Project Summary

Long-term atmospheric measurements over the past several decades have been used to document and understand chemical changes to atmospheric composition as a result of human activities (e.g. greenhouse gases, ozone and particulate pollution, acid rain, and ozone depleting substances). These examples demonstrate the power of continuous measurements spanning a decade or more at one location. However, our understanding of multiphase chemical processes remains incomplete. Human emissions also continue to evolve, even as ecosystems continue to respond to previously emitted pollutants. Thus, even while some crises appear to have been averted due to better understanding and interventions to reduce emissions of specific pollutants, we must remain vigilant to emerging threats to human and ecosystem health.

The proposed workshop will bring together researchers to develop a long-term plan for coordinating research effort at multiple high-elevation sites within the continental U.S., with the goal of assessing changes in atmospheric composition and developing better fundamental understanding of atmospheric processes. Combining measurements with different atmospheric chemistry foci (including the gas, aerosol, and aqueous phases) at high elevation sites across the U.S., along with measurements of the relevant photochemical and meteorological influences, will be key to developing better understanding of the sources, transformation and fate of atmospheric pollutants, so as to better understand and predict changes in atmospheric composition. Obtaining input and encouraging collaboration from the broader atmospheric sciences community will allow us to tackle complex emerging issues with greater competence.

Intellectual Merit: Workshop participants will help to identify and prioritize the most pressing research objectives and help develop a plan of action for the proposed high-elevation measurement network. Initial questions to spark this discussion:

i. *What are the domestic and international sources of key global pollutants such as ozone, mercury, and aerosols; and what are the impacts of these pollutants?*
ii. *What are the regional sources and sinks of aerosols and greenhouse gases in the U.S.?*
iii. *What determines the cloud-nucleating properties and radiative effects of aerosols?*
iv. *What is the current state of biosphere-atmosphere exchange processes, and how will these change in the future?*
v. *What is the role of snow and ice in the climate system? How will mountain snowfall, along with aerosol deposition on snow and ice, change in the future?*
vi. *What are the sources, sinks, and trends of reactive gases, and what drives atmospheric oxidation chemistry and cycles?*

Broader Impacts: The proposed network of mountaintop research observatories will elevate atmospheric sciences research in the U.S. by providing crucial infrastructure and transfer of knowledge that will advance the science currently being conducted independently by researchers at separate locations. This initiative will broaden the research objectives currently attainable by enabling a more complete set of measurements at each site and greater cooperation within the broader atmospheric science community, and will provide additional training opportunities for students and young scientists.

Workshop on the proposed MOCA Network

MOCA: Mountain Observatories for Composition of the Atmosphere

Introduction

This proposal is a request for NSF support for a science workshop dedicated to developing the research objectives and measurement/management strategy for a network of mountaintop observatories for coordinated measurements of atmospheric composition at high-elevations across the continental U.S. The first half of this proposal outlines the science motivation for such a network. The second half outlines the workshop goals, participants, venue and organization.

Motivation

Climatic changes in mountainous area can have serious impacts on human health and prosperity. Mountain snowpack is a primary freshwater source for numerous populous regions (Barnett et al. 2005). Smoke from mountain forest fires is increasingly responsible for poor air quality, especially in the western U.S. (Hallar et al. 2017, McClure and Jaffe 2018). Wildfires are anticipated to burn more frequently and over greater areas as climate continues to change (Westerling et al. 2006, Kurz et al. 2008, Pechony and Shindell 2010, Dennison et al. 2014, Abatzoglou and Williams 2016), further exacerbating the carbon cycle imbalance and worsening air quality.

In addition to sitting within an ecologically sensitive environment for which measurements are sparse (Hik and Williamson 2019), mountaintop observatories are well positioned to discover changes in atmospheric composition without incurring the considerable expense required for airborne sampling. The continuous sampling possible at mountain research observatories provides access to critical information about long-term changes in regional background atmospheric composition, as well as seasonal and diurnal processes and extreme episodes (e.g. biomass burning, radionuclide releases, volcanic ash, dust storms, stratospheric intrusions). A prime example is the long-term measurement of CO_2 obtained at Mauna Loa, which unambiguously demonstrated the degree to which human activities can alter atmospheric composition (Keeling et al. 1976), sounding a global alarm on climate change more than 50 years ago. Other more recent examples include long-term measurements obtained at Mount Bachelor in the northwestern U.S., which have been used to track changes in long range transport of wildfire smoke and pollutants arriving to the U.S. from Asia (Zhang and Jaffe 2017), and long-term measurements at Whiteface Mountain in the northeastern U.S., which have been used to track ongoing air quality improvements resulting from emissions reductions in the U.S. (Rattigan et al. 2017).

The National Science Foundation (NSF) and other federal and state government agencies have supported these sorts of research efforts within the U.S. for many years, but efforts like these have generally been led by individual researchers, independently, without a plan for coordinating activities between sites or for sustaining long-term operations. Numerous national reports have concluded that **there is a critical need to establish a network of coordinated, well-calibrated measurements to continually assess our fundamental understanding of atmospheric**

composition (NASEM 2016, NRC 2001,2010,2011). An important lesson learned through a lifetime of measurements at Mauna Loa: New discoveries about atmospheric processes will not happen in the absence of long-term observations (Sundquist and Keeling 2009). Even phenomena that are long thought to have been solved like the "ozone hole" and "acid rain" require continuous monitoring to assess and refine our fundamental understanding and assumptions (Palmer, 2020).

The Global Atmospheric Watch (GAW) program was established by the World Meteorological Organization in the 1960's in response to environmental concerns over rapid industrialization, with the goal of providing continuous, reliable observations and scientific assessment of global atmospheric composition to inform environmental policy. At present, the U.S. operates very few mountain sites for long-term observations of atmospheric composition. Not a single mountain site within the continental U.S. has reached the international distinction of a World Meteorological Organization Global Atmospheric Watch Global Station. For comparison, there are currently six such mountain sites in Europe. To be certified as a global station requires a commitment to an extensive measurement program within at least three of the six GAW focal areas: aerosols, greenhouse gases, reactive gases, stratospheric ozone and ultraviolet radiation, atmospheric deposition and urban pollution.

Figure 1. While there are 31 global stations operating within the GAW network, there are no high-elevation sites within the continental U.S. The only GAW global stations in North America are at Trinidad Head, Barrow and Alert (three sea-level sites).

In 2019, we submitted a proposal to NSF to establish a network of mountaintop observatories in the continental U.S. to monitor atmospheric composition across four broad themes: 1) reactive trace gases, 2) greenhouse gases, 3) aerosols, and 4) clouds and precipitation. The proposed Mountain Observatories for Composition of the Atmosphere (MOCA) network would build a common measurement infrastructure and data management structure across select sites to promote coordinated observations and to develop fundamental understanding of atmospheric composition, which has been identified as a national priority.

Some of the key questions we posed:

i. *What are the domestic and international sources of key global pollutants such as ozone, mercury, and aerosols; and what are the impacts of these pollutants?*
ii. *What are the regional sources and sinks of aerosols and greenhouse gases in the U.S.?*
iii. *What determines the cloud-nucleating properties and radiative effects of aerosols?*
iv. *What is the current state of biosphere-atmosphere exchange processes, and how will these change in the future?*
v. *What is the role of snow and ice in the climate system? How will mountain snowfall, along with aerosol deposition on snow and ice, change in the future?*
vi. *What are the sources, sinks, and trends of reactive gases, and what drives atmospheric oxidation chemistry and cycles?*

The proposed MOCA network spans across different climatic zones and ecosystems in the U.S. and includes four internationally recognized atmospheric research observatories, which have been in operation for decades and are well-positioned to observe both inflow to the continental U.S. from the west and outflow to the east.

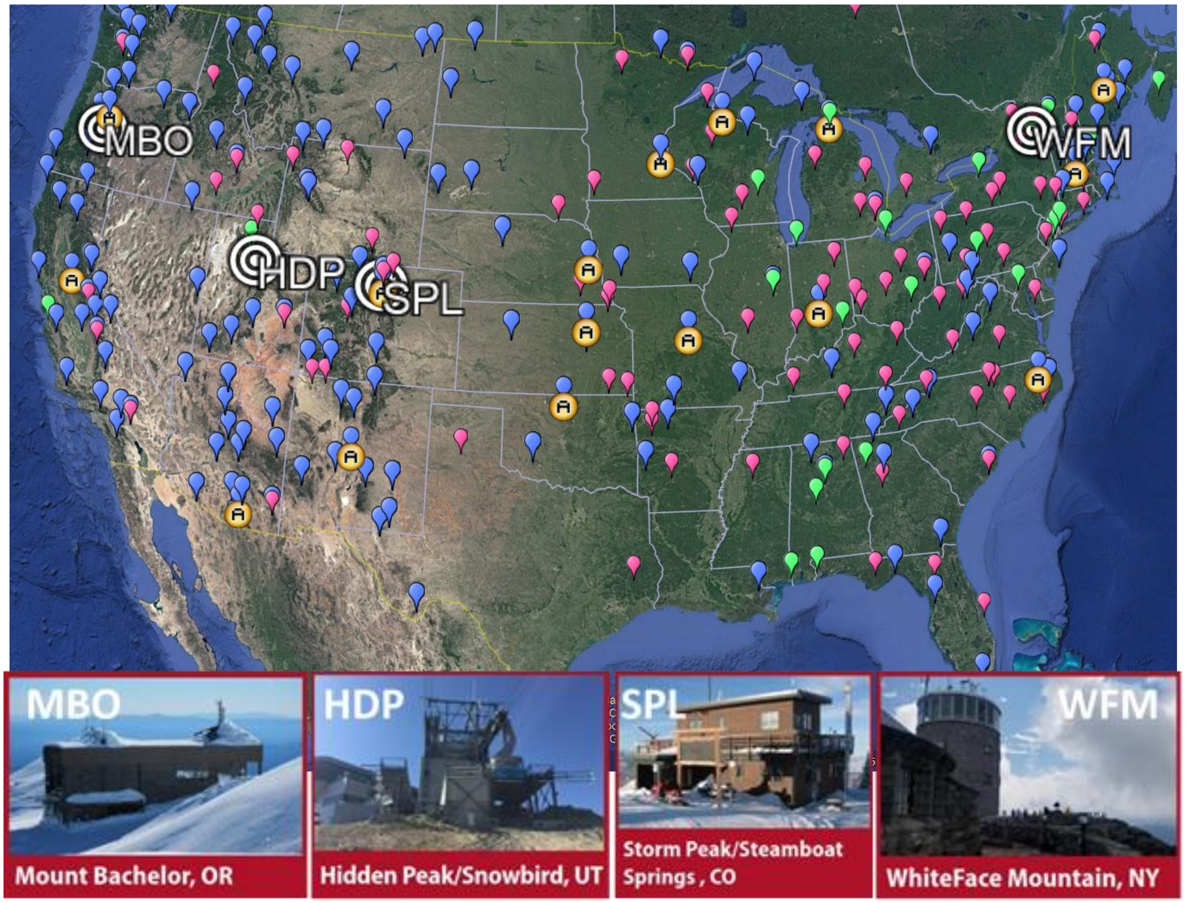

Figure 2. The proposed MOCA network, superimposed with the U.S. EPA IMPROVE Network PM2.5 monitors (blue markers), the National Acid Deposition Program (NADP) Ammonia (AMON) and Atmospheric Mercury (AMNet) networks (pink and green, respectively) and the AmeriFlux Network Core Sites (marked by the letter 'A'). The four high-elevation atmospheric observatories for the proposed MOCA Network (MBO, HDP, SPL and WFM) sit atop four prominent mountain peaks.

Historically, research at these four individual sites has primarily concentrated on different aspects of the atmospheric sciences: reactive trace gases at MBO, cloud and aerosol microphysics at SPL, cloud water chemistry at WFM, and greenhouse gases at HDP. We posit that pressing research questions cannot be fully addressed in isolation and require an understanding of the broader atmospheric context. For instance, aerosol and gas phase measurements are better understood in a meteorological context; cloud microphysical properties are better understood in the context of aerosol chemical and physical properties; and aerosol and cloud chemical properties are better understood in the context of reactive trace gases. Without this broader atmospheric context, data interpretation is subject to assumptions that limit the usefulness of any individual set of measurements. The proposed network design would broaden the focus of each site by fully aligning all locations with the four science themes, drawing on the expertise at each site to form a collaborative network for the exchange of knowledge across the United States.

Workshop Goals and Organization

The proposed 2-day workshop will bring together researchers to critically evaluate and further develop a plan of action for developing a coordinated long-term measurement network at high-elevation atmospheric observatories across the U.S.

Specific goals of the workshop will be to:

1. further develop and refine the research objectives within each of the proposed science themes:
    - reactive trace gases
    - greenhouse gases
    - aerosols and radiation
    - clouds and precipitation
2. identify critical measurements needed to meet these research objectives
3. identify strategies to leverage measurements within different science themes to address additional research objectives or to better address existing research objectives
4. identify research objectives that would especially benefit from a common set of measurements across sites
5. develop metrics to prioritize research objectives
6. evaluate the measurement infrastructure and data management needs for each site to be incorporated into the GAW network as a global station
7. encourage critical discussion about how best to implement and manage such a broad range of research objectives across multiple sites, including a data management plan
8. develop a timeline for each research objective, following initial instrument integration at each site
9. identify opportunities for meaningful and productive collaboration with other agencies and local/regional stakeholders
10. identify strategies for sustaining long-term, well-calibrated measurements at each site

Conclusions from the workshop will be disseminated through a workshop summary publication.

Recent Meetings

Several recent workshops have focused on scientific objectives that fall within the scope of our proposed measurement network, which will help lay the groundwork for the goals laid out above. The 2016 Cloud Chemistry workshop at WFM focused on chemical processing occurring within clouds and impacts on gas phase and aerosol composition. The 2018 CO_2-USA workshop at University of Utah focused on urban carbon cycle science. The 2019 Air QUAlity Research in the western US (AQUARIUS) workshop focused on wintertime air quality and greenhouse gas emissions in the western US. Three preceding international workshops, which have produced two special journal issues, led to the idea of the proposed MOCA network: The 2010, 2014 and 2017 Symposium on Atmospheric Chemistry and Physics at Mountain Sites, in Switzerland, Colorado and Japan (Hallar et al. 2016, Hatakeyama et al. 2019).

Workshop Organizing Committee

The organizing committee for this workshop includes the principal investigators and science theme leads for the MOCA network: Gannet Hallar (University of Utah, aerosols theme lead), Sara Lance (University at Albany, clouds and precipitation theme lead), Dan Jaffe (University of Washington, reactive trace gas theme lead), and John Lin (University of Utah, greenhouse gas theme lead). Two additional local organizers at the University at Albany, Justin Minder and Paul Casson, will help to plan out the field trip at WFM.

Workshop Location, Schedule and Participants

We propose for the workshop to take place in early fall (Late August/Early September) of 2020. Workshop outcomes will be used to revise our MOCA proposal in anticipation for re-submission to the next NSF mid-scale infrastructure call, with expected pre-proposal deadline of February 2021 and full proposal deadline of May 2021.

We propose to hold the 2-day workshop at the Campus Center Board room or Science Library Conference Room at the University at Albany in Albany NY (located near an international airport and in close proximity to inexpensive accommodations), and an optional 1-day trip to Whiteface Mountain (WFM) in Wilmington NY (2.5 hours drive away). The WFM field trip will provide an opportunity for researchers to visit this remote site (which will likely be the first time a majority of them will have the opportunity to do so) and allow researchers to assess the existing research infrastructure and future possibilities of the site. If possible, we will hold the field trip just prior to the workshop, to spark discussion and ideas relevant to the workshop goals.

We plan to invite approximately 30 participants to the workshop, with priority invitations sent to a select group of researchers who we expect to participate most directly in implementation of the MOCA network objectives (as science theme advisors and/or collaborators at each of the sites). We request funding so that we can offer travel support for the priority group to encourage their participation, especially junior researchers and those at smaller research institutions for whom travel support is needed. Invitations will be announced by email.

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Results from Prior NSF Support

<u>From the workshop PI</u>

PI: Dr. Lance, Award #:1753278, "RAPID: Supporting Cloud Water Collection at Whiteface Mountain Research Observatory. Pilot Study to Assess Chemical Processing of Organics within Clouds (CPOC)", $24,930, 5/1/17- 7/30/18.

Intellectual Merit: This grant supported the CPOC pilot study at Whiteface Mountain in Aug 2017. Measurements confirmed that organic matter is now a dominant component of both aerosol and cloud water, in contrast to the view from decades ago. Below-cloud and in-cloud chemical measurements were conducted, to provide insights into chemical processes leading to the observed long-term trend toward higher organic mass fractions. One manuscript has been published (Zhang et al., 2019). Another manuscript is in review (Lance et al, *submitted in 2019*). **Broader Impacts:** The CPOC pilot study demonstrated the ongoing value of cloud water sampling at WFM and its relevance to the broader scientific community, adding leverage to a subsequent request for funding from NYSERDA for continued support of cloud water collection and analysis, which was awarded in 2018 for five more years under the new guidance of Dr. Lance. Several graduate (4) and undergraduate (3) students gained research experience and contributed to research at all levels. Presentations and online showcasing of the pilot study helped Dr. Lance recruit two new graduate students for Fall 2018, the first students in Dr. Lance's tenure track career. All data/metadata are available at the ASRC Atmospheric Chemistry Server.

PI: Dr. Lance, Award #:1945563, "CAREER: Revitalizing aerosol-cloud-chemistry research at Whiteface Mountain", $683,701, 2/1/20- 1/31/25.

Intellectual Merit: This recently awarded grant will support ongoing research activities at the Whiteface Mountain (WFM) summit research observatory in New York State, particularly research focused on aerosol-cloud interactions. The targeted measurements will provide essential information for process-level understanding necessary to constrain air quality and climate modeling studies under current and future emissions scenarios. Central to the proposed research will be the installation of a ground-based Counterflow Virtual Impactor at the summit of WFM, which will allow for characterization of the cloud droplet residual composition in real-time using ion chromatography and water soluble organic carbon analysis, for direct comparison to the long-term measurements of cloud water composition. **Broader Impacts:** The biggest short-term impact of the proposed research will be re-invigoration of scientific activity at the WFM summit research facility, at a time when the site is celebrating its 50th anniversary, which will encourage substantial U.S. and international collaboration. The outreach activities will help establish a student pipeline from science and engineering disciplines within the Capital District to multi-disciplinary research opportunities in renewable energies, air quality, weather and climate through the Atmospheric Sciences Research Center (ASRC).

Lance, S., J. Zhang, J.J. Schwab, P. Casson, R.E. Brandt, D.R. Fitzjarrald, M.J. Schwab, J. Sicker, C.-H. Lu, S.-P. Chen, J. Yun, J.M. Freedman, B. Shrestha, Q. Min, M. Beauharnois, B. Crandall, E. Joseph, M.J. Brewer, J.R. Minder, D. Orlowski, A. Christiansen, A.G. Carlton, M.C. Barth, Overview of the Cloud Processing of Organics within Clouds (CPOC) Pilot Study at Whiteface Mountain, NY, *In review with the Bulletin of the American Meteorological Society.*

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From the Workshop Steering Committee Members

Co-PI: Dr. Hallar and Dr. Lin, Award #:1912664, " The AQUARIUS (Air QUAlity Research In the western US) Workshop; Salt Lake City, Utah", $25,779, 3/1/19- 2/31/20.

Intellectual Merit: This project supported a workshop, "The AQUARIUS (Air QUAlity Research In the western US) Workshop," held in Salt Lake City, Utah in September 2019. The workshop brought together experts to discuss and develop plans for a future aircraft campaign to investigate wintertime particulate matter (PM) in the mountain basins of the Western U.S. **Broader Impacts:** The outcome of the project will be a white paper outlining the scope and plan for a future aircraft and ground-based field campaign.

PI: Dr. Hallar, Award #:1749865, "Collaborative Research: An in situ Closure Study of Mixed Phase Clouds at Storm Peak", $215,465, 4/1/18- 3/31/21.

Intellectual Merit: Had an extremely successful first field campaign (January 2019) at Storm Peak Laboratory with deployment of new inlet and both warm and cold cloud chambers (SPIDER) to determine the particles on which the droplets and ice crystals in mixed-phase clouds form. Preparing two publications to result from the first year of sampling. One publication will focus on simultaneous measurements of water isotopes in both vapor and ice residual in mixed-phase clouds. The second instrument paper will focus on the inlet's technology. **Broader Impacts:** Two graduate students receive training via this grant and participated in the first field campaign. Two peer-reviewed papers in preparation. One has been submitted to JGR.

PI: Dr. Hallar, Co-PI: Dr. Lin, Award #:1851943, "REU Site: Inclusive Research Experience in ALpine Meteorology (REALM)", $368,992, 8/1/19- 7/31/22.

Intellectual Merit: Research Experience in ALpine Meteorology (REALM) will rely on the natural scientific laboratory, provided by the nearby Wasatch Mountains and adjacent urban areas, to enhance student awareness of societal challenges, such as water availability and air quality, that require understanding the influence of alpine terrain on weather and climate processes. The Department of Atmospheric Sciences at the University of Utah is recognized internationally for its expertise in atmospheric studies related to mountain environments. **Broader Impacts:** We received 143 applications for the first year of REALM, and applications closed on February 14, 2020.

PI: Dr. Jaffe, Award #:1749273, "Influence of Free Tropospheric Ozone and Particulate Matter on Surface Air Quality in the Western U.S.", $923,449, 3/15/15- 2/28/18.

Intellectual Merit: Here we summarize key findings from this project with reference to peer reviewed publications: 1) We identified a significant increase in summertime PM2.5 in the western U.S. associated with the increasing wildfire activity (McClure and Jaffe, 2018), 2) we found relatively modest impacts on photolysis rates even in significant wildfire smoke, with PM2.5 concentrations of 40 $\mu g\ m^{-3}$ (Baylon et al., 2018), 3) we evaluated a new method to measure peroxyacetyl nitrate (PAN) using a non-radioactive gas chromatography method (Zhang et al., 2018), 4) we identified and quantified the role of meteorological variations in controlling high O_3 seen in 2015 across large parts of the western U.S. (Jaffe and Zhang, 2017), 5) we compared a UV and chemiluminescence based O_3 measurements in strong wildfire plumes and found no significant bias in either method (Gao and Jaffe, 2017), 6) we identified significant positive trend in summertime O_3 and aerosol scattering based on MBO data from 2004-2015 and a positive trend in springtime O_3. The summer trends are due to recent increases in regional wildfire emissions (Zhang and Jaffe, 2017), 7) In spring of 2015, we identified a major Long-Range Transport event (using both data from MBO and the NOAA-SONGNEX aircraft campaign, associated with large wildfires burning in Siberia, mixed with dust and UTLS air (Baylon et al., 2017), 8) Laing et al. (2017) showed that the ΔPM2.5/ΔCO enhancement ratio is a robust indicator of wildfire smoke in urban areas, 9) Using data from MBO, we examined the relationship between aerosol size distributions, scattering and absorption enhancement ratios (to CO) in wildfire plumes and found a significant difference in the $\Delta\sigma ap/\Delta CO$ enhancement ratio between regional plumes and biomass burning plumes transported from Siberia (Laing et al., 2016), 10) Baylon et al. (2016) demonstrated the importance of inter-annual variations in background O_3 on air quality at sites in the western U.S., 11) McClure et al. (2016) examined three years of CO_2 data at MBO and found that free tropospheric air masses have higher average CO_2 concentrations compared to boundary layer air, due to surface uptake. This surface uptake can complicate the calculation of CO_2 enhancement ratios when an airmass is transported in the boundary layer, 12) We identified a wide range in wildfire enhancement ratios (e.g., O_3/CO, PM/CO, NOy/CO) and some of the controlling factors on these (Briggs et al., 2016; Baylon et al., 2015) based on MBO data from 2012-2015. **Broader Impacts:** Since its inception in 2004 there have been over 50 peer reviewed publications using MBO data and these have been cited more than 1500 times (excludes self cites). MBO data have also been collected by or used by scientists from 36 different institutions, including 17 U.S. universities, 8 U.S. government laboratories, 6 international universities and 5 private companies.

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