AGRICULTURAL AEROSOLS

The Impact of Farming Activity on Ice Nucleating Particles

ABSTRACT

Farming activities cause particles such as soil dust and plant material to be emitted into the air. Some of these aerosols can become ice nucleating particles (INPs), serving as seeds for ice and mixed-phase clouds. While there have been ground-based studies of these particles in the western Great Plains and a single air-based study in Indiana, there is a distinct lack of ground-based studies in the Midwest. In Indiana, over two-thirds of the state is farmland, with over 75% of land in Tippecanoe County used for agriculture. Despite farming being such an essential part of life in Indiana, the connection between agricultural activities and INP concentrations in the area has not been explored. Using field observations taken at the Purdue Agronomy Center for Research and Education (ACRE), we hope to study the impact of harvesting on INP concentrations in the midwestern United States. The field experiment took place from May to December 2021 at the ACRE site, but this study focuses on three days during the harvesting period. Data was collected via two instruments: the SPectrometer for Ice Nuclei (SPIN) and the Cloud Condensation Nuclei Counter (CCNC). It appears there is an increase in INP concentrations on days when harvesting occurs, most likely due to an increase in organic and biological particles. It is hoped that the data from this project will provide further insight into the composition and number concentrations of INPs from harvesting through ground-based field observations, as well as insight into INP concentration in the rural Midwest and its climatic impacts.

Keywords

INP, ice nucleation, agriculture, climatology, aerosols, particles

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Joseph Robinson is a recent alumnus of the College of Science and Honors College who majored in Atmospheric Science. Joseph has been working in the Purdue Atmospheric Chemistry Lab since January 2020, working with both cloud condensation nuclei and ice nucleating particles. While at Purdue, he has been an active member of various organizations, such as the Purdue University Meteorological Association, Global Science Partners, and the Purdue Varsity Glee Club. Joseph will be attending the University of Leeds to pursue a PhD studying ice nucleating particles in a project funded by the UK's National Environmental Research Council.

Mentors

Gouri Prabhakar is an Assistant Professor of Practice in the Department of Earth, Atmospheric, and Planetary Sciences. Her research interests are in studying aerosol-cloud interactions and impacts of atmospheric aerosols on climate and regional air quality. She uses ground and aircraft observations to understand chemical and physical processes in the atmosphere that dictate the formation of aerosols and their interaction with cloud droplets. Dr. Prabhakar teaches several courses in atmospheric science and enjoys mentoring undergraduate students.

Dan Cziczo is an atmospheric scientist interested in the interrelationship of particulate matter and cloud formation. His research utilizes laboratory and field studies to elucidate how small particles interact with water vapor to form droplets and ice crystals, which are important players in the Earth's climate system. Experiments include using small cloud chambers in the laboratory to mimic atmospheric conditions that lead to cloud formation and observing clouds in situ from remote mountaintop sites or through the use of research aircraft. Dan's current research interests include the chemical composition of atmospheric aerosols with an emphasis on their effect on cloud formation mechanisms, Earth's radiative budget, and meteoritic debris and launch vehicle emissions in the atmosphere.

Christopher Rapp earned his bachelor's in Atmospheric Sciences and Mathematics from the University of Utah, and then joined Professor Dan Cziczo's group at Purdue University to continue his passion for aerosol research. As a PhD student, his work focuses on experimentally determining the ice nucleating properties of secondary organic aerosols (SOAs). Chris's research is conducted using the SPectrometer for Ice Nucleation (SPIN) instrument, which obtains concentration and optical properties of ice nucleating particles. SOAs are a major component of total atmospheric aerosol; however, their impact on the radiative budget is not well understood. Investigating the physical and chemical properties of these aerosols improves our understanding of their effect on cloud formation and consequently their role in climate change.

INTRODUCTION

Farming activities cause particles such as soil dust and plant material to be emitted into the air. Particles suspended in the atmosphere have a significant impact on the Earth's climate system, mainly through their interaction with clouds. Depending on their chemical and physical properties, particles can become seeds for warm clouds made of liquid water (cloud condensation nuclei, CCN), cold clouds made of ice, and mixed-phase clouds. Mixed-phase clouds, consisting of both super-cooled liquid cloud droplets and solid ice particles, are a common occurrence in the atmosphere. Although rarer than liquid water droplets, ice particles are vital to various processes in the atmosphere, such as cloud formation, precipitation, and radiative balance (Cantrell & Heymsfield, 2005). For these ice particles to form in mixed-phase clouds, they require a particle to freeze on, which we call an ice nucleating particle (INP). While inorganic dust and volcanic ash are good INPs, organic and biological particles have also been proposed as INPs (Kanji et al., 2017). Despite years of research in this field, there continue to be significant uncertainties in the quantitative estimations of these impacts. The Midwest is a primarily agricultural region, which means it could be a potential source of INPs.

Particularly in the Midwest, few studies have investigated the relationship between farming activities and INP concentrations. An observational study carried out in the western Great Plains determined that there was an increase in INP concentrations after harvesting various crops such as soybean, sorghum, wheat, and corn (Suski et al., 2018). In addition, a recent aircraft study over Indiana suggested a slight increase in mineral-containing particles and organics at altitudes of 100–300 meters above agricultural fields (Tomlin et al., 2020). While an increase in these particles does not guarantee an increase in INPs, it is worth investigating. More than 80% of Indiana's 23 million acres are dedicated to farms, forests, and woodlands (ISDA, 2022), with farm operations on 14.8 million acres (USDA, 2021). Considering that most of the state is agricultural, there is the potential for a significant emission of INPs, particularly around the harvesting season. This study fills the gap in data and provides information on the impact of harvesting on the INP concentration in Indiana.

METHODOLOGY

Field Site Description

Measurements were collected from a plot located at the Purdue Agronomy Center for Research and Education, or ACRE (40.47°N, 86.99°W), approximately 5 miles northwest of Purdue University in West Lafayette, Indiana. The sampling location is surrounded by farm plots owned by Purdue University and private farms

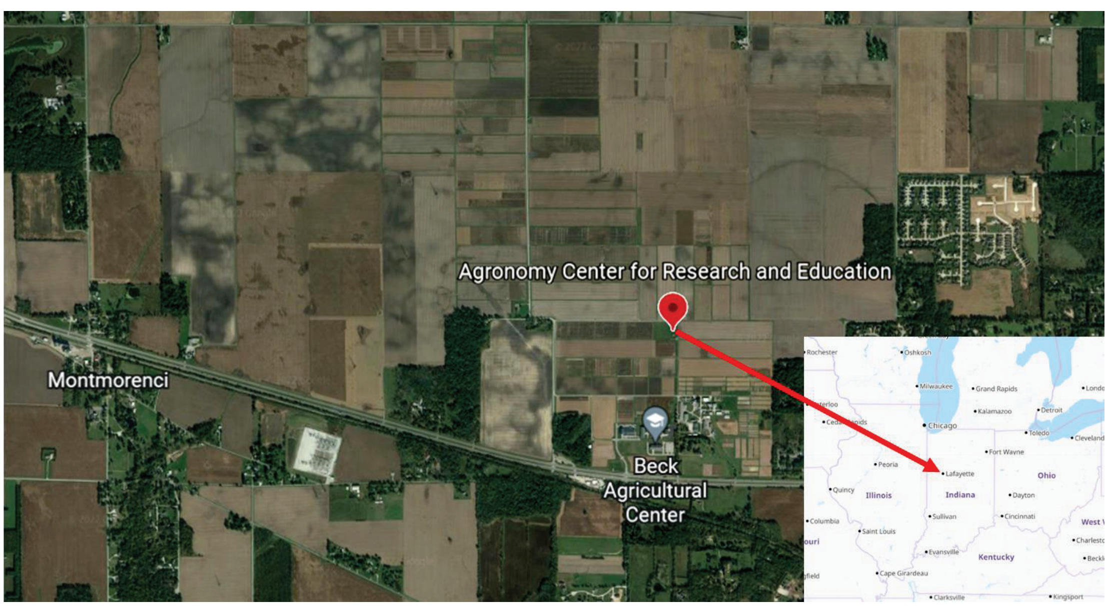

FIGURE 1. Location of ACRE field site within the local farm fields and Indiana.

(Figure 1). Measurements were taken from June 2021 through November 2021, encompassing most of the growing season and harvest. The aerosol measurement instruments were housed inside a climate-controlled shed with a 10-meter-tall sampling inlet attached to the exterior. The inlet utilized a blower, which pulled the sample air down to where the instruments were able to sample from the center of a laminar flow. A laminar flow is one that is smooth and free of turbulence, allowing little mixing and interaction between the flow and the surrounding air. The instruments housed in the shed included the SPectrometer for Ice Nuclei (SPIN) and the Cloud Condensation Nuclei Counter (CCNC). In addition, weather data hosted by the Indiana Climate Office was used to provide supplemental weather data throughout the study.

Data Description

The SPectrometer for Ice Nuclei (SPIN) is an ice nuclei counter created by Droplet Measurement Technologies in collaboration with MIT (Garimella et al., 2016). SPIN is pictured in Figure 2. SPIN is designed to detect ice nucleating particles (INP), the aerosols that ice particles form on. The instrument can detect particles from 1 to 20 μm. The nucleation chamber consists of 2 parallel plates that, when un-iced, are located 1 cm apart. During operation, the walls are kept at different temperatures, resulting in what is commonly referred to as a warm wall and a cold wall. When the instrument startup is complete, the chamber is iced, which creates an ice layer on the chamber walls that is approximately 1 mm thick. A laminar airflow travels equidistant from both sides of the chamber. The difference in temperature between the cold and warm walls allows for a linear profile of water vapor pressure and temperature between the walls. The environment causes the volume where the laminar flow is located to become supersaturated with respect to ice, allowing for the formation of ice particles. A sheath flow runs down the sides of both walls to ensure that the aerosol sample stays within the region of constant supersaturation. In the SPIN, the sheath flow is dry, filtered air that serves to keep the sample flow in place. While flowing through the supersaturated region of the chamber, both ice and water droplets begin to form on the aerosols. In order to be able to distinguish between water droplets and ice particles, the bottom fifth of the chamber is isothermal (meaning the walls are at the same temperature). This means that the air is subsaturated with respect to liquid water, leading to the evaporation of water droplets. Smaller water drops will evaporate completely, while larger drops will become significantly smaller than these ice particles. The sample air then flows through an Optical Particle

FIGURE 2. The SPectrometer for Ice Nuclei (SPIN).

Counter (OPC), which provides information regarding the size and concentration of particles passing through the counter (Garimella et al., 2016).

For this study, the temperature range of the laminar sample flow was kept between –23°C and –29°C, with a supersaturation relative to ice between 24% and 27%. The data processing stage assumed that any particle over 5 μm was an ice particle (Wolf et al., 2020). Each experiment was broken into 10-minute chunks, and the total number of activated ice nuclei was counted. The chunk with the smallest bin was considered the background count, which was subtracted from the rest of the data.

The Cloud Condensation Nuclei Counter (CCNC), seen in Figure 3, is another instrument by Droplet Measurement Technologies. The CCNC counts the number of aerosols that become cloud condensation nuclei (CCN). A CCN is a particle that water vapor condenses on to form a cloud droplet of liquid water. The model used for this study was the Dual-Column model, but only one column was utilized due to a technical issue. As in SPIN, the sample air is directed in a laminar flow through the chamber's center. The chamber walls are coated in water and heated, with the wall temperature increasing toward the chamber's exit at the bottom. The center of the column where the sample air flows is supersaturated with respect to water vapor. This is because heat diffusion in the air is slower than that of water vapor. Essentially, at a given point in the central laminar flow, the vapor pressure is from a warmer segment of the wall while the temperature is from a cooler segment, leading to supersaturated conditions (Roberts & Nenes, 2005). The central line through the chamber is designed to be kept at a quasi-uniform supersaturation, and the sample flow is surrounded by sheath flow to ensure the sampled air stays within the area of constant supersaturation. The sample air passes through an OPC upon exiting the diffusion chamber, which detects and sorts detected particles into 20 size bins, with bigger particles counted in higher number bins.

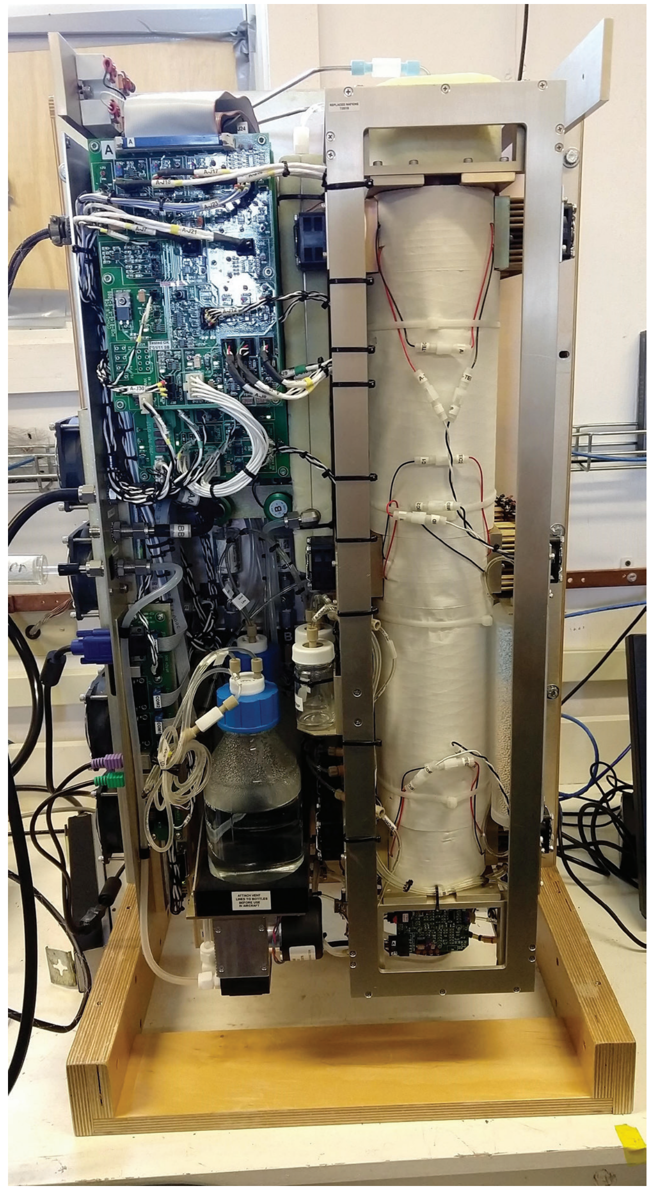

FIGURE 3. The Cloud Condensation Nuclei Counter (CCNC).

The CCNC can operate at supersaturations between 0.07% and 2%, and the OPC can detect activated particles between 0.75 μm and 10 μm. For the purpose of this study, the CCNC data from a supersaturation of 1% is used. At this supersaturation, it can be assumed that all aerosols are activated and detected, giving us the total number of aerosols in the sample. This is used to compare the number of detected INPs.

Co-located at the observation site is an automated weather station, part of the Purdue Mesonet. The Purdue Mesonet is a network of weather stations located on Purdue-owned farms that provide important agriculture information (Sheldon, 2020). Much of this information is also relevant to our study and provides us with a detailed weather report on a particular day. The data from the ACRE weather station served as valuable supplemental data to our experiment.

ANALYSIS

Although the field campaign occurred between June and November 2021, due to issues with SPIN, only three days of data (October 23, October 27, and November 6) were usable for analysis. Harvesting was observed to be actively taking place on October 23. Work at the granaries was observed on October 27, and no activity was observed on November 6. The weather and observed farming activity, as well as the mean temperature and ice supersaturation of the sample flow, are noted in Table 1.

Figure 4 shows various ice nucleation experiments and their thresholds (Garimella et al., 2016). The horizontal axis shows the temperature of the sample, while the vertical axis shows the supersaturation relative to ice. The blue line plotted shows the point of saturation for liquid water, meaning above the line is supersaturated and below the line is subsaturated. Our experiments operate within the red oval shown in the image. Since our experiments operated in a threshold that was subsaturated with respect to water and supersaturated with respect to ice, we can assume that all particles over 5 microns observed are indeed ice particles (Wolf et al., 2020).

Analysis Methods

The SPIN OPC reports data every second, reporting the number of particles in each bin as well as liquid and ice supersaturations, and metadata that were not used in the analysis. Since we assume that all particles larger than 5 microns are ice particles, we took a sum of the particles detected in bins 11 through 20. This information was then plotted alongside the ice supersaturation for an experiment. An example of this data for October 23 can be seen to the left in Figure 5. This data plots the number of particles greater than 5 microns detected by the OPC each second. Each day was broken into 10-minute intervals to compare the data from different days and make sense of the data. The mean INP concentration (in number of INPs per minute) was calculated for each

TABLE 1. Description of Weather Conditions, Farming Activities, and the Sample Flow's Mean Temperature and Ice Supersaturation for Each Date of Interest

Date	Weather	Farming	Sample Flow Mean Temp	Sample Flow Mean Ice SS
10/23/21	Clear and cool, drizzle previous day	Active harvesting at ACRE	−28.0°C	24.0%
10/27/21	Partly cloudy (cirrus)	Granary work	−29.0°C	24.9%
11/06/21	Calm and clear day	Harvesting to east on a private farm	−23.4°C	26.4%

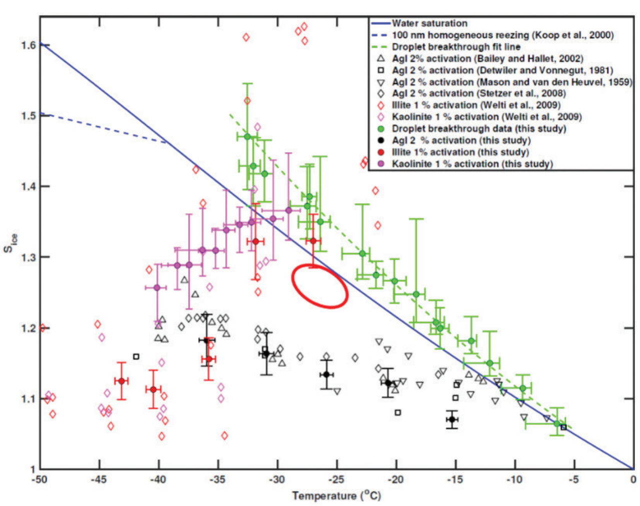

FIGURE 4. Relationship between ice supersaturation and sample flow temperature. The blue line represents saturation relative to liquid water (Garimella et al., 2016).

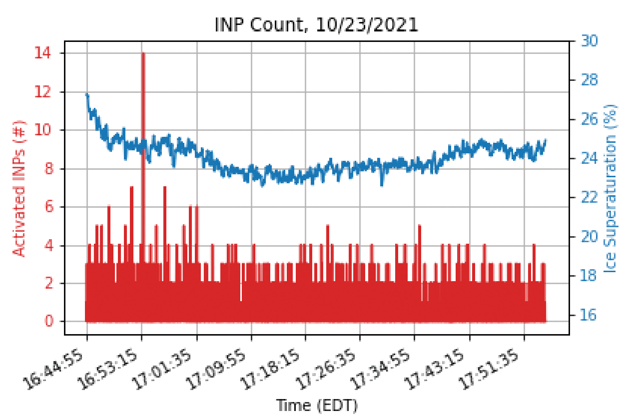

FIGURE 5. Number of INPs detected per second vs. ice supersaturation, October 23.

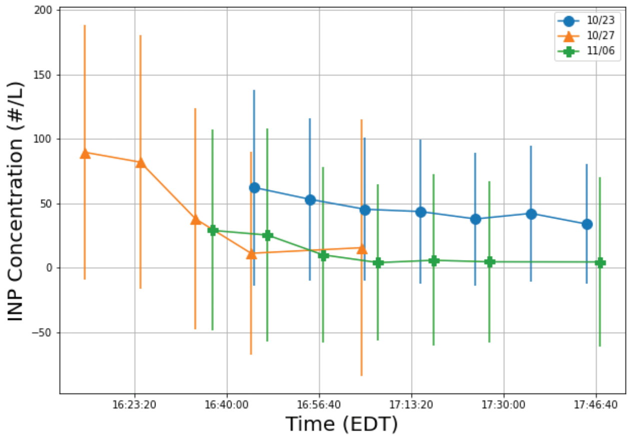

FIGURE 6. INP concentrations for all three days of interest.

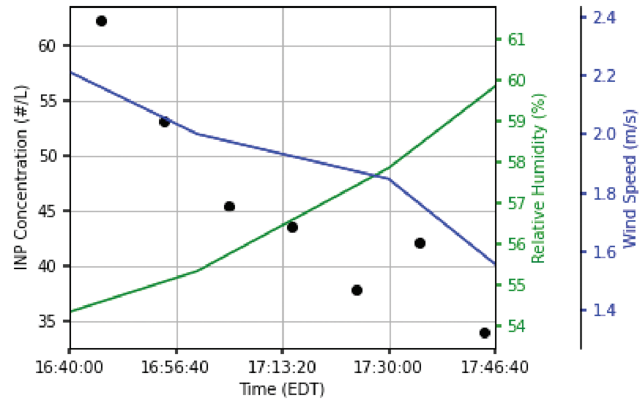

FIGURE 7. INP concentration, relative humidity, and wind speed, October 23.

interval. This was found by dividing the per-second INP count by the flow reported (in L/min) and multiplying by a conversion factor of 60 s/min. The mean and standard deviation of these concentrations was found for each 10-minute chunk. It was assumed that the interval with the lowest mean concentration was the "background" concentration for the dataset and was subtracted from all the other means. This gave us a standard deviation and adjusted mean for each interval for each experiment. Each day's mean and standard deviations were plotted together on the same time axis so comparisons could be drawn (Figure 6).

In order to visualize the relationship between weather and INP concentration during the experiments, 10-minute means for each day were plotted with the wind speed and relative humidity. The wind speed is a variable directly measured by the automated weather stations. However, the relative humidity had to be calculated by dividing the vapor pressure by the saturation vapor pressure and multiplying it by 100. This can be seen in Figure 7. The wind speed over the experiment time is depicted by the blue line, while the green line depicts the relative humidity.

INP concentrations are not enough to make assumptions about the relationship between harvesting and INP increases. It can be helpful to use a total particle count to see what percentage of all particles are measured to be activated INPs. Various instruments can be utilized to give the total number of particles. As mentioned above, when the CCNC is operated at high supersaturations, it can be assumed that the concentration of particles it detects is the total number of particles in the air. This is because, at extremely high supersaturations, water is eager to condense and will do so on any particle available, whether a good CCN or not (Roberts & Nenes, 2005). Due to this, we treat the total particle concentration recorded at a supersaturation of 1% as the total number of particles in the sample air. The CCNC cycles through various supersaturation levels and runs at a 1% supersaturation every 30 minutes for 5 minutes. Every second, the particle concentration (in particles per cubic centimeter) was reported. In order to compare the CCNC data to the SPIN data, an average of the concentration over the 5 minutes was taken and then converted to particles per liter. Since the CCNC data did not always align with the SPIN data, the CCNC data was interpolated. If the SPIN data preceded any of the CCNC data, the value of the earliest point of the total particle concentration was used. If the data fell between two data points of the CCNC data, the linear fit for the two points was interpolated and the total particle count was found. This can be seen in Figure 8. This was used to find the percentage of the total particles that became activated INPs, visible in Figure 9.

Findings

All three days used in the field campaign follow the same general pattern. The highest concentration is measured at the beginning of the collection period. As the experiment continues, the concentration tends to decrease but can fluctuate slightly toward the last few intervals recorded. This may be because any activity occurring

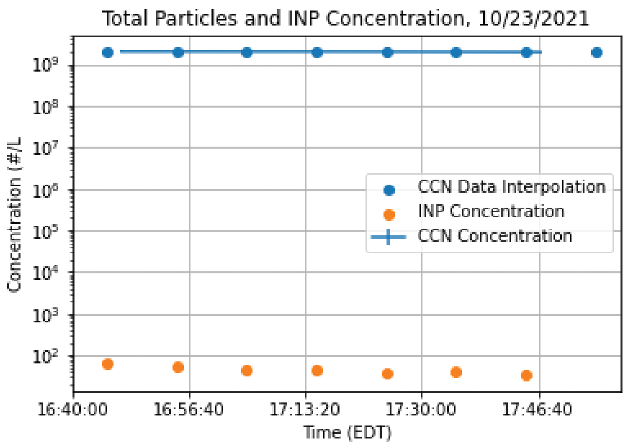

FIGURE 8. INP concentration and CCN concentration, including interpolation, October 23.

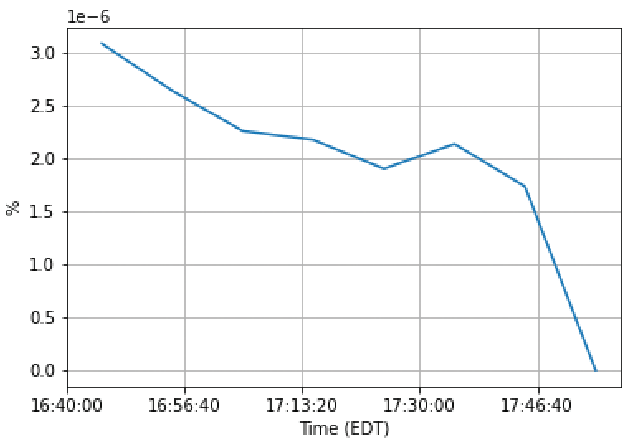

FIGURE 9. Percentage of the total particle concentration that became INPs, October 23.

typically happens during the day. By the time of the experiments in the late afternoon/early evening, the harvesting and other farming work had concluded for the day.

The day with the highest consistent concentrations of INPs was October 23. This was the day that active harvesting was observed on a nearby field. On this day, the concentrations vary from about 60 INPs/L to 35 INPs/L. The highest concentration is measured around 4:45 p.m. EDT. The concentration tends to decrease until about 5:30 p.m. EDT, where there is a spike before decreasing the lowest concentration around 5:45 p.m. EDT. Of the total number of particles measured by the CCN at 1% supersaturation, INPs made up between 0.000003% and 0.0000015% of these values, also decreasing similarly to the concentrations of the INPs (Figure 9). The decrease in INP concentration appears to correspond to the decreasing wind speed and increasing relative humidity. Although farming activity occurred earlier in the day, by the end of the experiment, the harvesting was completed for the day. It could be suggested that a combination of the conclusion of harvesting, decreasing winds, and increasing humidity led to a decrease in the overall INP concentration in the ambient air. During harvesting, particles are kicked up into the air by the motions of the farming equipment, leading to an increase of particles that could serve as the nucleus for ice particles. In addition, the higher the wind, the easier it is for particles to be picked up and transported in the air. With increased humidity, not as many particles may be able to remain suspended in the air.

While we did not observe any harvesting occurring on October 27, work occurred at the granaries, approximately 1,200 feet south of the field site. This could have caused particles (including INPs) to be emitted into the air, which our instruments could detect. This day had the highest recorded concentration at 90 INPs/L, but it was not consistent, dropping to 15 INPs/L by the end of the experiment about an hour later. As expected, the percentage of total particles that are INPs is on the same order of magnitude as on the 23rd but starts higher and rapidly diminishes. The wind speed was higher than on the previous day, fluctuating from 3.7 m/s at 4:30 p.m. to 3 m/s at 5:15 p.m. The relative humidity was significantly lower than on the 23rd, staying in the low 40s. Due to this, it is possible that the wind speed had an impact but that the relative humidity is relatively unimportant. It is possible that granary work produced an influx of particles that rapidly diminished once work was completed.

On November 6, there was no activity occurring at ACRE, but a distant field to the east was being harvested. On this day, we had consistently low INP concentrations ranging from 29 INPs/L at 3:35 p.m. EDT to less than 5 INPs/L by 4:40 p.m. EDT. Compared with the two days in October, the percentage of total particles that are INPs is an order of magnitude less, with values between 0.0000009% and 0.0000007%. The INP counts were lower for this day, but the total concentration was slightly higher than on the previous days (Figure 10). The wind was consistently blowing at about 3 m/s, and the relative

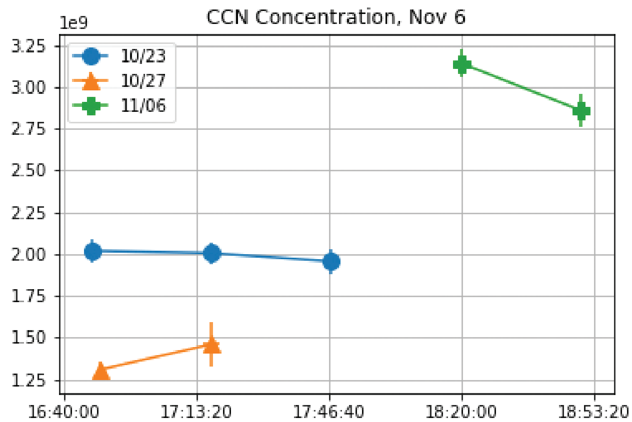

FIGURE 10. Total particle concentrations for the days of interest.

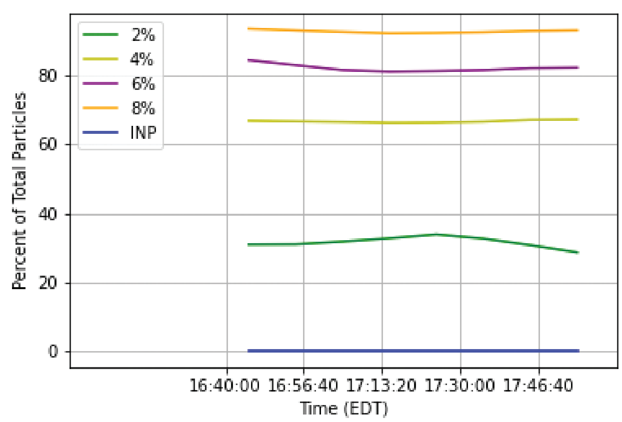

FIGURE 11. Percentage of total particles for various CCN supersaturations and INPs.

humidity was low, staying in the upper 30s. In this case, the wind might have had an effect, but the relative humidity was too low to have a noticeable impact.

Although this project's scope involved the concentration of INPs, the integration of the CCNC allowed us to take a glance at the CCN concentrations. Since most activity occurred on October 23, the CCN data from this day was selected to analyze more closely. Figure 11 shows the percentage of the total number of particles (determined by 1% supersaturation) than were activated CCNs at various supersaturations in the CCNC compared to the INPs. Compared to even the lowest supersaturations in the CCNC, the INP concentrations are 8 to 9 orders of magnitude less. This shows the rarity of these particles compared to those that make up the water droplets in warm clouds. Focusing on the CCN concentrations, there is a clear trend between supersaturation and CCN concentration, with higher supersaturations allowing more particles to serve as CCN. It is important to note that we typically do not see supersaturations this high and that typically, the relative humidity in clouds is much closer to 100% than 101% (representing a 1% supersaturation). This means that the 0.2% supersaturation most likely represents the most realistic environment. Even in this case, around 30% of the total amount of particles can serve as nuclei for liquid cloud droplets.

CONCLUSION

There is a measurable concentration of INPs on all three days of interest from the data collected and analyzed. When harvesting or other farm activities occur, it does appear that this leads to an increase in INP concentrations. This data seems to agree with western Kansas and Wyoming data in 2018. Suski et al. (2018) found that corn and soybean harvests led to counts of up to 200 INPs/L and 180 INPs/L, respectively. While these concentrations are slightly higher than those measured in this field campaign, this could be because the inlet was not positioned directly downwind of the harvesting and in the plume of harvest dust as it was in the Kansas/Wyoming study (Suski et al., 2018).

Not all particles can become INPs, with dust, organic, and biological particles as the best INPs. In an aircraft study conducted in the Greater Lafayette area, about 55% of particles were classed as inorganic, 30% classed as organic, and about 7% as biological (Tomlin et al., 2020). Due to the temperature range of our experiment, the organic and biological particles are most likely to be activated and become INPs. Our data is consistent with the fact that these organic and biological particles are present in the area due to harvesting.

Due to a small sample size, it is hard to make definite observations and conclusions. However, it appears that the data found in this study corresponds with previous literature on the topic. We recognize that this is only a case study and that more data is required to reach a statistically rigorous conclusion. However, the evidence collected in this study does point toward the fact that harvesting increases the concentration of ice nucleating particles. Unlike water droplets, ice particles in the atmosphere can lead to net warming. Shortwave ultraviolet radiation can pass through ice particles without much interaction, but the outgoing longwave infrared radiation is absorbed and reemitted by the ice. This can lead to the warming of the atmosphere. Studies show that the warming impact of INPs is greater at the middle and high latitudes (Zeng et al., 2009). With further study, this data could lead to implications that farming activity can impact climate and radiative balance, leading to global warming, particularly during the harvest season.

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