Title

Spatiotemporal Trends in Aerosol Physicochemical Properties at the Surface in the Contiguous United States and Enhancement of Student Preparation for Upper-Division Laboratory Courses

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Publication Date

2023

UNIVERSITY OF CALIFORNIA,
IRVINE

Spatiotemporal Trends in Aerosol Physicochemical Properties at the Surface in the Contiguous United States
and
Enhancement of Student Preparation for Upper-Division Laboratory Courses

DISSERTATION

submitted in partial satisfaction of the requirements
for the degree of

DOCTOR OF PHILOSOPHY

in Chemistry

by

Madison Marie Flesch

Dissertation Committee:
Professor Ann Marie Carlton, Chair
Professor Jim Smith
Professor Manabu Shiraiwa

2024

DEDICATION

To my parents, Bill and Cindy,
who have supported me since I was barely two pounds
and from whom I am continually inspired.
To Zac, for putting up with me these last few years
and still wanting to do life with me.
Thank you. I love you all dearly.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

ACKNOWLEDGMENTS

I would like to thank everyone who has been a part of my graduate school experience and has helped me complete this degree. The COVID-19 pandemic interrupted many things in the middle of my time at UCI, but everyone's support and desire to maintain connections even while we had to remain socially distant helped me get to where I am today. I am incredibly grateful for all of the collaborations and friends made along the way, as they made a difference every day in this process.

First and foremost, I would like to thank Ann Marie Carlton, my advisor, who provided me with so many interesting science questions to investigate both large and small during my graduate career. I am grateful for her guidance and mentorship over these last five years. Ann Marie, thank you for letting me explore what it means to be a woman in science under your leadership, for all of the good-natured ribbing on my Midwestern-isms, and for eating the baked goods I brought to the office as group meeting snacks.

Following closely behind are, of course, the members of the Carlton lab both past and present for encouraging me along the way and being such integral parts of my life at UCI. Without them proofreading for me, engaging in science discussions while snacking in the office, or even hanging out in our GatherTown virtual office to work together during quarantine, this experience would have been much lonelier and sadder. I must first thank Amy Christiansen for teaching me the basics of R and being my fellow Midwestern rock in the office when regional debates occurred, and for her equal baking contributions to the office. Amy, I have kept your snake plant alive and am following your professorship career with great interest. I am immensely grateful to Alyssa Burns and Yin Ting Chiu for their bright science ideas and connection during the pandemic through to today. Alyssa, I'll miss our frequent teaching discussions and your critical science questions. Yin Ting, I'll miss being desk neighbors and your sharp eye for presentation format and structure. I have to thank our postdoctoral scholar, Simon Rosanka, for collaborating and teaching me about CMAQ and other air quality models, along with our many discussions of U.S. versus German lifestyles. I'm inspired to see the ongoing progress and discoveries from everyone, especially newer minds like Ellie Smith and Tiffany Hu.

I must also thank Steve Mang, my teaching mentor. When I was assigned to be a teaching assistant for Advanced Analytical Chemistry in my first quarter at UCI, I had no idea what to expect. Steve's easygoing nature and desire for his TAs to also learn new things put me at ease immediately. His interest in constantly improving course instruction is what led to our publication together, and reminded me that I did, in fact, enjoy teaching quite a lot. Steve, your mentorship led me to teach Chem 1B as the Instructor on Record in Spring 2023, and I am incredibly grateful for the opportunity.

I also thank my other collaborators, Virendra Ghate, Thomas Surleta, and Nick Humphrey, for their insights and help on my research projects. I am grateful for the experience under Virendra's leadership in understanding satellites, atmospheric radiation and modeling, and improving my critical thinking skills about the combination of all of these things during the

COVID-19 pandemic and our projects together. Thomas, thank you for being so generous with answering all of my questions about the quirks of Python while I was in the DOE SCGSR fellowship program working with you and Virendra. Nick, it was a pleasure teaching with you and working to make the analytical chemistry class a little less boring during the pandemic for our students with the virtual lab activities.

I would also like to thank Jim Smith and Manabu Shiraiwa, both of whom served on my advancement committee and are now on my dissertation committee. They have both been very supportive of me during my time at UCI, especially with the curve ball the pandemic threw into some of my plans. Jim, thank you for being a calm and guiding presence in Rowland Hall, particularly when Ann Marie was on her big Washington D.C. adventure. Manabu, thank you for being supportive as I decided to diversify my dissertation research and for posing interesting and critical questions about my research during and after my advancement to candidacy exam.

I am so grateful for my family and friends who have supported me along this journey. I am forever indebted to my parents, who have always supported me in everything I do. They helped me move halfway across the country and always take time to check in with me since I live so far away from them, especially since 2020. Mom and Dad, thank you for always believing in me and giving me sage advice from two former educators. I miss you both so much and cannot wait until you can come out to visit Zac and I in our new place. Hunter and Kenzie, while you two stayed in Iowa for your careers, I still have to thank you both for listening to and supporting me from afar, and for letting me spoil your dogs when I'm home visiting. To my aunt, uncle, and cousins who live locally, thank you for letting me come over frequently for dinners and holidays, especially during my first year of grad school when I didn't know anyone yet. Having you in my corner while studying out here has been so important to maintaining my life outside of grad school.

Friends in graduate school come in a variety of shapes and sizes, and the ones I made at UCI are no different. To the cohort of folks that I entered this program with, thank you all for collectively agreeing to be friends and for choosing to maintain those connections during the global pandemic that forced us all into our apartments for many months. I have to give a special shoutout to my fellow atmospheric ladies Cynthia Wong, Lia Dam, and Mooji Boldbataar. You three roped me into a community I never imagined having. Thank you for all of the late nights studying together and exchanging our latest baking adventures and for showing me how cool it is to be in science with women like you. I cannot wait to see what the future holds for all of us. I also want to thank Brian Miller, Marcus Marracci, Jenn Campos-Ayala, Rachel Alvelais, and Amanda Kummeth for our three years of playing Dungeons & Dragons together. Having that space to escape to a different world for a few hours every week or two was truly necessary during the pandemic and onwards. Let's keep telling silly stories with each other in the future.

I am beyond thankful for Zac Dietrich and his relentless support and patience with me during my graduate career. Being long-distance for two years and helping you move halfway across the country in the middle of a pandemic was a challenge, but I knew I could count on you no matter what. Your unfailing smile and jokes always bring me out of a bad mood and have been instrumental in getting me through this program. Thank you for being willing to move out here to California, and thank you for wanting to stay now that you have hobbies that require an ocean nearby.

I must also acknowledge my funding sources. The NASA grant #80NSSC19K0987 supported Chapter 2 and Chapter 3. Chapter 2 was supported, in part, by the NSF-AGS grant #2024170. Chapter 4 was supported by the University of California, Irvine Department of Chemistry. I also acknowledge fellowship funding from the U.S. Department of Energy Office of Science Graduate Student Research Program.

The text of Chapter 2 in this thesis is a reprint of the material as it appears in *ACS Earth and Space Chemistry* used with permission from the American Chemical Society under the Creative Commons Attribution 4.0 License (`https://www.creativecommons.org/licenses/by/4.0/`). The co-authors listed in this publication are Amy E. Christiansen, Alyssa M. Burns, Virendra P. Ghate, and Ann Marie G. Carlton. The last co-author in this publication directed and supervised research which forms the basis for this dissertation.

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PUBLICATIONS

In review. **Flesch, M.M.**; Rosanka, S.H.; Chiu, Y.T.; Carlton, A.G. The ratio of $PM_{2.5}$ to aerosol optical depth in humid and arid regions of the U.S.: Impacts of aerosol liquid water. *Submitted to Environmental Science: Atmospheres in November 2023.*

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PRESENTATIONS

Flesch, M.M.; Christiansen, A.E.; Ghate, V.P.; Carlton, A.G. (2020) ***Oral:*** "Investigating the Clear Sky Bias: Cloud-Relevant Aerosol Chemistry" at the American Association for Aerosol Research Annual Conference, held virtually October 5th-9th, 2020.

Flesch, M.M.; Christiansen, A.E.; Ghate, V.P.; Carlton, A.G. (2020) ***Oral:*** "Investigating the Clear Sky Bias: Cloud-Relevant Aerosol Chemistry" at the AirUCI Internal Symposium, Irvine, CA, March 12th, 2020

Flesch, M.M.; Christiansen, A.E.; Ghate, V.P.; Carlton, A.G. (2019) ***Poster:*** "Fine Particles: Now You See Them, Now You Don't" at the UC Irvine 3rd Annual Environmental Research Symposium, Irvine, CA, December 4th, 2019

Flesch, M.M.; Hanson, C.D., (2018) ***Oral:*** "Analysis of Vibrational Modes of Chemically Modified Tone Wood" at the UNI Honors Thesis Symposium, Cedar Falls, IA, April 14th, 2018

Flesch, M.M.; Hanson, C.D., (2018) ***Poster:*** "Analysis of Vibrational Modes of Chemically Modified Tone Wood" at the American Chemical Society National Meeting, New Orleans, LA, March 18-22nd, 2018

Flesch, M.M.; Hanson, C.D., (2017) ***Poster:*** "Method Development for Analyzing Vibrational Modes of Unattached Violin Plates" at the Iowa Academy of Science Annual Meeting, Cedar Falls, IA, April 21-22nd, 2017

Flesch, M.M.; Hanson, C.D., (2017) ***Poster:*** "Method Development for Analyzing Vibrational Modes of Unattached Violin Plates" at the American Chemical Society National Meeting, San Francisco, CA, April 2-6th, 2017

Flesch, M.M.; Hanson, C.D., (2016) ***Poster:*** "Development of a Semiconductor Based Quadrupole" at the Iowa Academy of Science Annual Meeting, Des Moines, IA, April 22-23rd, 2016

Flesch, M.M.; Hanson, C.D., (2016) ***Poster:*** "Development of a Semiconductor Based Quadrupole" at the American Chemical Society National Meeting, San Diego, CA, March 12-16th, 2016

ABSTRACT OF THE DISSERTATION

Spatiotemporal Trends in Aerosol Physicochemical Properties at the Surface in the Contiguous United States
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Doctor of Philosophy in Chemistry

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Professor Ann Marie Carlton, Chair

The atmospheric aerosol burden affects air quality and climate, and is characterized with a variety of techniques, including optical and filter-based mass measurements. Aerosol optical properties are largely a function of particle size, which is influenced by chemical composition. Notably, aerosol composition that affects hygroscopicity plays a determining role in particle size by affecting water uptake. Aerosol liquid water (ALW) is an important and ubiquitous particle constituent. However, it is poorly characterized by regulatory surface monitoring networks that measure fine particulate matter ($PM_{2.5}$) mass. Satellite measurements of aerosol optical depth (AOD) observe aerosols *in situ* with ALW intact. Surface measurements of $PM_{2.5}$ mass concentrations are often compared to satellite AOD. This work seeks to reconcile filter-based mass and optical measurements of ambient aerosols through fundamental chemistry. I apply a thermodynamic model to ambient meteorological and particle chemical composition data across the contiguous United States (CONUS) to estimate ALW mass. I perform statistical analyses to establish significant associations among ALW and optical aerosol measurement techniques. The topics of air quality and climate often create an interested but sometimes misinformed public. Scientific literacy begins in the classroom by instructing students in fundamental science and chemistry so they can later apply these concepts in their chosen careers.

My dissertation aims to 1) reconcile ambient aerosol measurements made via different mass and optical techniques and platforms through understanding aerosol chemical properties and 2) improve student learning of contextualized analytical chemistry concepts via the development of interactive virtual prelab activities for an instrumental analysis course. To address these aims I 1) investigate the differences in aerosol physicochemical properties on cloudy and clear sky days at a surface monitoring location in the Midwestern U.S. from 2010 to 2019 (945,796 samples), 2) analyze decadal trends in the $PM_{2.5}$-to-AOD ratio (η) across the U.S. as a function of chemical composition from 2006 to 2015 (5,193,448 samples), and 3) assess changes in student learning outcomes and laboratory preparation after the addition of choose-your-own-adventure (CYOA) virtual prelab activities in an upper-division instrumental analysis chemistry course. In Bondville, Illinois, I apply cloud-screening quality assurance information in the AERosol RObotic NETwork (AERONET) data record in seasonal comparison of aerosol optical and physical properties on cloudy and clear sky days. With optical measurements, I find that ambient aerosols are physically larger on cloudy days and that ALW estimated from chemically speciated $PM_{2.5}$ mass measurements is a plausible contributing explanation. Through surface observations of AOD and $PM_{2.5}$, I calculate η at multiple locations across the CONUS and assess chemical and optical properties over time. I compare surface observations of η in August 2015 and January 2016 to Community Multiscale Air Quality (CMAQ) model simulations. I find the spatial and temporal variation in η across humid and arid locations is influenced by chemical constituents that affect ALW mass concentrations, particularly on peak days. Comparison with two representative month-long CMAQ simulations for summer and winter reproduce similar spatial and temporal variability in η to surface observations. I statistically compare laboratory report rubric learning outcomes between quarters with and without virtual prelab activities and conduct Likert opinion surveys across three offerings of an analytical chemistry laboratory class. I find that the addition of CYOA prelab activities improve student mastery of analytical chemistry concepts in laboratory reports and lessened student concerns about preparing for complex laboratory experiments. Future directions of this research include the need for quantitative understanding of ambient ALW and varied cloud-cover conditions, along with better preparing students via improving scientific literacy and contextualizing learning in the classroom to apply fundamentals in environmental chemistry measurements and analyses.

Chapter 1

Introduction

1.1 Background

1.1.1 Atmospheric Aerosol

Aerosols are solid or liquid particles suspended in a gas. In the atmosphere, they impact climate, air quality, and human and ecosystem health. Atmospheric particles have inorganic and organic components that influence their physical, chemical, and optical properties. They encompass a wide range of chemical composition, volatility, phase state, size, hygroscopicity, and partitioning behavior between the gas and condensed phase.[1,2] Primary aerosols are directly emitted (e.g., sea spray, dust, biomass burning), while secondary aerosols form through chemical reactions of gaseous precursors in the atmosphere followed by partitioning to a preexisting matrix or via new particle formation.[3–5] Directly emitted aerosols are often physically generated and span a large range in size, from tens of nm to tens of μm in diameter. Secondary aerosol generally tend to be smaller and exist in the fine mode. The U.S. Environmental Protection Agency (EPA) defines fine particulate matter, particles with an aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$), as an air quality criteria pollutant, as it has been linked to respiratory illnesses, cardiovascular disease, and premature mortality.[6–10] The EPA began measuring 24-hour integrated $PM_{2.5}$ mass concentrations in 1980, and implemented hourly measurements beginning in 2008. Excessive concentrations of $PM_{2.5}$ in the atmosphere worsen air quality and can cause both acute and chronic health effects in humans.[11–13]

Ambient $PM_{2.5}$ mass concentrations and chemical composition vary spatially and temporally across the United States.[14,15] Additionally, atmospheric aerosols have direct (light scattering and absorption) and indirect (perturbation of cloud properties) effects on the Earth's radiation budget.[4,16] They create haze in the atmosphere, reducing visibility due to increased light scattering.[4,17,18] Some aerosols higher in the planetary boundary layer (PBL) tend to uptake water vapor and can also serve as cloud condensation nuclei (CCN), with chemical composition, phase state, and droplet size modulating this ability.[19–23] Increased aerosol concentrations from anthropogenic and biogenic sources lead to greater numbers of CCN in the atmosphere with smaller droplet sizes, which can lead to increased cloud reflectivity and lifetimes.[24–27] However, aerosol-cloud interactions remain poorly understood, particularly regarding climate and radiative effects.[28] The composition of atmospheric aerosols is complex and changes under different conditions, with only a fraction of the possible constituents well characterized.

1.1.2 Chemical Composition of Aerosol

Some of the major chemical constituents of atmospheric particles in the contiguous United States (CONUS) include the inorganic species such as sulfate, nitrate, ammonium, dust, organic species, and water. Particulate sulfate, nitrate, and ammonium typically form from chemical reactions of gaseous precursors in the atmosphere. These precursors include SO_2

and NO_x (NO and NO_2), which primarily originate from fossil fuel combustion.[3] SO_2 is also emitted to the atmosphere from volcanic eruptions and from the oxidation of dimethyl sulfide near ocean water,[3,4] and NO_x from lightning strikes.[29] Ammonia (NH_3) enters the atmosphere from animal waste, wildfires, and other biological sources across the United States and converts to ammonium in the particle phase.[29,30] Oxidation of SO_2 and NO_x in the atmosphere leads to the formation of two dominant particle phase species, sulfate and nitrate. NH_3 can form salts with these acidic species, particularly in the Midwestern U.S. where ammonium concentrations can exceed those of nitrate and sulfate.[31,32] The hygroscopicity of sulfate and nitrate can also affect particle phase water concentrations, though relative humidity (RH), temperature, and particle number concentration also play a role. Sulfate-containing aerosols contribute significantly to haze in the atmosphere,[33,34] particularly in the southeastern U.S. where sulfate is more prevalent. In the western U.S. nitrate dominates aerosol composition.[35,36] Seasonal cycles in both sulfate and nitrate concentrations occur across the country, where sulfate typically peaks in summer months and nitrate peaks in winter months, though absolute concentrations vary spatially.[15,37,38] Regional spatiotemporal trends in these inorganic constituents are well-studied, while detailed understanding of organic constituents remains primarily confined to short-term, intensive field campaigns or long-term, routine monitoring network broad categorization of carbon-containing organic species.

Organic species are an important, complex component of atmospheric aerosols. Oxidation of atmospherically abundant volatile organic compounds (VOCs) from biogenic and anthropogenic sources generate many secondary oxidation products that are difficult for routine monitoring network measurement methods to individually quantify. In the atmosphere, these VOCs react and partition into the particle phase, often via aerosol liquid water (ALW).[4,39,40] As sulfate and ALW due to inorganic species mass concentrations decline over the CONUS, the organic fraction of aerosols are becoming larger contributors to total $PM_{2.5}$ mass concentrations, particularly in the southeastern U.S.[41–43] Regulatory networks that measure

$PM_{2.5}$ chemical composition typically report total organic species mass concentrations as an organic carbon (OC) value. However, organic species are not comprised of only carbon, and estimates of the total organic matter (OM) from measured OC concentrations are often reported in the literature.[42] An accepted standard OM to OC ratio of 1.8 is widely used for the CONUS, though many studies indicate seasonal variability in this ratio is significantly different from this value.[44–46] OM also affects particle hygroscopicity, particularly in areas with high biogenic VOC emissions such as the southeastern U.S.[47]

1.1.3 Long-Term Aerosol Chemical Speciation Measurements

Federal routine monitoring networks such as the Interagency Monitoring of PROtected Environments (IMPROVE) network and the EPA's Chemical Speciation Network (CSN) have measured $PM_{2.5}$ mass concentrations and chemical composition at sparsely spaced locations across the United States for decades.[48] Both networks operate on a 1-in-3 day schedule, where filter samples of particulate matter are taken once every 3 days as a 24 hour integrated sample. The IMPROVE network measures PM_{10} and $PM_{2.5}$ mass concentrations and $PM_{2.5}$ chemical speciation at 160 monitoring stations across the CONUS, with a record dating back to 1988.[49] The EPA CSN record begins in 2000 with several hundred monitors across the CONUS, some of which are co-located with an IMPROVE monitor.[50]

The IMPROVE Network uses four modules with separate inlets and different filters for a range of characterization techniques.[51] Modules A, B, and C collect only fine particulate matter ($PM_{2.5}$) and Module D collects both fine and coarse particulate matter (PM_{10}). Module A uses a Teflon filter and measures the gravimetric mass of $PM_{2.5}$, elemental concentration, and light absorption. Module B has a carbon denuder on the inlet and uses a nylon filter that is analyzed for the sulfate, nitrate, nitrite, and chloride anions via ion chromatography. IMPROVE uses X-ray fluorescence (XRF) to analyze other, lower concentration species measured on this filter such as sodium, calcium, magnesium, and potassium. Module C uses a quartz fiber filter to analyze organic and elemental carbon (OC and EC, respectively) by thermal optical reflectance.[52] Lastly, Module D has a PM_{10} inlet and utilizes a Teflon filter similar to Module A, though only gravimetric mass of PM_{10} is collected. EPA's CSN measures $PM_{2.5}$ after collecting particles for 24 hours on either Teflon, nylon, or quartz filters, performing the same type of analysis as the IMPROVE network for species present.[50] These networks measure many components of aerosol well, though they frequently miss the direct measurement of ALW, an important $PM_{2.5}$ constituent.

1.1.3.1 Aerosol Liquid Water

ALW is a ubiquitous contributor to aerosol mass that routine monitoring networks do not frequently measure. It efficiently scatters visible light, impacting visibility and remotely sensed measurements of aerosol optical depth (AOD).[15,18,53,54] A study by Nguyen et al. in 2016 estimates that ALW can make up as little as 3% and as much as 80% of aerosol mass at aerosol mass spectrometer field campaigns across the world.[55] Secondary organic aerosol (SOA) formation is traditionally thought to occur via the partitioning and reaction of water-soluble gases onto dry organic aerosol.[56] However, it is also well-established that semi-volatile water-soluble organic compounds can partition into the aqueous phase of aerosols and cloud droplets,[57] undergoing chemical reactions to form lower volatility species that remain in the particles,[58] contributing significantly to SOA mass.[59] RH plays a significant role in the fractional contribution of ALW to particulate mass, especially near clouds.[60–62] ALW mass concentrations are also a function of chemical composition, temperature, and particle number concentration.

Although ALW is a widespread and important component in ambient atmospheric aerosols, it is often not directly quantified. Most laboratory studies are performed at dry RH conditions,[63] and only a few field studies have measured ALW in ambient conditions.[64,65] The U.S.

EPA Federal Reference Method (FRM), Federal Equivalence Method (FEM), and CSN monitoring stations that measure $PM_{2.5}$ mass and its chemical constituents on filters typically equilibrate them at an RH of 30-40% and 20-23 °C,[66,67] though the IMPROVE network does not strictly adhere to these standards. This often does not represent the ambient conditions during sampling, particularly in places such as the humid southeastern U.S.[68] The difference between the equilibration RH and temperature and ambient conditions can lead to the loss of nitrate, ALW, and semi-volatile organic species before filter analysis.[35,68] When ALW is the focus of studies, most often it is to investigate hygroscopic particle growth factors or in relation to visibility in national parks.[69–71] Previous work indicates ALW varies diurnally, similar to trends in hygroscopic growth factors.[64] As ALW is difficult to measure in ambient conditions, it is frequently estimated using a variety of models, such as the thermodynamic equilibrium model ISORROPIA.[72] ISORROPIA only considers inorganic components (e.g., sulfate, nitrate, ammonium) in the calculation as water uptake due to these species largely determines the amount of ALW compared to contributions from organic species. Hygroscopicity parameters (κ_{org}) for the organic fraction of aerosol are typically low, though they can vary greatly between urban and rural locations and overall aerosol composition,[73] and these can be used to estimate organic ALW mass concentrations.[19] In general, ALW due to organic species contributes a small fraction to overall $PM_{2.5}$ mass concentrations compared to ALW due to inorganic species but is still an important component of particulate matter.

1.1.4 Long-Term Aerosol Optical Measurements

Aerosol optical properties within the vertical column have been measured by the AErosol RObotic NETwork (AERONET), managed by the National Aeronautics and Space Administration (NASA), since 1993 across the CONUS.[74] AERONET measures AOD, a summation of the total refraction and absorbance of aerosols, using sun photometers stationed at or near the surface looking upwards. The light scattering of particles limits visibility with a hazy appearance and is largely due to Mie scattering.[3] Sun photometers measure the total optical depth of the column from the surface to the top of the atmosphere, subtracting Rayleigh scattering due to trace gases (e.g., water vapor, ozone, nitrogen dioxide, etc.) and other species to determine the optical depth due to aerosols at a series of discrete wavelengths from 340 to 1640 nm.[75] These measurements occur once every few minutes during daylight hours at hundreds of surface stations around the Earth. AERONET also calculates a measure of the average size distribution of aerosols within the vertical column, the Ångström exponent (α), from observed AOD using the spectral dependence of aerosols.[76–79] An $\alpha \geq 2$ typically indicates the population is more heavily comprised of chemically generated fine mode particles, while an $\alpha \leq 1$ tends to indicate evenly mixed coarse and fine mode particles in the column.[79–81] Size is a determining factor in the light scattering of particles, which is also modulated by chemical composition, including ALW. Ångström exponents are often expressed within the visible light range (most often 440 to 870 nm) as the shorter wavelength of 440 nm is more sensitive to the fine mode volume fraction.[80] This sensitivity is helpful when utilizing AOD to estimate near-surface $PM_{2.5}$ mass concentrations.

Surface measurements of AOD from AERONET are frequently used to quality check satellite observations.[75,82] Similarly to AERONET sun photometers, the Moderate Imaging Resolution Spectroradiometer (MODIS) onboard the Aqua and Terra satellites observes ambient aerosol radiative properties unperturbed once or twice daily across the Earth.[83] MODIS AOD measurements are often paired with near-surface daily $PM_{2.5}$ mass concentration measurements or are used to estimate $PM_{2.5}$ mass concentrations at a greater spatial resolution than surface monitors can provide. Epidemiological studies often apply satellite AOD to estimate near-surface $PM_{2.5}$ mass to determine human health endpoints.[84,85] These studies occur primarily in the eastern U.S. where AOD and $PM_{2.5}$ mass concentrations correlate well. Closely tied to the measurement of AOD is the measurement of visibility and haze, similar optical indicators of ambient particulate matter,[86] which is monitored by the IMPROVE network. Determination of particulate matter composition via optical methods is difficult as aerosols are complex and particle chemical composition is not uniform throughout the vertical column. Some components reflect and scatter light (ALW, sulfate, nitrate) while other species (organic carbon, dust) absorb light, all of which can coexist in the particle phase.

1.1.4.1 The Clear Sky Bias

Multiple federal agencies have monitored air quality aloft for decades using remote sensing techniques that can be validated with surface networks such as AERONET. The MODIS AOD data product is frequently used to study aerosol optical properties and estimate near-surface $PM_{2.5}$ mass concentrations. These columnar observations provide a wealth of information about the composition of our atmosphere, but there are limitations. Satellite observations frequently screen out measurements taken during cloudy periods[87,88] due to increased uncertainty of these retrievals.[89,90] As the Earth has approximately 60% of its surface covered by clouds at any given time,[91] this is a key knowledge gap. This also limits the applicability of satellite findings to clear sky periods only. Previous work shows that aerosols have different chemical, physical, and optical properties in the presence of clouds compared to a clear sky day.[92,93] Similarly, the indirect effect of aerosols on clouds contribute to the largest uncertainties in climate projections.[28,56] Where agreement between $PM_{2.5}$ mass and AOD is poor, this cloud-free sampling bias is a contributing factor to increased uncertainties in these retrievals.[94,95] In the presence of clouds, AOD enhancement due to physical growth from water uptake[60,62,96,97] and aqueous reactions[98] can occur. Cloud processing of atmospheric particles can further affect the vertical profile of the chemical species present.[59,99,100] The lack of measurements taken during cloudy periods impacts greater understanding of aerosol-cloud interactions.

1.1.5 Surface Air Quality Measurement Gaps

Surface air quality monitor spatial coverage of the United States is mostly concentrated where the majority of people live, in coastal cities. The EPA has been monitoring weather, air quality, and environmental contaminants at these stations since 1980, with many stations reporting data records of more than twenty or thirty years. The Clean Air Act of 1963 and its subsequent amendments in 1970, 1977, and 1990 paved the way for the EPA to establish monitors specifically to measure air quality and $PM_{2.5}$ mass concentrations, with thousands put into operation during the 1970s through the 1990s. There are now several thousand monitors in operation around the CONUS from multiple regulatory networks, of which a select few are shown in Figure 1.1 pertaining to the studies performed in this dissertation. The majority of these monitors are located near moderate to highly populated areas. IMPROVE network stations are predominantly located in rural national parks, which increases the overall monitor coverage. However, the employed 1-in-3 day sampling method at IMPROVE and occasionally FRM and FEM monitors impedes near-real time $PM_{2.5}$ analysis. Peak aerosol-emitting events, such as wildfires, emit significant amounts of primary organic aerosol aloft that are not always well characterized by surface monitors that could improve understanding of acute health effects and immediate air quality impacts due to these events.[101–103] Air quality has improved across the U.S. since implementation and continual updating of the National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ mass concentrations.[7] The eastern U.S. has had the greatest success in reaching and maintaining $PM_{2.5}$ mass concentrations below the NAAQS standards, primarily due to a significant reduction in sulfate mass concentrations.[33,41,55,104,105] The western U.S., where wildfires are common, has had improvements in air quality as well, though such acute exposure events are becoming more prevalent and have the potential to limit the current trend in air quality. Improving regulatory monitor coverage and effectively informing the public about air quality as it relates to health is increasingly important in a changing climate.

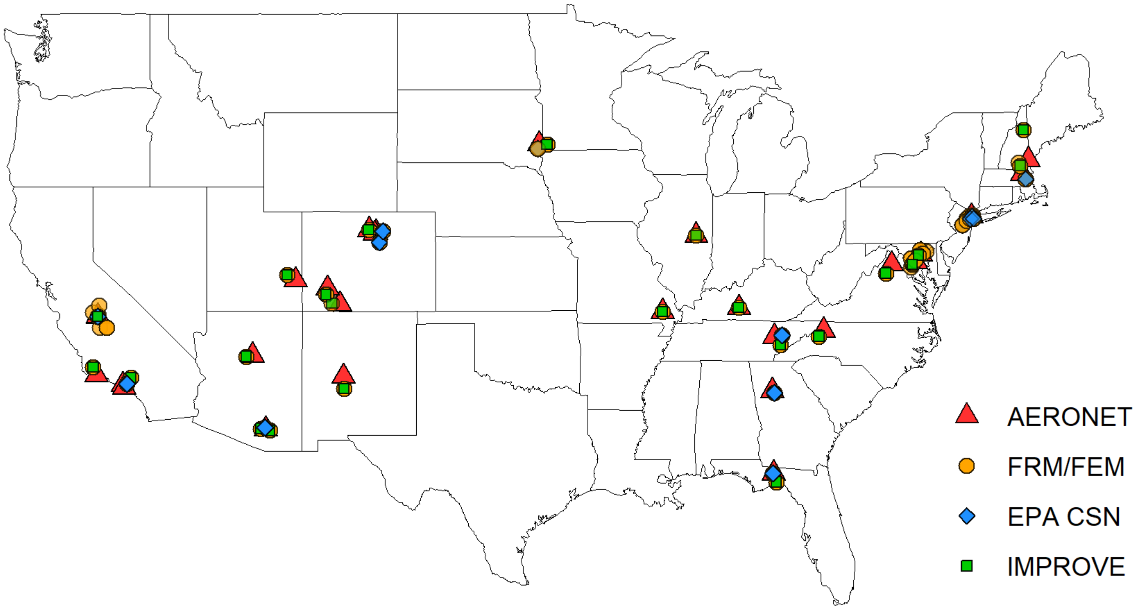

Figure 1.1: AERONET, EPA, FRM, FEM, and IMPROVE monitoring stations across the contiguous United States that measure aerosol optical properties and ambient $PM_{2.5}$ mass concentrations and chemical composition used in this work.

1.2 Informing Effective Science Communication

Understanding the impact of $PM_{2.5}$ exposure on human health is important for all members of society. With information about air quality, pollution events, and more readily available on the internet, it is important that such information be communicated effectively. Post-secondary and graduate education often focuses on sharing scientific results to those also in the scientific community, where complex topics and jargon are commonly used in peer-reviewed journals and conference presentations. Promoting training in more diverse types of science communication throughout undergraduate education is important to ensure everyone can effectively understand and disseminate information about air quality to a variety of audiences. Climate change affects everyone, and improvements in both measurements and understanding the impacts from those measurements is necessary to safeguard the environmental prospects of future generations.

1.2.1 Chemical Education in a Changing Environment

As technology and the needs of the workforce evolve and change, so too must the way we train and educate students. The ways in which chemistry is taught have shifted significantly over the last several decades, as studies show improvements to a variety of metrics that represent student learning, retention, and application of skills gained during their collegiate educational experience.[106,107] New graduates are expected to have more than basic knowledge gained from coursework in their program's field of study; career listings often want potential applicants to show that they have applied the knowledge in a meaningful or impactful way, whether that be via presenting at conferences, performing outreach in the local community, or even teaching or tutoring in the subject area. In other words, students must be able to effectively communicate about chemistry to a variety of audiences. Chemistry courses often employ written laboratory reports for students to practice their communication skills,[108] though the academic laboratory as structured in today's education environment is still a relatively new concept. The addition of a laboratory component to chemistry courses to provide practical instruction in important concepts has been a tenet of teaching chemistry in post-secondary education for over a century, though considerations for health and safety were not often considered in early iterations of chemical laboratories.[109] Early iterations had students simply observe the instructor and possibly an assistant perform demonstrations at the front of the lecture hall, with the "classical" laboratory set up as we know it not becoming popular until the 1860s. With the widespread use of electricity, ability to pump gas and air into building spaces, and free public education in the 1950s, the modern laboratory space began to take shape. Significant improvements in health and safety considerations and development of instrumentation in the 1980s and onward led to a much greater number of students able to take chemistry classes and learn practical laboratory skills.[109] Providing students with hands-on experience that is directly applicable to the real-world is necessary to not only improve their suitability for certain career paths, but also broaden their knowledge outside of the classroom laboratory experience.

Properly preparing students for performing experiments in the laboratory is essential to maintain safety and ensure they conceptually understand the experiment and any instrumentation being used. Chemistry courses often use pre-laboratory (prelab) exercises to perform this function. Traditionally, prelab exercises prepare students to perform experiments via assigned readings (e.g., laboratory manuals) ahead of time, though students often struggle to engage with this type of assignment,[110] particularly if the experiment encompasses unfamiliar concepts and skills.[111] This type of prelab exercise often adds to the amount of time students spend reading extraneous material before the lab, which is not the intended outcome. Effective prelab exercises that still cover important procedural and safety information should reduce the amount of reading material and cognitive overload students face before an experiment.[112] Implementing more active-learning style prelab exercises shows higher levels of student engagement with the material and feelings of preparation, with mixed results for overall academic performance.[110,111,113] While students seem to prefer active methods over passive assignments for pre-laboratory preparation, there is no definite solution to improve both student attitudes and assessment scores.

The COVID-19 pandemic forced instructors to convert entire course offerings, include laboratories, to online virtual options. Many instructors had little time to do so at the beginning of the pandemic, often choosing to film themselves or a teaching assistant performing the experiment and having students answer questions during or after passively watching the video. With this change in instruction format, some instructors instead updated their laboratory components with virtual activities to better engage students and augment learning during class.[111,114,115] Students often found these additions to be more engaging than passively watching an instructor video or completing an embedded quiz, even if overall grades did not change significantly.[110,113] The lack of student decision making in these virtual options led other instructors to create "Choose Your Own Adventure" (CYOA) style activities, where students make choices and critically analyze their selected outcomes similar to hands-on experimental data.[115–117] Virtual prelabs where students actively participate in determining their optimal instrument conditions or expected data facilitate greater engagement and can better connect laboratory experiments to the lecture material.[32,111,115]

Students benefit more from active-learning style classrooms, particularly in disciplines with complex concepts such as chemistry.[118] This includes providing opportunities for students to improve upon their technical writing skills via laboratory reports,[107,108] as writing peer-reviewed publications or technical reports is common in both academia and the private sector. Students also benefit from operating instrumentation themselves during laboratory classes and performing research studies.[106,112] It bridges the fundamental concepts behind analysis methods and techniques to the physical operation of those instruments that are often difficult for students to put together via traditional learning methods. Hybrid Laboratory Action-Based Pedagogy (HLAB) links non-lab and in-lab activities to maximize student learning experiences with a focus on virtual replacements for in-person instruction during the COVID-19 pandemic.[119] Kelley et al. (2021) compares outcomes with and without in-person laboratory instruction finding the loss of hands-on laboratory experiences to be detrimental to student engagement and competences, though student performance was similar or somewhat worse to in-person experiments.[119] Retention of some aspects of virtual activities for laboratory courses can improve student attitudes and better prepare them for engaging in critical thinking while performing the experiment.[106,110,120] In upper-division chemistry laboratory courses, experiments can provide opportunities for students to ask questions and form hypotheses of their own rather than following a prescribed experimental procedure and achieving the expected outcome with no activation of critical thinking skills. The impact of effective teaching strategies in the lecture and the laboratory that acknowledge multiple learning styles leads to better-prepared future citizens who can continue working to understand our atmosphere and improve air quality at all levels.

1.3 Research Questions

The studies in this dissertation were guided by the following questions:

1. How are aerosol physicochemical properties different on cloudy and clear sky days in the rural, humid Midwestern United States?
2. What factors are driving trends in the $PM_{2.5}$-to-AOD relationship, and are they different across the United States?
3. What effect does aerosol liquid water have on aerosol physicochemical properties over time in the United States?
4. Are virtual replacements for in-person undergraduate chemistry laboratory experiments effective in delivering instruction on fundamental analytical chemistry concepts?
5. How do interactive virtual prelab exercises impact students' conceptual understanding and assessment outcomes for an upper-division analytical chemistry laboratory course?

1.4 Specific Aims

I address these research questions in two parts. I approach Questions one through three via spatiotemporal analysis of large federal $PM_{2.5}$ composition and aerosol optical depth data sets and model outputs across the CONUS. I examine questions four and five via quantitative assessment of survey responses and comparison of student assessment outcomes. In this dissertation, I 1) investigate differences in fine particulate matter composition and particle size on cloudy and clear sky days as defined by a surface monitor, 2) investigate temporal and spatial patterns in the ratio of $PM_{2.5}$ to AOD across the United States and the role of ALW, and 3) analyze how interactive virtual prelab activities improve student outcomes for an upper-division chemistry laboratory course using assessment scores and student attitude survey information.

1.5 Structure of the Dissertation

The study described in Chapter 2 aims to understand the impact of cloud presence on atmospheric aerosol optical properties and chemical composition at a rural location in the Midwestern U.S. using a new method of daily cloudiness determination. At the collocated AERosol RObotic NETwork (AERONET) and Interagency Monitoring of PROtected Visual Environments (IMPROVE) network monitoring stations in Bondville, Illinois, I utilize AERONET's cloud-screening algorithm to bin sun photometer observations as predominantly "cloudy" or "clear sky" and compare measured aerosol physicochemical properties from 2010 to 2019. I perform daily comparisons of AOD, Ångström exponents, and $PM_{2.5}$ chemical composition mass concentrations. Results from this case study suggest meteorology alone is insufficient to explain differences in aerosol optical properties on cloudy and clear sky days, though chemical speciation inclusive of ALW may provide some insights.

Chapter 3 discusses the ratio between near-surface $PM_{2.5}$ mass concentrations and surface-measured columnar AOD (η) from 2006 to 2015 at multiple locations across the CONUS using surface observations and models. I utilize measurements from AERONET, IMPROVE, EPA, and CSN to assess η from the surface, including with ALW and nitrate losses from $PM_{2.5}$ filters accounted for in the total $PM_{2.5}$ mass concentrations. Comparison of these measurements to a winter and a summer simulation from the Community Multiscale Air

Quality (CMAQ) model is described for monthly averages across the CONUS and hourly analysis at four selected locations. This work suggests that accounting for sampling artifacts in surface monitors may improve agreement with model predictions and remote sensing of $PM_{2.5}$ mass concentrations.

The study in Chapter 4 analyzes the effects of virtual prelab exercises on student attitudes and assignment outcomes for an upper-division chemistry laboratory course with complex experiments. The COVID-19 pandemic forced instructors of chemistry lab classes to develop a variety of alternative materials for remote delivery of traditionally hands-on content. One solution to this challenge was virtual choose-your-own-adventure (CYOA) laboratories that allowed students to practice realistic decision-making and collect authentic data. Four virtual CYOA labs were adapted into prelab exercises for an upper-division instrumental analysis course to prepare students for experiments in chromatography method development and electronic circuit design. Findings from this study suggest students felt better prepared to come into the laboratory and perform the experiment with greater understanding of its purpose and concepts, which also showed in laboratory report assessments.

This thesis concludes in Chapter 5, wherein I summarize the main findings from Chapters 2 through 4 and describe the design of an environmental chemistry course intended for upper-division chemistry majors with a lecture and laboratory component. The course will feature applications of my research findings and experiences as part of the curriculum, including several laboratory exercises and small group research projects. Designing this course bridges my research and teaching interests with the intent for future implementation.

Chapter 2

Ambient Aerosol is Physically Larger on Cloudy Days in Bondville, Illinois

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2.1 Abstract

Particle chemical composition affects aerosol optical and physical properties in ways important for the fate, transport, and impact of atmospheric particulate matter. For example, hygroscopic constituents take up water to increase the physical size of a particle, which can alter the extinction properties and atmospheric lifetime. At the collocated AERosol RObotic NETwork (AERONET) and Interagency Monitoring of PROtected Visual Environments (IMPROVE) network monitoring stations in rural Bondville, Illinois, we employ a novel cloudiness determination method to compare measured aerosol physicochemical properties on predominantly cloudy and clear sky days from 2010 to 2019. On cloudy days, aerosol optical depth (AOD) is significantly higher than on clear sky days in all seasons. Measured Ångström exponents are significantly smaller on cloudy days, indicating physically larger average particle size for the sampled populations in all seasons except winter. Mass concentrations of fine particulate matter that include estimates of aerosol liquid water (ALW) are higher on cloudy days in all seasons but winter. More ALW on cloudy days is consistent with larger particle sizes inferred from Ångström exponent measurements. Aerosol chemical composition that affects hygroscopicity plays a determining impact on cloudy versus clear sky differences in AOD, Ångström exponents, and ALW. This work highlights the need for simultaneous collocated, high-time-resolution measurements of both aerosol chemical and physical properties, in particular at cloudy times when quantitative understanding of tropospheric composition is most uncertain.

2.2 Introduction

Aerosol interactions with liquid water impact air quality and regional climate.[121] Most aerosol mass forms in the atmosphere from reactions of gas-phase precursors and partitions into a condensed phase, including into liquid water.[4,56] Aerosol effects on Earth's radiation balance occur through complex mechanisms, and the magnitude of these impacts is highly uncertain.[28] The aerosol direct effect (perturbation of solar insolation via light scattering and absorption) is a function of particle shape and size, among other variables such as particle composition and refractive index. Atmospheric liquid water uptake by hygroscopic particle-phase chemical constituents and subsequent aqueous-phase reactions has a determining effect on aerosol size.[18,122] Liquid water efficiently scatters visible light, and its presence in the vertical column contributes to aerosol optical depth (AOD) and extinction.[15,53,123,124] Improved air quality in recent decades over the United States, largely due to decreases in mass concentrations of the hygroscopic sulfate species in fine particulate matter ($\mathrm{PM}_{2.5}$–particles with an aerodynamic diameter of 2.5 μm or less), has helped improve visibility and reduce aerosol extinction.[37,41,104,105] Aerosol indirect effects (perturbation of cloud properties) contribute the largest uncertainties in climate projections.[28,56] Christiansen et al. (2020) find that $\mathrm{PM}_{2.5}$ chemical composition, important for overall particle hygroscopicity and water uptake, is significantly different on clear and cloudy days across the contiguous U.S. (CONUS).[92] This suggests that accurate prediction of aerosol-cloud interactions requires quantitative understanding of aerosol properties during cloudy periods. However, this is when aerosol physicochemical properties are least understood due to a clear sky bias in atmospheric sampling. This key limitation may become increasing problematic, in part, because global cloud cover is changing in response to climate change.[28,125,126]

Much of the chemical characterization of atmospheric aerosol in the CONUS is derived from surface networks that sample $\mathrm{PM}_{2.5}$ at sparsely spaced locations such as the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network and the U.S. Environmental Protection Agency's Chemical Speciation Network.[49,50] The employed sampling and analysis techniques can remove aerosol liquid water (ALW)[68] and other species during sampling and filter equilibration under laboratory conditions, which differ from the field and best characterize non-volatile particle constituents.[88] Radiometers such as the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the polar-orbiting Aqua and Terra satellites observe aerosol radiative properties unperturbed from the ambient environment and improve upon spatial and temporal resolution of surface networks. Comparison of remotely sensed aerosol radiative properties with the surface measurements of $PM_{2.5}$ mass is restricted to clear sky conditions[87,127] because remotely sensed observations frequently screen out measurements taken during cloudy periods due to increased uncertainty.[74,128–130] Relationships between near-surface-point measurements of $PM_{2.5}$ and columnar AOD differ by season and location.[17,68,131] For the CONUS, correlation between $PM_{2.5}$ and AOD in the east is generally stronger than for the west.[94,95] The cloud-free sampling bias in AOD is a contributing factor when agreement is poor, especially in the western CONUS.[95] Particulate nitrate, a hygroscopic constituent, is more abundant in the western CONUS and is not well captured in filter-based collection methods.[35,132] During cloudy periods, AOD can be enhanced due to physical growth from water uptake[60,62,96,97] and aqueous phase accretion reactions.[98] Further, cloud processing affects the vertical profile of particulate species.[36,59,99]

NASA's surface-based AERosol RObotic NETwork (AERONET) supports and provides quality assurance for satellite observations[74] and screens final data products for cloud contamination, similar to satellite retrievals.[75,82] The IMPROVE network shares six monitoring stations with AERONET sun photometers in the CONUS. Two were temporary AERONET campaigns and four are permanent installations for both networks. The Bondville, Illinois location, in a Midwestern agricultural area, is the only site with data for both networks prior to 2013, and this area of the CONUS records suitable cloud frequency for statistical analysis year-round. AERONET computes AOD at a given wavelength from the total optical depth measured y the sun photometer at discrete spectral wavelengths from the surface to the top of the atmosphere through remove of contributions from Rayleigh scattering optical depth and spectrally dependent atmospheric trace gases.[75] Specifically, Ångström exponents are calculated using least-squares regression of AOD for each non-polarized wavelength measured between two channels. AERONET uses the 440, 500, 675, and 870 nm channels to determine the 440-870 nm Ångström Exponent. Ångström (1929) represents this spectral dependence of aerosols by a power law relationship (eq 2.1).[76]

$$\tau(\lambda) = \tau_1 \lambda^{-\alpha} \tag{2.1}$$

Here, $\tau(\lambda)$ is the AOD at a specific wavelength λ, τ_1 is the approximated AOD at 1 μm, and α is the Ångström exponent. From this, the Ångström exponent can be derived to qualitatively estimate the size distribution of aerosols within the vertical column.[76,78,79] Literature-reported Ångström exponents often use wavelengths within the range of 440 and 870 nm, as the former is sensitive to the fine mode radius and the latter is sensitive to the fine mode volume fraction.[80] Chemically generated fine mode particles typically have values of $\alpha \geq 2$, while physically generated coarse mode particles or evenly mixed fine and coarse mode populations have values of $\alpha \leq 1$.[79–81,133] Several previous studies pair near-surface $PM_{2.5}$ mass measurements with satellite-based total column AOD observations via MODIS and demonstrate statistically significant correlation during cloud-free conditions. However, aloft aerosol, when present, confounds comparison because it is not captured in point measurements at the surface.[95,134–136] By comparing chemically speciated surface $PM_{2.5}$ mass concentrations with columnar AOD and Ångström exponent estimates, we investigate associations among physicochemical properties under clear sky and cloudy conditions.

We employ a combination of surface measurements, satellite products, and thermodynamic modeling to analyze seasonal trends at a collocated AERONET and IMPROVE network site in rural Bondville, IL that has a 10 year record for both networks and frequent cloud cover during all seasons. We capitalize on AERONET's cloud screening process to study differences in aerosol mass, chemical composition, and optical properties as a function of sky cloudiness conditions as observed from Earth's surface. The surface-based AERONET station is ideal for identifying low-level clouds that interact with boundary layer aerosol. We quality check this method with geostationary operational environmental satellite (GOES) observations of cloud top temperature over a representative year of data for this location. We explore seasonal trends in Ångström exponents, AOD, meteorology and $PM_{2.5}$ chemical composition on days that are predominantly cloudy or clear sky from 2010 to 2019 within the context of AERONET cloud flagging. We seek to infer differences in atmospheric particle size, a key determining factor in aerosol lifetime, that is attributable to plausible chemical explanation.

2.3 Experimental

We analyze differences in aerosol optical properties and particle-phase species mass concentrations for a collocated AERONET and IMPROVE network monitoring location in rural Bondville, IL. We use all available surface air quality measurements from January 2010 to December 2019 from the IMPROVE network public archives for the monitoring station (40°05'20" N, 88°37'33" W).[49] IMPROVE surface $PM_{2.5}$ mass concentrations of sulfate, nitrate, organic carbon (OC), sea salt, and crustal species such as calcium, potassium, and magnesium are available as 24 h integrated samples every 3 days. Sulfate and nitrate concentrations are determined using ion chromatography, OC fractions via thermal/optical reflectance, and crustal and sea salt species are determined by X-ray fluorescence (XRF). We assume all XRF species are present as fully water-soluble concentrations in our particle water estimates, as described below, although these species may be part of non-water-soluble matrices such as silicates and dust. While this introduces some uncertainty, sulfate and nitrate largely control overall estimated ALW mass concentrations. For quality assurance, we estimate ALW with and without crustal species, finding that ALW is insignificantly higher with these species excluded (Figure A.1). AERONET public archives provide surface based estimates of columnar AOD at 440, 500, 675, and 870 nm, and Ångström exponents estimated for the 440-870 nm spectral range at the Level 1.0 and Level 2.0 quality levels.[137] We define winter as December, January, and February; spring as March, April, and May; summer as June, July, and August, and fall as September, October, and November. A cloud determination method assigns surface measurements into two bins, "predominantly cloudy day" and "predominantly clear sky day" (discussed below). We use the Mann-Whitney U test, a non-parametric statistical test, in R statistical software[138] to compare to non-normal population distributions. The Mann-Whitney U test determines the probability that a sample from the clear sky population will be smaller or larger than a sample from the cloudy population. We determine statistical significance of $p < 0.05$ for differences in the populations for Ångström exponents, AOD, and $PM_{2.5}$ chemical composition.

We estimate mass concentrations of particle-phase water using the inorganic thermodynamic equilibrium model ISORROPIAv2.1 in the reverse, open-system direction.[72] We assume metastable particles with ammonium nitrate and ammonium sulfate in the aqeuous phase[139] and a well-mixed boundary layer. We estimate inorganic ALW using all available IMPROVE-reported mass concentrations of fine particulate matter chemical constituents. IMPROVE does not measure ammonium ion concentrations at the Bondville location, and neglecting this species in an agricultural area such as Bondville adds uncertainty to ALW estimates. Ammonia is abundant in agricultural areas and facilitates the formation of particulate nitrate, a hygroscopic species with substantial losses from filter measurements, particularly during summer.[35,132] Seasonal cycles and temporal trends of ALW estimates are similar with and without ammonium from 2010 through 2015 for IMPROVE sites across the CONUS.[92] At the Bondville, location, seasonal average ALW estimates with weekly aggregated ammonium filter measurements from a nearby CASTNET site[140] are not significantly different at the 95% confidence level (Figure A.2). We use daily averaged temperature and relative humidity (RH) from the European Centre for Medium range Weather Forecasting (ECMWF) reanalysis model (ERA5) meteorological outputs for estimating ALW mass concentrations.[141] The ECMWF ERA5 reanalysis model yields hourly averages of surface temperature, dewpoint temperature, and planetary boundary layer height. We take daily averages of both temperature values before calculating surface RH using the formula in Huang (2018).[142] We screen RH values $> 95\%$ due to constraints within the model for ALW estimates, although no daily averages were above this threshold for the study period. As in Christiansen et al. (2019) and Nguyen et al. (2015), we estimate organic ALW using the κ-Kohler theory and the Zdanovskii-Stokes-Robinson mixing rule:[43,54]

$$V_{w,o} = V_o \kappa_{org} \frac{a_w}{1 - a_w} \tag{2.2}$$

Briefly, we assume water activity (a_w) to be equivalent to RH. V_o and $V_{w,o}$ are organic matter volume and the associated water volume, respectively; κ_{org} is the assumed organic hygroscopicity parameter of 0.3 employed for rural aerosol.[64,73,92] We use a mass balance method to calculate organic mass (OM) from IMPROVE-measured OC fractions with site- and season-specific (annually) OM/OC ratios, as described in greater detail elsewhere.[44,92] Note that estimated OM/OC ratios at IMPROVE sites across the CONUS, including Bondville, exhibit an upward trend over the last 20 years,[44] as do gravimetric mass measurements since 2011.[143] We divide OM by an assumed density of 1.4 g cm^{-3} to determine V_o.[55,144]

The time resolution of the data products employed creates a limitation in this analysis. AERONET measures aerosol optical properties every 15 (or less) minutes during daylight hours, while IMPROVE provides 24 h integrated chemical measurements every 3 days. However, directly measured *in situ* ALW mass concentrations change over the course of a day in response to changing meteorology and particle chemical composition.[64] Therefore, differences in ALW mass estimates during cloudy versus clear sky conditions described here may be different than we are able to quantitatively assess with existing data sets and limitations of current measurement techniques. This likely results in understated differences for the predominantly clear sky and predominantly cloudy days in these findings because daily averages obscure diurnal patterns in meteorology and particle hygroscopicity that have a direct impact on ALW mass concentrations on a diurnal time scale.

We assess a cloudiness classification method at Bondville, IL: interpretation of the AERONET cloud screening algorithm evaluated with the spatiotemporal pairing of GOES cloud top temperature measurements. AERONET Level 1.0 products, Level 2.0 products, and related cloud information are available approximately every 15 minutes. This improves upon temporal limitations with the MODIS Cloud Mask used in a previous analysis,[92] which is only available once or twice daily. We match all AERONET observation days to IMPROVE measurement days. For quality assurance of the cloudy versus clear classification method, we evaluate half-hourly GOES meteorological observations from 2017, a representative year during the study period, and match them to the nearest AERONET observational times within a 30 minute window around the GOES observation. We derive a cloud mask from the AERONET quality assurance (QA) cloud screen algorithm to assess impacts of retrieval cloudiness category on the measured AOD and Ångström exponents.[75]

We use the AERONET QA algorithm that identifies each observation made by the Sun photometer with specific data quality metrics, provided upon request for this analysis, which are available in the associated data repository for this publication. Pre-processing removes observations due to instrument errors or full cloud obscuration of the field of view (marked as a "0" in the meta data). AERONET releases all other observations makred on a scale of 1 to 4 in their public data products, indicating the maximum quality level of the retrievals.[75] A "1" is likely cloudy, a "2" has most clouds removed, a "3" has nearly all clouds removed, and a "4" has nearly all clouds removed and final calibration applied. For the study period at the Bondville site, no "3" observations are recorded. The Level 1.0 data product retains all observations marked "1" to "4", while the Level 2.0 data product retains only those marked "4". For both data products, we analyze all days with 10 or more non "0" retrievals during AERONET observational hours. We deem an individual day in the Level 1.0 product "predominantly cloudly" if the number of observations marked "1" and "2" meets or exceeds 50% of the total number of measurements for the day (Figure 2.1). We employ "1" and "2" (cloud-impacted) observations only to determine daily cloudiness classification. In the statistical comparison between cloudy and clear categories, we use days with 10 or more quality-assured "4" points only. Thus, while we use the Level 1.0 product for determining an implicit daily cloud flag, our statistical evaluation uses only observations that qualify for the Level 2.0 data product.

All AOD measurements pass the triplet variability quality assurance step in the algorithm and have approximate uncertainties of 0.02.[75] The AERONET QA algorithm uses solar au-

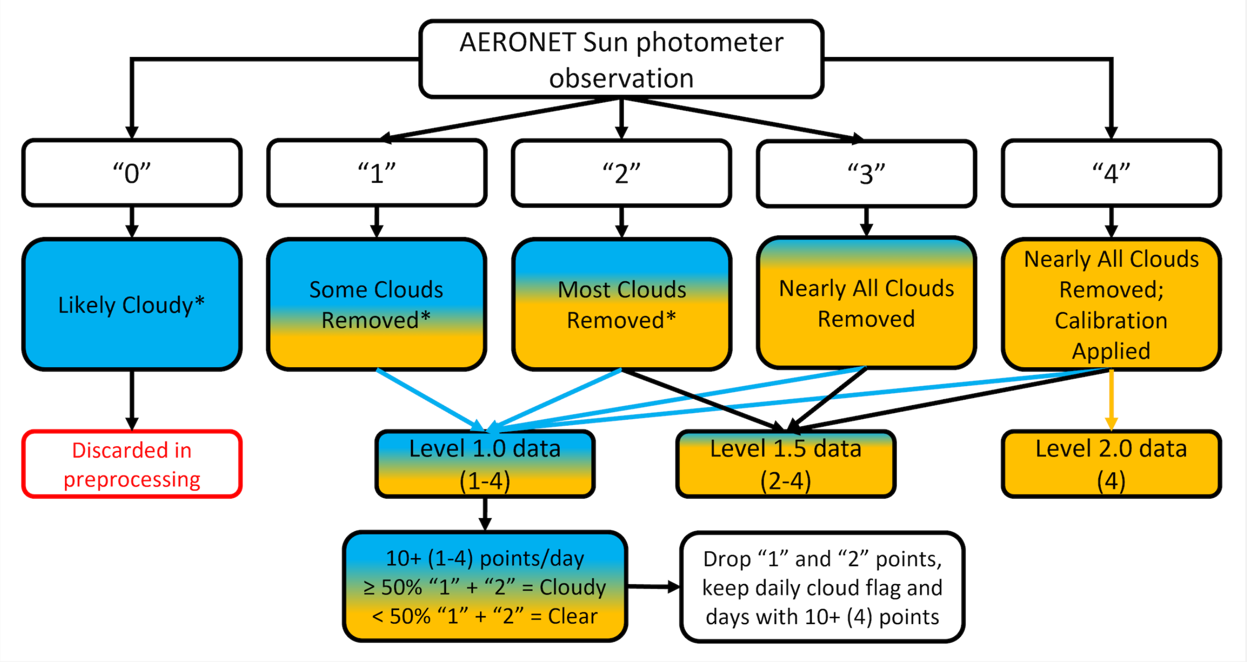

Figure 2.1: Flow chart of the cloud processing method for sun photometer observations by the AERONET quality assurance algorithm and subsequent categorization and analysis performed within this chapter. Note that these metadata flags are not available in AERONET public data products. Each number is indicative of the maximum achieved quality level and those demarked with * can be impacted due to instrument operation issues or measurement anomalies. With this, "0" and "1" are "cloudy", "2" as "some clouds", and "3" and "4" as "clear sky".

reole radiance with respect to the scattering angle to account for and remove high thin cirrus clouds, but cloud contamination for low optical depth (AOD < 2) clouds is possible.[75,89,145] The Sun photometer can find the sun during broken cloud conditions during some of the days binned as "predominantly cloudy," and the differences presented here represent a lower boundary for distinction. For brevity, legends and figure captions will refer to the cloudiness bins defined above as "cloudy" and "clear sky". We cross-check instrument downtime and periods of collimator tube obstructions (e.g., spider webs or water droplets), removing any retrievals that passed pre-processing steps by the QA algorithm but fit either of these criteria, as well as AOD measurements with values above 1.5 to retain only physically realistic measurements for this location. We bin observations by day to be consistent with the 24 h chemical speciation measurements from IMPROVE.

Agreement of sky conditions via this method is supported with measured cloud top temperatures from the GOES historical archive and lends support to the suitability of our AERONET-derived categorical cloud determination method.[129,130,146] GOES observes a range of meteorological variables every half hour, and we use the cloud top temperature measurement[82,129,130] to determine how frequently both GOES and AERONET identify clouds over the Bondville site. We sample the year 2017 and match AERONET observations from the Level 1.0 data product and pair in space and time within 15 min of the corresponding GOES measurement. For example, a GOES measurement of cloud top temperature at 12:45 PM local time is paired with valid AERONET observations (”1” to ”4”) within the time frame of 12:30-12:59 PM. We bin each GOES observation as ”cloudy” if a cloud top temperature is recorded, and ”clear sky” if GOES does not observe clouds at the Bondville location. We deem a GOES-based day as ”predominantly cloudy” if 50% or greater of the GOES observations record cloud top temperatures. GOES detects high-level thin cirrus clouds, but the GOES-13 lacked the 12 μm channel to detect this cloud type well.[89,147] The spatial and temporal resolutions of the GOES data are 9 km and 30 min, respectively. It is therefore possible for GOES to miss boundary layer shallow cumulus clouds with a smaller spatial extent and shorter lifetime. In addition, cloud top temperatures are not retrieved during broken cloud conditions. GOES retrievals are not available for our entire study time frame. Direct comparison of the methods for 2017 finds that the daily predominantly cloudy or predominantly clear day bin definitions for AERONET and GOES agree for approximately 76% of the paired comparisons (Figure A.3). We rely on the AERONET method in this analysis.

2.4 Results and Discussion

During the study period in Bondville, population distributions for values of AOD at all measured wavelengths of 440, 500, 675, and 870 nm are significantly different on clear sky days than on cloudy days in every season (Figure 2.2). On clear days, Ångström exponents (AE) are significantly larger, indicating a smaller average physical size for the ambient aerosol distribution (recall the inverse relationship in 2.1), with the sole exception of winter (Figure 2.3). In spring and summer, median AOD values are greatest at all examined wavelengths, and differences between cloudy and clear sky days are most pronounced (Table A1.1). There are more predominantly clear days than predominantly cloudy days in every season, with only 16-27% of days binned as "cloudy" (Table A1.2). The difference in medians is small, yet more often than not, is more than the 0.02 instrumental uncertainty.

Meteorological variables of temperature, RH, and PBL height affect AOD and are insufficient to fully explain the predominantly clear and cloudy period AOD and AE differences. Integration over the PBL height, where most aerosols reside, is a primary driver of AOD. In spring and summer when the boundary layer is the highest, AOD is greatest. However, during spring and summer, PBL height is significantly higher on clear sky days (Figure 2.4), while, in contrast, AOD is larger on cloudy days. While particle growth through humidification is consistent with elevated AOD and smaller AE values on predominantly cloudy days,[148] differences in RH are not significant for any season. Temperature does not exhibit significant differences on clear sky and cloudy days in most seasons at Bondville, apart from higher temperatures on cloudy days in the spring (Table A1.3). Wintertime meteorology seems to be similar regardless of cloudiness bin for all variables, although this season also has the fewest cloudy days (Table A1.2). Surface-sensible and latent heat fluxes or large-scale subsidence could induce seasonality in meteorology, including the PBL height, rather than the boundary layer temperature and humidity that are evaluated here. This indicates that factors other than the meteorological variables we evaluate here (i.e., T, RH, and PBL)

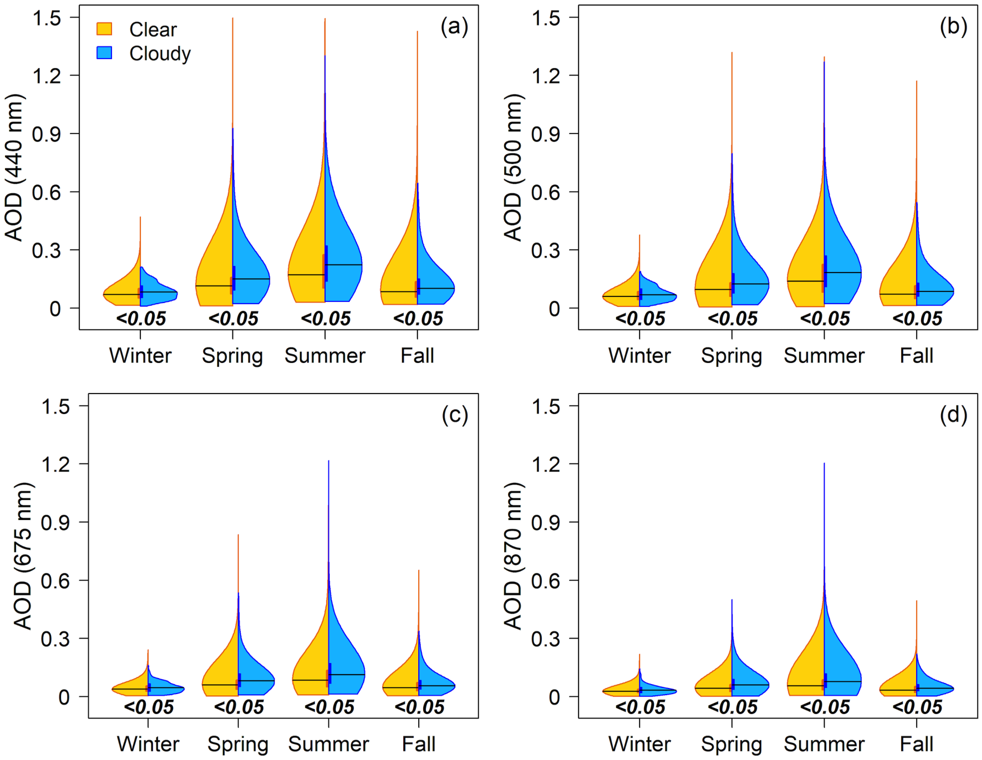

Figure 2.2: Seasonal distributions from 2010 to 2019 of AOD at (a) 440, (b) 500, (c) 675, and (d) 870 nm on clear sky (gold) and cloudy (blue) days as binned by the AERONET quality assurance algorithm method. Vertical gold (clear sky) and blue (cloudy) bars represent the 25^{th} to 75^{th} quartile of each distribution, with the black horizontal line as the median. Black numbers are p-values from two-sided Mann–Whitney U tests, with bold italicized font indicating statistical significance between distributions where a p value < 0.05 indicates the two populations are not equal.

may play a role in the observed differences in aerosol optical properties.

Chemical composition of $PM_{2.5}$ provides a plausible explanation for AOD and AE observations at the Bondville monitoring site, should surface measurements adequately represent the boundary layer column. Particle composition affects AOD and AE via intrinsic properties such as refractive index and through influences on particle size via changes in hygroscopicity and water uptake.[16,149] Cloudy versus clear sky differences in overall $PM_{2.5}$ and ALW mass are sharpest during spring and summer, exhibiting similar patterns to AOD and AE measurements for those categories. In the summer when $PM_{2.5}$ and ALW mass are highest, AOD is also the highest. IMPROVE-measured $PM_{2.5}$ mass concentrations that include ALW

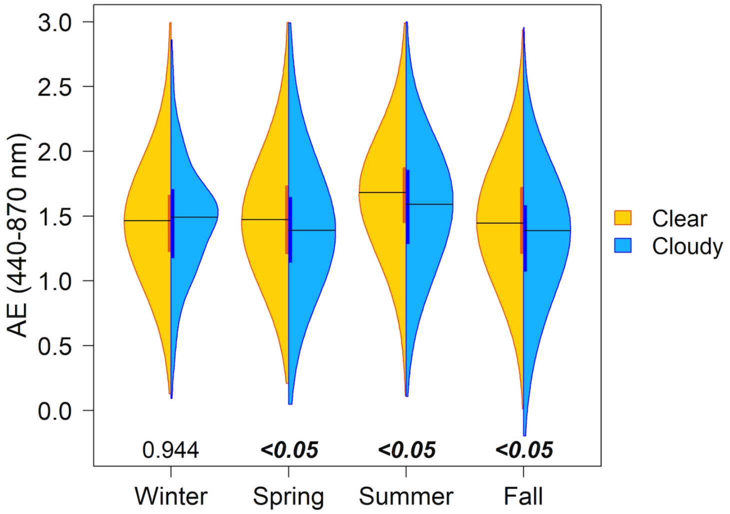

Figure 2.3: Seasonal distributions from 2010 to 2019 of AE for the 440–870 nm wavelength range on clear sky (gold) and cloudy (blue) days. Cloud bins, coloring, numbers, and statistical significance are as in Figure 2.2

are the highest on cloudy days in every season but winter (Figure 2.5). This is similar to patterns in AOD and is physically consistent with an abundance of hygroscopic particles that take up water to grow in size on cloudy days. Aqueous-phase processing of air parcels is documented to shift accumulation mode aerosol to a larger population size.[150,151]

$PM_{2.5}$ chemical speciation determines particle hygroscopicity, water uptake, ALW mass concentrations, and subsequently aerosol size. Christiansen et al. (2020) found significant differences between cloudy and clear sky distributions of particle chemical consitutents, especially ALW, using the MODIS Cloud mask approach paired with IMPROVE $PM_{2.5}$ chemical speciation over a continuous 5 year period across the CONUS.[92] Springtime distributions in sulfate, ALW, and organic matter are significantly different on predominantly clear and cloudy days in Bondville. Distributions in RH for cloudy and clear sky days do not exhibit significant differences in any season and therefore cannot be the sole explanation for clear versus cloudy patterns in ALW mass concentrations. In Bondville, sulfate concentrations are highest in summer and spring on cloudy days, when ALW mass concentrations are estimated to be greatest and AOD is observed to be highest. Elevated sulfate and ALW mass

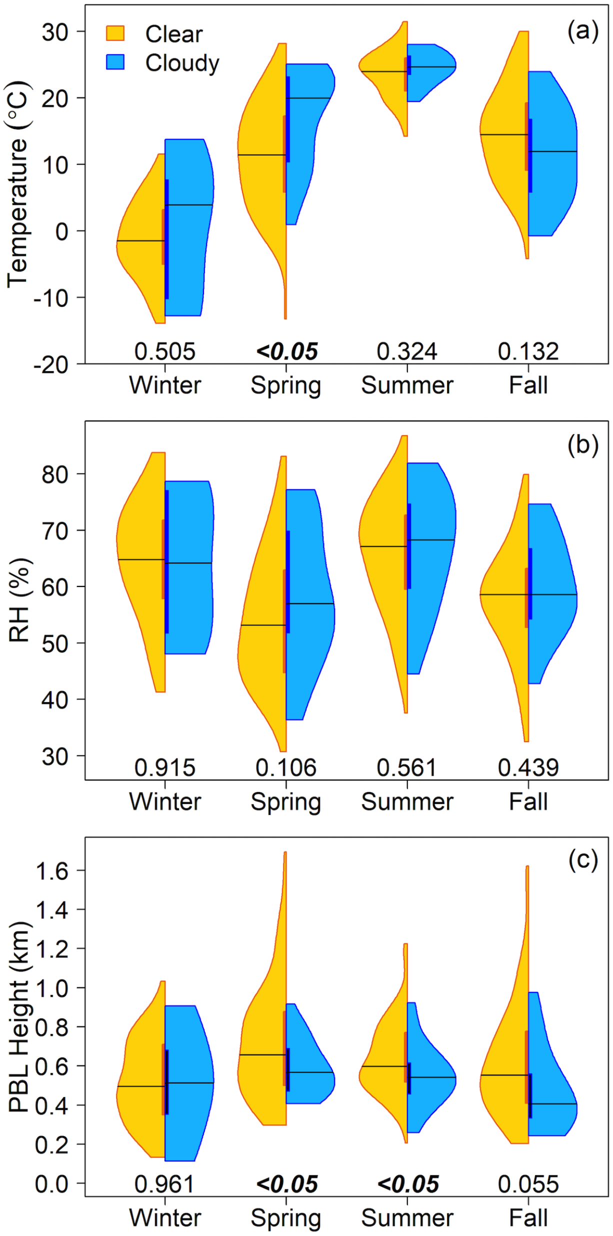

Figure 2.4: Seasonal distributions of (a) temperature, (b) relative humidity (RH), and (c) planetary boundary layer (PBL) height on clear sky (gold) and cloudy (blue) days. Cloud bins, coloring, and statistical significance are as in Figure 2.2.

on cloudy days are consistent with the hypothesis that an abundance of hygroscopic aerosol in the boundary layer can serve as cloud condensation nuclei to affect cloud systems.[27,152] In contrast, ALW mass concentrations during wintertime are highest on clear days when nitrate is most abundant. In the Po valley, an agricultural region of Italy, nitrate was found to

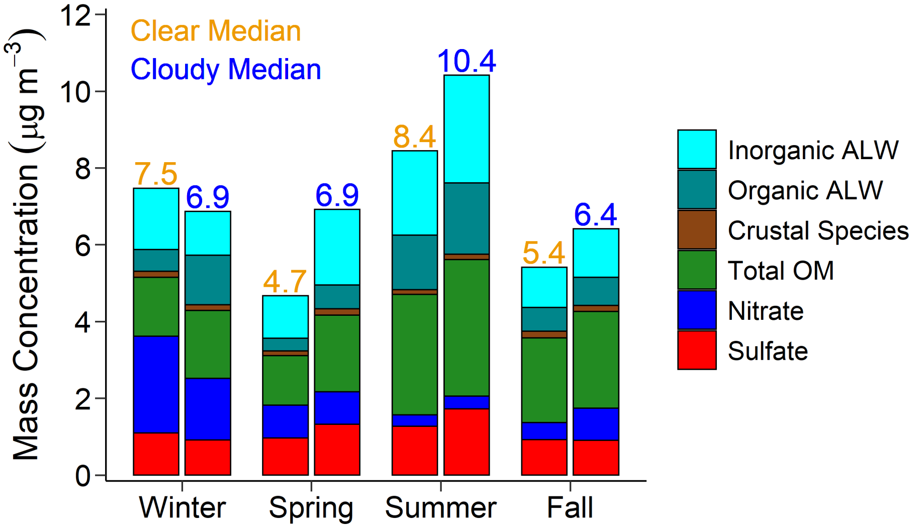

Figure 2.5: Stacked bars of median total $PM_{2.5}$ chemical composition and ALW observation-based estimates during clear sky (left bars, gold numbers) and cloudy (right bars, blue numbers) days as binned using the AERONET QA algorithm for 2010–2019. Crustal species include calcium, potassium, magnesium, and sea salt species of sodium and chloride. Total OM was calculated from IMPROVE-measured OC using OM/OC ratios defined in the Experimental Section. Table S4 identifies statistical significance for individual chemical constituents.

have a determining impact on ALW.[153] Organic mass varies seasonally and is a key source of uncertainty. Organic species can alter intrinsic volumetric absorptive properties important for AOD, and the associated hygroscopicity is poorly understood relative to inorganic salts. The chemical composition that controls water uptake in Bondville during winter (nitrate) versus spring and summer (sulfate) may contribute to seasonal patterns regarding clear versus cloudy optical measurements of AOD and AE.

Decadal analysis indicates that sulfate and inorganic ALW mass concentrations decreased in Bondville (Figure A.4), similar to trends in sulfate and ALW reported for the southeast region of the U.S.[33,43] This is suggestive that with less water uptake, smaller particles would exhibit larger AE values over time. However, there is not a clear trend in AE. This may be due to increasing organic mass, or nitrate mass, which initially declines then dramatically increases (by a factor of 8) in recent years (Figure A.4). Also, an increase in coarse-mode aerosol (PM_{10})–particles with aerodynamic diameters of 10 μm–that are not evaluated here, may affect the AERONET-measured AOD and the reported AE. Hand et al. (2017) and Malm et al. (2007) suggest increasing PM_{10} over the CONUS in recent years.[154,155] The AE value on clear sky days in larger, suggesting an aerosol population more dominated by fine mode aerosol;[133] however, this may not be observed in every circumstance.[98] Previously in agricultural Midwest locations, investigators find several factors, including organic species, control AOD and extinction.[134,135] Mass concentrations of OM and organic ALW (estimated from OM, not OC) do not decline over the decade. The ratio of OM/OC varies substantially by season (Figure A.5), indicating substantial changes in organic constituents. Extinction properties of ambient carbon may vary as the OM/OC ratio does, and this may also influence these trends.[44] A higher OM/OC ratio indicates more oxidized organic aerosol, which can be more hygroscopic. Over the studied decade because sulfate mass is decreasing while OM is not, the fractional contribution of organic species to particle dry mass and influence on overall particle hygroscopicity is increasingly important. Critical open questions regarding water uptake by particle-phase organic species remain.

2.5 Conclusions

From 2010-2019 at the collocated AERONET and IMPROVE network monitoring stations in rural Bondville, IL, median AOD at 440, 500, 675, and 870 nm is higher on cloudy days in every season. Ångström exponents are smaller on cloudy days in every season except winter when nitrate mass concentrations are highest. Meteorological variables of temperature, RH, and PBL height are insufficient to fully explain the statistical significance for differences in AOD, Ångström exponents, and ALW mass concentrations on predominantly clear sky versus cloudy days. Aerosol chemical composition that alters particle hygroscopicity to affect water uptake and growth is a plausible explanation consistent with observations that suggest physically larger particles and higher AOD measured by AERONET on predominantly cloudy days. Size largely determines aerosol extinction and lifetime, critical parameters that define particle impacts on air quality, regional radiation budgets, and surface temperature. Our findings here suggest that aerosol size is different on cloudy days, when tropospheric composition is least understood. This warrants further study and highlights the need for collocated chemical, optical, and physical aerosol measurements at high time and vertical resolution, including at cloudy times, when quantitative understanding of boundary layer aerosol is least robust.

Chapter 3

Chemically specific sampling bias: the ratio of $PM_{2.5}$ to surface AOD on average and peak days in the U.S.

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3.1 Abstract

Accurate quantitative description of the atmospheric fine particulate matter ($PM_{2.5}$) burden requires an understanding of aerosol amounts and properties that transcends measurement platforms. For example, air quality studies often seek to describe ambient $PM_{2.5}$ with columnar aerosol optical depth (AOD), point measurements of mass, or some combination. $PM_{2.5}$

chemical constituents affect such measurements differently. We investigate the ratio of $PM_{2.5}$-to-AOD (η) from 2006 to 2015 at multiple surface locations across the contiguous U.S. using observations and models, and quantitatively account for $PM_{2.5}$ sampling bias of nitrate and aerosol liquid water (ALW). We find η peaks during winter and is lowest in summer at all locations, despite contrasting seasonality in $PM_{2.5}$ mass and AOD. Accounting for loss of nitrate and ALW from $PM_{2.5}$ monitors improves consistency among η calculations in space and time, most notably for wintertime peak values. Co-occurrence of extreme $PM_{2.5}$ mass concentrations and AOD events declined in the eastern U.S. but not in the west. On peak days in all locations there are higher concentrations of ALW relative to average chemical composition. This suggests an increased fraction of ambient $PM_{2.5}$ is detectable via optical methods but not well described by surface networks on peak days. In 2006 AOD is more strongly correlated with chemical constituents than with $PM_{2.5}$ mass. The Community Multiscale Air Quality (CMAQ) model reproduces similar spatial and temporal variability in η to surface observations in winter and summer simulations. Accounting for sampling artifacts in surface monitors may improve agreement with model predictions and remote sensing of $PM_{2.5}$ mass concentrations.

3.2 Introduction

Globally, exposure to ambient fine particulate matter ($PM_{2.5}$) is a leading cause of non-communicable disease such as worsened cardiovascular health and increased mortality risks.[9,13] Long term, chronic $PM_{2.5}$ exposure is harmful to human health, as is acute exposure at high concentrations.[11] Accurate, quantitative understanding of $PM_{2.5}$ trends and peak concentrations is therefore critical to the management of air resources to safeguard human health. The U.S. Environmental Protection Agency (EPA) and other regulatory bodies in the U.S. employ surface monitoring networks to measure $PM_{2.5}$ mass and characterize chemical composition. For example, the EPA's Chemical Speciation Network (CSN) measures 1-in-3 or 1-in-6 daily aggregated $PM_{2.5}$ chemical composition predominantly in and near urban areas.[50] The Interagency Monitoring of PROtected Visual Environments (IMPROVE) network measures fine aerosol mass and chemical composition predominantly in national parks around the U.S. on a similar 1-in-3 daily schedule.[49] EPA's chief $PM_{2.5}$ monitoring network measures daily-aggregated and/or hourly concentrations across the country, primarily located near populated areas.[156] These networks record improved air quality and visibility at many locations over past decades, most notably in the eastern United States and in localized sites of California.[15,30] Recently, to improve upon the spatial limitations of surface networks and increase global coverage, satellite observations of columnar aerosol optical depth (AOD) are frequently employed to understand near-surface $PM_{2.5}$.[8] AOD products advance understanding of the troposphere's $PM_{2.5}$ burden and facilitate epidemiological studies that link $PM_{2.5}$ mass concentrations to health endpoints related to exposure.[10,157–160]

Application of once-or-twice a day satellite-derived clear sky AOD to understand surface $PM_{2.5}$ and its impacts are limited. During cloudy times, quantitative understanding of tropospheric composition from satellites is most uncertain and often not possible. At any given time most of the Earth's surface is covered by clouds. Even during clear sky conditions, it is difficult to accurately assess surface $PM_{2.5}$ with remotely retrieved AOD in some locations. In the arid western contiguous United States (CONUS), surface reflectivity induces large uncertainty in AOD.[14,161] Further, the chemical speciation of atmospheric particles via satellite is challenging, and toxicology and epidemiology studies suggest $PM_{2.5}$ chemical constituents, including water soluble species, contribute differently to adverse health impacts.[162–165] Babila et al. (2020) hypothesize that including ALW in $PM_{2.5}$ mass concentrations may reconcile epidemiological studies that find positive statistical associations among sulfate in ambient $PM_{2.5}$ and health endpoints in the humid eastern U.S. but not in the arid western U.S.[68] ALW mass is largely controlled by sulfate in the eastern U.S.,[92] while nitrate, which is more hygroscopic, is more abundant in the western U.S. $PM_{2.5}$ mass surface monitors exhibit negative artifacts for nitrate, organic species, and ALW from filter measurements,[35,68,166] while satellites observe aerosols *in situ* with the condensed phase of these chemical species intact.

Peak mass concentrations of $PM_{2.5}$ and some constituents are frequently not captured by satellite AOD products due to satellite pass over times. The Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua and Terra satellites measures AOD and has nominal overpass times on a sun-synchronous near-polar orbit of 10:30 a.m. (descending) and 1:30 p.m. (ascending) local time,[83,167] which does not correspond to peak concentrations.[168,169] During six weeks of direct surface measurements during the Southern Oxidant and Aerosol Study (SOAS),[64] hourly ALW mass concentrations and particle hygroscopicity peak at approximately 8 a.m. Throughout the CONUS, peak values in ALW and nitrate mass are perceived to occur on cloudy days[92,170] when remotely sensed AOD is least reliable. AOD and ALW mass concentrations are both greater in the eastern CONUS, though reported $PM_{2.5}$ mass concentrations are often higher in the west.[40,55,171] Over the CONUS correlations between satellite derived AOD and surface $PM_{2.5}$ mass concentrations are more strongly positive in the east.[14,95,172] Several factors affect the $PM_{2.5}$-to-AOD (η) relationship, such as vertical distribution of particles and their intrinsic physicochemical properties that impact extinction, in addition to surface reflectivity.[173,174] ALW mass and potentially other chemical constituents not well characterized by surface monitors also provide a plausible contributing explanation for the differences among $PM_{2.5}$-to-AOD ratios.[68,175]

Recently, Jin et al. (2020) assessed η for the eastern CONUS from 2003 to 2017.[134] They employed once-a-day MODIS AOD observations and daily integrated surface $PM_{2.5}$ mass reported at EPA regulatory monitors. Their findings demonstrate declining correlation between filter-based surface $PM_{2.5}$ mass and remotely sensed AOD, in addition to decreasing frequency of co-occurrence for peak values of AOD and $PM_{2.5}$ mass. $PM_{2.5}$ mass concentrations decreased dramatically in the eastern U.S. during their study period, most notably as a co-benefit of acid rain rules that also reduced sulfate concentrations.[33,176,177] Mass concentrations of ALW and detection of its optical extinction also decreased in the eastern CONUS, contributing to improved visibility.[41,43,54,178]

For most locales on a clear sky day with a well-mixed boundary layer, $\mathrm{PM}_{2.5}$ and AOD are directly linked:

$$\mathrm{AOD} = \mathrm{PM}_{2.5} \times \mathrm{H} \times f(RH) \times \frac{3 \times Q_{ext,dry}}{4 \times \rho \times r_{eff}} \tag{3.1}$$

Where H is the height (m) of the planetary boundary layer (PBL), f(RH) (dimensionless) is the ratio of ambient-to-dry extinction coefficients as a function of RH, $\mathrm{Q}_{ext,dry}$ is the Mie extinction efficiency of the dry particle (cm^{-1}), ρ is the mass density of aerosols (g m^{-3}), and r_{eff} is the effective particle radius (m). Equation 3.1 is often rearranged and presented as Equation 3.2, and η is used to estimate near-surface $\mathrm{PM}_{2.5}$ mass concentrations from satellite AOD.[134,174,179]

$$\frac{\mathrm{PM}_{2.5}}{\mathrm{AOD}} = \eta \tag{3.2}$$

ALW is implicit in η. f(RH) is an estimate of the ALW-induced change in aerosol radius, and the amount of liquid water affects both ρ and r_{eff} directly. $\mathrm{Q}_{ext,dry}$ describes optical properties of dry particle mass; however, the metastable state of ambient aerosol is ubiquitous,[139] and ALW is found in most ambient aerosol.[55] Consideration of ALW can reconcile AOD and surface $\mathrm{PM}_{2.5}$ mass. Nguyen et al. (2016) find that in the southeastern U.S., seasonality in AOD is more consistent with seasonality in ALW[55] than surface $\mathrm{PM}_{2.5}$ mass.[180] Additionally, Bergin et al. (2000) find better agreement between surface-measured and inferred AOD from micropulse lidar profiles of aerosol backscatter and extinction when accounting for ambient RH and particle hygroscopicity.[181] Thus, accounting for ALW and other semi-volatile $\mathrm{PM}_{2.5}$ constituents that exhibit negative mass artifacts in surface monitoring can facilitate improved understanding of the relationship between near-surface $\mathrm{PM}_{2.5}$ mass and columnar

AOD and how it changes in space and time.

In this work, we evaluate hourly, seasonal, and interannual trends in η from 2006 to 2015 at multiple surface monitoring locations across the CONUS, including the western U.S. where satellite retrievals are more uncertain. Hourly measurements of $PM_{2.5}$ and AOD at surface locations facilitate analysis of η over a day, which is useful due to large diurnal changes in ALW and other variables that affect η. We investigate the plausibility of nitrate and ALW, $PM_{2.5}$ chemical constituents lost by regulatory surface $PM_{2.5}$ monitors, to reconcile $PM_{2.5}$-to-AOD relationships across the CONUS over a recent ten-year period during average conditions and at peak times. We also examine the ability of the EPA's Community Multiscale Air Quality (CMAQ) model, which is used to develop and assess policy related to $PM_{2.5}$,[182] to reproduce observed spatial and temporal trends in η.

3.3 Data and Methods

We analyze η, AOD, and mass concentrations of $PM_{2.5}$ and chemical constituents at surface sites across the CONUS in areas we classify as "humid" or "arid" based on average RH and simulated ALW mass concentrations (Figure 3.1, Table B.1) from 2006 to 2015. This date range encompasses declining $PM_{2.5}$ mass concentrations.[33,177] Figure 3.1a denotes 8 AERONET locations paired with long-term $PM_{2.5}$ chemical speciation measurements within 50 kilometers in red outlines. All other sites outlined in black are studied in August 2015 and January 2016 only, and compared with CMAQ as discussed below. The 8 AERONET sites studied from 2006 to 2015 are depicted in Figure 1b and 1c with their respective paired $PM_{2.5}$ mass concentration monitoring stations. We use all available surface air quality measurements for these 8 locations from January 2006 to December 2015 from AERONET,[137] IMPROVE,[49] and CSN[50,156] public archives. Selected sites are chosen so that all network data records have at least 85% completeness for paired measurement days during the study

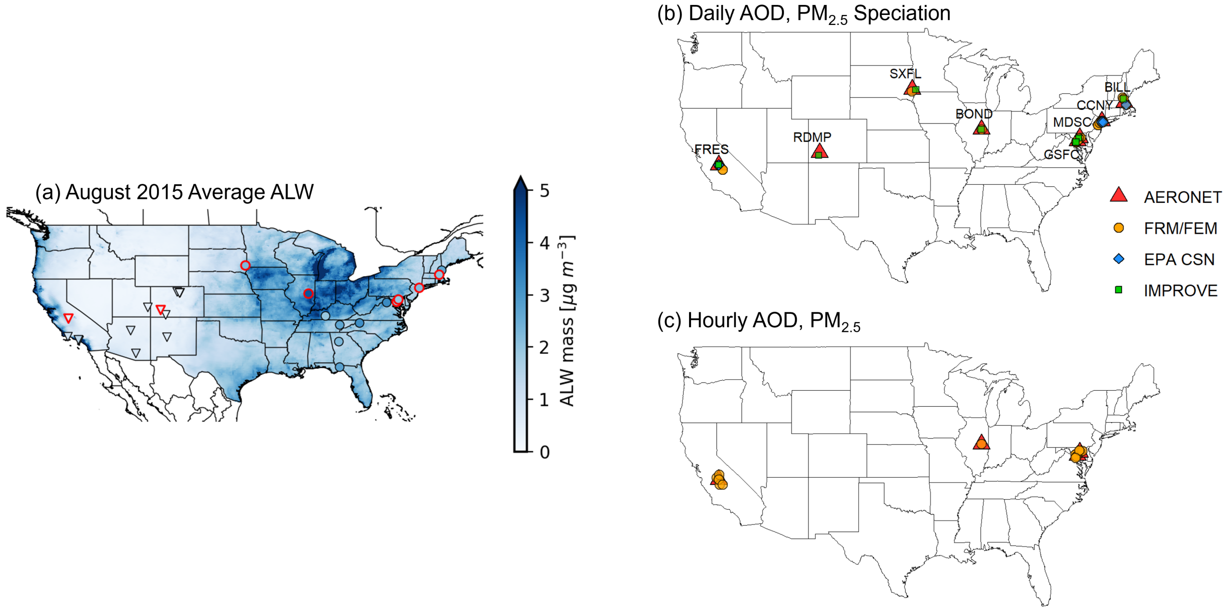


Figure 3.1: (a) Surface monitoring locations and average aerosol liquid water (ALW) mass concentrations simluated by the regional air quality model CMAQ for August 2015 paired in time with observational estimates. Triangles indicate sites in the "arid" region and circles indicate "humid" sites. Black markers indicate AERONET stations used for η comparisons with CMAQ in 2 simulations. Red triangles indicate AERONET stations matched with $PM_{2.5}$ mass and chemical composition monitors analyzed from 2006 to 2015 for (b) daily ambient $PM_{2.5}$ measurements from the IMPROVE Network (green squares), EPA's CSN (blue diamonds), Federal Reference Method (FRM), and Federal Equivalence Method (FEM) monitors (orange circles; Table B.1), and (c) hourly measurements.

period and represent areas with different physical and chemical climatology. We assume consistent surface and aloft aerosol during a given measurement for the matched stations. This assumption is a limitation that adds uncertainty. We employ AERONET surface-based estimates of columnar AOD at 440 nm in the Level 2.0 quality data product with 15 minute (or finer) time resolution as this wavelength has the greatest data completeness at all sites.[75] AERONET measurements are aggregated to temporally match available hourly and daily $PM_{2.5}$ mass concentrations and chemical speciation measurements. IMPROVE and CSN report surface, filter-based $PM_{2.5}$ mass concentrations of sulfate, nitrate, organic carbon (OC), and other species as 24-h integrated samples measured once every 3 or 6 days. We use meteorological categories for seasonal analysis and define winter as December, January, February; spring as March, April, May; summer as June, July, August; and fall as September, October, November.

At IMPROVE and CSN locations, we estimate mass concentrations of ALW using the inorganic thermodynamic equilibrium model ISORROPIAv2.1 in the reverse, open-system direction.[72] The method is described in detail elsewhere;[170] briefly, we assume metastable particles and inorganic chemical species are fully water-soluble. We employ hourly average values of surface temperature, dew point temperature, and PBL height at monitor locations from the European Centre for Medium range Weather Forecasting (ECMWF) ERA5 reanalysis product.[141] We use daily averages of both surface and dew point temperatures to calculate surface RH as in Huang (2018).[142] We remove data points when RH values exceed 95%, which excludes less than 1% of all measurement days. Nitrate is a dominant, hygroscopic, and semi-volatile $PM_{2.5}$ constituent in the western U.S. well documented to exhibit negative sampling artifacts in Federal Reference Method (FRM) and Federal Equivalence Method (FEM) $PM_{2.5}$ monitors.[35,183] We estimate nitrate volatilization from $PM_{2.5}$ filters with relevant chemical equilibrium reactions that describe partitioning between the gas and condensed phase, accounting for the deliquescence relative humidity (DRH, 62% at 25°C).[184] Application of a similar approach to filter measurements in southern California was shown to describe nitrate losses from Teflon filters well.[35] We add the estimated loss to the nitrate values in ALW calculations. Chemical composition data is only available for 24 h integrated samples. Six of the eight CSN locations measure the ammonium ion. We estimate ALW with and without ammonium as a sensitivity at these locations, finding the inclusion of ammonium reduces predicted ALW mass concentrations by 9%. Overall decadal trends do not change (Figure B.1). As the IMPROVE network does not report ammonium concentrations, ALW mass concentrations reported here are calculated excluding ammonium for consistency of analysis across networks.

We use a mass balance method to calculate organic mass (OM) from total OC mass with site- and season-specific OM:OC ratios.[44,45,185] OM:OC ratios are calculated for every paired measurement day with available OC data, then averaged by season per site and applied to organic matter (OM) and organic ALW calculations. Some sites did not begin measuring OC until partway through the time period studied, preventing holistic analysis of the organic contribution to ALW and total $PM_{2.5}$ mass. We assess differences in peak and average mass concentrations at four individual sites where OC is available for the entire study period separately (Figure B.2). ALW due to organic constituents is estimated at IMPROVE locations in a sensitivity analysis using a lower bound hygroscopicity parameter (κ) of 0.05 to represent urban, anthropogenic aerosol and an upper bound κ of 0.3 to represent rural, biogenic aerosol. We find organic ALW contributes, on average, 1-9% of the total $PM_{2.5}$ mass (Figure B.3). For consistency of analysis across networks, the lack of OC data at some sites, and the relatively small contribution to overall $PM_{2.5}$ mass, organic ALW is not included in this analysis.

We calculate the ratio of $PM_{2.5}$-to-AOD (η) at 440 nm at AERONET sites with the network reported $PM_{2.5}$ mass concentrations and with $PM_{2.5}$ adjusted for losses calculated individually for nitrate and ALW for each valid measurement pair (i.e., AOD and $PM_{2.5}$ mass) from 2006 to 2015. We calculate Pearson correlations (ρ_P) among measured values of $PM_{2.5}$ mass concentrations, sulfate, nitrate, AOD, and estimates of PBL height in and from 2006 to 2015 for locations in the humid and arid regions independently. We also examine the annual "hit rate" of extreme events for $PM_{2.5}$ mass and AOD at each location. As described by Jin et al. (2020), a hit rate is defined as the number of hits (days with peak $PM_{2.5}$ and peak AOD) divided by the sum of hits and misses (days with only peak AOD).[134] We compute an annual hit rate (θ) at each location to determine the co-occurrence of extreme AOD and $PM_{2.5}$ mass concentration events for paired measurements:

$$\theta = \frac{\text{days with AOD} \, > \, 75\% \text{ and } [PM_{2.5}] \, > \, 75\%}{\text{days with AOD} \, > \, 75\%} \tag{3.3}$$

Where the numerator is the number of days when both AOD and $PM_{2.5}$ mass exceed their locally determined 75^{th} percentile and the denominator is the number of days when only AOD exceeds its 75^{th} percentile, regardless of $PM_{2.5}$ mass concentration on those days. The 75^{th} percentile is determined locally at each site for the entire 2006 to 2015 study period. We perform a linear regression to determine the change in θ over time. Values of θ closer to 1 indicate frequent co-occurrence of extreme $PM_{2.5}$ mass concentrations and extreme AOD events, while values closer to 0 indicate less co-occurrence of these events.

We evaluate the Community Multiscale Air Quality (CMAQ, version 5.3.3)[186] model's representation of η compared to surface measurements during a representative summer and winter month. CMAQ simulations for the CONUS employ a 12 km x 12 km horizontal grid resolution using meteorological inputs from the Weather Research and Forecasting model (WRFv4.1.1). Anthropogenic and biogenic emissions are based on the EPA's Air Quality Time Series (EQUATES).[187] In our simulation, we represent gas-phase chemistry with the Carbon Bond (version 6.3) mechanism, and the particle microphysics with the aero7 submodel.[188] We perform a summer and winter simulation, each for two months. For summer, we simulate July to August 2015, and for winter, December 2015 to January 2016, where July and December are regarded as spin up periods, respectively, and are not included in the analyses presented here. August is used to describe a representative summer month and January a representative winter month. CMAQ-derived η is calculated for each day and hour of the month matched to surface sampling days in space and time. The version of CMAQ utilized does not account for nitrate losses in the $PM_{2.5}$ mass concentrations, and the paired surface observations similarly only utilize the network-reported $PM_{2.5}$ mass concentrations for this analysis. We use all available data from all sites and present monthly averages. We compare CMAQ-simulated AOD available at 550 nm with AERONET AOD at 440 nm. Lower AOD values are expected at 550 nm than 440 nm due to the spectral dependence of AOD.[75] Four of the AERONET stations (Fresno (FRES), Bondville (BOND), Goddard Space Flight Center (GSFC), Maryland Science Center (MDSC)) have nearby stations that measure hourly $PM_{2.5}$ mass concentrations in August 2015 and January 2016 (Figure 1c), which we compare to diurnal patterns during daylight hours in CMAQ model simulations. All data processing and significance tests are conducted with R statistical software[138] and relevant code and data are available at the related repository.[189]

3.4 Results and Discussion

3.4.1 Temporal and Spatial Analysis

All estimates of η calculated with reported or adjusted $PM_{2.5}$ mass concentrations exhibit the same seasonal pattern at both humid and arid locations from 2006 to 2015, despite contrasting patterns in $PM_{2.5}$ and AOD (Figure 3.2). η peaks in winter and is lowest in the summer. $PM_{2.5}$ mass concentrations are highest at locations in the western arid region and peak in the winter, up to a factor of 4 times greater than summertime values. At sites in the humid eastern U.S., $PM_{2.5}$ mass peaks in the summer and seasonal differences are considerably less. Conversely, AOD is highest at humid locations, with pronounced seasonality (factor of 3) not observed at western sites. At the arid sites, the wintertime peak in η is driven by greater $PM_{2.5}$ mass concentrations and lower AOD. At the humid locations, the wintertime peak in η is driven primarily by lower AOD.

ALW and nitrate are plausible contributors to the spatial and temporal patterns in η. AOD observational techniques observe ambient aerosol unperturbed, while $PM_{2.5}$ surface monitors exhibit negative sampling artifacts for nitrate, organic species, and largely remove ALW.[68,166] In the eastern U.S., non-volatile sulfate mass peaks during summer and plays a predominant role in aerosol physicochemical properties, in particular water uptake. The seasonality in AOD and ALW mass concentrations at humid locations is similar. Further, $PM_{2.5}$ mass concentrations and AOD decline at eastern surface sites over the time period, consistent with

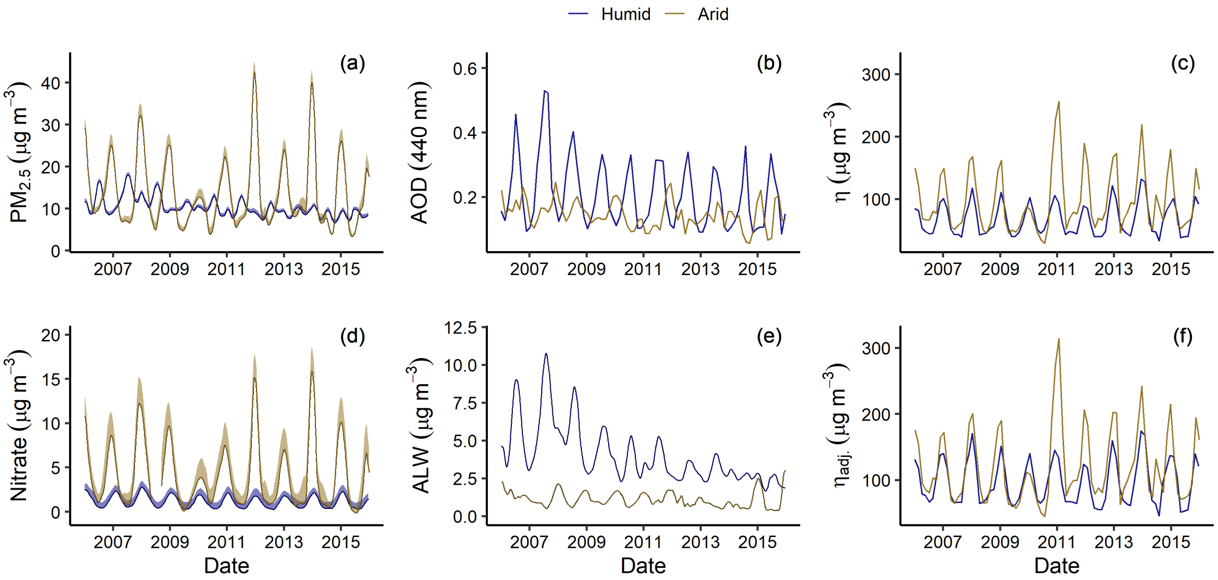


Figure 3.2: Time series of (a) monitor-reported $PM_{2.5}$ mass concentrations (lower line) with nitrate corrections applied (shaded region) (b) aerosol optical depth (AOD, 440nm), (c) η calculated using reported $PM_{2.5}$ mass, (d) nitrate mass concentrations (lower line) with nitrate corrections applied (shaded region), (e) aerosol liquid water (ALW) mass concentrations, and (f) η calculated with ALW and nitrate corrections applied to reported $PM_{2.5}$ mass for the 8 AERONET locations in the humid (dark blue) and arid (dark gold) regions studied from 2006 to 2015.

previously noted observations (Figure B.4).[33,41,43,54,178] Western arid sites do not exhibit a decline in sulfate, ALW, or AOD from 2006 to 2015. Nitrate contributes substantially to particle mass and hygroscopicity in the western U.S., especially during winter when thermodynamics favor the condensed phase. Consistency among η estimates for the humid and arid locations is noted with these adjustments applied, most notably in wintertime peak values (Figure 3.2c, f). Accounting for ALW, Nguyen et al. (2016) could better reconcile surface $PM_{2.5}$ collected on filters with remotely sensed AOD,[175] similar to findings here where the addition of ALW to $PM_{2.5}$ mass concentrations improves correlation with surface AOD (Figure B.5).

At sites in the eastern humid U.S., average AOD values and surface mass concentrations of $PM_{2.5}$, nitrate, sulfate, and ALW estimates significantly decline. Only nitrate mass significantly declines at arid western sites (Table 3.1). Estimated ALW mass concentrations, and how they differ for arid versus humid locations seasonally and over the studied decade, is consistent with the observed trends in AOD. Liquid water is a plausible contributor to these trends.

Table 3.1: Annual changes in aerosol optical depth (AOD) and surface mass concentrations of $PM_{2.5}$ and several chemical constituents at sites in the eastern and western U.S. from 2006 to 2015. Statistically significant linear regression coefficients ($p < 0.05$) are denoted with asterisks.

Region	Parameter	Rate of Change
East	AOD	-6.7×10^{-3} (yr^{-1})*
	$PM_{2.5}$	-0.38 ($\mu g\ m^{-3}\ yr^{-1}$)*
	Nitrate	-0.06 ($\mu g\ m^{-3}\ yr^{-1}$)*
	Sulfate	-0.23 ($\mu g\ m^{-3}\ yr^{-1}$)*
	ALW	-0.40 ($\mu g\ m^{-3}\ yr^{-1}$)*
West	AOD	-3.5×10^{-3} (yr^{-1})
	$PM_{2.5}$	-0.13 ($\mu g\ m^{-3}\ yr^{-1}$)
	Nitrate	-0.06 ($\mu g\ m^{-3}\ yr^{-1}$)*
	Sulfate	-0.01 ($\mu g\ m^{-3}\ yr^{-1}$)
	ALW	-0.03 ($\mu g\ m^{-3}\ yr^{-1}$)

Particle chemical composition varies at the humid and arid sites to affect the amount of ALW, which influences particle size and subsequently AOD. At the arid locations, AOD correlates positively with nitrate, sulfate, and $PM_{2.5}$ mass concentrations in 2006. In 2015, all species exhibit weaker but positive correlation with AOD (Figure 3.3). At the humid locations, positive correlations between AOD with sulfate and $PM_{2.5}$ also decline from 2006 to 2015. Nitrate is slightly anti-correlated with AOD in both years and across the decade. Negative correlation for all variables with PBL height is noted across all analyzed sites. PBL height is greatest in summer and lowest in winter across the CONUS (Figure B.4). The western sites demonstrate greater variability in and stronger anti-correlation of all variables to PBL height relative to locations in the eastern U.S. These spatial and seasonal trends over

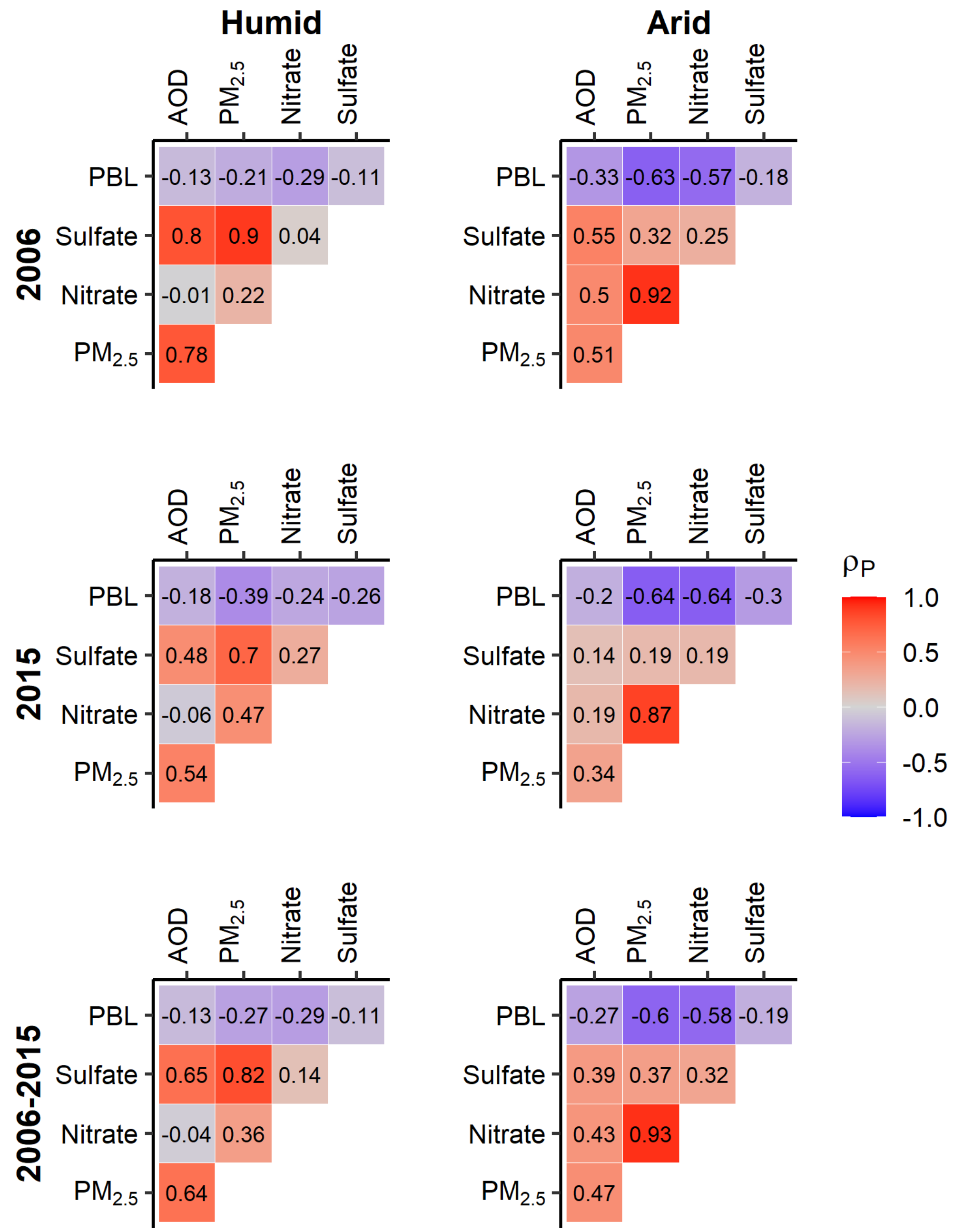


Figure 3.3: Pearson (ρ_P) correlations for aerosol optical depth (AOD) at 440 nm, planetary boundary layer (PBL) height, $PM_{2.5}$, nitrate, and sulfate mass concentrations for sites in the (left) humid and (right) arid regions in (top) 2006, (center) 2015, and (bottom) interannually from 2006 to 2015. Red and positive numbers indicate positive correlation, blue and negative numbers anti-correlation, and gray little to no correlation between two variables. From 2006 to 2015 for the 6 humid locations N=25,211 days, and for the 2 arid sites N=4,858 days.

the decade in the fractional contribution of aerosol chemical constituents at eastern sites is consistent with earlier findings regarding trends in sulfate mass, haze, and visibility, optical indicators of ambient particulate matter burden similar to AOD detection, with changing optical properties detected with AOD measurements.[17,86]

Mean mass concentrations of ALW, nitrate, and sulfate are higher on peak (extreme $PM_{2.5}$ mass and extreme AOD) days at humid and arid locations alike (Figure 3.4). Comparison of average and peak day $PM_{2.5}$ chemical composition for each site illustrates the impact of semi-volatile species and their detection *in situ* with satellites versus regulatory filters. Over the studied decade, $PM_{2.5}$ mass and chemical composition are relatively similar for average days at individual sampling sites. Peak day concentrations decline by 50% or greater at sites in the humid region. On peak days when $PM_{2.5}$ mass concentrations and AOD are elevated, the fractional contributions of ALW and sulfate are larger. For the two arid sites, average and peak day chemical composition mass is nearly identical in both 2006 and 2015 for the Red Mountain Pass (RDMP) and Fresno (FRES) locations. The contribution of nitrate is greater for the Fresno site compared to all other sites. At the four locations where OC is measured throughout the decade, contributions from OM are similar on average days in 2006 and 2015, but peak day fractional contribution of OM increases at all sites, with the exception of the Bondville (BOND) site (Figure B.2). The precise sampling artifacts of filter-collected OC are difficult to quantify and may contribute to variation in $PM_{2.5}$-to-AOD (η) values.

Collectively and individually, all humid locations exhibit a significant decline in annual hit rate (θ) from 2006 to 2015, while the arid locations do not (Figure 3.5). The decrease in co-occurrence of extreme reported and adjusted $PM_{2.5}$ mass concentrations and AOD values suggests diminishing ability of satellite AOD to estimate peak values of near-surface $PM_{2.5}$ at eastern locations.[134] Addition of ALW and nitrate to daily $PM_{2.5}$ mass concentrations at BOND renders the change statistically insignificant. For the arid west locations, peak

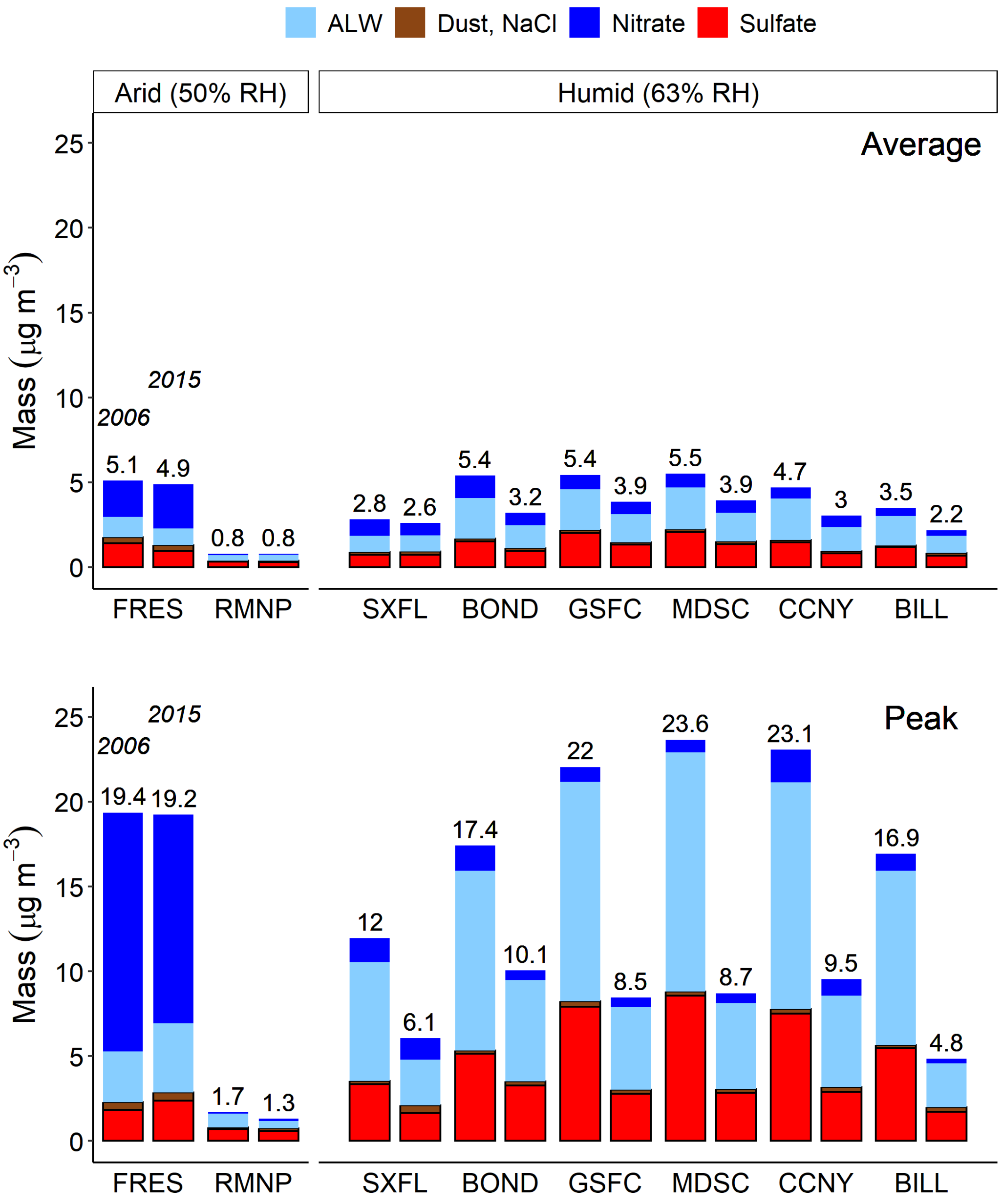


Figure 3.4: Sum of measured species that contribute to $PM_{2.5}$ chemical composition consisting of non-volatile species (sulfate, dust (calcium, magnesium, potassium), NaCl) and semi-volatile species (nitrate, aerosol liquid water (ALW)) on average (top) and peak days (bottom, where AOD and $PM_{2.5}$ are both above their locally determined 75th percentile) for the 8 AERONET stations analyzed in 2006 (left bars) and 2015 (right bars).

AOD and surface $PM_{2.5}$ mass concentrations co-occur at similar rates from 2006 to 2015, with no significant change in annual hit rate with or without ALW and nitrate adjustments. The sites with significantly decreasing θ had hit rates of nearly 100% co-occurrence of peak

AOD and $PM_{2.5}$ events in 2006 and declined to approximately 55% co-occurrence or less in 2015. Additionally, sites with declining hit rates had fewer days with elevated $PM_{2.5}$ mass concentrations later in the decade compared to earlier in the decade. The number of peak AOD days varied less. Aloft aerosol is becoming more prevalent due to wildfires and contributes to AOD and surface $PM_{2.5}$ differently,[101–103,190] consistent with declining hit rates in the east.

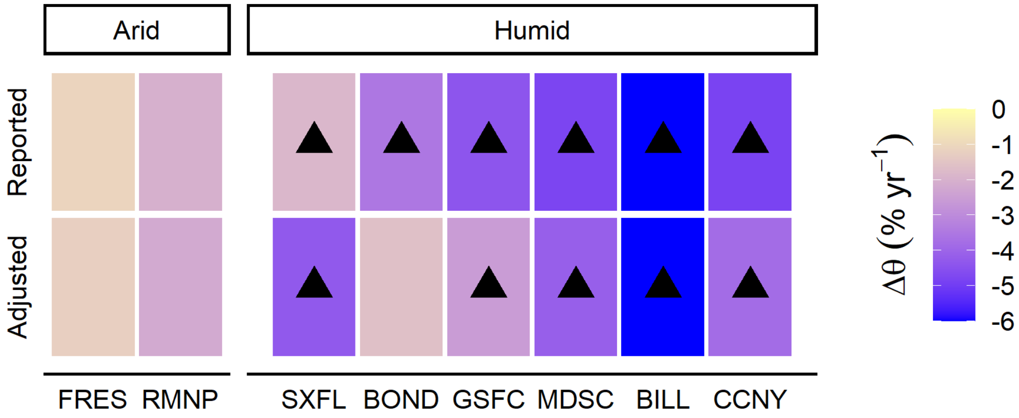


Figure 3.5: Annual rate of change in hit rates (θ) for peak values of $PM_{2.5}$ mass concentrations and AOD for the 8 AERONET stations analyzed from 2006 to 2015 with reported $PM_{2.5}$ mass concentrations (top) and aerosol liquid water and nitrate adjustments (bottom). Blue indicates a decrease in θ and yellow little to no change. Black triangles represent statistical significance ($p < 0.05$).

3.4.2 Comparison of Observations with CMAQ

Spatial and seasonal patterns in CMAQ-simulated and AERONET-observed η are identified in the summer and winter simulations (Figure 3.6). In both the observations and CMAQ predictions, η peaks in winter and is lowest in the summer across the CONUS, similar to the seasonal patterns in Figure 3.2. Localized peaks in η in January at surface network sites in the California Central Valley and the Colorado Plateau near Denver are also present in the CMAQ prediction. CMAQ's replication of both localized and regional patterns in surface observations in summer and winter simulations highlight a usefulness for the regulatory model when either satellite AOD retrievals or $PM_{2.5}$ monitors are scarce.

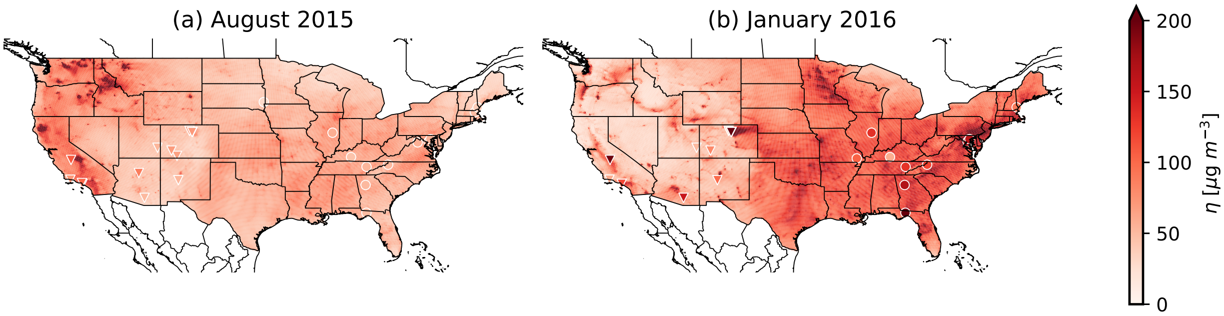


Figure 3.6: Monthly averaged eta (η) simulated by CMAQ with monthly averaged surface network observations overlaid in white outlined triangles ("arid" region) and circles ("humid" region) across the CONUS for (a) August 2015 and (b) January 2016.

Diurnal patterns in simulated and observed η are similar at all sites, with the largest differences occurring at humid locations during the summer in the morning (Figure 3.7). CMAQ predictions of η are offset higher relative to observations, consistent with the utilization of a smaller wavelength used for AOD in CMAQ versus AERONET due to the spectral dependence of AOD.[75] Observed and modeled η peaks at or before local 9:00 a.m. in both seasons at all locations, prior to Aqua and Terra pass over times. The greatest differences between measured and modeled hourly η values occur for the sites with larger ALW mass concentrations (MDSC, GSFC, BOND; Figure B.6). Summertime differences in η are minima near midday. ALW mass concentrations during SOAS were highest from 8:00 to 9:00 a.m. and lowest during midday,[47] similar to trends in the differences between CMAQ-simulated and surface estimated η at humid locations. In the winter when ALW mass concentrations are low for all sites, differences between the model and observations throughout the day are small.

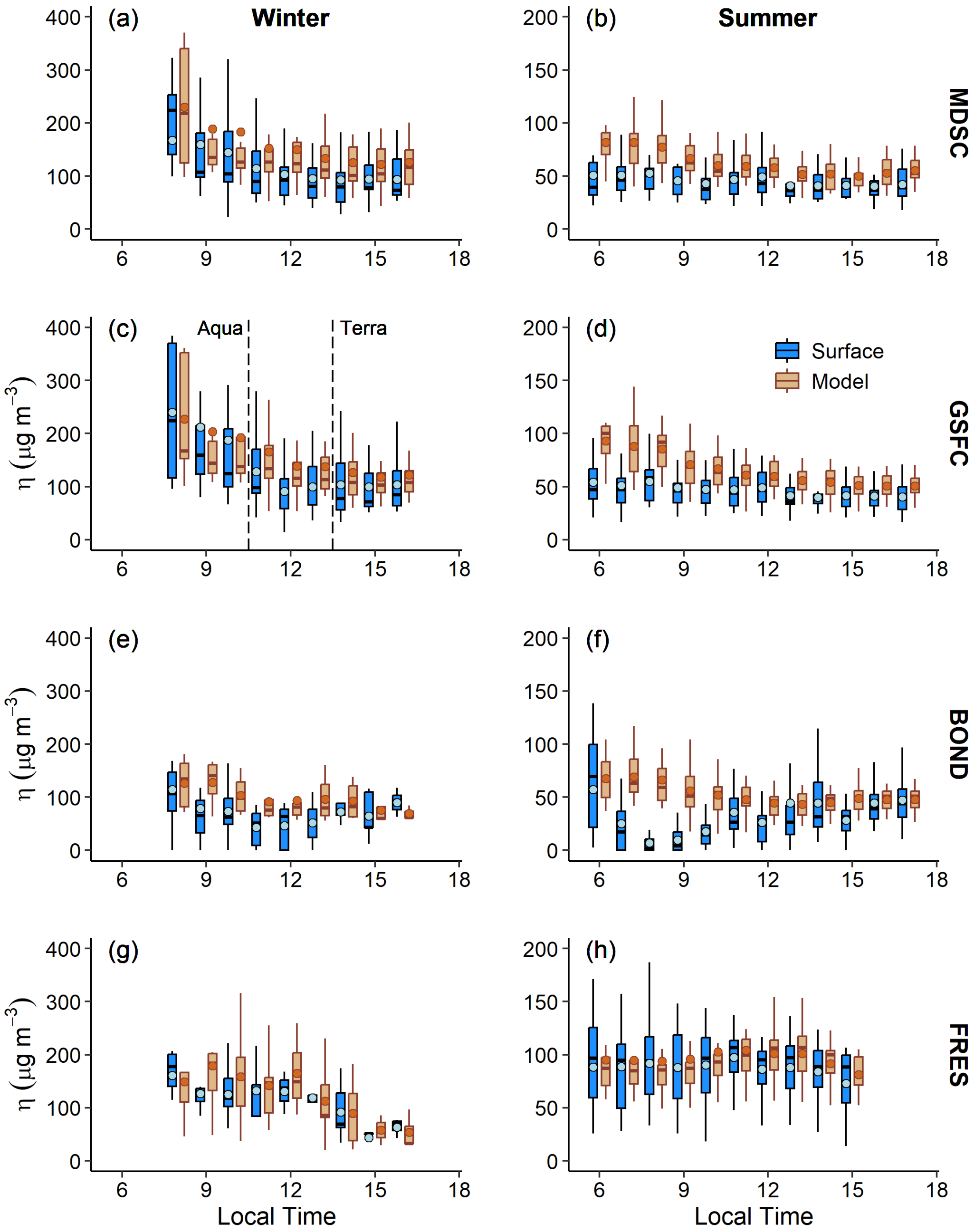


Figure 3.7: Diurnal distributions of eta (η) for January 2016 ("Winter" column) and August 2015 ("Summer" column) of surface-based hourly $PM_{2.5}$ mass concentrations and AERONET AOD at 440 nm (blue boxes, points) and CMAQ model predictions using AOD at 550 nm (brown boxes, points) for Fresno (a,b), Bondville (c,d), GSFC (e,f), and MDSC (g,h). Points indicate the mean. Boxes represent the 25th to 75th percentile with the center horizontal line as the median and whiskers as the 10th and 90th percentile. Note the winter and summer y-axis scales differ. Vertical dashed lines in (c) indicate nominal overpass times for MODIS onboard the Aqua (10:30 a.m.) and Terra (1:30 p.m.) satellites.

3.5 Conclusions

Changing chemical composition of semi-volatile species not well-characterized in reported $PM_{2.5}$ mass measurements is a plausible contributing explanation for spatial, diurnal, seasonal and decadal patterns in the $PM_{2.5}$-to-AOD relationship at multiple locations across the CONUS from 2006 to 2015. ALW and nitrate are semi-volatile $PM_{2.5}$ constituents for which we can estimate losses. Accounting for their negative mass artifacts in $PM_{2.5}$ surface mass improves agreement in peak η values across arid and humid surface-based sampling locations. Patterns in the associations among AOD and hygroscopic $PM_{2.5}$ constituents that differ in space and time are consistent with particle hygroscopicity and water uptake exhibiting a determining impact on AOD. There are other semi-volatile species known to influence AOD, such as organic $PM_{2.5}$ constituents, but their sampling biases are difficult to quantitatively describe. EPA's chief air quality modeling tool, CMAQ, replicates average summertime and wintertime surface-based geospatial patterns in observed η in the winter and summer across the CONUS. The co-occurrence of peak AOD and $PM_{2.5}$ events decline at humid sampling locations, but not arid. Chemical composition during peak events, most notably for sulfate and ALW, declined substantially from 2006 to 2015 in those locations, but not at western arid sites. Specific accounting of semi-volatile species can be useful to reconcile $PM_{2.5}$ measurements across sampling platforms.

Chapter 4

Virtual Choose-Your-Own-Adventure Prelabs Improve Student Understanding of Analytical Chemistry Concepts and Instrumentation

Reprinted with permission from the *American Chemical Society Journal of Chemical Education*. Flesch, M.M.; Humphrey, N.J.; Mang, S.A., Virtual Choose-Your-Own-Adventure Prelabs Improve Student Understanding of Analytical Chemistry Concepts and Instrumentation. Submitted for publication to *ACS Journal of Chemical Education* (*under review*).

4.1 Abstract

The COVID-19 pandemic forced instructors of chemistry lab classes to develop a variety of alternative materials for remote delivery of traditionally hands-on content. One solution to this challenge was virtual choose-your-own-adventure (CYOA) laboratories that allowed students to practice realistic decision-making and collect authentic data. With the return to in-person instruction, these CYOA labs can be modified into prelab exercises to support and supplement in-person experiments. Four virtual CYOA labs were adapted into prelab exercises for an upper-division instrumental analysis course to prepare students for experiments in chromatography method development and electronic circuit design. Students were surveyed to gauge their opinions on the prelabs and their comfort in the laboratory overall. Survey results indicated that students enjoyed the CYOA prelab activities and found them helpful in feeling prepared for experiments and understanding the source and quality of their experimental data. Students who completed the virtual prelabs met more learning objectives in several important categories on lab reports for two electronic circuit design experiments. Virtual CYOA prelabs may be a good way for instructors to expand the depth of their lab curriculum, improve student learning in the lab, and reduce the anxiety their students feel before complex experiments.

4.2 Introduction

Pre-laboratory exercises are important for preparing students to perform experiments and help avoid cognitive overload caused by focusing on unfamiliar content and new practical skills.[111] Many labs assign reading (such as textbook chapters or lab manuals) ahead of time, but students may find these assignments more difficult to engage with than active models that prompt questions.[110] When done effectively, prelab exercises should work to reduce the amount of reading material rather than add to it, reducing rather than contributing to cognitive overload.[112] Prelab videos offer a more active option for student preparations and improve student attitudes and feelings of preparation in the course.[113] Videos with embedded quizzes show an even higher level of attitude and score performance than traditional narrative videos, a change that is attributed to the more active learning environment.[110,111] Students seem to prefer more active methods of pre-laboratory preparation, but many of the strategies reported in the literature have little to no impact on students' overall academic performance.

During the remote instruction part of the COVID-19 pandemic, we developed several virtual laboratory activities for an upper-division combined lecture-laboratory instrumental analysis course using the free, open-source software Twine. Twine is a non-linear storytelling platform that allows users to create interactive games, stories, and learning activities.[191] Other instructors have created virtual learning activities using Twine for laboratory replacements,[114,192,193] including colleagues in the chemistry department at our institution.[117] These virtual offerings often take the form of a "Choose-Your-Own-Adventure" (CYOA) style activity, allowing students to make choices and determine if the outcome of those choices was correct or if they should try a different procedure instead.[116] For the period of remote learning, seven CYOA labs were generated with Twine, and the resulting HTML files were embedded in pages on our university's Learning Management System. The CYOA format provided a way to keep the emphasis on decision making and analysis of preliminary data in a remote format. Each Twine lab guided the students through a series of instrument optimization steps, giving students realistic data that incorporated their decisions. For example, if a student selected a nonoptimal mobile phase composition for their HPLC experiment, they obtained unsatisfactory peak separation or peak shape. The virtual lab workflow contained frequent prompts for students to evaluate their data and reconsider their previous decisions.

While these Twine labs served as a replacement while physical labs could not be conducted, there have been recent calls to use virtual labs as a supplementary tool for in-person courses even when students can be in the lab.[115,120] Hybrid Laboratory Action-Based Pedagogy (HLAB) focuses on a link between in-lab and non-lab activities in order to complement student learning and maximize student experiences.[119] HLAB suggests that the cognitive overload and anxiety students experience about complex experiments might be lessened by the virtual prelabs because students can review nontechnical skills (conceptual questions about the operation of instruments, ideal parameters, chemistry concepts, etc.) before coming to lab, leaving them free to focus on technical skills (physically conducting and troubleshooting experiments, processing data, and interpreting results) in the laboratory.

Beginning with the return to in-person instruction, we modified the full virtual labs into shorter prelab exercises to familiarize students with equipment, help them perform initial method development steps that they confirmed in the laboratory, and prepare them to collect and analyze data in the laboratory. Our course is structured such that students attend two lectures and a discussion section with the instructor and a seven-hour laboratory period supervised by a teaching assistant (TA) every week. Students taking the course are chemistry majors in their third or fourth year of study who have learned basic analytical chemistry techniques and laboratory report writing skills in prerequisite chemistry courses. Student performance on lab report learning outcomes was compared to offerings of this course where the CYOA prelabs were not used in order to investigate the effect of the new prelabs on student learning. Learning outcomes were chosen to reflect student learning of chemistry content and the principles of instrument operation. We assessed the impact of these prelabs using student survey data that asked them about their attitudes toward the experiments and their opinions of the CYOA prelabs.

4.3 Creating and Assessing CYOA Twine Prelab Activities

Four virtual CYOA prelabs were converted into prelab exercises for the return to in-person instruction. These experiments were selected because they contained content and equipment that was new to almost all students and therefore posed a high risk of cognitive overload. Two of them centered on developing chromatography methods, and the other two focused on different types of operational amplifier circuits.

4.3.1 Chromatography Method Development Experiments

The first two experiments for which virtual CYOA prelabs were developed dealt with chromatography method development using HPLC and GCMS. The original in-person versions of these experiments lasted two laboratory periods and required students to develop a method for their separation, then use that method to perform a quantitative analysis. For example, in the HPLC experiment, students worked in groups to identify an ideal mobile phase flow rate and composition for the separation of dihydrocapsaicin and capsaicin, followed by the quantification of these two molecules in several hot sauces. For an in-person chromatography lab, students worked in groups to identify the ideal mobile phase parameters during the first lab meeting, then used those parameters to perform the quantitative analysis during the second meeting. For remote instruction, both parts of the lab were performed virtually in the same lab period because it was much faster to simulate a chromatogram than it was to physically collect one. JavaScript code embedded into Twine was used to simulate chromatograms and generate .csv files for students to download and analyze. The simulations used student choices to generate data and were designed to replicate the results when the experiments were performed on our lab's HPLC system (Thermo Ultimate 3000, Waters

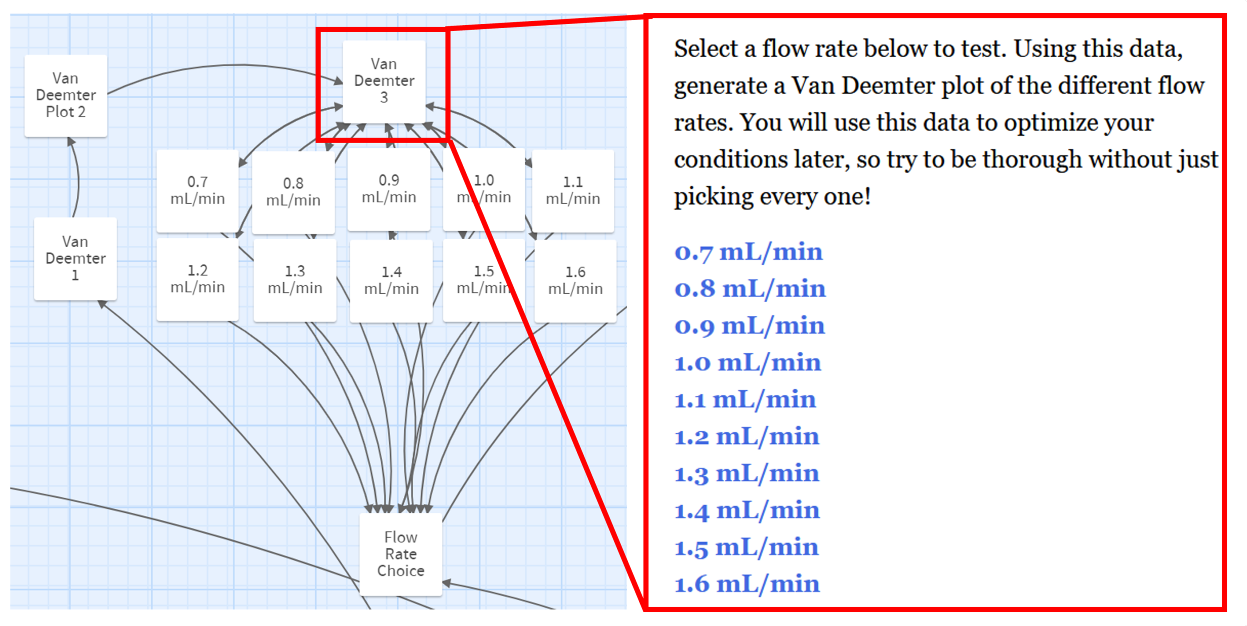
Spherisorb ODS2 column, PDA detector) or GCMS system (Thermo TSQ Duo, Agilent VF-5ms column), and were unique to each student.

The method development portions of the full virtual lab were converted into the CYOA prelabs. For both experiments, students went step-by-step through the process of determining the ideal flow rates and either mobile phase composition (HPLC) or column temperature (GCMS). Figure 4.1 shows the Twine creator view and a partial student view for the part of the HPLC prelab exercise where students find the optimal flow rate for the separation using an optimized mobile phase composition that they found earlier. Students were required to submit van Deemter plots and answers to questions about the method development procedure as part of their online prelab quiz, due before the in-lab portion of the experiment. During the in-lab experiment, their first task was to confirm the method parameters they had identified in the virtual prelab, after which they used their method to perform a quantitative analysis.

4.3.2 Electronic Circuit Design Experiments

The other two virtual CYOA labs that were converted into prelab exercises were related to instrument design using operational amplifiers (op amps). Op amps are an important part of the instrumental analysis curriculum because of their ubiquity in chemical instrumentation,[194] but they are not covered in any of our students' prerequisite courses. This leaves students susceptible to cognitive overload because they are learning an unfamiliar concept (the theory of operation of op amp circuits), while at the same time navigating unfamiliar physical tasks (the process of assembling an op amp circuit on a solderless breadboard).

In the two circuit labs, students built non-inverting and difference amplifiers to amplify the signal from a thermocouple, and a transimpedance amplifier to convert a photocurrent from a photodiode into a voltage. Students build and test these circuits and then use them to



Select a flow rate below to test. Using this data, generate a Van Deemter plot of the different flow rates. You will use this data to optimize your conditions later, so try to be thorough without just picking every one!

0.7 mL/min
0.8 mL/min
0.9 mL/min
1.0 mL/min
1.1 mL/min
1.2 mL/min
1.3 mL/min
1.4 mL/min
1.5 mL/min
1.6 mL/min

Figure 4.1: The HPLC Twine prelab activity from the creator/instructor view (left) depicting potential paths students can take in the process of optimizing a flow rate. The expansion outlined in red (right) depicts the interface a student would see as they choose which flow rates to use to simulate chromatograms. The cards called “Van Deemter 1” and “Van Deemter 2” are text-only screens that introduce students to the process of optimizing the flow rate. Students choose their preferred flow rate and enter it on the card called “Flow Rate Choice.”

construct an adiabatic calorimeter and visible light colorimeter, respectively. The full CYOA virtual labs were designed to give students visual references to the process of building the circuits. YouTube videos were embedded into the Twine virtual labs so that when a student chose to test a given circuit, they first watched the instructor building that circuit. Based on their choices of resistors for a given circuit, they received simulated data from a JavaScript code running in the Twine page. The simulated data was designed to be slightly noisy so that students could do calculations related to data acquisition, and to accurately reproduce the phenomenon of the op amp’s output saturating if the circuit was designed with too large of a gain for the input voltage, a common problem for students performing these experiments. Once students virtually tested their circuits, they were guided to choose a combination of circuit elements to design the full instruments, then given simulated data from an analysis (an adiabatic calorimetry experiment to find the enthalpy of solution of a salt and the construction of a Beer's law plot to quantify Red 40 dye in a sports drink, respectively).

For the CYOA prelab versions of these experiments, we again used the first half of each virtual experiment. In this case, that meant preserving the parts where students chose circuits to make and watching videos of these circuits being built, after which they received simulated data from testing those circuits. They were also asked to perform calculations to predict the output of a given circuit before getting the simulated data. Their responses to these questions were tracked using Twine variables and exported to a Google Sheet as described above.

4.3.3 Monitoring Student Progress on the Virtual Prelabs

While interacting with both the full virtual labs and CYOA prelabs, students made choices about which instrument settings to use and performed calculations meant to check their understanding of various concepts. Variables in the Twine code were used to track the student responses, count the number of tries to get a correct answer or how many sets of sample data the student generated, and recorded important choices the students made while completing the exercise. When a student completed the prelab, these variables were exported to a Google Sheet to give instructors data on how students were interacting with the virtual prelab.

The original reason for collecting this data was to see whether students were performing calculations correctly and whether they were collecting enough data to draw meaningful conclusions and prepare for the in-person experiment. For example, in the thermocouple electronics experiment, students were asked to calculate a calorimeter constant based on simulated voltage data from measurements using the circuit they chose earlier in the prelab. In Winter 2023, every student calculated their answer to within 5% of the true value as required to move on in the prelab, but the average number of tries it took them to enter that answer was 3.6. This suggests that students in future classes may need more practice on how to do these calculations in the lecture part of the course before they perform the lab. Adding variables to the Twine code is trivial, meaning that the prelabs can be a convenient way to collect data that helps an instructor monitor student understanding of a topic.

4.3.4 Assessment of Student Outcomes and Attitudes

To understand the effect of the CYOA prelabs on student lab report scores, we compared student mastery of learning outcomes on grading rubrics between two offerings of this course (Fall 2022 and Winter 2023) for three different experiments. No Twine prelabs were used in Fall 2021. The Fall 2022 course did not require Twine prelabs for the electronic circuit labs, while the Winter 2023 course did. We compared the circuit labs to an unrelated fluorescence experiment, without a Twine prelab component, performed in both quarters as a control. The GCMS and HPLC labs required Twine prelabs in both the Fall 2022 and Winter 2023 quarters, so we did not perform a specific analysis on laboratory report outcomes for these two experiments. There were 59 lab reports analyzed from Fall 2022 and 54 from Winter 2023. Rubrics for the lab reports and lab manuals for each experiment are available in the Supporting Information.

We qualitatively assessed student knowledge, comfort, and other metrics relating to analytical chemistry techniques, concepts, and laboratory preparedness using the Motivated Strategies for Learning Questionnaire (MSLQ)[106] survey tool along with Likert scale and open-ended questions about the CYOA exercises for the Fall 2021, Fall 2022, and Winter 2023 quarters (Table C.3). The Fall 2021 quarter was included as a "no Twine prelabs" control; during that quarter, students were required to read the lab procedures and complete a short pre-lab quiz before coming to the lab. The Fall 2022 and Winter 2023 quarters required completion of the Twine prelabs in addition to reading the lab procedures and completing the pre-lab quiz before the lab period. For all three quarters, students were surveyed in week one of the quarter as a “pre-quarter” assessment, and again in week ten as a “post-quarter” assessment. They were incentivized to take these surveys via the awarding of intangible tokens to spend on various assignment resubmissions or grade improvements as laid out in the course syllabus. Collection of student survey data was approved by UCI’s Institutional Review Board (Exempt Protocol #2231). In total, 115 students took the pre-quarter surveys, and 123 students took the post-quarter surveys. Note that students were not required to complete either survey, and response rate varied during and between quarters due to the grading structure and token economy of the course. The CYOA prelabs applied only to the Fall 2022 and Winter 2023 quarters, with a total of 81 students answering survey questions regarding them. We performed two-sided Mann-Whitney U tests and independent t-tests (95% confidence interval) using R statistical software[138] to compare lab report scores and survey responses between quarters.

4.4 Lab Report Outcomes and Survey Responses

4.4.1 Impact of Virtual Prelab Exercises on Assessments

Students generally improved scores on lab report rubric categories related to content knowledge of op amp circuits for both electronics experiments in the Winter 2023 class with Twine prelabs compared to the Fall 2022 class without Twine prelabs (Table 4.1). Lab reports were graded using specifications grading rubrics, on which each rubric criterion corresponds to one student learning outcome for the assignment and each criterion is graded as either “met” or “not met”. Each report was graded by three graders, whose determinations of the numbers of students meeting each learning outcome differed by an average of 8%. Rubrics for the experiments discussed here are available in the Supporting Information. The learning outcomes included in Table 4.1 were chosen because they relate to chemistry content and the principles of instrument operation, topics that we hoped would be covered by the CYOA virtual prelabs instead of being only introduced during the lab sessions. The number of rubric criteria (that is, learning outcomes) met by students for the calorimetry experiment lab report increases the most for the thermochemistry categories, indicating students had a better understanding of why they were building the circuit and the application of its theoretical principles to calorimetry.

Students struggled with correctly identifying and explaining concepts relating to circuit behavior in the colorimetry experiment in both quarters, though scores improved in the light measurement and determining absorbance (interpreting the colorimetry data to create a Beer's law plot and determining the concentration of dye in a colored sports drink sample) category in the Winter 2023 class. For both circuit experiments, students performed similarly on correctly explaining their operational amplifier circuit setups, but the Winter 2023 section struggled more with explaining the application of the Nyquist theorem and signal modulation to real-world circuit measurements. On a Fluorescence laboratory report that did not have a Twine prelab component in either quarter, students in both quarters performed similarly on rubric items relating to important fluorescence concepts. These results suggest students benefited the most when applying the circuit principles first displayed in the prelabs to the "unknown" part of the calorimetry and colorimetry experiments, where they had to combine previous knowledge of thermochemistry and UV-Visible spectroscopy to their circuit builds. In the Winter 2023 electronic circuit reports, student responses to questions specifically asking about calorimetry and colorimetry were written with a higher level of analysis and connections to relevant lecture material than in the Fall 2022 quarter. Furthermore, TA and instructor discussions during laboratory periods with students showed they made more connections between the circuit labs and the later experiments as related to the fundamental operation of the instruments they were using, which ties into an overall course learning objective. Twine prelab activities for complex or otherwise time-consuming experiments

Table 4.1: Concept-based learning outcomes met by students for three experiments for the Fall 2022 and Winter 2023 quarters. Percentages are averages of results from three graders. Statistically significant ($p < 0.05$) differences are denoted with asterisks.

Experiment	Rubric Learning Outcome Category	Fall '22 % of Students Meeting Outcome (N=59)	Winter '23 % of Students Meeting Outcome (N=54)	T-test p-values, Fall '22 vs. Winter '23
Thermocouples and Adiabatic Calorimetry	Operational Amplifier Operation and Calculations	87%	85%	0.350
	Seebeck Coefficient Determination and Thermocouple Calibration	62%	71%	0.134
	Calibrating and Using the Calorimeter	53%	72%	0.001*
Transimpedance Amplifiers and Colorimetry	Circuit Building and Calculations	80%	81%	0.752
	Nyquist Theorem and Signal Modulation	68%	55%	0.005*
	Light Measurement and Determining Absorbance	58%	74%	0.005*
	Circuit Behavior	47%	55%	0.180
Fluorescence	Excitation Wavelength Choice	91%	91%	0.647
	Analysis of Spectra	86%	82%	0.507
	Quantum Yield Determination	68%	70%	0.601
	Quinine Quantification	93%	94%	0.478

increased the number of students achieving satisfactory learning outcome scores, though their use may not be suitable for all types of chemistry laboratory experiments.

4.4.2 Student Perceptions of CYOA Prelab Exercises

Student opinions on the usefulness of the Twine prelabs differed significantly between the Fall 2022 and Winter 2023 quarters. The Fall 2022 quarter only used Twine for the chromatography labs, while the Winter 2023 course also used it for the two circuit labs at the beginning of the quarter. An overwhelming majority of students in both quarters agreed that the Twine prelabs helped them understand the laboratory procedures, sources and appearance of experimental data, and the concepts involved in the experiments, with a statistically greater portion of students agreeing with the latter two items in Winter 2023 (Figure 2). While both quarters similarly enjoyed the Twine prelabs, the Winter 2023 students particularly said that they would prefer to do the Twine prelabs instead of or in addition to traditional prelab exercises more than their Fall 2022 counterparts. Mann-Whitney U tests determined no statistical differences in student responses about the Twine prelabs better preparing them for the lab, helping them understand the laboratory procedures, or in their enjoyment of the prelabs (Table C.1). Five of the seven questions asked about the Twine prelabs had no "disagreement" responses in the Winter 2023 quarter while the Fall 2022 quarter did, suggesting that the students taking the course in the later offering generally found the Twine prelabs more useful. This may be due to student unfamiliarity or confusion with circuits and related concepts, giving them an appreciation for the embedded videos in the prelabs. We often observed students watching these videos during the lab period while building their circuits. The circuit labs occur in weeks two and three of the quarter and are the first in-person experiments students perform, while the two chromatography experiments happen between weeks five and nine on a rotating basis. Student understanding of the purpose behind the Twine prelabs after finding them helpful with circuit building in the first two weeks of the course may have improved their attitudes towards the "additional" prelab requirements for those experiments in the Winter 2023 quarter compared to the Fall 2022 quarter, where they were only introduced starting in week five.

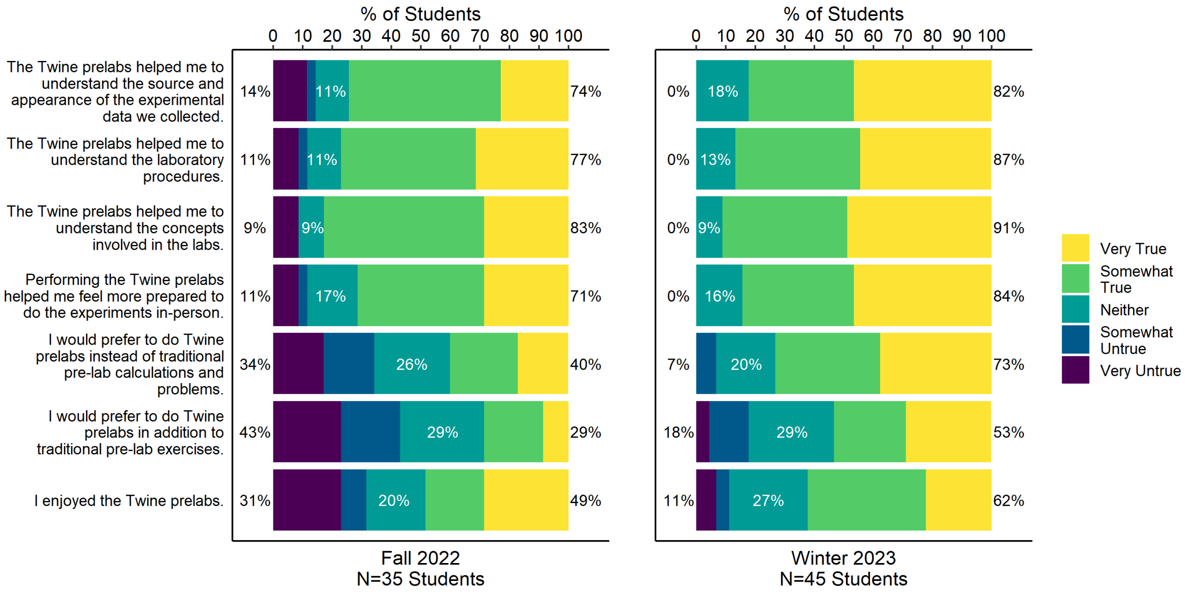


Figure 4.2: Student responses to (left) pre-quarter and (right) post-quarter MSLQ survey questions pertaining to feelings about preparation before laboratory experiments for the Fall 2021 (top), and combined Fall 2022 and Winter 2023 quarters (bottom). From left to right, percentages represent the combined negative responses, neutral responses, and combined positive responses. Note that N values change due to varying course enrollments and student participation in the surveys.

In an open-ended question asking students to explain their thoughts on whether or not the virtual Twine prelabs helped them understand the experiment better, an overwhelming majority indicated that the CYOA prelabs did better prepare them for the experiment. Many respondents said that these prelabs "helped to set the right instrument parameters" for the chromatography experiments. Another student specifically said that the prelab helped their "understanding [of] the lab…decreas[ing] the actual lab time" they spent doing the experiments, a sentiment echoed in multiple responses to this question. Students also mentioned the visual component of the simulated chromatography data and circuit construction videos multiple times as being helpful for understanding instrument operation and what to expect the data would look like when they collected it during the in-person experiments. We observed many students returning to these videos during the in-person lab as they worked to assemble and troubleshoot their circuits before asking the TA or instructor for assistance.

One student wrote that the Twine prelabs “provided the necessary information and setup needed to do the lab in an interesting way”, suggesting that this type of interactive prelab activity may improve students’ understanding and confidence about difficult experiments. For example, when asked in the lab to explain why they chose a specific flow rate and eluent mixture for the HPLC experiment, students who completed the Twine prelab correctly were able to relate the concepts of peak separation and peak broadening to these parameters. Negative responses indicated that the CYOA prelabs were too time consuming, or that they did not like how they could sometimes continue through the prelab without knowing whether or not they chose the optimal parameters. Overall, student opinions were mostly positive for the Twine prelabs because they provide a different way for students to learn about relevant analytical chemistry concepts and techniques than traditional teaching methods offer.

Student responses to MSLQ survey questions asking about comfort in the laboratory before and after the quarter changed significantly in their distribution with the addition of Twine prelab activities in the Fall 2022 and Winter 2023 quarters compared to the Fall 2021 quarter (Figure 3). Student anxieties around having enough time to complete the lab, the time available for the experiment, and working with chemicals showed statistical differences in the pre- and post-quarter response populations for the Twine-inclusive quarters only. In the pre-quarter surveys, 69%, 64%, and 32% of students agreed with these statements, which declined to 47%, 39%, and 20%, respectively, in the post-quarter surveys (Table C.2). The “control” Fall 2021 pre- and post-quarter response populations are very similar for all questions, with similar divisions in agreement type among respondents. This suggests that the addition of the Twine prelabs may lessen student anxieties around preparing for laboratory activities for this course, particularly as it relates to concern about time management while in the lab with unfamiliar concepts like circuit building and instrument operation. Comparison of the Fall 2021 to Fall 2022 and Winter 2023 pre-quarter surveys indicate comparable levels of anxiety about the lab before the course began. The post-quarter surveys had similar response distributions between the quarters as well, indicating that some proportion of students will

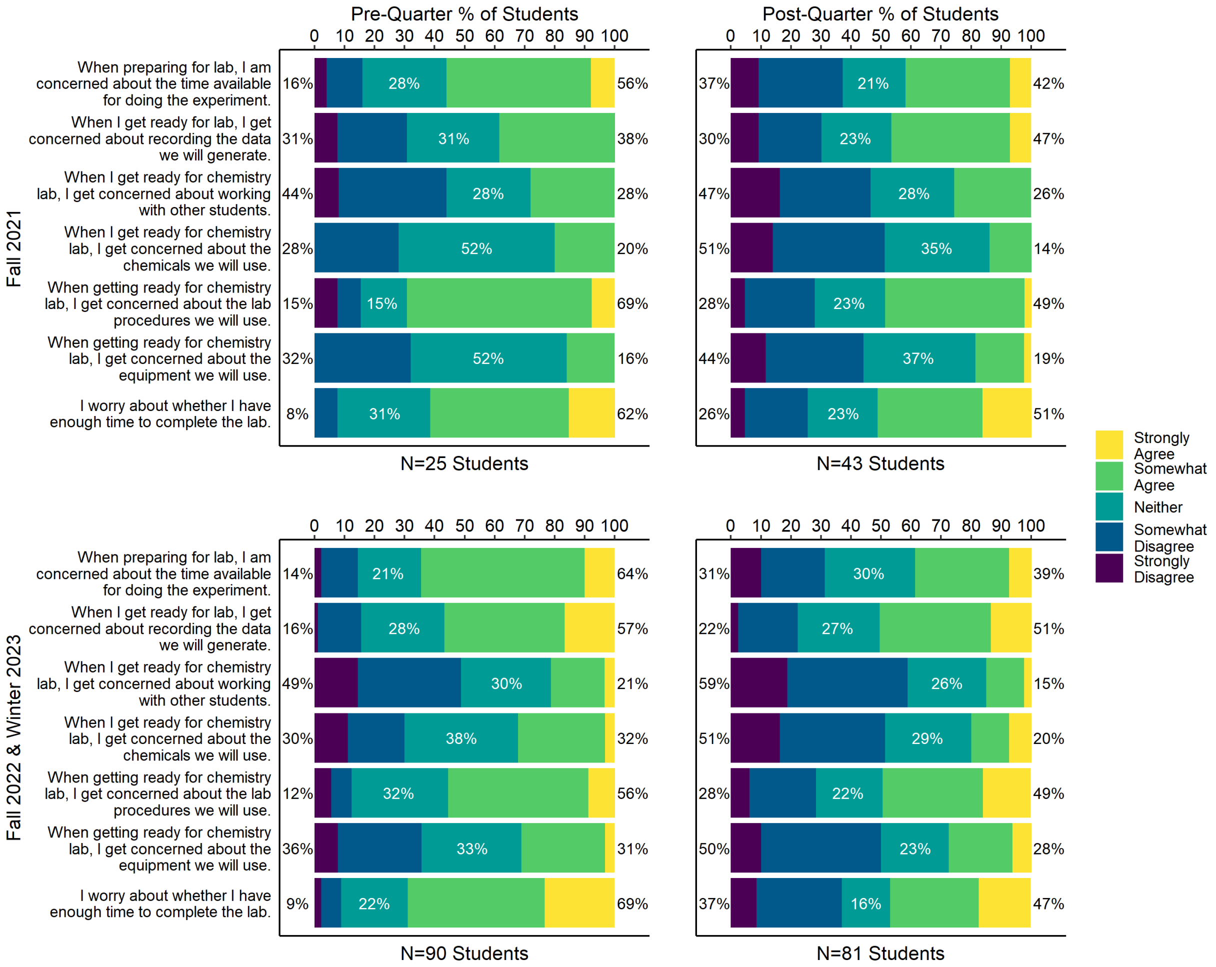


Figure 4.3: Student responses to (left) pre-quarter and (right) post-quarter survey questions pertaining to feelings about preparation before laboratory experiments for the Fall 2021, Fall 2022, and Winter 2023 quarters combined. From left to right, percentages represent the combined negative responses, neutral responses, and combined positive responses.

always be anxious around the lab, regardless of interventions meant to reduce such anxieties or worries about performing experiments with unfamiliar concepts and instrumentation.

4.5 Conclusions

Virtual choose-your-own-adventure labs had several benefits for students in an upper-division instrumental analysis course. There was an increase in the number of students meeting learning objectives in several important categories on lab reports for two electronic circuit design experiments when Twine prelab exercises were implemented. The largest benefit observed was an improvement in student attitudes towards the laboratory, including a reduction in anxiety about several aspects of the lab course. A majority of students surveyed reported feeling more prepared after the CYOA prelabs, in particular for the Winter 2023 quarter with the addition of the electronic circuit Twine prelabs. When asked about whether or not the Twine prelabs were useful, many students reported that it better prepared them for what to expect from the experiment and decreased the amount of time they spent in the lab, which agreed with instructors' informal observations. The CYOA prelabs also reduced the anxiety associated with working with chemicals and equipment, preparing for the lab, and recording data that many students reported feeling before the quarter. We suggest this is likely due to the prelabs introducing students to the procedures and equipment used in the experiments before they enter the laboratory. Application of this type of prelab activity to similarly complex advanced chemistry laboratory courses can potentially better prepare students for in-person experiments and reduce their stress surrounding complex techniques and instrumentation.

Chapter 5

Summary and Future Directions

5.1 Summary

5.1.1 Ambient Aerosol is Physically Larger on Cloudy Days in Bondville, Illinois

At the collocated AERosol RObotic NETwork (AERONET) and Interagency Monitoring of PROtected Visual Environments (IMPROVE) network monitoring stations in rural Bondville, Illinois, I used AERONET's quality assurance flags to bin their AOD and Ångström exponent measurements as predominantly cloudy or clear sky for a given day, then performed analysis on points that passed the highest calibration steps to compare measured aerosol physicochemical properties on predominantly cloudy and clear sky days from 2010 to 2019. I hypothesized that aerosols were larger on cloudy days than clear sky days due to greater ALW mass concentrations on cloudy days. Aerosol liquid water was estimated using ECMWF ERA5 model meteorological variables and IMPROVE-measured $PM_{2.5}$ chemical composition mass concentrations. I present findings that show that median AOD at 440, 500, 675, and

870 nm is higher on cloudy days in every season. Ångström exponents are smaller on cloudy days in every season except winter, when nitrate mass concentrations are highest. Mass concentrations of ALW-inclusive $PM_{2.5}$ are higher on cloudy days in all seasons but winter, consistent with larger particle sizes inferred from Ångström exponent measurements. Meteorological variables of temperature, RH, and PBL height are insufficient to fully explain the statistical significance for differences in AOD, Ångström exponents, and ALW mass concentrations on predominantly clear sky versus cloudy days. This highlights the need for simultaneous collocated, high-time-resolution measurements of both aerosol chemical and physical properties, in particular at cloudy times when quantitative understanding of tropospheric composition is most uncertain.

5.1.2 Chemically-specific sampling bias: the ratio of $PM_{2.5}$ to surface AOD on average and peak days across the U.S.

I investigated the $PM_{2.5}$-to-AOD relationship (η) at surface locations across the contiguous United States via surface observations and CMAQ model simulations of AOD and $PM_{2.5}$ chemical composition. I hypothesized this relationship would decline from decreasing $PM_{2.5}$ mass concentrations due to sulfate and ALW in the east but not in the west. I accounted for sampling biases of nitrate and ALW from filter measurements, finding this improved agreement in peak η values across arid and humid surface-based sampling locations. Changing chemical composition of semi-volatile species not well-characterized in reported $PM_{2.5}$ mass measurements is a plausible contributing explanation for spatial, diurnal, seasonal, and decadal patterns in the $PM_{2.5}$-to-AOD relationship at multiple locations across the CONUS from 2006 to 2015. Patterns in the associations among AOD and hygroscopic $PM_{2.5}$ constituents that differ in space and time are consistent with particle hygroscopicity and water uptake exhibiting a determining impact on AOD. I also find that the EPA's chief air quality modeling tool, CMAQ, replicates average summertime and wintertime surface-based geospatial patterns in observed η in the winter and summer across the CONUS. Specific accounting of semi-volatile species can be useful to reconcile $PM_{2.5}$ measurements across sampling platforms, particularly when satellite retrievals or surface monitors are sparse.

5.1.3 Virtual Twine Prelabs Improve Student Understanding of Analytical Chemistry Concepts and Instrumentation

The COVID-19 pandemic required development of alternative materials for remote delivery of hands-on chemistry laboratory experiments. One solution to this challenge was virtual choose-your-own-adventure (CYOA) laboratories that allowed students to practice realistic decision-making and collect authentic data. I created some of the course's CYOA labs using the free, open-source platform Twine, then modified them for application as prelab exercises for an upper-division instrumental analysis course to prepare students for experiments in chromatography method development and electronic circuit design. I assessed student lab report outcomes with and without these Twine prelabs along with surveying their attitudes towards laboratory comfort and the CYOA prelab exercises across multiple quarters of the course offering. I present findings that suggest student outcomes improved on both electronic circuit lab reports when the Twine prelabs were included compared to a report with no Twine prelab in either quarter. Survey results indicated that students enjoy the CYOA prelab activities and find them helpful in feeling prepared for experiments and understanding the source and quality of their experimental data. Virtual CYOA prelabs may be a good way for instructors to 1) expand the depth of their lab curriculum and 2) reduce the anxiety their students feel before complex experiments or working with unfamiliar instrumentation and techniques.

5.2 Future Directions: Designing an Environmental Chemistry Course

5.2.1 Background

Effects of the COVID-19 pandemic continue to reverberate in higher education institutions, but also offer insights into making courses more accessible to a wider range of students. This shift in instruction and thus student engagement and feedback with courses meant instructors had to pivot from their usual methods, particularly for laboratory-heavy classes such as Advanced Analytical Chemistry (Chem 152L) at UCI. Typically, Chem 152L is offered in both Fall and Winter quarters, but it was delayed for the 2020-2021 academic year in order to provide enough time to translate the laboratory-intensive course into a virtual offering for the Winter 2021 quarter. I was a Development TA for Chem 152L during the Fall 2020 quarter, where we transformed the eight in-person laboratory experiments to fully virtual "choose-your-own-adventure" (CYOA) style experiments. This experience in taking the curriculum and student learning objectives (SLOs) for an in-person, laboratory-focused course and determining how to achieve as many of them as possible with a fully remote course granted many important insights into maintaining student-centered learning even when isolated. Our efforts continue to be implemented in the course today as shortened virtual prelab exercises for four of the experiments, part of which culminated in the study discussed in chapter four of this dissertation. Based on my research experience, I would like to create and teach an upper-division collegiate environmental chemistry course.

Course and curriculum development is ever-changing and advancing as chemistry and related fields continue to branch out into new topics of study. Many versions of environmental chemistry courses exist already, whether as lecture only, lab only, or a combination of both. Often, these courses focus on the overarching concepts of the field: the atmosphere, lithosphere, hydrosphere, and then the pollution and remediation of these three spheres. My research and teaching experience provides me with a perspective suited to teaching an environmental chemistry course with a special focus on implementing coding and data analysis techniques for a range of environmental data not typically taught in undergraduate education courses such as regulatory network emissions and air quality data. Proficiency with computer programs and software is also becoming increasingly important for new graduates as they enter the workforce, and providing more opportunities for students to build those skills is imperative. The environmental chemistry course I hope to teach in the future combines the rigor of data analysis and laboratory experiments with in-depth coverage of important environmental concepts and how students can directly apply the knowledge and skills learned in this course.

5.2.2 Course Description and Student Learning Objectives

This course uses the fundamental properties of molecules and bonds as a framework for understanding chemicals and their sources, reactions, transport, and fate in the environment. It will focus on the three main areas of the environment: the atmosphere, hydrosphere, and lithosphere, with particular emphasis on atmospheric composition and pollution. Students in the chemistry or biochemistry major could take this course after finishing the organic chemistry sequence as part of their upper division elective and/or laboratory requirements for a chemistry or biochemistry bachelor's degree. Due to the laboratory component, this course is ideally structured for between fifteen to thirty students per section. The primary goal of this class is to describe the complex workings of the chemical world using concepts such as chemical characterization, thermodynamics, solubility, and kinetics. Opportunities for research projects and journal-style writing in chemistry courses are often scattered, leading students into graduate school or the workforce inadequately prepared for the expectations of these fields. Purposeful demonstration and explanation of these skills through my own research publications will help bridge the disconnect between typical laboratory report and peer-reviewed journal publication writing.

Lecture content will primarily rely on landmark peer-reviewed journal publications, with supplementation for other topics from the textbook ”Basic Concepts of Environmental Chemistry” by Des W. Connell,[195] as necessary. Some in-class assignments will require students to find their own readings on a given subject. In general, publications relating to each lecture topic will be used to explain past and current understanding in the field, with bi-weekly reading assignments diving deeper into either an assigned article or dedicated topic for that week. I will assess students within a framework of student learning outcomes (SLOs), which are meant to illustrate the knowledge and skills individual students should possess and can demonstrate after finishing a given learning experience. Each assessment within the course has specific learning outcomes, all of which serve to meet the course SLOs.

Upon successful completion of this course, students will be able to:

- Explain the interconnections between the atmosphere, hydrosphere, and lithosphere and the chemical principles behind each of them.
- Understand the chemical processes behind the sources, fates, transportation, and transformation of pollutants in the environment.
- Apply previous knowledge of analytical chemistry, organic and biological structures, kinetics, and thermodynamics to environmental chemistry problems.
- Prepare samples and conduct analyses using relevant chemical instrumentation and interpret those results using modern software (Excel/Google Sheets and R)
- Discover, interpret, analyze, and present results from experiments and peer-reviewed journal articles related to current environmental chemistry problems.
- Communicate key takeaways from all of the above to a variety of audiences.

These SLOs will be achieved through a combination of assignments, quizzes, exams, laboratory experiments, and reading of current environmental science literature. More specifically, these objectives will be achieved via solving problems and writing about environmental chemistry, and collecting and interpreting data from the laboratory experiments and small group projects. Affective learning goals of this course are to improve students' communication skills (both written and oral), critical thinking, and problem-solving skills. The weekly assignments will be designed to improve student understanding of the discussed topics and their ability to express that understanding in written form. A general goal of this course is also to give students a greater appreciation of the importance of environmental chemistry and related policy at the local, federal, and global scale.

5.2.3 Course Structure

A weekly course schedule is listed below in Table 5.1 if this course were to be offered at a semester-based institution with sixteen weeks of instruction. I propose three lecture hours of face-to-face instructional time with a single three- or four-hour laboratory period once a week to provide hands-on experience and examples of the types of environmental analysis performed by scientists and regulatory agencies across the country. Due to the nature of some of the proposed experiments requiring either extensive instrument sampling or analysis times or learning a new skill (such as a new technique or time-intensive coding), some experiments and the small group projects will take multiple laboratory periods to complete. A brief summary of what each laboratory experiment will cover conceptually and which SLOs are addressed along with the techniques and instrumentation used are listed below. Where appropriate, citations for the basis of each experiment are noted after the experiment title. Not all of the experiments will be ”in-lab” experiments (e.g., the Data Analysis, Air Quality Patterns experiments), but will still occur during the laboratory time.

Table 5.1: Proposed weekly schedule for a semester-length version of lecture material covered and laboratory experiments performed during this course. Student familiarity with fundamental chemistry concepts and general and organic chemistry lab skills is presumed.

Week of Semester	Lecture Topics	Laboratory Activity
1	Atmospheric structure & air pollution chemistry	Lab 1
2	Human health and atmospheric particles	Lab 2
3	Regulatory networks and remote sensing	Lab 3
4	Water-soluble chemistry & water treatment	Exam 1
5	CO_2 toxicity & ocean acidification	Lab 4
6	Toxic heavy metals & persistent organic pollutants	Lab 5
7	Heavy metal remediation of Superfund sites	Lab 6
8	Chemistry of pesticides, fungicides, & herbicides	Exam 2
9	Agriculture and animal emissions	Project 1
10	Nuclear fusion & uranium fuel waste	Project 1
11	Project 1 presentations	Lab 7
12	Oil drilling & hydraulic fracturing,	Exam 3
13	Coal mining & automobile emissions	Project 2
14	Chemical transport & emissions/waste regulations	Project 2
15	Environmental chemistry in the media	Project 2
16	Project 2 presentations	Final Exam

5.2.3.1 Experiment and Project Descriptions

Laboratory Experiments

Students will perform each experiment in groups of three to four, depending on the overall class size. To minimize the amount of time spent waiting for instrument availability and to allow for greater flexibility should an instrument break down, a rotation of the experiments in weeks three through seven and eleven may be implemented. All lecture material relating to instrumentation and topics covering experiments would be made available before the beginning of these lab rotations even if it has not yet been covered in the lecture to ensure students have a baseline understanding before coming to the lab. Additional guidance would be provided to groups performing experiments early on with concepts related to material being covered later in the semester to ensure fair grading practices and contextual understanding of the experiment.

Lab 1 - Data Analysis: This will be a fully virtual activity I create that will happen during week one as a complement to a safety/equipment check-in day in the lab. It will familiarize and/or refresh students with the type of statistical analysis techniques I expect them to perform on the various kinds of data they will collect during the course. The lab will have two parts. The first is an Excel-based portion to familiarize them with calibration curves and associated LINEST features and functions, along with basic statistical calculations such as confidence intervals, propagation of error, and standard deviation. The second is an R-based portion to familiarize them with graphically displaying results and performing more robust statistical analyses such as non-parametric population statistics and model comparisons to real data.

Learning Outcomes:

- Prepare data and perform statistical analysis.
- Create and interpret data tables and figures in Excel and R.
- Understand why performing statistical analyses and determining uncertainties and errors are important in environmental analytical chemistry.

Lab 2 - Air Quality Patterns: Students will analyze a year's worth of measurements from an EPA $PM_{2.5}$ monitor in R to contextualize large data set manipulation and draw conclusions about air quality trends at a given location. Detailed instructions for reading and inputting the data files into R will be provided, including example code from my own research projects. Students will perform analyses covering the whole year, seasons, and each month. Students will compare and contrast their figures to the regional figures from the EPA.

Learning Outcomes:

- Organize and investigate air quality measurements via statistical analysis in R.
- Construct and interpret appropriate figures and tables in R.
- Evaluate results and draw conclusions about trends in air quality.

Lab 3 - Acidity & Alkalinity of Drinking Water:[196] Using simple acid-base titration techniques, students will determine the acidity or alkalinity of real-world water samples from various locations (local creek water, well water, ocean water, etc.), with the additional step of ensuring all samples and solutions are (mostly) CO_2-free via nitrogen gas bubbling techniques. Class data will be collected and shared for students to analyze in R, with emphasis on location comparisons between samples using the mapping capabilities in R.

Learning Outcomes:

- Apply knowledge of acid-base titration techniques to acidic and alkaline water samples.
- Apply volumetric and quantitative transfer laboratory skills in sample preparation.
- Investigate potential spatial patterns using mapping tools in R.
- Connect lecture topics about water pollution and the importance of pH on aquatic environments to this experiment.

Lab 4 - Identification of FD & C Dyes by Paper Chromatography & Visible Spectroscopy:[196] Two parts comprise this experiment, where students will use two different techniques to identify the dyes present in real food, drug, and cosmetics (FD & C) samples. They will perform paper chromatography using filter paper as the chromatography plate and petri dishes as the developing chambers, similar to TLC plates in organic chemistry. Visible spectrophotometry will be used to qualitative identify dyes as well, with multiply-dyed items being purified before absorption spectra are taken.

Learning Outcomes:

- Apply knowledge of thin-layer paper chromatography and visible spectroscopy techniques to common food, drug, and cosmetic dye samples.
- Perform separations with multiple liquid chromatography techniques.

- Understand why certain dyes are harmful to human and environmental health via structural and potential reactivity information.

Lab 5 - Determination of Nitrate Ion in Water:[196] Students will first prepare a calibration curve of nitrate standards via Beer's Law from visible spectroscopy measurements. Absorbance measurements of real water samples they collect from various sources will be matched to the absorbance curve to determine the original nitrate concentrations in the samples. Students will perform statistical comparison of water types (drinking, stream, ocean, etc.) and sample locations in R with the class data.

Learning Outcomes:

- Prepare standard solutions and real water samples and operate visble spectrophotometers.
- Apply knowledge of calibration curves and Beer's Law to absorbance measurements to determine the concentration of nitrate in various water samples.
- Connect results to water contamination and treatment topics discussed during lecture.

Lab 6 - Retention of Metal Cations in Soil:[197] Spiked solutions of pH-controlled copper and zinc are used to "contaminate" different soil samples, then an ammonium solution is added for visible spectrophotometer analysis of the copper amine complex formed. Students will then determine an appropriate wavelength to measure absorbance of aqueous Cu^{2+} and create a Beer's law plot (calibration curve). Based on these values and the resulting color changes of solutions, they will determine the concentration of Cu^{2+} retained in the various soil samples.

Learning Outcomes:

- Prepare realistic soil samples and associated calibration standards.
- Operate and interpret output from visible spectrophotometers.
- Determine the retention of Cu^{2+} in various soil types and connect their results and hypothesized findings to topics discussed during lecture.

Lab 7 - Detection of Microplastics in Soil and Sediment:[198] Students will collect soil samples from nearby local water sources and prepare them for analysis using a density separation technique before the samples are disgested via a Fenton reaction. Microscopic techniques will then be applied to quantify microplastics and microfibers in their samples. Class data will be collected and shared for students to perform various spatial and qualitative location type analysis in R.

Learning Outcomes:

- Perform the Fenton reagent reaction to digest organics from soil samples.
- Apply knowledge of density and filtration techniques for soil analysis.
- Identify microplastics and microfibers via microscopy techniques.
- Propose and justify reasoning for spatial variability in contaminant concentrations across sampled locations.

Small Group Projects

Students will complete these projects with their regular laboratory groups. Both projects require some literature review to contextualize the research they are performing. After the completion of each project presentation session, I will have students peer review their group members to ensure work was divided evenly amongst all members as a rubric category for the project. These projects will also serve as opportunities for low stakes public speaking practice in sharing their science to a varied audience.

Project 1 - Superfund Contamination Sites: Each group will be given a unique Superfund contamination scenario with varying contaminants, geographies, geologies, etc., and propose a remediation plan for the hypothetical location with the knowledge and techniques covered in the course so far. Students will be expected to reference relevant literature on their contaminants and remediation techniques. Groups will present their findings in a short presentation (5-8 minutes) the following week in lecture. The specifications grading rubric will include categories for scientific quality of their research and suggested remediation plans along with presentation skills.

Learning Outcomes:

- Research relevant environmental contamination and remediation literature.

- Prepare and organize a short group presentation with helpful visuals and the most important contamination findings and suggested remediation plans.

- Work with other students in their group and build communication skills with each other and potential audiences via collaborative presentation.

- Connect their scientific reasoning and treatment plans to topics discussed during lecture.

Project 2 - Air Quality Assessment: Students will be paired and given a variety of fine particulate matter ($PM_{2.5}$) mass concentrations, meteorology conditions (temperature, relative humidity), and other data sets from a given location in the continental United States and asked to write a short technical report (4-5 pages) explaining and presenting the data as if to fellow scientists to practice their scientific communication skills. The data will provide broad guiding questions with a brief example using the publication in chapter two of this dissertation[170] to provide additional context. They will also share their results with the class during the last week of lab in an 8-10 minute presentation. Students will practice their public speaking skills once more, though this time with a focus on peer-to-peer guidelines, where more technical language and specific chemical reasoning is encouraged. The specifications grading rubric will include categories for scientific quality of their analysis and support for conclusions with peer-reviewed references along with presentation and writing skills.

Learning Outcomes:

- Apply earlier course knowledge to create and interpret data tables and figures in R.

- Research relevant background information and determine good references to include in support of their findings for their location.

- Write a well-structured and detailed report of their findings with an appropriate level of scientific and technical language, with connections to lecture topics and peer-reviewed atmospheric literature.

- Summarize and present their findings with their partner via a collaborative presentation.

5.2.4 Assessments and Grading Scheme

5.2.4.1 Assessments

I propose six different types of assignments and assessments for this combined lecture-laboratory course. The lecture portion will have thirteen homework assignments of varying content. The laboratory component will have nine experiments with associated pre- and post-lab quizzes and lab notebooks, with reports written after the experiment is completed and all necessary data is gathered. Two small group research projects will happen partially during the laboratory and partially outside the laboratory. Lastly, there will be four exams: three that occur during the semester and one final cumulative exam. The lab reports, lab projects, and lecture assignments will all be graded on a mastery-based scale, the quizzes and exams graded on a traditional percent-based scale, and the lab notebooks graded on a complete/incomplete basis. Further detail about the grading scheme can be found in the appropriate subsection below.

Lecture Assignments. The lecture assignments will assess students' understanding of the concepts covered during the lecture portion of the course in a variety of ways. There will be mathematical and chemical reaction problems, critical thinking problems asking them to evaluate a situation and explain their thoughts behind the solution they come up with, and summaries of a peer-reviewed journal article related to the current lecture topic. Each week will alternate the type of assignment; odd weeks will be problem sets and even weeks will be journal article summaries, with weeks one, eleven, and sixteen excluded. The problem sets will contain a variety of chemical reactions or equation-based problems in addition to some open-ended logical reasoning questions. The article summaries will serve the instructor to quickly gather student comprehension of the reading material based on their written explanations. The first peer-reviewed journal article will be on the publication in chapter three of this thesis, with specific call-outs to guide student summaries. These assignments address all but the fourth SLO, with the intent of reinforcing the information learned during lecture and improving their skill in interpreting environmental chemistry literature.

Exams. The exams will assess student comprehension and retention of concepts and terms learned throughout the semester. Exam 1 will cover everything from the beginning of the semester through water-soluble chemistry. Exam 2 will cover topics from wastewater treatment to heavy metal remediation of Superfund sites. Exam 3 will cover the chemistry of pesticides, fungicides, and herbicides through radioactivity. The final exam, while cumulative, will focus primarily on the remaining topics of oil drilling and hydraulic fracturing through environmental chemistry in the media. Exams address the first three SLOs in a more traditional assessment format.

Small Group Projects. In two group projects, students will perform a variety of tasks including literature review, data analysis, problem-solving, and presentation of findings. Two lab periods will be dedicated to work time for each project, though students will need to meet with their group outside of class to finalize presentation information and review literature. Students will be given minimal frameworks within which to work aside from specifications grading rubrics to encourage critical thinking and greater collaboration. These projects address SLOs two, three, five, and six, with emphasis on the second and sixth SLOs within the context of Superfund contamination sites and air quality monitoring data.

Lab Reports. For each of the seven experiments, students will write a laboratory report requiring some elements to be in the style of a journal article.[107,199,200] Students will be expected to answer a series of questions, some of which will be in the style of a results and discussion section or have them practice writing an abstract or experimental section for the experiment they performed.[201] This addresses the last three SLOs, with a focus on the fourth and fifth SLOs as the experiments will improve their laboratory and data analysis skills, in addition to the scientific writing practice the reports will provide.

Lab Quizzes. Each of the five in-lab experiments will require students to complete a pre-lab and post-lab quiz. The pre-lab quizzes will serve to ensure they have prepared for the experiment and have begun filling out their lab notebooks with the relevant safety and procedural information. The post-lab quizzes will check student comprehension of the experiment just performed, occasionally asking for figure or data uploads depending upon the experiment. Students will be able to take these with their lab groups as they perform them in groups, but each member must submit their own quiz. The lab quizzes address SLOs four and five.

Lab Notebooks. The five experiments performed in the lab will require students to keep a laboratory notebook for each experiment, with an easy to follow layout and organization. Students will be expected to fill in experimental objectives, important materials, safety information, and procedures before setting foot in the laboratory space. As they perform experiments, marking of changes to specific procedural information and data collection will be required, along with any calculations performed when analyzing data, where necessary. These will address SLOs three, four, and five, with particular emphasis on SLO four due to the importance of maintaining a laboratory notebook as a detailed record-keeping of experimental information in graduate and professional scientific study.

5.2.4.2 Specifications Grading

For this course, I would like to utilize the specifications grading system[202] over a traditional grading scale. Specifications grading allows students to know exactly what grade they need to get on each kind of assessment in order to achieve a specific grade in the course overall. This grading scheme typically goes hand in hand with a scale of ”mastery” for each outcome on a given rubric for a given assignment, which is accounted for in determining letter grade cutoffs.

This is a three point scale from zero to two, where a zero is "Needs Revision", a one is "Low Pass" and a two is "High Pass". In other words, if a student receives a "Needs Revision" grade, that means they did not display satisfactory mastery of the required concepts. A "Needs Revision" does not mean a student has failed the assignment; students are given the opportunity to revise many assessments if they earn this score. If they receive a "Low Pass", they showed sufficient mastery, and if they receive a "High Pass", they displayed complete (or near complete) mastery of the material for that assignment. Specifications grading thus permits for greater flexibility in determining student mastery of the course learning outcomes to set their overall grades rather than flat point thresholds. When letter grade requirements are set to simple point thresholds, students can (and often do) pass a course without demonstrating real understanding or mastery of the material, which can lead to them struggling more in later courses which will likely have more challenging material.

Some assessments such as the exams and pre- and post-lab quizzes will apply the standard percentile grading scale as those assessments may be done with their lab group or do not fit the structure for specifications grading as readily. Specifications grading will apply to assessments with associated rubrics, such as the lab reports, lecture assignments, and group projects. As this scheme is not commonly used at the collegiate level, I will apply ways in which students can resubmit assignments that they perform poorly on, particularly early in the semester as they are still learning how the grading system works. This resubmission protocol will be an intangible token economy[203] that students can gain and spend tokens in throughout the semester via the completion of multiple surveys before, during, and near the end of the course. The specifications grading scheme combined with the token economy is a model that I have seen others implement and used myself in both lower and upper division chemistry courses taught at UCI with great success.[117,204,205]

For each assessment category, the following requirements for each letter grade are described (Table 5.2). In order to achieve an overall B grade, for example, a student must meet the minimum requirements for a B in all assessment categories; otherwise, they will receive a C grade. If a category does not list all three types of specification grade options (High Pass, Low Pass, Needs Revision), that means the student can have any combination of the other options to achieve that grade. For example, if students are aiming for a B on the lab reports, they need at least 5 High Pass reports, with the remaining 2 reports designated as any option (Needs Revision, Low Pass, High Pass); the combination of scores on these 2 reports does not disqualify them from achieving the B requirements for that category.

Table 5.2: Proposed specifications grading scheme letter grade requirements.

Assessment Type (number)	**Minimum to earn D***	**Minimum to earn C**
Lecture Assignments (13)	7 Low Pass	7 High Pass AND 4 Low Pass OR 10 Low Pass
Lab Reports (7)	4 Low Pass	3 High Pass AND 1 Low Pass
Group Projects (2)	1 Low Pass	2 Low Pass
Exams (4)	Average score 50%	Average score 60%
Lab Quizzes (10)	Average score 55%	Average score 65%
	Minimum to earn B	**Minimum to earn A**
Lecture Assignments (13)	9 High Pass AND 2 Low Pass	11 High Pass AND 0 Needs Revision
Lab Reports (7)	4 High Pass AND 1 Low Pass	6 High Pass AND 0 Needs Revision
Group Projects (2)	1 High Pass AND 1 Low Pass	2 High Pass
Exams (4)	Average score 70%	Average score 80%
Lab Quizzes (10)	Average score 75%	Average score 85%
Add a Plus:**	”Complete” 4+ Lab Notebooks	
Add a Minus:	”Complete” $\leq$3 Lab Notebooks	

* If the requirements for a D grade is not met, a grade of F is earned.

**Exception: A+ grade requires High Passes on all lab reports, group projects, & lecture assignments, average quiz scores >90%, and 5 lab notebooks completed.

Incorporating a mixture of specifications grading and traditional assessment styles into this course allows the introduction of a new grading method to students while providing familiar anchor points. As I would like to focus this course most heavily on the lecture assignments, lab reports, and small group projects, this is where specifications grading can shine due to the opportunities for a more holistic approach to determining students' mastery of course content, SLOs, and laboratory skills. The traditional assessments (exams) will serve as knowledge checkpoints throughout the semester. The pre- and post-lab quizzes serve two purposes: 1) ensure students are prepared for the experiment before entering the laboratory space, and 2) examine students' understanding of the experiment they just performed.

My experience with this grading scheme reports that students often prefer it over the traditional scale, as they feel it is very clear what students must achieve or complete in order to receive a particular letter grade in the course. This includes the potential pathway of students not necessarily completing or attempting all assessments if it does not negatively impact their grade, which can be a relief for students who would otherwise feel overwhelmed with the course workload by the end of the semester. This type of combined lecture and laboratory course means the required time spent outside of class working on material is significant. Therefore, by setting grade minimums rather than grade maximums, I hope to assuage some student fears and/or coursework burnout by accounting for some breaks in due dates and setting reasonable grade threshold bounds that do not compromise the quality of work required to achieve them. Using the specifications grading scheme will also minimize the need for final grade adjustments. This practice is frequently employed for traditional grading scales, particularly if a section of students performs extremely poorly on an exam compared to the instructor's expectations and thus skews course grade distributions lower than the instructor desires. At any point, students may also check their current likely grade in the course by calculating the number of complete and passing assessments and exam scores they currently have and projecting forward to the end of the class as in Table 5.2.

5.2.4.3 Traditional Grading Scale

Most institutions continue to use the traditional grading scale that assesses students via completion of a certain percentage of course material to assign them an A to F letter grade. This typically means an A is 90% and up, a B is 80-89%, a C is 70-79%, a D is 60-69%, and below a 60% is an F. While this is still the primary grading scheme used, many instructors acknowledge the scale is not necessarily indicative of true mastery of material, and sometimes adjust letter grade cutoffs based on class statistical distributions. If this type of grading scheme is required, a general guideline for the various assignment and exam grade breakdown is listed in Table 5.2. Ideally, I want students to work hard on all aspects of the course, but emphasize the lab reports, lecture assignments, and group projects. My goal with the selected experiments is to give them context for real-world applications of the skills they learn from their undergraduate chemistry labs. Additionally, the lecture assignments will be a mixture of problem-solving, critical thinking, and interpreting scientific literature. By weighing more of the course grade on these types of assessments over exams, I aim for students to care less about the number of points they earn in the course and more about the concepts and skills they are learning. Rather than set each assessment category (and subsequently each assignment within) to a specific amount of points, I hope to minimize the amount of ”point grubbing” students tend to do when they are near a letter grade cutoff. Students can easily

Table 5.3: Proposed traditional grading scheme breakdown for various assessment types.

Assessment Type (amount)	Percent of Grade
Lecture Assignments (13)	15%
Exam 1	10%
Exam 2	10%
Exam 3	10%
Lab Reports (7)	25%
Lab Quizzes (10) and Notebooks (5)	5%
Group Projects (2)	15%
Final Exam	10%
Total	**100%**

determine their current standing in the course by taking their current scores in each category and multiplying it by its weighted value then summing them all together. Letter grade cutoffs would originate at the standard percents as previously described, though based on class size and statistical distribution at the end of the course adjustments to lower end boundaries may occur.

5.2.5 Conclusions

The overarching purpose of this course is to provide real-world context for the chemistry students learn about in the classroom, and the ways in which we can measure, mitigate, and respond in the environment. Students will build connections between many types of chemistry by integrating applications of techniques and instrumentation to pollution problems that cross multiple areas of study. The laboratory portion of the course provides another opportunity for students to improve upon or develop new laboratory skills including higher-order data analysis techniques. Experiments with direct connections to real-world testing for pollutants and assessment of environmental health provides greater context and exposure to scientific processes in the regulatory and research fields. Additionally, as public speaking continues to be a large source of anxiety for many students as they progress through their collegiate careers, adding two opportunities for them to practice sharing their science is incredibly important for several reasons. It requires them to fundamentally understand what they are meant to be sharing, how they need to frame it for their audience, and imposes practice with public speaking and scientific communication. As an upper division course, student learning is focused primarily on improving critical thinking and complex analysis skills, particularly in student ability to interpret and understand results and conclusions from their own experiments and broader scientific literature. With a variety of activities and assessments based on student-centered learning, I hope to achieve impactful outcomes beyond the classroom.

With the combination of research performed and courses taught while at UCI, I want to prepare burgeoning chemists for a range of careers where knowledge of technical information and broader context can be applied. Science is not done in a vacuum, and neither should coursework with direct connections to the environment. The inclusion of some components of scientific research with literature reviews and written reports will better prepare students for graduate-level study and careers in the field. With my atmospheric chemistry research, a large part of the research process was spent determining the best way to analyze large data sets to try to answer my hypotheses about aerosol physicochemical properties. In this course, the two group projects that require students to come up with their own hypotheses, determine procedures, analyze data, and draw conclusions perform a similar function on a smaller scale. Students who take this course will improve their knowledge of important environmental chemistry concepts and leave with a fundamental understanding of why these techniques, instruments, and methods are necessary to be good environmental scientists and informed citizens. Fostering a space where students are free to make mistakes and can fix them via showing their learning is also a tenet of performing research, where many experiments often fail until one does not, and that success is strengthened because of the failures and knowledge gained from them. I also hope it promotes the concept of performing research in a research group either in undergraduate study or at the graduate level, particularly if they wish to learn more about a specific subset of the field discussed during this course. My goal as an instructor is to not only send students who are capable and competent into chemistry careers, but also provide them with the tools to succeed in a variety of life paths.

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Appendix A

Supporting Information for Chapter 2

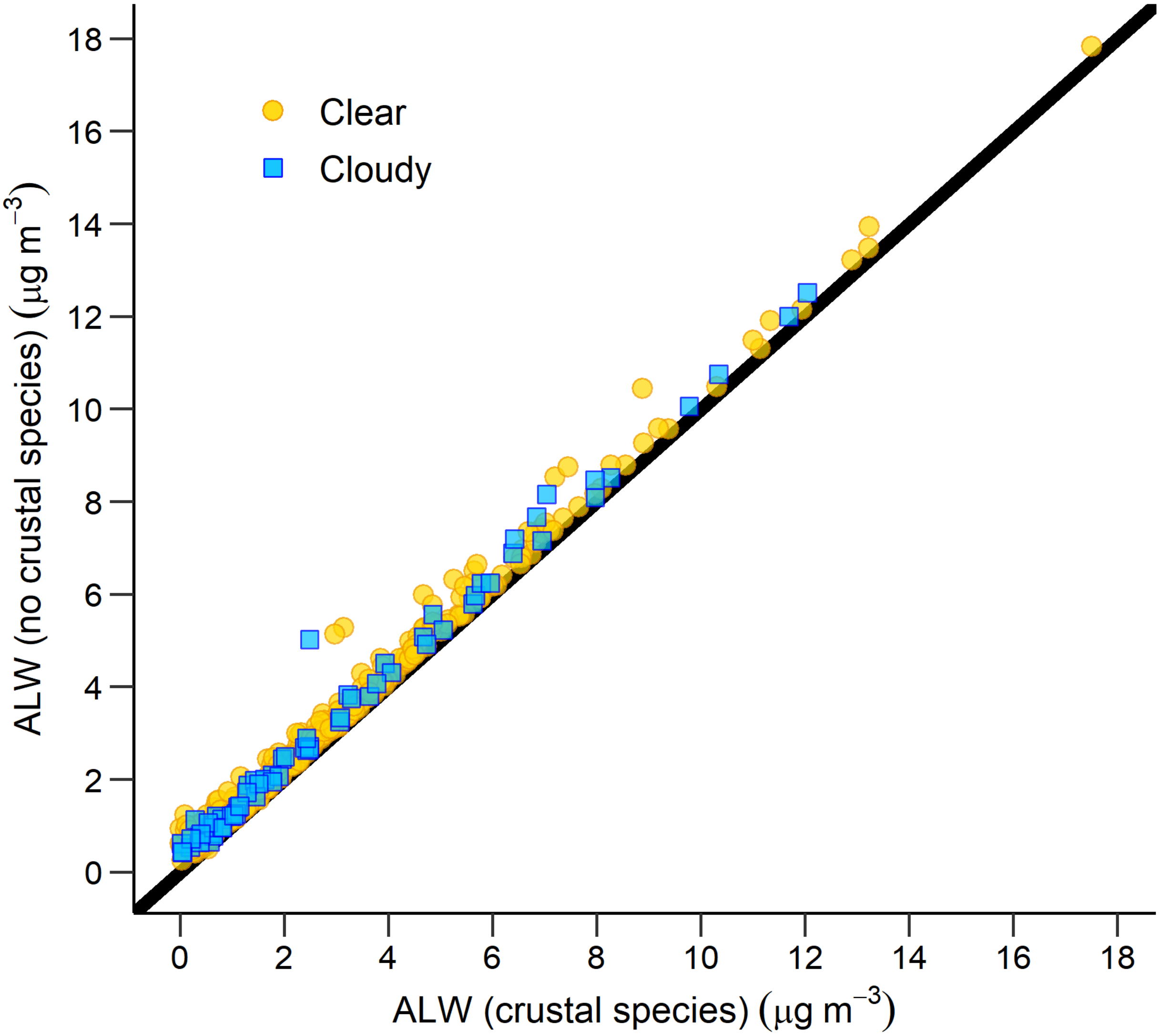

Figure A.1: Comparison of ISORROPIA model outputs of aerosol liquid water (ALW) mass concentrations from 2010-2019 at Bondville on predominantly clear sky (gold circles) and predominantly cloudy (blue squares) days. Crustal species include calcium, potassium, magnesium, and sea salt constituents sodium and chloride. Temperature, relative humidity, and mass concentrations of sulfate and nitrate were kept constant. The black line is a 1:1 line for reference. ALW estimates average 14% lower when crustal species are included.

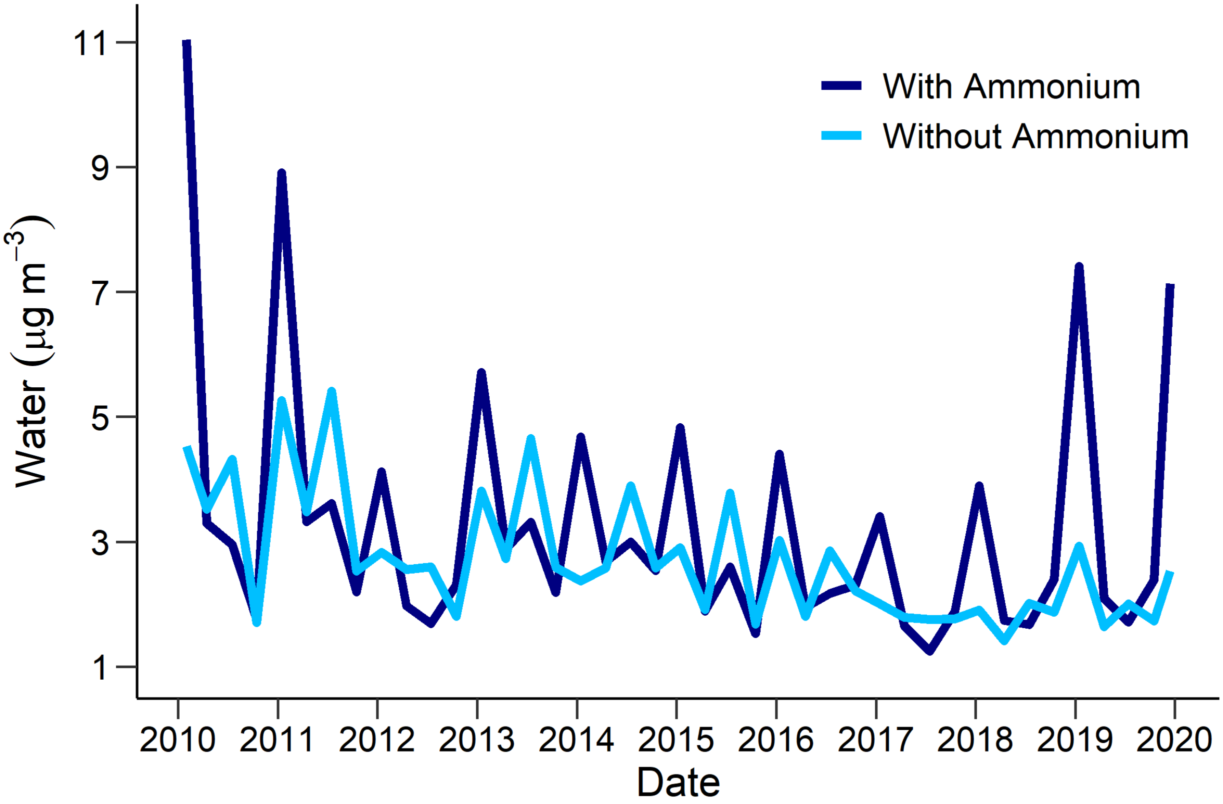

Figure A.2: Comparison of seasonal average aerosol liquid water (ALW) mass concentrations from 2010-2019 at Bondville. Temperature, relative humidity, and mass concentrations of sulfate, nitrate, and crustal species were kept constant. Ammonium values are taken from a collocated CASTNET site, and seasonal aggregates are used as inputs to ISORROPIA. The dark blue trace includes ammonium, while the bright blue trace excludes ammonium. ALW estimates are approximately 21% lower without ammonium, those this is not statistically significant for a T-test with 95% confidence.

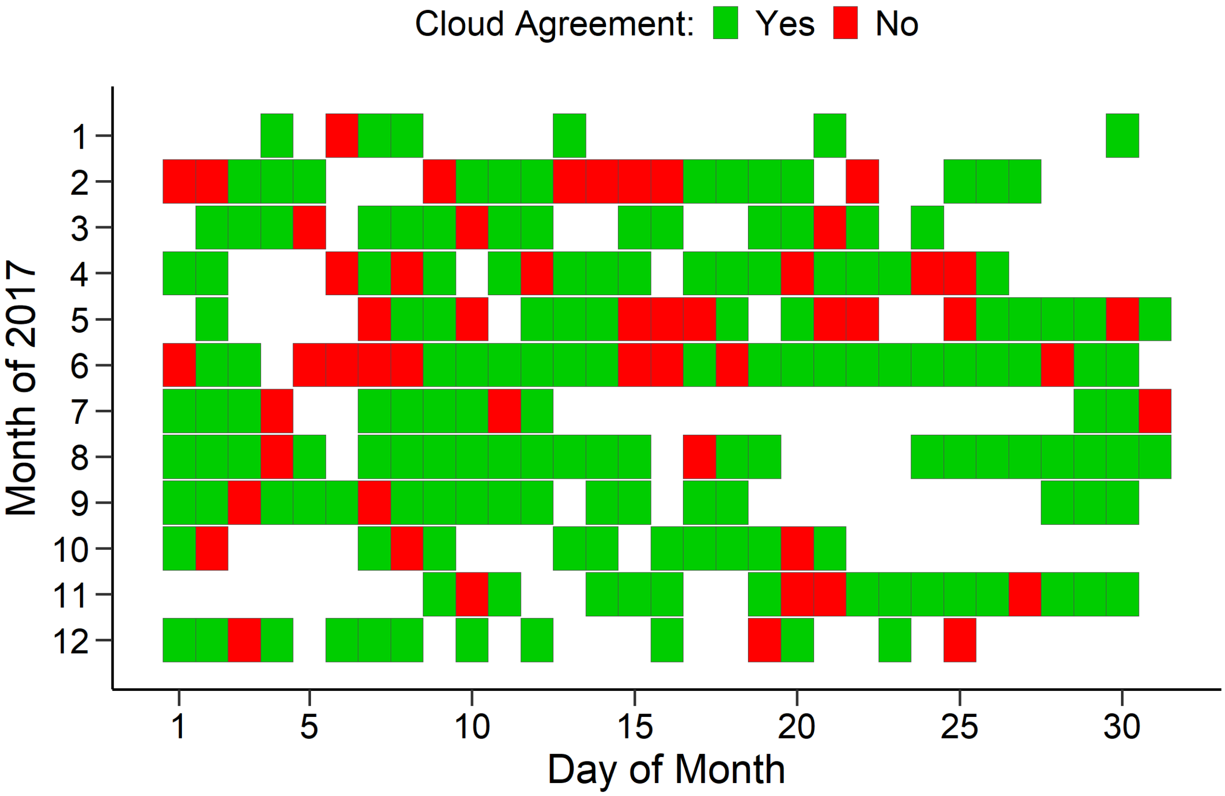

Figure A.3: Comparison of the AERONET-based cloudiness determination method to GOES-measured cloud top temperature at the Bondville location for daily aggregate AERONET observations for 2017. Green indicates both methods agree and red both methods disagree, with agreement occurring approximately 76% of the time. GOES-13 did not have the 12 μm channel available for thin cirrus cloud detection, which AERONET's algorithm screened better at the time, plausibly contributing to most of the disagreement occurring from AERONET measurements binned as cloudy and GOES retrievals binned as clear sky.

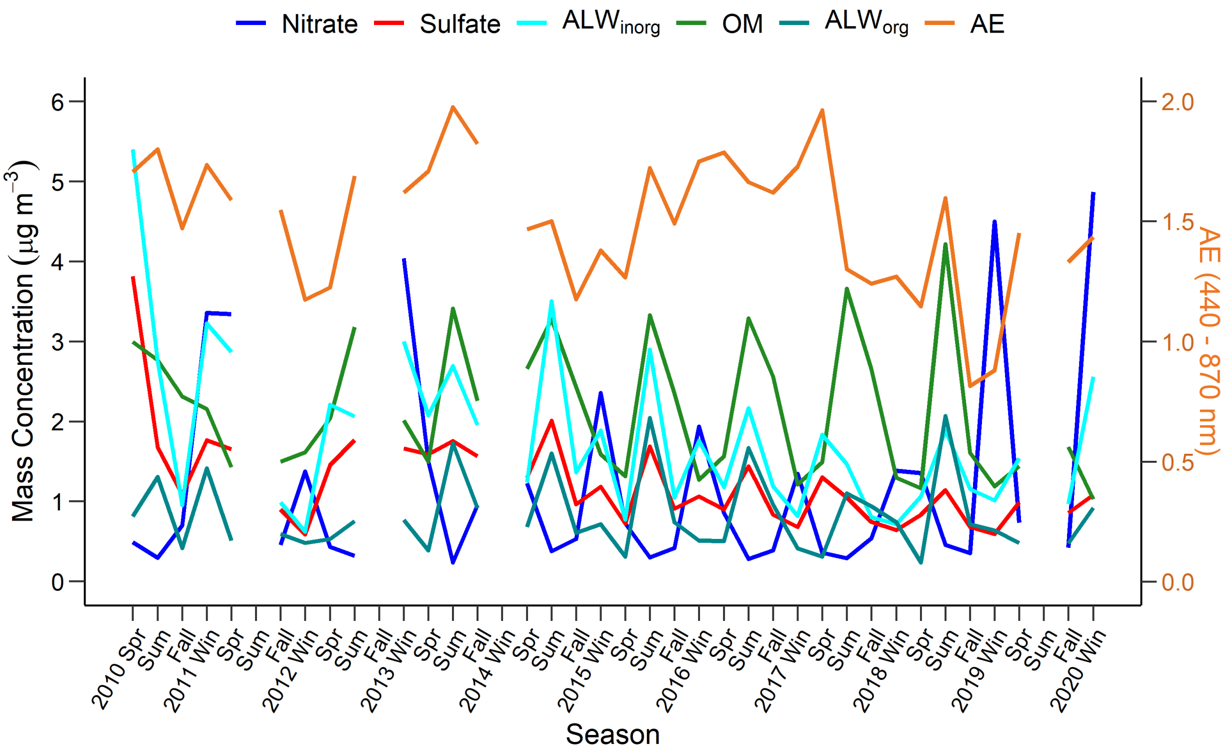

Figure A.4: Ångström exponents (AE, orange) and mass concentrations of nitrate (blue), sulfate (red), inorganic aerosol liquid water (ALW_{inorg}, cyan), organic matter (OM, green), and organic ALW (ALW_{org}, dark turquoise) over time at Bondville. Gaps indicate a lack of quality assured data from AERONET for that season matched to IMPROVE measurement days. Note that "2020 Win" contains data from December 2019 only.

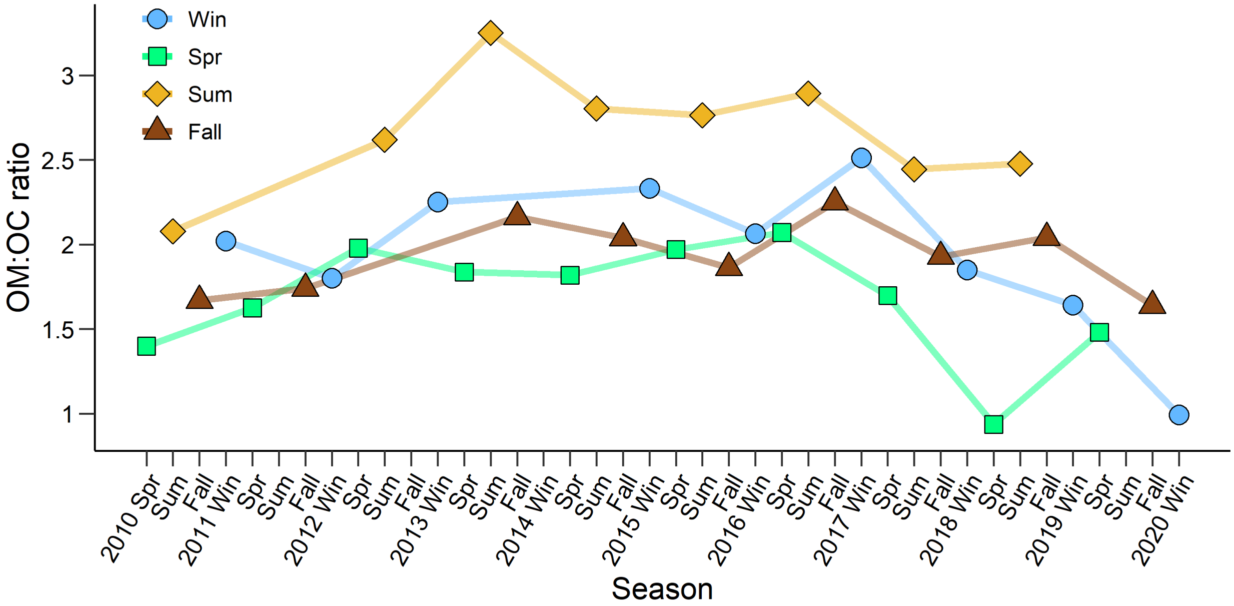

Figure A.5: Time series of the ratio of organic matter (OM) to organic carbon (OC) calculated annually for winter (blue circles), spring (green squares), summer(gold diamonds), and fall (brown triangles) at Bondville. Gaps in points for each season indicate missing data for that season based on a lack of quality assured data from AERONET for that season matched to IMPROVE measurement days. Note that "2020 Win" contains data from December 2019 only.

Table A.1: Predominantly clear sky and cloudy medians and two-sided Mann-Whitney U test p values of aerosol optical depth (AOD) and Ångström exponents. For clarity, p values significantly less than 0.05 are listed as "<0.05" and statistically significant seasons are bolded.

Parameter	Season	Clear Median	Cloudy Median	Mann-Whitney U test p value	Medians > 0.02 Experimental Uncertainty
Ångström exponent (440-870 nm)	Winter	1.46	1.49	0.944	Y
	Spring	**1.47**	**1.39**	**<0.05**	Y
	Summer	**1.68**	**1.59**	**<0.05**	Y
	Fall	**1.45**	**1.39**	**<0.05**	Y
Aerosol optical depth (440 nm)	**Winter**	**0.07**	**0.08**	**<0.05**	N
	Spring	**0.11**	**0.15**	**<0.05**	Y
	Summer	**0.17**	**0.22**	**<0.05**	Y
	Fall	**0.08**	**0.10**	**<0.05**	Y
Aerosol optical depth (500 nm)	**Winter**	**0.06**	**0.07**	**<0.05**	N
	Spring	**0.10**	**0.12**	**<0.05**	Y
	Summer	**0.14**	**0.18**	**<0.05**	Y
	Fall	**0.07**	**0.09**	**<0.05**	N
Aerosol optical depth (675 nm)	**Winter**	**0.04**	**0.05**	**<0.05**	N
	Spring	**0.06**	**0.08**	**<0.05**	Y
	Summer	**0.08**	**0.11**	**<0.05**	Y
	Fall	**0.04**	**0.06**	**<0.05**	N
Aerosol optical depth (870 nm)	**Winter**	**0.03**	**0.03**	**<0.05**	N
	Spring	**0.04**	**0.06**	**<0.05**	N
	Summer	**0.06**	**0.08**	**<0.05**	Y
	Fall	**0.03**	**0.04**	**<0.05**	N

Table A.2: Total days binned as "clear sky" or "cloudy" using the AERONET quality assurance algorithm method for each season before matching with IMPROVE dates at Bondville from 2010-2019 and related days with GOES cloud top temperature observations for 2017.

	AERONET (2010-2019)			GOES (2017)		
Season	Clear	Cloudy	Total	Clear	Cloudy	Total
Winter	183	31	214	33	9	56
Spring	232	87	319	44	16	60
Summer	305	80	385	53	14	67
Fall	245	62	307	45	5	50

Table A.3: Predominantly clear sky and cloudy medians and two-sided Mann-Whitney U test p values for meteorological variables of temperature, relative humidity, and planetary boundary layer height. For clarity, p values significantly less than 0.05 are listed as "<0.05" and statistically significant seasons are bolded.

Parameter	Season	Clear Median	Cloudy Median	Mann-Whitney U test p value
Temperature (°C)	Winter	-1.3	4.0	0.505
	Spring	**11.5**	**20.1**	**<0.05**
	Summer	24.1	24.8	0.324
	Fall	14.6	12.1	0.132
Relative Humidity (%)	Winter	64.7	64.2	0.915
	Spring	53.2	57.0	0.106
	Summer	67.1	68.2	0.561
	Fall	58.6	58.6	0.439
Planetary Boundary Layer Height (km)	Winter	0.49	0.51	0.961
	Spring	**0.66**	**0.57**	**<0.05**
	Summer	**0.60**	**0.54**	**<0.05**
	Fall	0.55	0.41	0.055

Table A.4: Predominantly clear sky and cloudy medians and two-sided Mann-Whitney U test p values for IMPROVE-measured $PM_{2.5}$ chemical species and aerosol liquid water estimates. For clarity, p values significantly less than 0.05 are listed as ”<0.05” and statistically significant seasons are bolded.

Parameter (μg m^{-3})	Season	Clear Median	Cloudy Median	Mann-Whitney U test p value
Inorganic Aerosol Liquid Water	Winter	1.60	1.14	0.329
	Spring	**1.10**	**1.97**	**<0.05**
	Summer	2.19	2.81	0.093
	Fall	1.05	1.27	0.183
Organic Aerosol Liquid Water	Winter	0.58	1.29	0.898
	Spring	**0.34**	**0.62**	**<0.05**
	Summer	1.42	1.86	0.116
	Fall	0.62	0.73	0.235
Nitrate	Winter	2.52	1.60	0.650
	Spring	0.56	0.85	0.621
	Summer	0.30	0.33	0.443
	Fall	0.45	0.84	0.052
Sulfate	Winter	1.10	0.92	0.301
	Spring	**0.97**	**1.32**	**<0.05**
	Summer	1.27	1.72	0.113
	Fall	0.45	0.90	0.599
Organic Mass	Winter	1.55	1.77	0.961
	Spring	**1.31**	**1.99**	**<0.05**
	Summer	3.13	3.55	0.110
	Fall	2.20	2.52	0.536
Potassium	Winter	0.032	0.038	0.595
	Spring	**0.032**	**0.041**	**<0.05**
	Summer	0.031	0.031	0.387
	Fall	0.036	0.037	0.496
Chloride	Winter	0.058	0.038	0.258
	Spring	0.020	0.026	0.093
	Summer	0.016	0.016	0.425
	Fall	0.020	0.016	0.546
Calcium	Winter	0.024	0.033	0.602
	Spring	0.035	0.031	0.907
	Summer	0.052	0.041	0.743
	Fall	0.066	0.058	0.193
Magnesium	Winter	0.007	0.006	0.549
	Spring	0.012	0.017	0.073

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Table A.4 – continued from previous page

Parameter (μg m^{-3})	Season	Clear Median	Cloudy Median	Mann-Whitney U test p value
	Summer	0.014	0.007	0.112
	Fall	0.013	0.007	0.256
Sodium	Winter	0.020	0.018	0.816
	Spring	0.012	0.014	0.149
	Summer	0.010	0.013	0.249
	Fall	0.010	0.006	0.300
$PM_{2.5}$ + Aerosol Liquid Water	Winter	10.20	10.20	0.746
	Spring	**6.22**	**10.47**	**<0.05**
	Summer	10.30	13.40	0.083
	Fall	7.18	8.81	0.415

Appendix B

Supporting Information for Chapter 3

Text B1

From Equation B.1, we estimate f(RH) using mass concentrations of reported dry PM2.5, PM2.5 chemical constituents, estimates of ALW, and aerosol density:

$$f(RH) = \frac{(\frac{[ALW]}{\rho_w})^{1/3} + (\frac{m_{d,dry}}{\rho_d})^{1/3}}{(\frac{m_{d,spec}}{\rho_d})^{1/3}} \tag{B.1}$$

Here, [ALW] is the mass concentration of ALW (μg m^{-3}), ρ_w is the density of water (1 g mL^{-1}), m$_{d,dry}$ is the monitor-reported PM$_{2.5}$ mass (μg m^{-3}), m$_{d,spec}$ is the summed mass of the reported chemical constituents (sulfate, nitrate, OM, sodium, chloride, calcium, potassium, and magnesium; μg m^{-3}), and ρ_d is the density of dry aerosol as a mass-weighted average dependent on measured aerosol composition (g cm^{-3}). Note that f(RH) is typically measured at arbitrary "humid" and "dry" values, and in this work, we estimate particle diameters accounting for particle composition and ambient RH. Reported PM$_{2.5}$ mass concentrations average 5.3 μg m^{-3} greater than the speciated sum PM$_{2.5}$ totals. This is primarily due to missing chemical species measurements at some sites and limiting the calculation of the summed speciation mass to only the species used to determine dry aerosol density for consistency.

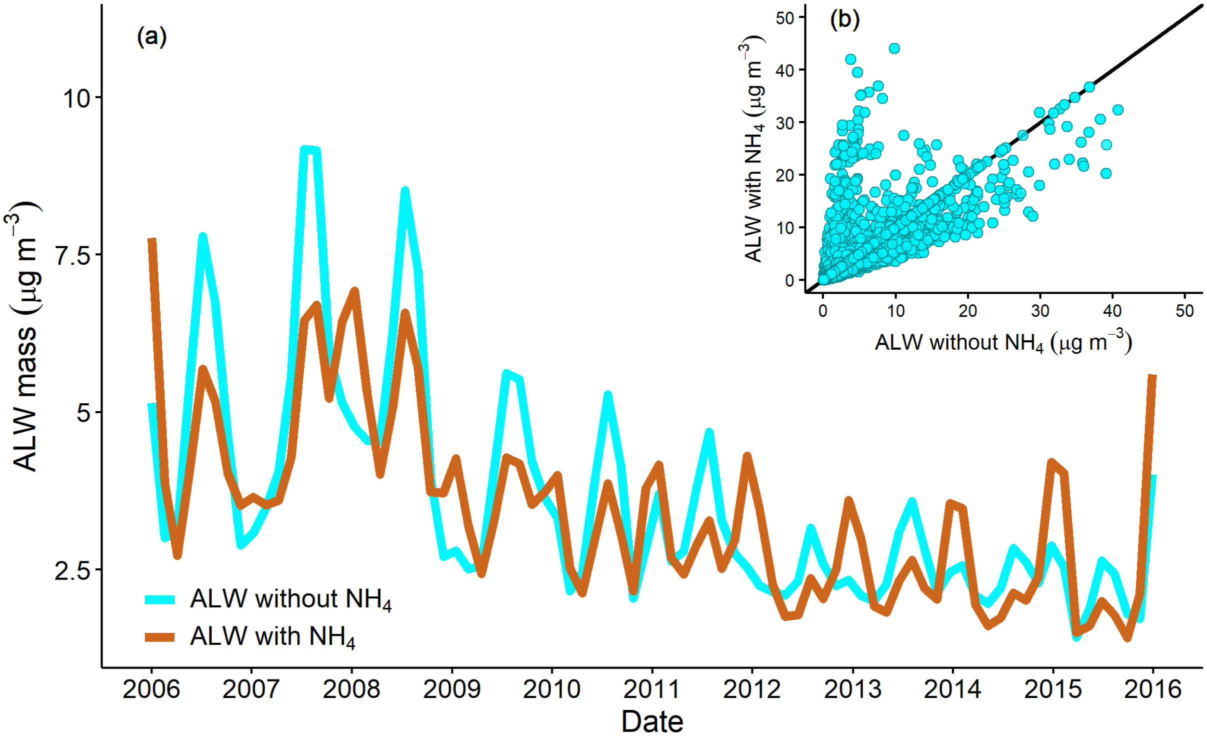

Figure B.1: Sensitivity study of aerosol liquid water (ALW) with and without ammonium (NH_4) included in estimations via ISORROPIAv2.1 at 8 AERONET stations studied from 2006 to 2015 as a function of time (main figure) and against each other (inset). In the time series, the light blue fit excludes ammonium, and the brown fit includes ammonium. The black line in the inset represents the 1:1 line.

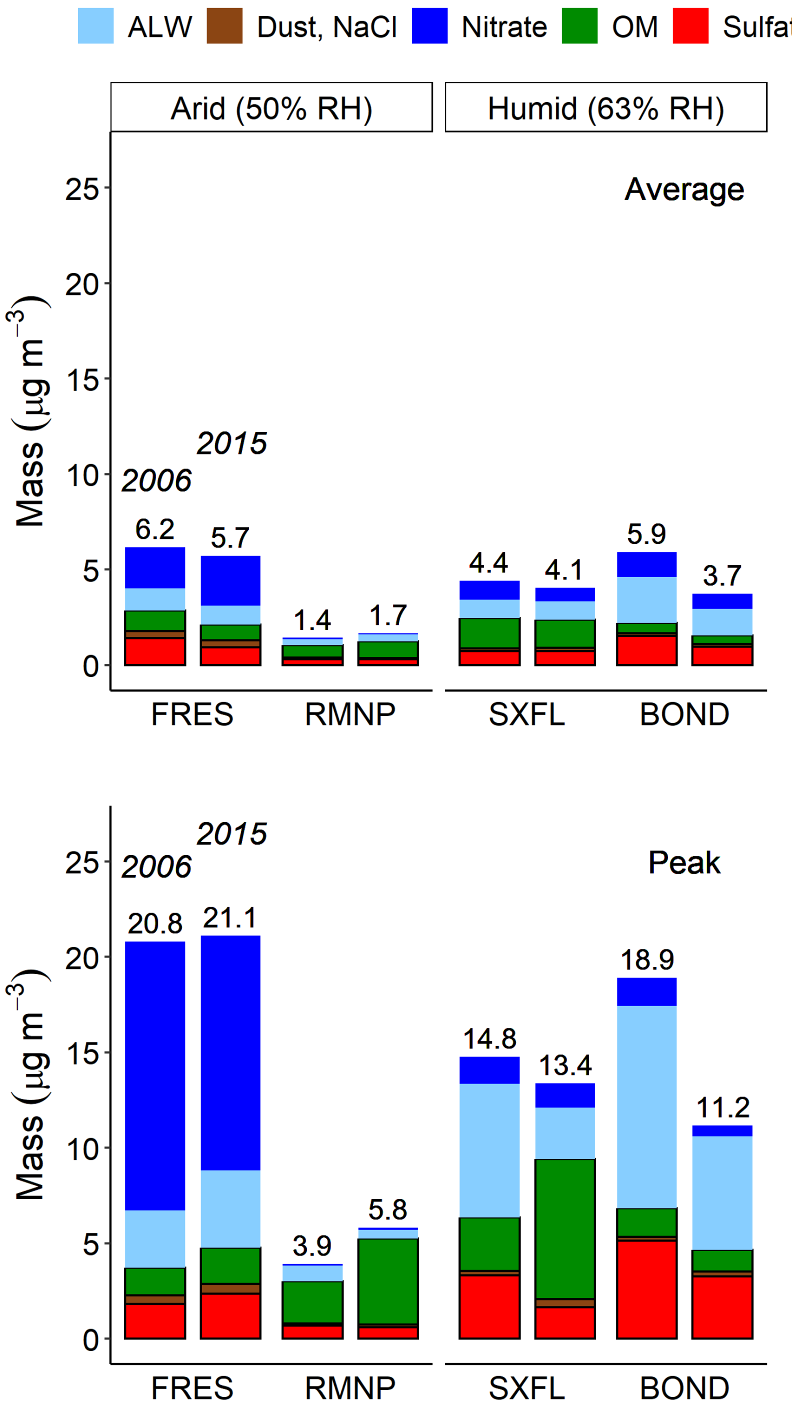

Figure B.2: $PM_{2.5}$ chemical composition consisting of non-volatile species (sulfate, dust (calcium, magnesium, potassium), NaCl, and organic matter (OM)) and semi-volatile species (nitrate, aerosol liquid water (ALW)) on average (top) and peak (bottom, where AOD and $PM_{2.5}$ are both above their locally determined 75^{th} percentile) days for the 4 stations with consistent organic species measurements in both 2006 (left bars) and 2015 (right bars).

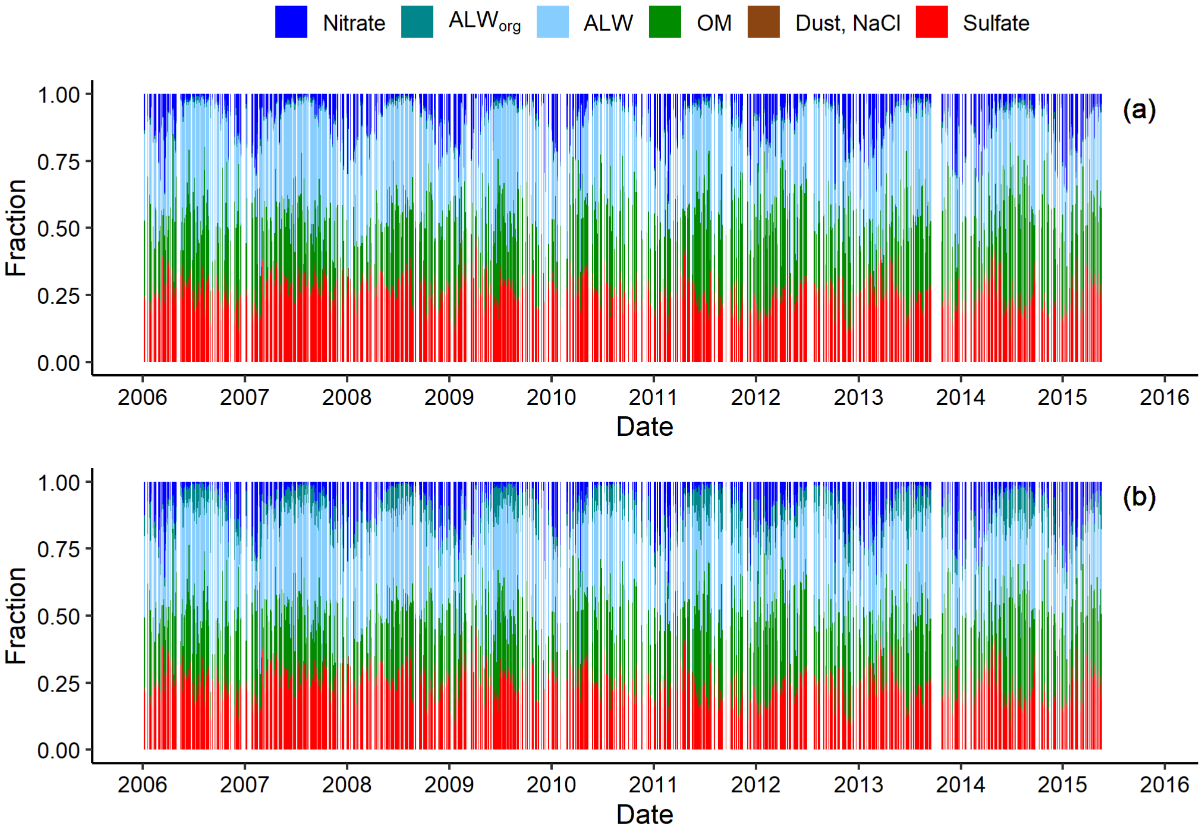

Figure B.3: Sensitivity study of the fractional contribution of organic aerosol liquid water (ALW$_{org}$, teal) to PM$_{2.5}$ mass concentrations using hygroscopicity parameters of (a) $\kappa = 0.3$ and (b) $\kappa = 0.05$ at the GSFC monitoring location with IMPROVE chemical composition from 2006 to 2015. Dust species include calcium, potassium, and magnesium.

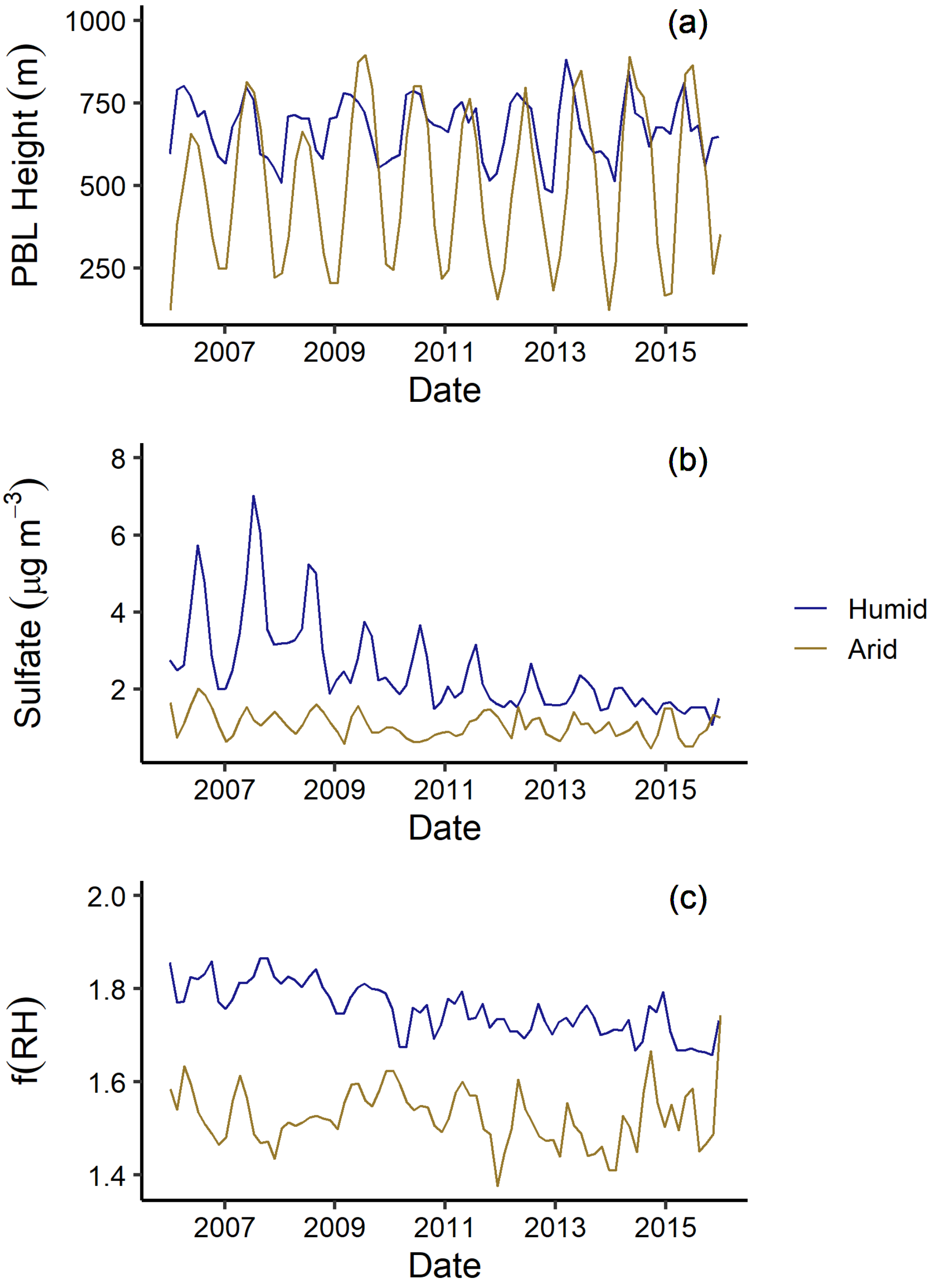

Figure B.4: Time series of (a) planetary boundary layer (PBL) height, (b) sulfate mass concentrations, and (c) f(RH) for 8 AERONET locations in the humid (dark blue) and arid (dark gold) regions from 2006 to 2015.

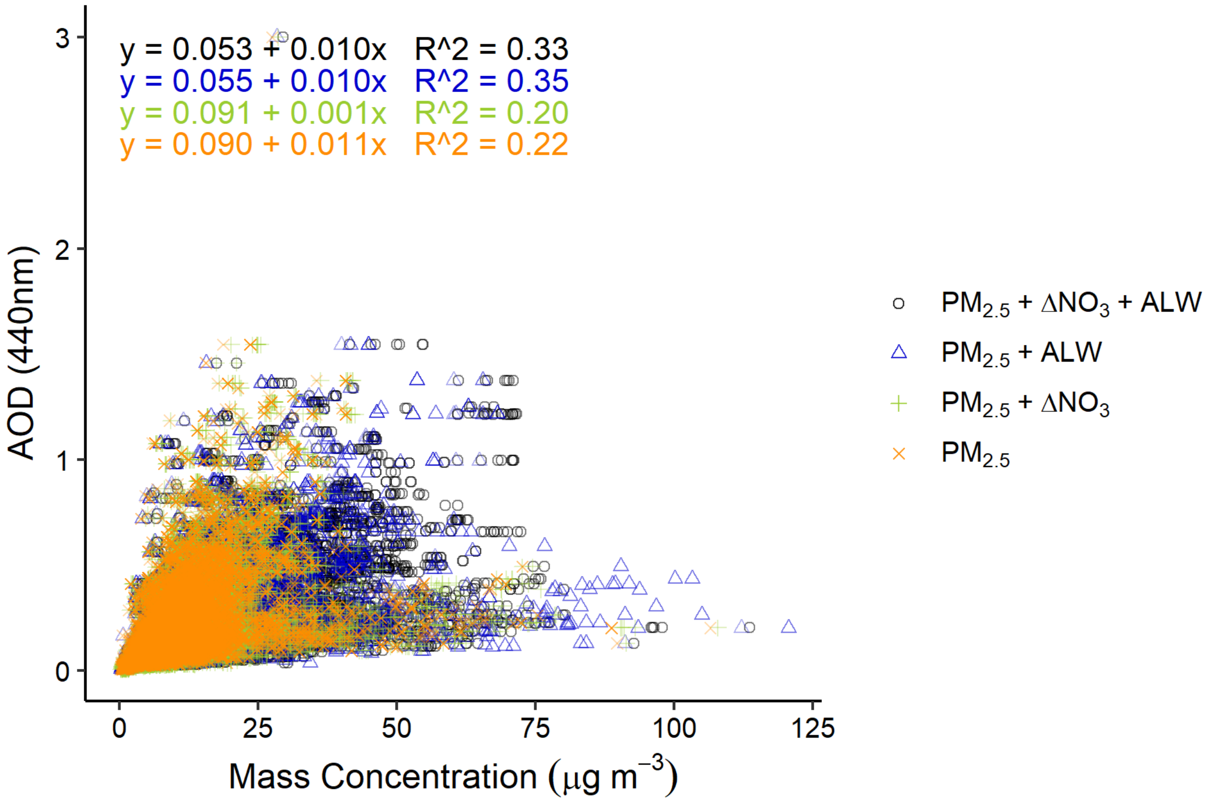

Figure B.5: Comparison of various combinations of $PM_{2.5}$, aerosol liquid water (ALW), and nitrate (NO_3) correction mass concentrations against aerosol optical depth (AOD) at 440 nm with their respective equations of best fit and R^2 values.

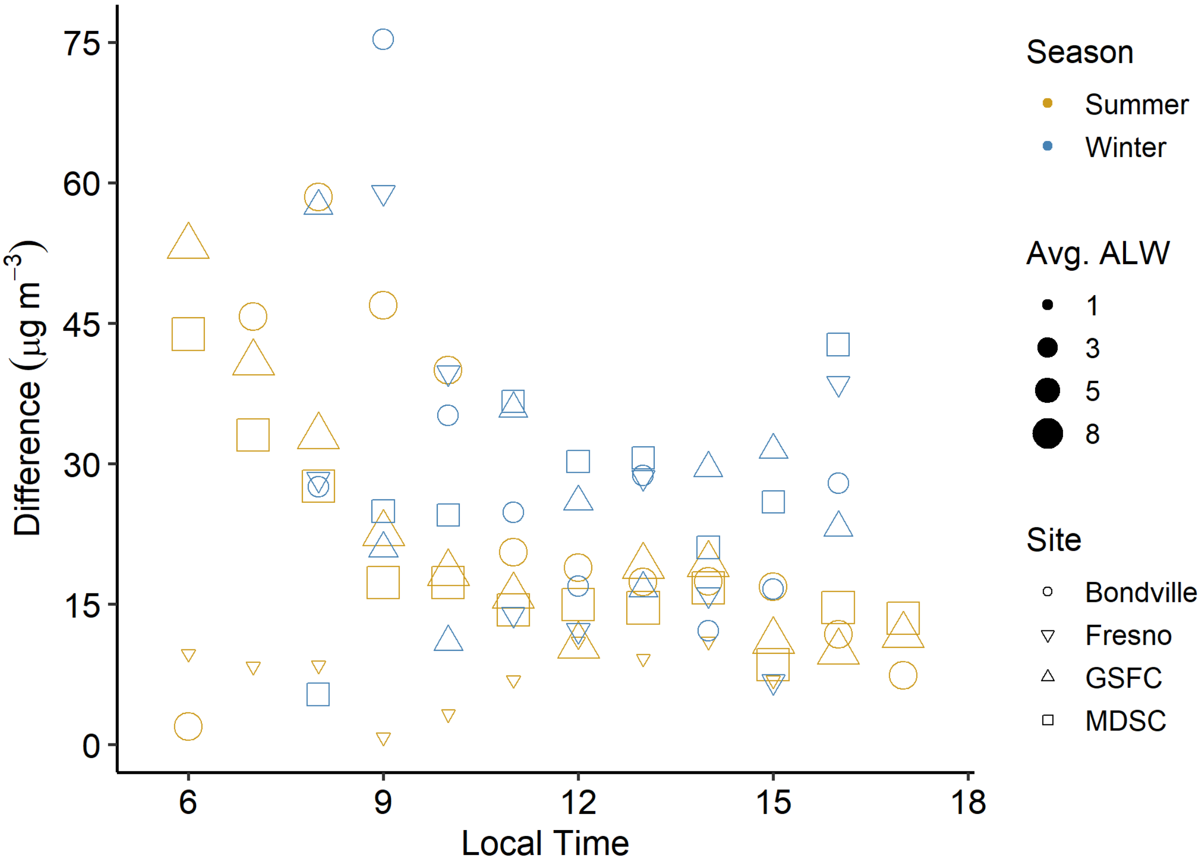

Figure B.6: Differences in hourly CMAQ-simulated and surface observation median η values for the Bondville, Fresno, GSFC, and MDSC sites (point shape) in winter (blue) and summer (tan), grouped by each site's average winter and summer aerosol liquid water (ALW; μg m^{-3}; point size), respectively.

Table B.1: Site information for all AERONET, IMPROVE, EPA CSN, and ambient $PM_{2.5}$ mass concentration monitoring stations used in this study. $PM_{2.5}$ mass monitors include SLAMS (State or Local Air Monitoring Stations), SPM (Special Purpose Monitors), and Tribal monitors. Note that not all SLAMS or SPM stations may have associated site names.

Site Name/Code	Monitoring Network	Latitude	Longitude
Appalachian_State	AERONET	36.2146	-81.6938
Billerica	AERONET	42.52835	-71.2686
BONDVILLE	AERONET	40.05333	-88.3719
El_Segundo	AERONET	33.9125	-118.382
Fresno	AERONET	36.78173	-119.773
Fresno_2	AERONET	36.78537	-119.773
IMPROVE-MammothCave	AERONET	37.1318	-86.1479
NEON_CVALLA	AERONET	40.16063	-105.167
NEON_MOAB	AERONET	38.24833	-109.388
NEON_ORNL	AERONET	35.96413	-84.2826
NEON_RMNP	AERONET	40.2759	-105.546
NEON_SCBI	AERONET	38.89292	-78.1395
Pagosa_Springs	AERONET	37.27028	-107.05
Red_Mountain_Pass	AERONET	37.90745	-107.711
Santa_Monica_Colg	AERONET	34.01685	-118.471
Sevilleta	AERONET	34.35472	-106.885
Sioux_Falls	AERONET	43.73648	-96.626
Table_Mountain	AERONET	40.125	-105.237
Tallahassee	AERONET	30.44583	-84.2994
Thompson_Farm	AERONET	43.10877	-70.9483
Tucson	AERONET	32.233	-110.953
UCSB	AERONET	34.41543	-119.845
USGS_Flagstaff_ROLO	AERONET	35.21484	-111.634
CCNY	AERONET	42.52835	-71.2686
Georgia_Tech	AERONET	33.7802	-84.3995
GSFC	AERONET	38.9925	-76.8398
MD_Science_Center	AERONET	39.28123	-76.6121
LIGO1	IMPROVE	35.9723	-81.9331
LOND1	IMPROVE	42.8624	-71.3801
BOND1	IMPROVE	40.052	-88.3733
SAGA1	IMPROVE	34.2969	-118.028
FRES1	IMPROVE	36.7818	-119.773
MACA1	IMPROVE	37.1318	-86.1479
ROMO1	IMPROVE	40.2783	-105.546
CANY1	IMPROVE	38.4587	-109.821
GRSM1	IMPROVE	35.6334	-83.9416
SHEN1	IMPROVE	38.5229	-78.4348
SHMI1	IMPROVE	37.3038	-107.484

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Table B.1 – continued from previous page

Site Name/Code	Monitoring Network	Latitude	Longitude
WEMI1	IMPROVE	37.6594	-107.800
BOAP1	IMPROVE	33.8695	-106.852
BLMO1	IMPROVE	43.7158	-96.1913
SAMA1	IMPROVE	30.0926	-84.1614
SAGU1	IMPROVE	32.1746	-110.737
SAWE1	IMPROVE	32.2486	-111.218
RAFA1	IMPROVE	34.7339	-120.007
SYCA1	IMPROVE	35.1406	-111.969
ATLA1	IMPROVE	33.6878	-84.2905
WASH1	IMPROVE	39.31083	-76.4744
Clovis-Villa	SLAMS	36.81945	-119.71643
HU-Beltsville	SLAMS	39.05528	-76.87833
Rockville	SLAMS	39.11431	-77.10688
Oldtown	SLAMS	39.29773	-76.60460
Madera-City	SLAMS	36.95326	-120.03420
3425 N FIRST ST, FRESNO	SLAMS	36.78133	-119.77319
Picayune Rancheria	TRIBAL	37.21360	-119.69907
MCMILLAN NCore-PAMS	SLAMS	38.92185	-77.01318
Edgewood	SLAMS	39.41019	-76.29695
Hanford-Irwin	SLAMS	36.31567	-119.64345
Fresno – Garland	SLAMS	36.78538	-119.77321
ISWS CLIMATE STATION	SLAMS	40.05278	-88.37251
Padonia	SLAMS	39.46048	-76.63354
Visalia-Church	SLAMS	36.33218	-119.29123
Lee District Park	SLAMS	38.77335	-77.10468
HAINS POINT PM-2.5 AND IMPROVE SITE ON ROOF OF PARK POLICE BLDG.	SLAMS	38.87626	-77.03406
Essex	SLAMS	39.31083	-76.47444
DUDLEY SQUARE ROXBURY	SLAMS	42.32950	-71.08260
PEARL ST MUNICIPAL PARKING LOT	SLAMS	42.99578	-71.46253
MOOSEHILL SCHOOL	SLAMS	42.86253	-71.38014
	SPM	40.81722	-74.04376
	SLAMS	40.81681	-74.04367
	SLAMS	40.83311	-74.04346
Newark Firehouse	SLAMS	40.72099	-74.19289
New Brunswick	SLAMS	40.47283	-74.42240
Rutgers University	SLAMS	40.46218	-74.42944
Elizabeth Lab	SLAMS	40.64144	-74.20837
MORRISANIA	SLAMS	40.83606	-73.92009

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Table B.1 – continued from previous page

Site Name/Code	Monitoring Network	Latitude	Longitude
BOTANICAL GARDEN	SLAMS	40.86585	-73.88083
IS 52	SLAMS	40.81600	-73.90200
DIVISION STREET	SLAMS	40.71436	-73.99518
QUEENS COLLEGE 2	SLAMS	40.73614	-73.82153
SD School for the Deaf	SLAMS	43.54792	-96.70077
Fresno – Garland	EPA CSN	36.78538	-119.77321
DUDLEY SQUARE ROXBURY	EPA CSN	42.32950	-71.08260
IS 52	EPA CSN	4081600	-73.90200
DIVISION STREET	EPA CSN	40.71436	-73.9518
QUEENS COLLEGE 2	EPA CSN	40.73614	-73.82153

Appendix C

Supporting Information for Chapter 4

Table C.1: Mann-Whitney U test p-values for the comparison of questions about the usefulness of the Twine prelabs between the Fall 2022 and Winter 2023 quarters. Statistical significance ($p < 0.05$) is indicated with bold and italicized font.

Question	Mann-Whitney U p-value
The Twine prelabs helped me to understand the laboratory procedures.	0.137
The Twine prelabs helped me to understand the concepts involved in the labs.	***0.049***
I would prefer to do Twine prelabs instead of traditional pre-lab calculations and problems.	***0.009***
Performing the Twine prelabs helped me feel more prepared to do the experiments in-person.	0.050
I would prefer to do Twine prelabs in addition to traditional pre-lab exercises.	***0.003***
I enjoyed the Twine prelabs.	0.298
The Twine prelabs helped me to understand the source and appearance of the experimental data we collected.	***0.032***

Table C.2: Mann-Whitney U test p-values for the comparison of Likert scale student responses to MSLQ questions about comfort in the laboratory in pre- and post-quarter surveys for the Fall 2021 quarter (no Twine prelabs) and the combined Fall 2022 and Winter 2023 quarters (2 and 4 Twine prelabs, respectively). Statistically significant ($p < 0.05$) differences are indicated with asterisks.

Question	Fall 2021 Mann-Whitney U p-value	Fall 2022, Winter 2023 Mann-Whitney U p-value
When getting ready for chemistry lab, I get concerned about the lab procedures we will use.	0.205	0.392
I worry about whether I have enough time to complete the lab.	0.413	0.001*
When I get ready for lab, I get concerned about recording the data we will generate.	0.655	0.296
When preparing for lab, I am concerned about the time available for doing the experiment.	0.141	1.34×10^{-5}*
When I get ready for chemistry lab, I get concerned about the chemicals we will use.	0.053	0.018*
When getting ready for chemistry lab, I get concerned about the equipment we will use.	0.394	0.202
When I get ready for chemistry lab, I get concerned about working with other students.	0.643	0.176

Table C.3: Survey questions regarding the Twine prelabs asked as part of the MSLQ.

Question	Response Type
The Twine prelabs helped me to understand the laboratory procedures.	Likert Scale
The Twine prelabs helped me to understand the concepts involved in the labs.	Likert Scale
I would prefer to do Twine prelabs instead of traditional prelab calculations and problems.	Likert Scale
Performing the Twine prelabs helped me feel more prepared to do the experiments in person.	Likert Scale
I would prefer to do Twine prelabs in addition to traditional prelab exercises.	Likert Scale
I enjoyed the Twine prelabs.	Likert Scale
The Twine prelabs helped me to understand the source and appearance of the experimental data we collected.	Likert Scale
The Twine prelabs helped me to understand the laboratory procedures.	Likert Scale
For the chromatography lab(s) that you did, do you feel like the virtual Twine prelab helped you understand the experiment better? Why or why not?	Free Response

Table C.4: Rubric for the Thermocouples and Adiabatic Calorimetry lab report.

Criteria	Meets Expectations	Does Not Meet Expectations
Expression of Numbers	All numbers are reported with the correct number of significant figures, and also have uncertainties and units where appropriate.	
Figures	All figures are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. All figures have a figure number and descriptive caption.	
Tables	All tables are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. The columns and rows are aligned correctly so that the table is legible and visually appealing. All tables have a table number and descriptive title.	Tables that are not created with the Word/Docs table tool will not meet expectations. Screenshots from Excel or Google Sheets are not appropriate. Tables with very small cells that are not legible will also not meet this criterion.
Propagation of Error	The author clearly and correctly explains the process for finding the uncertainty in theoretical gain values based on the uncertainties in individual resistors.	
The Science - Op Amp Circuit Math	All calculations of expected voltages from op amp circuits are performed correctly. Explanations of predicted voltages, where required, are written clearly and correctly.	
The Science - Seebeck Coefficient	Questions on the Seebeck coefficient (predicted voltage change for a given temperature change, calculated vs. expected Seebeck coefficient) are answered correctly and written in such a way that everything can be understood.	

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Table C.4 – continued from previous page

Criteria	Meets Expectations	Does Not Meet Expectations
The Science - Thermocouple Calibration Curve	The thermocouple amplifier is calibrated correctly, with the correct variable on the correct axis of the calibration curve. The experimental Seebeck coefficient is calculated correctly from the gain of the circuit and the calibration curve.	
The Science - Calorimeter Calibration	The calorimeter calculation is performed correctly, and the calculations are explained clearly. The author makes a comparison to the heat capacity of pure water.	
The Science - Using the Calorimeter	The calorimeter constant is used correctly to determine the enthalpy of solution of the salt. The result is compared to the theoretical value, and errors that might have affected the result in the observed direction from the true value are identified.	
Scientific Writing - Support for Conclusions	All conclusions that are reached are supported with evidence and references to figures and data. The explanations are clear and understandable and are based on a sound understanding of the science.	
Writing - Grammar and Spelling	The report demonstrates the correct use of Edited Standard Written English and seems to have been proofread. There are fewer than four mistakes in grammar, spelling, and word use throughout.	
Citation	The author includes a relevant citation for the enthalpy of solution of the salt they used in the experiment. The citation is formatted correctly according to ACS guidelines.	The source is not peer-reviewed or otherwise trustworthy, or the citation is not formatted correctly according to the ACS guidelines.

Table C.5: Rubric for the Transimpedance Amplifiers and Colorimetry lab report.

Criteria	Meets Expectations	Does Not Meet Expectations
Expression of Numbers	All numbers are reported with the correct number of significant figures, and also have uncertainties and units where appropriate.	
Figures	All figures are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. All figures have a figure number and descriptive caption.	
Tables	All tables are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. The columns and rows are aligned correctly so that the table is legible and visually appealing. All tables have a table number and descriptive title.	Tables that are not created with the Word/Docs table tool will not meet expectations. Screenshots from Excel or Google Sheets are not appropriate. Tables with very small cells that are not legible will also not meet this criterion.
Propagation of Error	Propagation of error calculations for the RC circuits are performed correctly, and the correct absolute uncertainty is derived from the relative uncertainty in each case. The propagated error is used correctly to compare the measured time constant to the experimental time constant.	
The Science - Circuit Math	All calculations of expected voltages from RC circuit filters and op amp circuits are performed correctly. Explanations of predicted voltages, time constants, cutoff frequencies, etc. are written/graphed clearly and correctly.	

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Table C.5 – continued from previous page

Criteria	Meets Expectations	Does Not Meet Expectations
The Science - Circuit Behavior	Questions that ask the writer to explain how a circuit (low-pass filter, transimpedance amp, etc.) are answered correctly and the explanations are clear. The questions are obviously answered in the writer's own words and do not rely too much on definitions from websites, articles, etc.	
The Science - Determining Absorbance	The author clearly and correctly explains how absorbance values were determined using voltage measurements from the transimpedance amplifier. The explanation is complete and includes all necessary information.	
The Science - Calibration Curve	The Beer's Law plot is made correctly and fitted to a line. The slope and intercept are given with their uncertainties. The plot is used correctly to determine the amount of Red 40 in Gatorade.	
The Science - Nyquist Theorem and Signal Modulation	The author suggests a use for signal modulation in an analytical chemistry application. The reason that signal modulation is used in that application is clearly in the author's own words and makes sense.	
Scientific Writing - Support for Conclusions	All conclusions that are reached are supported with evidence and references to figures and data. The explanations are clear and understandable and are based on a sound understanding of the science.	
Writing - Grammar and Spelling	The report demonstrates the correct use of Edited Standard Written English and seems to have been proofread. There are fewer than four mistakes in grammar, spelling, and word use throughout.	

Table C.6: Rubric for the Fluorescence lab report.

Criteria	Meets Expectations	Does Not Meet Expectations
Expression of Numbers	All numbers are reported with the correct number of significant figures, and also have uncertainties and units where appropriate.	
Figures	All figures are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. All figures have a figure number and descriptive caption.	
Tables	All tables are formatted appropriately according to the guidelines in the Writing the Journal Style Lab Report document. The columns and rows are aligned correctly so that the table is legible and visually appealing. All tables have a table number and descriptive title.	Tables that are not created with the Word/Docs table tool will not meet expectations. Screenshots from Excel or Google Sheets are not appropriate. Tables with very small cells that are not legible will also not meet this criterion.
Propagation of Error	Propagation of error calculations for the RC circuits are performed correctly, and the correct absolute uncertainty is derived from the relative uncertainty in each case. The propagated error is used correctly to compare the measured time constant to the experimental time constant.	
The Science - Excitation Wavelength	The choice of the ideal excitation wavelength is justified by reference to spectra presented in the lab report. The chosen wavelength makes sense based on the absorption and excitation spectra and the constraints of the experiment.	
The Science - Spectra Explanation	The author correctly explains what information can be obtained from an excitation spectrum (or EEM) that can't be obtained from an absorption spectrum alone.	

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Table C.6 – continued from previous page

Criteria	Meets Expectations	Does Not Meet Expectations
The Science - Quantum Yield	The quantum yield of quinine is calculated correctly from the calibration curves. The calibration curves in this part of the report are plotted with the correct variables on the correct axes and are fitted and interpreted correctly.	
The Science - Quinine Quantification	The amount of quinine present in tonic water is calculated correctly from the calibration curves. The calibration curves in this part are plotted with the correct variables on the correct axes and are fitted and interpreted correctly.	
The Science - Signal Modulation	The author suggests a use for signal modulation in an analytical chemistry application. The reason that signal modulation is used in that application is clearly in the author's own words and makes sense.	
Scientific Writing - Support for Conclusions	All conclusions that are reached are supported with evidence and references to figures and data. The explanations are clear and understandable and are based on a sound understanding of the science.	
Writing - Grammar and Spelling	The report demonstrates the correct use of Edited Standard Written English and seems to have been proofread. There are fewer than four mistakes in grammar, spelling, and word use throughout.	

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Table C.6 – continued from previous page

Criteria	Meets Expectations	Does Not Meet Expectations
Scientific Writing - The Experimental Section	The Experimental section is written in the correct format, as specified in the ”Writing a Journal-Style Lab Report” document. The section is written in the third person, past tense. The section does not give instructions to the reader. The level of detail is correct; only details that would affect the results of the experiment are mentioned, and superfluous or minor details are not included.	

Twine Prelab Exercises
Thermocouples and Adiabatic Calorimetry - https://stephenamang.github.io/DC-Circuits-PreLabVersion.html
Transimpedance Amplifiers and Colorimetry - https://stephenamang.github.io/AC-Circuits-PreLabVersion.html
GCMS - `https://stephenamang.github.io/GCMS-PreLabVersion.html`
HPLC - `https://stephenamang.github.io/HPLC-PreLabVersion.html`

The CYOA prelab exercises linked here will not collect user data into a Google Sheet. To add this functionality to these Twine exercises, edit the HTML files in Twine and follow the instructions below:

In Twine
Throughout the CYOA exercise the code contains variables that can be used to track student choices. For example, in the HPLC prelab, the card "Mobile Phase Determination 2" contains the line:
(set: $flowRate to 1.0)

Which sets the variable flowRate to the value of 1.0. This value can be changed or used in calculations in any other card during the Twine simulation. For this variable, you can see it being used in JavaScript code in the "90% Water, 10% Methanol" card or any other card where a chromatogram is simulated.

Even if you're not using a variable to calculate simulated data, it may be useful to keep track of it to find out how students are interacting with the prelab. To do this, we use the last card in the simulation, which is always named "Post Data". This card contains a "thank you" message to users, and also some JavaScript code that they don't see, but that is used to save the data to Google Sheets. This code has been left intact in the simulations accessed with the links above, except that the link to a Google Sheet in the "url:" field has been deleted. All you need to do is add a link to your own Google Sheet using the directions below after editing the code above that field to add or remove any variables from the data to be saved.

In Google Sheets
Create a Google Sheet with the first column "Timestamp" and the rest of the columns labeled with the names, in order and typed exactly the same way (case sensitive, etc), as the Twine variables in the Post Data card. Share the Google Sheet so that anyone with the link can Edit.

From the Google Sheets Extensions menu, choose AppScript. Update the name of the App, as necessary. Next, from the top menu, choose "setup" and click "Run".

Allow permissions, as necessary, then click Deploy and choose New Deployment. In the menu, give it a name and the following settings (your email should show up instead of mine).

Get the web link from the resulting window and paste it in the URL field of the Post Data card in the Twine script. The script will only run if a user makes it all the way through the simulation and accesses the Post Data card.