Instrument Acquisition for Determination of Size-Resolved, Individual Particle, Chemical Composition for the Study of Multi-component, Atmospheric Chemical Processes

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Description of the Research Instrument

The CIA[2]CIA (Figure 1) is a powerful instrument to study the interaction of atmospheric aerosols and clouds. It is an Aerosol Mass Spectrometer (AMS) based system, specifically, consisting of: (1) The Aerodyne High Resolution (HR), Time-of-Flight (ToF), Aerosols Mass Spectrometer (AMS) instrument (HR-ToF-AMS) has a mass range of approximately 1000 amu and a mass resolution of up to 5000. The range of aerodynamic particle size measurements is between 40 and 2,500 nm with a resolution of between 5 and 10 (Daero/ΔDaero). Chemical analysis is performed by thermal vaporization and electron impact ionization (EI) high resolution, time of flight mass spectrometry. The vaporization temperature is adjustable from 200 to 900 °C. The HR-ToF-AMS has a time resolution of seconds with a data rate of up to 100 Hz. The software provides data acquisition and chemical analysis. The HR-ToF-AMS is coupled with modules that include the Soot Particle (SP) module that enables the detection of particles containing absorbing materials such as black carbon and with the Brechtel Manufacturing Inc (BMI) Model 1205 Ground Counterflow Virtual Impactor (GCVI), which dries hydrometeors (cloud droplets and ice crystals) to produce residue particles. Hydrometeors are accelerated within a wind tunnel located upstream of the GCVI inlet tip. The GCVI inlet has a carefully controlled counterflow (16-30 lmp) of filtered/heated air that pushes air out the inlet tip against oncoming hydrometeors. The hydrometeors pass into a chamber where water evaporates so the chamber's exit airstream only contains residue particles. An integrated anti-icing system provides the ability to obtain samples in supercooled clouds. Robust software provides system protection from clog inlets, overheating and power losses that can occur during field measurements. The software provides a real-time display of important parameters and allows users to change the hydrometeor cut size to meet sampling needs.

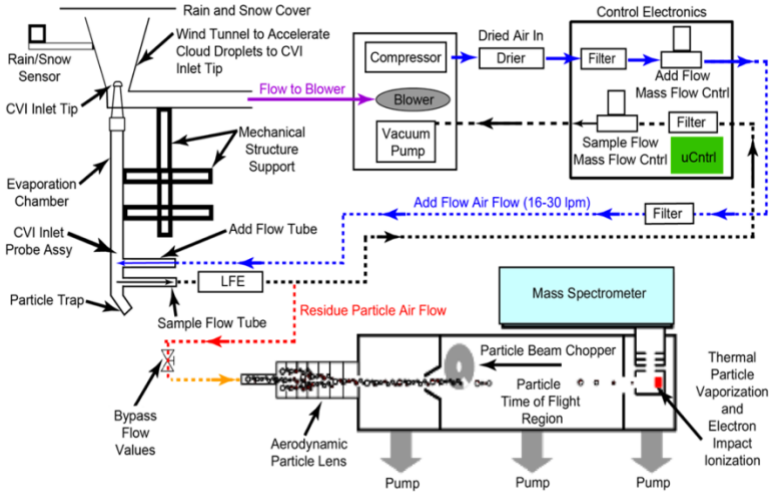

Figure 1: Illustration showing the sample flow in the CIA2CIA composed of the Counterflow Virtual Impactor, and High Resolution, Time-of-Flight, Aerosol Mass Spectrometer).

[1] https://www.energy.gov/fecm/carbon-dioxide-removal-and-conversion
[2] https://climate.nasa.gov/vital-signs/carbon-dioxide/ Accessed August 28, 2023

Previous Experience

During August 2020, the Atmospheric Sciences Department host Aerodyne Research Inc. and WMI to conduct experiments related to cloud seeding flares (Figure 2). At the conclusion of the week long flare testing experiments, the HR-ToF-AMS (Full AMS) was used to measure ambient atmospheric aerosols during overnight sampling on 14 August 2020. Air samples were pulled into instruments from an inlet on the roof of Clifford Hall on the University of North Dakota campus. The measurements show that organic aerosols are a large mass component, with a couple of interesting spikes evident in the time series (Figure 3). The presence of high concentrations of organic compounds can impact the aerosol hygroscopicity and thus the number of activated aerosols that form clouds. Future projects could extend these initial AMS measurements to understanding the chemical processes that influence aerosols activation, which is important for precipitation development and fog formation.

Figure 2: The left image shows the Aerosol Mass Spectrometer (HR-ToF-AMS) at the University of North Dakota's Clifford Hall 423, Instrumentation and Chemistry Laboratory during August 2020. The right image shows aerosol instruments own by the University of North Dakota (UND) and Weather Modification International (WMI) that sampled concurrently with the AMS. The aerosol instruments shown include (middle of the wood top bench) a Scanning Mobility Particle Spectrometer Particle (SMPS), which is required for AMS calibrations.

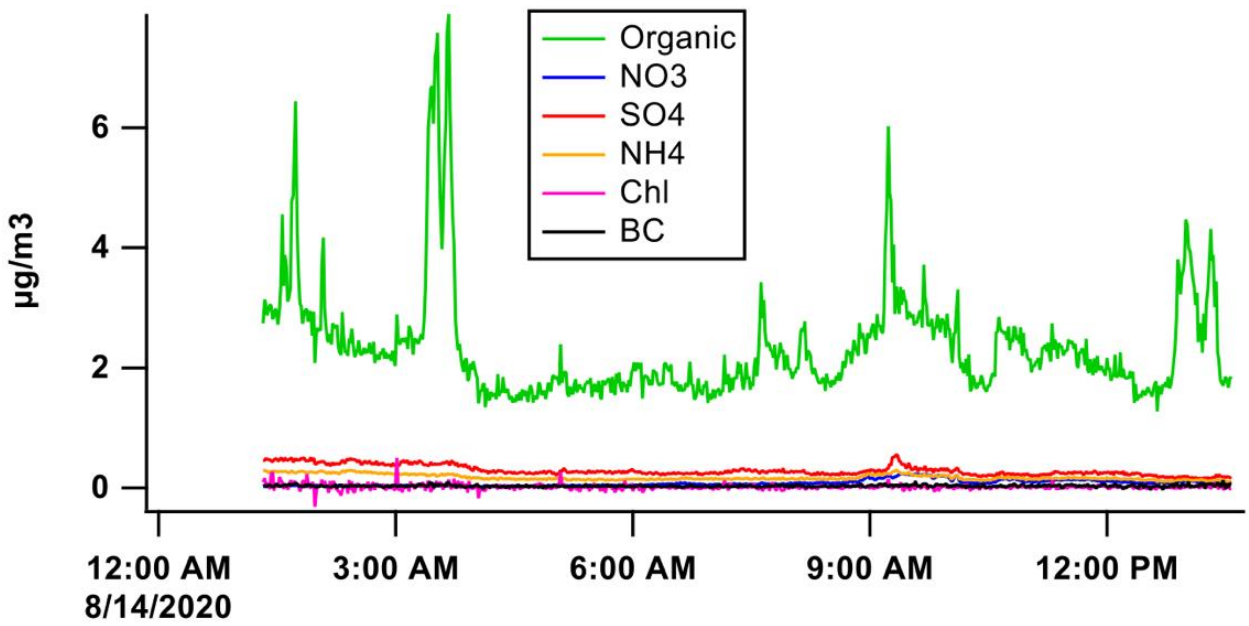

Figure 3: Time series of aerosol species detected by the Full AMS during an overnight sample of ambient air on 14 August 2020 using inlet on the roof of Clifford Hall on the University of North Dakota campus in Grand Forks, North Dakota.

Enabled Research Proposals and Projects

Below, we provide details on several research projects that we (likely PIs noted) foresee submitting proposals on using the CIA^{2}CIA.

Fog Microphysics (Majdi, Delene):

Cloud condensation nuclei (CCN) are particles that enable the condensation of water vapor and the formation of droplets at a given supersaturation. CCN constitute an important fraction of atmospheric aerosol. Their variation can impact the microphysical properties of clouds, fog, precipitation, and climate (Ramanathan et al. 2001; Rosenfeld et al. 2008). At a given supersaturation, the CCN activation ability depends strongly not only on the size of aerosols but also on their chemical composition and mixing state through changes in their hygroscopic capacity (Zaveri et al. 2010). Several studies highlighted the importance of the particle size in CCN activation (Dusek et al. 2006; Ervens et al. 2007). However, few studies investigated the impact of the aerosol chemical composition and mixing state on CCN activation ability. Moreover, an accurate prediction of CCN concentration is crucial to accurately forecast fog using numerical models. In CCN predictions, the particles' internal mixing assumption generally overestimates the CCN concentration (J. Wang et al. 2010), whereas the external mixing assumption underestimates it (Zhang et al. 2010). These two mixing assumptions can result in up to ~40 % relative error in CCN predictions, thus, the aerosol mixing state definitely needs to be taken into consideration in CCN predictions.

Most of the numerical models (such as WRF-Chem) consider the instantaneous internal mixing assumption. This assumption can alter the optical and chemical properties of particles in WRF-Chem simulations (Zhang et al. 2014) and therefore has the potential to influence aerosol-cloud interaction (i.e., CCN activation). In reality, particles are emitted with unique chemical composition and only become internally mixed over a period of hours to days depending on atmospheric conditions. Therefore, it is important to characterize the chemical composition of the

aerosol to quantify the impact of particle mixing assumptions on predicted CCN formation under a fog event. To that end, the CIA^{2}CIA will be used to characterize the chemistry of the atmospheric hydrometeors while also measuring the size distribution and CCN spectrum of particles that produced the hydrometeors to understand the how particles are mixed in the earth's atmosphere. The main goals of this research are to derive accurate estimations of the fog droplet concentrations and to investigate the impact of aerosols composition and mixing state on fog microphysics. With the use of the CIA^{2}CIA, this project will provide a useful data set for determining the role of aerosol chemical composition, aerosol mixing states, and hygroscopic capacity for the formation and dissipation of fog, which enable more accurate fog forecasting.

Lead Particulate Matter from UL94 Fuel Switch (Delene, Majdi, Pierce, Wilson, Bowman):

Lead (Pb) is a highly toxic metal polluted that has a sufficient economic cost (Grosse et al. 2002) and impacts human health due to accumulation in the blood, which reduces full-scale intelligence quotient (IQ) scores (Galiciolli et al. 2022). Due to lead's environmental impacts, vehicles have been engineered to no longer required tetraethyl lead as a gasoline additive. The unleaded gasoline transition started in the 1970s with new vehicles engineered to run on unleaded gasoline, and by the mid-1980s most gasoline used in the United States was unleaded. A similar transition is now occurring in general aviation (Kessler 2013), with newer build aircraft able to operate with currently available, unleaded aviation fuel; however, older (legacy) aircraft generally have engine systems that develop maintenance issues if operated with unleaded fuels. A major issue holding back unleaded fuel usage by general aviation with newer aircraft is a fundamental understanding of the amount of lead pollutants within the local, airport environment. There is a need to qualify airport pollution to enlighten cost-benefit discussion on unleaded fuel adaption in general aviation.

The project's objective is to obtain a quantitative observation of the change in particulate matter lead near the airport due to the UL94 switch. While lead observations have been made near airports (Carr et al. 2011), near airport observations of lead concentration changes due to switching a majority of the airport's aircraft to unleaded fuel has not been done previously. The project's research team have started conducting some observations of the UL94 switch by the John D Odegard School-Aerospace aircraft fleet during summer 2023. We deployed available facilities, instruments and personnel that can be realigned to conduct measurements before and after the UL94 switch. The project's primary measurement is particles filter samples. Bi-weekly filters were obtained using the low volume sampler and daily filters were obtained using the high-volume sampler. Quartz fiber filters were weighted and stored in a freezer until analyzed. The CIA^{2}CIA will be used to analyze the stored filters and collect high quality measurements of near airport observations of lead, and other heavy metal and multi-component chemistry evolution near airport.

Solar Eclipse and Aerosols Photochemical Reactions (Delene, Majdi)

Solar eclipses provide a rare opportunity to investigate the photochemical responses to abrupt changes in incident solar radiation. Solar eclipses have a significant impact on oxidant production, which can cause sudden changes in the gas-to-particle conversion process. New particle formation is a significant source of atmospheric particles that affect climate and radiative balance. Photochemical cycles control the concentrations of reactive oxidants, such as hydroxide radicals and ozone. Organics, such as terpene, are one of the most important drivers of new particle formation in the Earth's atmosphere. However, significant gaps remain in how atmospheric processes produce new particles and contribute to aerosol growth. Growth is mainly by condensation of low-volatile, highly-oxidized multifunctional compounds from terpene oxidation,

and the low volatility of some highly-oxidized compounds are responsible for initial particle formation. Three-dimensional chemistry-transport models, such as the Weather Research and Forecasting model coupled with Chemistry regional model (WRF-Chem), have been used to simulate new particle formation and aerosol growth to forecast atmospheric aerosol mass concentrations. Yet, modeling of secondary organic aerosols remains a difficult task since newly formed particles are not well represented in WRF-Chem due mainly to uncertainties in particle nucleation involving organic compounds and growth rate. A major challenge is accurately simulating particle growth of newly formed nuclei from 1 nm to the measurable size range of 3-10 nm.

The project's objective is to conduct measurements during total solar eclipse related to terpene's photochemical reactions to improve model's nucleation parameterizations of new particle formation in a location of low existing particle concentrations, such as in the upper troposphere. The study investigates the connection between ultraviolent light, production and composition of different low-volatility compounds, and new particle formation involving processes related to hydroxide radical and ozone radical driven reactions. The eclipse event provides a unique and challenging opportunity to collect measurements of particle concentrations, amount of solar radiation, trace gas concentration, and standard meteorological parameters. The challenge of obtaining eclipse measurements is mitigated by extensive testing and having several preselected locations for possible instrument deployment. A metrological trailer houses all field project instruments and equipment, which enables re-deployment to one of a number of secondary sites if the 3-5 day weather forecast indicates a high probability of clouds at the primary site. The CIA^{2}CIA will be used to quantify the new particles from terpene's oxidation reactions.

WRF-Chem is used to study the effects of sunlight on meteorological parameters and the new particle formation from terpene's oxidation reactions. Comparison to observations is conducted to evaluate the model's performance to capture the effects of the total solar eclipse on the nucleation processes related to new particle formation and to improve the model's representation of the newly formed particles from nucleation before, during and after the total eclipse. The project aims to provide a unique data set of atmospheric observations that includes the April 8, 2024 total eclipse event to investigate new particle formation and particle growth, which has previously only been observed in controlled laboratory experiments. The project is an important step toward understanding the connection between radiation driven photochemical oxidation processes, new particle formation and secondary aerosol formation, which are needed for improving models to quantify the global particle load and radiative balance.

Aerosols from Rockets Emissions (Majdi, Delene, Fernandez-Tous)

Space transportation plays an important and growing role in Earth's economic system. Rockets uniquely emit gases and particles directly into the middle and upper atmosphere where exhaust from hundreds of launches accumulates changing therefore the atmospheric radiation patterns. Particulate emissions from rockets, have an important impacts on climate and ozone (Ross and Sheaffer 2014). Rocket engines emit various amounts of submicrometer-sized particles of soot (or black carbon, BC) and alumina (aluminum oxide) directly into the stratosphere. Because of the unique nature of their combustion chemistry, rocket engines emit large amounts of BC when compared to, for example, a modern jet engine. Because particles emitted by rockets are small, they reside for 3 to 4 years in the stratosphere, where they accumulate. The BC particles absorb solar radiation and slightly reduce Earth's albedo. The alumina particles reflect solar radiation and so increase the albedo slightly. Both have the same consequence for the underlying atmosphere: a reduction in the intensity of solar flux entering the top of the troposphere. Solar flux reductions

caused by stratospheric particles are well understood to cool the lower atmosphere (Caldeira, Bala, and Cao 2013). Therefore, rocket launch emissions contribute to cooling of Earth's lower atmosphere and surface. This project aims at quantifying particle emissions from rockets and hypersonic aircrafts. The use of the CIA^{2}CIA enables to conduct ground-based measurements of chemical properties of rocket exhausts during environmental chamber tests. Quantifying of the various rocket emissions and associated radiative forcing leads to elucidate the most important processes by which rocket exhaust cause climate forcing.

Aerosol Chamber Experiments (Kubatova, Bowman, Delene)

Atmospheric particles are a complex and continuously evolving mixture of organic and inorganic compounds. Their impacts on climate, visibility, and human health depend on particle size, number, chemical composition, and mixing state. Laboratory experiments allow measurement of aerosol formation and growth processes under controlled conditions. A new laboratory aerosol chamber is being developed jointly by researchers in Chemistry and Chemical Engineering and is on schedule to begin operation in summer 2013. The chamber consists of a 20 m3 Teflon bag within a ventilated, reflective enclosure and surrounded by UV lights. Current instrumentation includes a variety of gas-phase analyzers, particulate samplers, and on-line particle equipment. CCN and condensation particle counter (CPC instruments from Dr. Delene's laboratory add the capability for measurements of particle hygroscopicity and cloud droplet formation).

There are two main areas the chamber studies will be focused on using the proposed instrument: 1) the characterization of aerosols based on their volatility; 2) online assessment of currently studied reactions of PAHs.

Currently, a number of studies recognize the significance of volatile organics participating more readily atmospheric reactions. However only recently (Huffman et al. 2009; May et al. 2012), and others demonstrated the occurrence of volatile components in primary organic aerosols (POA), which is in contrast to typical atmospheric model representation assuming POA to be non-volatile). These studies have noted the discrepancy between the chamber studies and field measurements. We have recently shown (Beránek et al. 2013) with detailed off-line characterization using thermal desorption/pyrolysis with gas chromatography/mass spectrometry (TD/Py-GC/MS) that the TD may be strongly affected by matrix interactions when using graphite or silica as model matrices. Here we propose to explore the role of aerosol matrix interactions further generating online-data from the Full AMS with GCVI and with previously developed temperature programming (50–230 °C) by Huffman et al. (2009) allowing studying the aerosol volatility. The CIA^{2}CIA provides an overall concentration of organics through the generation of common electron ionization fragments from organics present, however since all compounds are analyzed simultaneously speciation is limited. Thus detailed speciation will be enabled with the off-line TD/Py-GC/MS measurement using comparable temperature programming. We are intent to apply both methods to samples generated in chamber studies with various seed aerosols (organic such as α-pinene, inorganic such as SO_4^{2-} etc.) and reactants as well as field samples. The proposed work will allow studying the matrix effects on the desorption of the volatile species. Further, the detailed speciation of all temperature fractions including pyrolysis may reveal additional marker ions that may be used in future AMS studies.

Dr. Kubatova's NSF CAREER grant supports the first project to use the aerosol chamber. The experiment exposes particles generated by a diesel engine to reactive atmospheric species (O_3 and NO_2) and measures polycyclic aromatic hydrocarbons (PAHs) and their oxidation products embedded within the diesel exhaust particles. PAHs are extensively studied promutagens; however, recently we have observed in our preliminary work a potential proinflammatory role of

hydroxy-PAHs and other polar PAHs derivatives. Currently planned measurements of PAHs and their derivatives use particle samples collected on filters and gas chromatographic (GC) analysis of the extract particles or thermal desorption with pyrolysis GC. However, while this type of measurement is valuable, providing detailed information on targeted species, it is also limited to longer reaction time scales, requires relatively large amounts of sample, and has some sampling artifacts. The Full AMS instrument will resolve these issues as it adds the ability to measure particle chemical composition with high time resolution, as a function of particle size, even for single particles. Thus the instrument will complement the advanced chemical characterization (Cochran et al. 2012; Beránek et al. 2013) methods recently developed by Dr. Kubatova's research group together providing the unique capability to define chemical composition based on individual compounds as well as elemental and ion-fragment ratios (e.g., O:C, H:C, f43:f44). This will allow us to better define the relationship between bulk and single particle compositions, particle hygroscopicity, and the influence of particle mixing state.

Both tasks will be carried out in collaboration with Drs. Kubatova, Bowman and Delene. All three investigators will be involved in the setup of the chamber, Dr. Delene will be in charge of the particle measurements, Dr. Kubatova's group will focus on the chemical characterization and Dr. Bowman on the modeling. Both tasks are in line with the projects currently carried out in Dr. Kubatova lab. The graduate student expected to pursue this work is currently supported through CAREER award and is also applying for UND doctoral dissertation award, however if no other funding available, the student will be supported through teaching the assistant position after expiration of the CAREER grant. The undergraduate students will be involved either through their research capstone projects or REU summer research experience.

Point Source Emissions (Nasah)

Carbon dioxide capture from major point sources (PSC) is considered a key approach for mitigating the effects of climate change attributed to growing carbon dioxide concentrations in the atmosphere[(1)(2)].These systems employ solvents for the capture of carbon dioxide which could result in trace emissions of the capture solvent, its degradation products, or reaction products with other species in the flue gas. Recent research activities that focused on studying the stability of a specific solvent, monoethanolamide, reported multiple degradation compounds with unknown structures (Buvik et al. 2021). With multiple PSC projects announced targeting over 140 million tons per year of carbon dioxide (Fahs et al. 2023) and considering these capture systems are expected to use even more complex amine-based proprietary chemicals, it is important to not only evaluate the trace emissions of these commercial systems as they are commercialized, but also understanding their impact on aerosol formation in the region surrounding these facilities. The CIA[2]CIA unique abilities for identifying aerosol composition, and their interactions with atmospheric aerosols and clouds, present a unique opportunity to lead research in a new field.

Aerosols from Crop Residue Burning (Majdi, Delene)

As one of the main form of biomass burning in North Dakota, open field agricultural crop residue burning is a huge source of a seasonal intensive production of fine particles in the atmosphere (Akagi et al. 2012; Andreae and Merlet 2001). The aging of these fine particles leads to the production of the Secondary Organic Aerosols (SOA) which can contribute to regional haze formation (Zhang et al. 2015).

[1] https://www.energy.gov/fecm/carbon-dioxide-removal-and-conversion
[2] https://climate.nasa.gov/vital-signs/carbon-dioxide/ Accessed August 28, 2023

Biomass burning with controlled start and aging conditions can be provided by laboratory experiments and the obtained results can help to improve the prediction for field studies and enhance the performance of the models. Several recent chamber experiments studies have investigated the volatile organic compound and aerosol properties for crop residue burning in laboratory (Li et al. 2016; Wang et al. 2014; Stockwell et al. 2015). These studies have determined the emission factors, quantified and identified the SOA formed during the aerosol aging processes. They have also investigated the variation in physico-chemical properties of particles from crop residue burning (chemical composition, density, volatility, etc.).

Although the great progress that these studies have made in determining the emissions factors and physico-chemical properties of aerosols from crop residue burning, considerable uncertainties remains concerning the effect of the SOA formed from biomass burning on the weather as Cloud Condensation Nuclei (CCN) activities of the aerosols from biomass burning vary significantly with the fuel type, the burning conditions as well as the atmospheric aging processes (Reid et al. 2005). Several previous studies have shown that biomass burning is an important source of CCN (Pierce et al. 2007; Spracklen et al. 2011). They have also highlighted that the hygroscopicity and CCN activation ability of aerosols from biomass burning increase significantly with the photooxidation reactions (Martin et al. 2013; Vakkari et al. 2014). However, some observations have shown that aerosol ageing processes can lead to alter and decrease CCN activity. Therefore, further work is needed to study the variability of CCN activity of aerosols from biomass burning by investigating the impact of the aerosol aging processes on CCN activation during the biomass burning.

In this project, the photochemical aging of particles from the burning of the common crop residue in North Dakota will be investigated. The particle size distribution, the chemical composition and CCN concentrations will be quantified simultaneously at the beginning and during the aging processes. Moreover, the relationship between the SOA chemical composition and CCN activation properties will be studied. To that end, the CIA^{2}CIA will be used to analyze the chemical composition of particles from crop residue burning at the beginning and during the aging processes while measuring the CCN concentrations and the aerosol size distribution. With the use of CIA^{2}CIA, we will enrich the database for numerical models to better predict CCN during biomass burning. Considering crop residue particles, their physico-chemical aging processes are critical to improve the prediction of CCN activation and the hygroscopicity of atmospheric aerosols.

Seeding Flare Chemistry and Cloud Microphysics (Delene)

Studying the physical processes involved in the chain of events that results in rain formation is difficult because there is currently no robust method that links in-situ measurements of aerosols and clouds. Therefore, it is difficult to directly quantify how aerosols produced by burning cloud seeding flares affect cloud microphysical properties. Without quantitative relationships, it is impossible to model the effects that different cloud seeding techniques have on precipitation development. An AMS instrument sampling the chemistry of particles from evaluated droplets enables direct determination of the relationship between aerosols used to seed clouds and corresponding cloud properties with the use of special tracer flares containing the element indium, which is used to distinguish seeding aerosols from naturally occurring aerosols. A counter-flow virtual impactor is used to evaporate droplets in seeded clouds to obtain only aerosols (cloud condensation nuclei) involved in the nucleation process. An AMS system monitors the aerosol chemical composition in real-time for indium. Detection of indium is a direct indication that the cloud parcel is affected by seeding since indium is not a detectable component of atmospheric aerosols. Cloud droplet size distribution measurements, concurrent with indium detection, provide

observations of seeded parcels, while periods without indium detection provide observations of non-seeded parcels. Comparisons of seeded and non-seeded parcels provide direct evidence of how concurrently burned hygroscopic seeding flares change the microphysics of clouds. This innovative observational methodology allows for the first time the direct determination of changes in cloud properties under atmospheric seeding conditions. A mesoscale numerical weather model that uses the observed difference in cloud mass and cloud droplet concentration between seeded and non-seeded clouds can quantitatively determine the impact on precipitation and will be a significant advancement in precipitation development research.

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