeast	4	9	2	15
Southeast	0	7	5	12
Northwest	3	19	8	16
Southwest	5	8	3	30
Total	12	43	18	73

Table 1: The Total of Optimal, Suboptimal, and No Slope days per Wind Direction.

The wind data was broken up by direction (Table 1). A total of 73 days were reported, 18 of which had no slope, 43 had a suboptimal slope, and 12 days had an optimal slope. This indicates that HAB was able to detect an uptake approximately 75% of the time.

The results of the statistical tests are listed in Table 2. Each of the main variables, daily max temperature, ozone slope, ozone value, and wind angle, were tested for normality with the Shapiro-

Wilk test. Daily max temperature, ozone slope, and wind angle were found to be not normal. Ozone was found to be normal.

Ozone slope appears to be slightly lower when the wind is coming from the southeast (Figure 5), however this difference is not statistically significant (p=0.1646) (Table 2). A lower ozone slope indicates a lower dC/dZ value (Equation 1), which implies a lower flux. A lower flux from the east is contrary to our hypothesis, which predicted higher ozone flux from the agricultural fields to the east. This led to the exploration of temperature. There should be no correlation between temperature and wind direction. If the temperature of Boulder was lower on days when the wind came from the southeast, it could indicate the possibility of a synoptic-scale meteorological effect driving the change in both ozone slope and temperature. A Kruskal-Wallace test was run between daily max temperature and wind angle bins, and a significant relationship was found between the wind direction and daily max temperature. A Dunn's Test for Multiple Comparisons was run to determine the relationship. Wind from the southeast was found to have a significantly different temperature compared to wind from the northeast, northwest, and southwest (Figure 6). A Spearman's rank correlation test was then run between daily max temperature and ozone slope, to test for a correlation. A slight positive correlation was found, with a rho value is 0.285, and a p value of 0.03659.

We tested whether days that had optimal ozone slopes preferred a certain wind direction. When the data was limited to only days with optimal ozone slopes the results were again not significant (p values of 0.42 and 0.97, respectively). There was only a total of 13 days with optimal ozone curves, and none of which were on days with wind from the southeast, limiting the significance of statistical tests due to the small sample size (Table 1).

Table 2: Results and Significance of Statistical Tests between Variables.

	Variables	**Statistical Test**	**P Value**	**Significance**
1	Daily Max Temperature	Shapiro-Wilks	0.01802	Data not normal
2	Ozone Slope	Shapiro-Wilks	0.0004151	Data not normal
3	Ozone Value	Shapiro-Wilks	0.1013	Data normal
4	Wind Angle	Shapiro-Wilks	$2.048*10^{-5}$	Data not normal
5	Daily Max Temperature vs Direction	Kruskal-Wallace Test	0.001444	Temperature varies with wind angle bins
6	Ozone Slope vs Wind Direction	Kruskal-Wallace Test	0.1646	No relationship
7	Daily Max Temperature vs Ozone Slope	Spearman's rank correlation	0.03659	Rho = 0.285, slight positive correlation
8	Optimal Days Daily Max Temperature vs Wind Direction	Kruskal-Wallace Test	0.3278	No relationship
9	Optimal Days Ozone Slope vs Wind Direction	Kruskal-Wallace Test	0.6953	No relationship

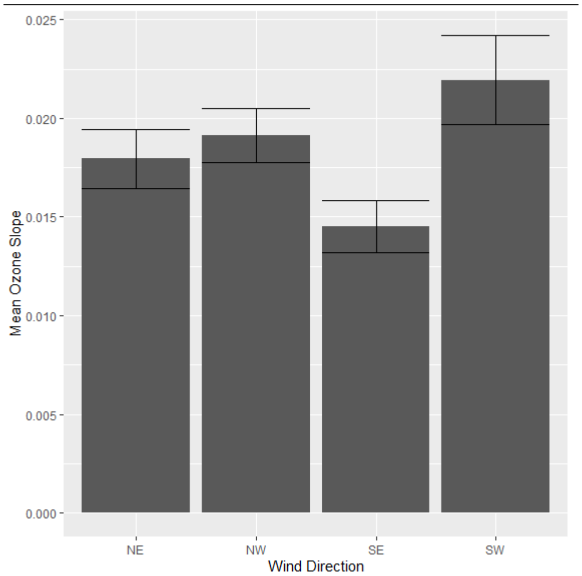

Figure 5: A graph of Ozone Slope by Wind Direction, with standard error. While the mean slope from the southeast looks lower than the mean slope from other directions, the difference is not significant (p=0.1646)

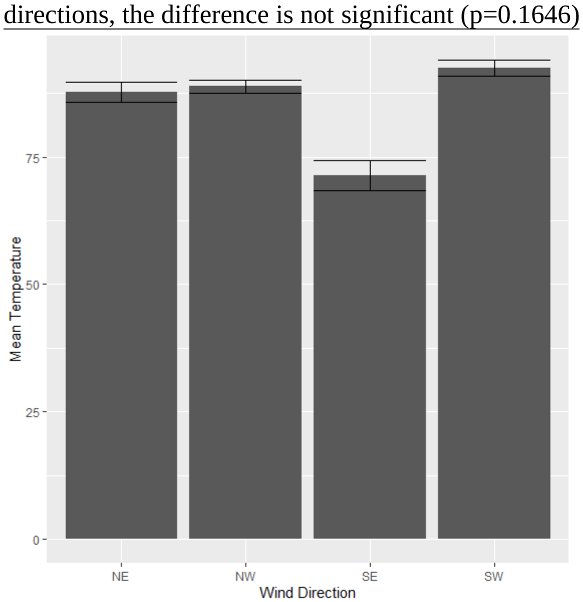

Figure 6: A graph of Mean Daily Max Temperature by Wind Direction, with standard error. The mean temperature on days with wind blowing from the southeast is significantly different than the temperature on days with wind blowing from the northeast, northwest, and southwest (p=0.001444)

DISCUSSION

Our analysis yielded mixed results regarding our hypothesis. The Ozonesonde data for some days had a clear uptake slope at the surface. This is a promising result and supports using ballooning as a method of observing surface uptake. However, the wind direction data did not support our hypothesis. Only 22% of days resulted in an optimal uptake result, and there was no increase in uptake on days where the wind blew from the east, blowing over the crop fields in the Great Plains (Figure 1). The lower average ozone slope from the southeast was contrary to our hypothesis of higher uptake from the east and agricultural crops. This led to the exploration of temperature as a confounding variable. For a valid test of the hypothesis, the temperature of the wind should not vary with wind direction.

However, temperature was found to be significantly different when the wind came from the southeast. Additionally, the ozone slope was found to have a slight correlation with temperature. This confounds our results and obfuscates any true correlation between wind direction and ozone slope. One possible source of interference could be the close proximity of Denver. The production of NO from fossil fuel combustion in the city has a strong impact on the cycling of tropospheric ozone (Figure 7).

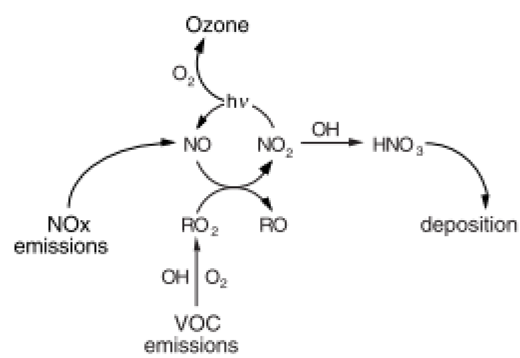

Figure 7: A simplified diagram of the effect of VOCs on ozone levels in the lower troposphere. (Adapted from Ryerson et al. 2001.)

Fossil fuel combustion in Denver is a source of NO, which could chemically quench ozone (Equation 3). NO2 would later photochemically dissociated back into NO and produce O (Equation 4), which would combine with diatomic oxygen to form O3 (Equations 5). While NO emissions ultimately lead to ozone production (Figure 7), a decrease of ozone is initially observed proximate to NO sources (Ryerson et al, 2001).

$$\mathrm{NO} + \mathrm{O_3} \rightarrow \mathrm{NO_2} + \mathrm{O_2} \qquad (3)$$

$$\mathrm{NO_2} + h\nu \rightarrow \mathrm{NO} + \mathrm{O} \qquad (4)$$

$$\mathrm{O} + \mathrm{O_2} \rightarrow \mathrm{O_3} \qquad (5)$$

Denver is situated approximately 35km from the Ozonesonde launches in Boulder, so this could explain the reduction in ozone. Ultimately, the necessity to include wind direction as a factor ultimately confounds our experiment. While Boulder had all the required data for our experiment, it is a suboptimal location. The close proximity of a major city interferes with observing surface-uptake ozone processes. Additionally, the only close farmland is located to the east, requiring the wind to blow from the east for data to be relevant. Furthermore, the mountains situated to the west of Boulder could also impact the local meteorology, further confounding the data gathered. As wind blows over the mountains from the west and down into Boulder, it increases turbulence mixing the atmosphere more than usual. This results in the ozone profiles that are driven by vertical mixing rather than surface uptake, resulting in abnormal ozone concentration slopes. Despite these complications, the presence of optimal ozone curves in some of the data is promising and shows support HAB as a method for measuring surface level ozone uptake.

The location of the ozone profiles in Boulder introduced two potentially confounding variables: ozone reactivity and mountain meteorology. The initial hypothesis relied on the wind to have moved across crop fields, but the geography of Boulder violates this assumption to the west. This introduced the complication of relying on wind direction, which both introduced bias and limited the data available. While the problem of nearby mountainous terrain would be moot with a proper launch location, the issue of chemistry associated with NO emissions could be more problematic. Denver's urban environment is a source of NO and this could lead to ozone quenching (Equations 3-5). A quarterly air quality report done by the City of Denver shows hourly NO2 emissions on average between 20 and 40 ppb (Figure 8). NO emissions are generally included in NO2 emissions, as the two chemicals are largely produced from the same sources and are present together. The observed increase of ozone with altitude could be due to chemical loss near the ground due to NO sources, not direct surface deposition into plants (Clifton et. al, 2020).

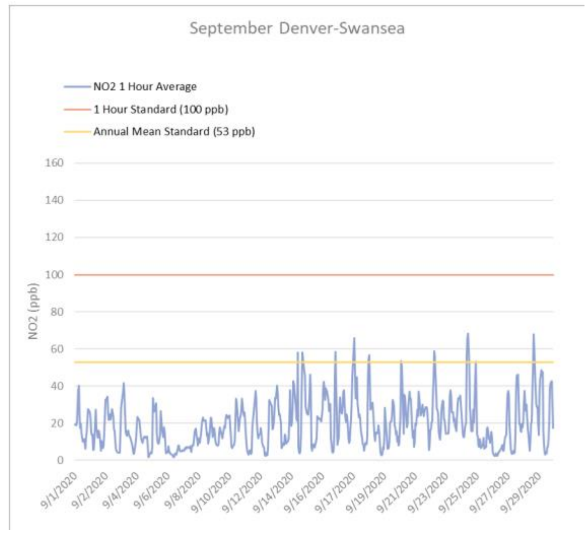

Figure 8: A report of NO2 emissions in Denver in September 2020. The blue line indicates average hourly emissions, ranging from 0 to 40 ppb, with occasional spikes to 60. The yellow line is the year mean ambient NAAQS (National Ambient Air Quality Standard). The red line is the 1 hour NAAQS, 100 ppb.

However, this method does show promise. One of the main issues of using a Flux-Gradient method for determining ozone flux is that the primary method of calculating the eddy diffusivity for ozone is by calculating the eddy diffusivity for sensible heat using the Monin-Obukhov Similarity Theory (Clifton et. al, 2020). However, this theory does not strictly hold in the layer of air right above the vegetation where it is often utilized (Clifton et. al, 2020). This is one major advantage of using a HAB platform, as Monin-Obukhov Similarity Theory would still hold. Additionally, because a ballooning platform can hold multiple sensors, it would be possible to calculate the eddy diffusivity using another gas. This is what we had previous planned prior to COVID-19; the diffusivity of carbon dioxide is much easier to calculate than sensible heat, so using it as a substitute would be an easy alternative to traditional Flux-Gradient methods.

If HAB worked, this would provide much more knowledge about ozone uptake over crop fields. HAB is comparatively cheaper and easier than traditional methods used to calculate ozone uptake. This could be used to gather data over larger areas of crop fields, furthering our understanding of ozone interactions with crop health. HAB data could also be paired with satellite crop health data, to look at the correlation between crop health and plant ozone uptake

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