Title

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Journal

Bulletin of the American Meteorological Society, 0(10)

ISSN

0003-0007

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Publication Date

2021-06-25

DOI

10.1175/bams-d-20-0017.1

Peer reviewed

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May 23, 2021

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Early Online Release: This preliminary version has been accepted for publication in *Bulletin of the American Meteorological Society*, may be fully cited, and has been assigned DOI 10.1175/BAMS-D-20-0017.1. The final typeset copyedited article will replace the EOR at the above DOI when it is published.

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Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

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Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

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Abstract:

Wintertime episodes of high aerosol concentrations occur frequently in urban and agricultural basins and valleys worldwide. These episodes often arise following development of persistent cold-air pools (PCAPs) that limit mixing and modify chemistry. While field campaigns targeting either basin meteorology or wintertime pollution chemistry have been conducted, coupling between interconnected chemical and meteorological processes remains an insufficiently studied research area. Gaps in understanding the coupled chemical-meteorological interactions that drive high pollution events make identification of the most effective air-basin specific emission control strategies challenging. To address this, a September 2019 workshop occurred with the goal of planning a future research campaign to investigate air quality in Western U.S. basins.

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

Approximately 120 people participated, representing 50 institutions and 5 countries. Workshop participants outlined the rationale and design for a comprehensive wintertime study that would couple atmospheric chemistry and boundary-layer and complex-terrain meteorology within western U.S. basins. Participants concluded the study should focus on two regions with contrasting aerosol chemistry: three populated valleys within Utah (Salt Lake, Utah, and Cache Valleys) and the San Joaquin Valley in California. This paper describes the scientific rationale for a campaign that will acquire chemical and meteorological datasets using airborne platforms with extensive range, coupled to surface-based measurements focusing on sampling within the near-surface boundary layer, and transport and mixing processes within this layer, with high vertical resolution at a number of representative sites. No prior wintertime basin-focused campaign has provided the breadth of observations necessary to characterize the meteorological-chemical linkages outlined here, nor to validate complex processes within coupled atmosphere-chemistry models.

1. Introduction:

Winter episodes of high aerosol concentrations occur frequently in urban and agricultural basins and valleys across the globe (e.g., Yamuna Basin, India (Tiwari and Kulshrestha, 2019); Tokyo Basin, Japan (Osada et al. 2019); Taiyuan Basin, China (Miao et al. 2018); and the San Joaquin and Salt Lake Basins, USA (Whiteman et al. 2014; Zhang et al. 2020)). These episodes may last from several days to several weeks and often arise due to the development of persistent cold-air pools (PCAPs), within which lateral and vertical mixing are inhibited due to sheltering by

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surrounding topography and a stable temperature profile (Dorninger et al. 2011; Reeves et al. 2011; Lareau et al. 2013; Sheridan et al. 2014; Holmes et al. 2015; Sun and Holmes, 2019; Ivey et al. 2019; Sun et al. 2020). While a number of field campaigns targeting either wintertime basin meteorology (e.g., Lareau et al. 2013; McCaffrey et al. 2019), or wintertime pollution chemistry (e.g., Brown et al. 2013; Franchin et al. 2018; Young et al., 2016) have been conducted in the U.S., only a few of these campaigns have explicitly considered coupling between interconnected chemical and meteorological processes (e.g., e.g., Baasandorj et al. 2017; Prabhakar et al., 2017; Salvador et al. 2021). The upcoming ALaskan Pollution And Chemical Analysis (ALPACA) is specifically targeting this knowledge gap in cold and dark conditions.

Current gaps in our understanding of the coupled chemical-meteorological interactions that result in high pollution events in many basins worldwide make identification of the most effective air-basin specific emission control strategies challenging. Meteorological processes (thermodynamic, radiative, and dynamical) influence both pollution accumulation, dispersion, and transport, and aerosol pollution chemistry; while chemical processes in turn influence radiative transfer, cloud formation, and mixing processes. Figure 1 presents a graphical illustration of some of the coupled chemical-meteorological processes that occur in basins. Some key meteorological processes that control the formation, duration, and breakdown of PCAPs include: synoptic drivers such as high pressure and associated subsidence, which can precipitate elevated thermal inversions; warm air advection aloft and large-scale winds and turbulent mixing, alongside local drivers such as the surface energy and radiation budget, which strongly influence formation and dissipation of surface-based thermal inversions; characteristics of the underlying surface (e.g., snow cover, water, urban or non-urban landscape); low clouds and fog; and local boundary-layer flows near the surface. In turn, the location and types of urban

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emissions, aerosol formation and growth processes, and chemical cycling processes influence and are influenced by the ambient meteorology (Fig. 1). The unique basin topography (e.g., slope, how enclosed the basin is, and the size of basin) also play an important role in modulating the rate of pollutant build-up and vertical profiles of temperature and moisture. The interactions between these numerous meteorological processes regulating the frequency, location, and speed of chemical processes in PCAPs, and complex wintertime chemistry, has not yet been observed with sufficient detail to provide satisfactory understanding of these complex pollution episodes and their evolution in time and space

Ammonium nitrate (NH_4NO_3) is a major component of aerosol mass in many polluted boundary layers during wintertime episodes (e.g. Womack et al. 2019; Fu et al., 2020; Kelly et al., 2018; Kim et al., 2014; Aksoyoglu et al, 2017; Schaap et al., 2004). Organic aerosol (OA) can also contribute substantially (Chen et al., 2018). In some basins, the NH_4NO_3 contribution increases with the total aerosol loading while in others OA dominates at high loadings, often with differences between day and night. Understanding the factors that govern the evolution of pollution within a basin is critical for implementing effective emission control policies. Precursor emissions and chemical transformations leading to NH_4NO_3 formation are an area of ongoing research, as are a number of processes controlling OA production and loss. While OA formation has been extensively studied in warm seasons with large emissions of volatile organic compounds (VOCs, precursors to secondary organic aerosol) often from biogenic sources, characterization for winter urban environments is lacking. Slower photochemistry occurs in the winter, and biogenic and evaporative emissions tend to decrease with temperature, suggesting that these emissions play a smaller role in wintertime OA formation. However, recent evidence of rapid and widespread OA formation (Shah et al. 2019; Schroder et al. 2018) indicate that

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

urban wintertime OA formation is an important contributor to $PM_{2.5}$ mass. Sulfate is a small fraction of $PM_{2.5}$ mass during winter in basins such as the Salt Lake Valley but is more important elsewhere (Wang et al., 2016). Mechanisms leading to winter sulfate oxidation are a topic of current interest for which detailed studies in the western U.S. may serve as a test bed. Consequently, it is imperative to better understand the mechanisms that drive local nitrate, organic and sulfate formation, alongside the emission sources of key precursor gases—e.g., nitrogen oxides, ammonia, SO_2 and VOCs. This is particularly important given recent findings showing the importance of urban VOC emissions from evaporative sources relative to those from

{Figure 1 here}

fuel use (McDonald et al. 2018). Process-level understanding also requires investigation of radical cycling involving VOCs and NO_x in the winter and its relationship to high pollutant levels in stagnant boundary layers.

In recent decades, overall aerosol concentrations have declined in the U.S. owing to emissions changes driven by regulatory policies (Bennett et al. 2019). However, a recent study has conclusively demonstrated that particulate levels across the U.S. are associated with mortality impacts and loss of life expectancy, with the highest rates observed within the San Joaquin Valley in California (Bennett et al., 2019). Furthermore, particulate matter with diameters less than 2.5 microns ($PM_{2.5}$) within major urban areas across the U.S. has declined more slowly in winter than in summer, and average winter levels are now higher in winter than in summer (Chan et al. 2019). These trends are particularly pronounced in basins across the Western U.S., where wintertime aerosol concentrations regularly approach or exceed regulatory standards (Green et al. 2015). Gaps in understanding the coupled chemical-meteorological interactions that result in high pollution events may preclude identifying the most effective

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emission control strategy for a given air basin. With this challenge at the forefront, a workshop took place in September 2019 at the University of Utah with the goal of planning a future winter research campaign to investigate mountain basins of the western U.S. With funding from the National Science Foundation (NSF) Atmospheric Chemistry Program and the National Oceanic and Atmospheric Administration (NOAA) Atmospheric Chemistry, Climate and Carbon Cycle Program, the workshop brought together ~120 air quality experts and meteorologists from across the globe, representing 50 institutions and 5 countries.

As summarized in this article, the workshop outlined the rationale and design for a comprehensive study that couples atmospheric chemistry and meteorology for wintertime poor air quality episodes in mountain basins across the western U.S. The campaign framework is laid out in Section 2. The existing uncertainties and opportunities for this campaign are summarized in Sections 3-7, organized by science sub-themes. Finally, an integrated perspective on measurements and modeling is presented, along with next steps, in Section 8.

2. Design of the Research Study

The two western U.S. regions with the most severe winter aerosol pollution are Northern Utah (comprised of the Salt Lake, Utah and Cache Valleys) and the San Joaquin Valley (SJV) in California. The workshop concluded that while winter meteorology and a mix of urban and rural emissions affect basins across the western U.S. (see Figure 2), a comprehensive study should focus on these two regions, which exhibit contrasting aerosol chemistry, basin topography, climate, agricultural practices and meteorological-chemical interactions. In the Salt Lake City area, ammonium nitrate is the major contributor to wintertime PM, whereas in the SJV primary

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and secondary organic aerosol (SOA) are also major contributors (Baasandorj et al. 2017; Lurmann et al. 2006; McDuffie et al. 2019; Chow and Watson, 2002). Furthermore, historical databases and previous studies provide context for an air quality study in both of these regions, as PM regulatory standard exceedances in wintertime are common to both. Finally, strong science capacity is available in these areas to conduct this research. Figure 2 shows a long-term record of $PM_{2.5}$ at the Hawthorne monitoring site in Salt Lake City, the SJV (Bakersfield), and, for comparison, a site in California's South Coast basin (Riverside-Rubidoux). The decreasing trend in $PM_{2.5}$ in the South Coast basin is visually apparent from Figure 2, in contrast to the SJV and Salt Lake City sites, which still regularly experience regulatory exceedances during winter, despite modestly decreasing $PM_{2.5}$ trends (Green 2015).

{Figure 2 here}

The onset and evolution of wintertime pollution episodes in the SJV of California differs in many respects from those in the colder, smaller and frequently snow-covered Intermountain West basins. Additionally, western U.S. mountain valleys show significant variability in emissions. Emissions of nitrogen oxides (NO_x), VOCs, and ammonia (NH_3) are highly dependent on the nature of the agricultural and industrial sectors in each valley (Kelly et al. 2013; Wang et al. 2015), and their chemical transformations are dependent on many meteorological and topographical factors (Green et al. 2015; Wang et al., 2015; Kleeman et al. 2005; Pusede et al. 2016).

The proposed field program focuses on understanding how variations in coupled meteorological and chemical processes contribute to the production, transformation, cycling, and destruction of chemical species in each locale. This paper highlights the need for acquisition of chemically and meteorologically comprehensive datasets using airborne platforms with sufficient

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

range, coupled to extensive surface-based measurements that provide continuous, chemically detailed data at ground level where human exposure occurs. Sampling of the boundary layer at high vertical resolution will provide representative profiles that reflect regional topographical, meteorological, and emissions variability. The chemical measurements will be combined with comprehensive meteorological measurements to characterize the influences of a wide range of meteorological and land surface processes and parameters (e.g, transport and mixing, surface albedo) and topography on chemical processes during the PCAP episode.

3. Understanding the Coupling between "Cold-Air Pool" Meteorology and Air Quality

The complex cold-air pool basin meteorology that impacts pollutant dispersion and air pollution chemistry remains an active area of research worldwide (Giovannini et al. 2021). Many of the meteorological processes and their effects on air pollution transport and chemistry are still not well documented, understood, or adequately modelled despite having contributed to poor air quality in western U.S. basins for over a century (Fig. 1: see Lareau et al. 2013; Giovannini et al. 2021; Lighthall and Capitman, 2007; Mitchell, *in prep*). Figure 1 illustrates the coupling of chemical and meteorological processes wherein the atmospheric state impacts the atmospheric chemistry and, in some cases, results in feedbacks between the two (e.g., aerosols affect cloud properties and lifetime and can absorb shortwave solar radiation, which leads to alteration of the vertical temperature profile and hence pollutant vertical transport). Transport and mixing processes, insolation, and microphysical cloud processes all affect the type and extent of different chemical processes. In turn, weaker feedbacks between aerosol loading and cloud chemistry can have significant impact - via radiative feedbacks (e.g., longwave energy transfer

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252 back to the surface from low clouds, net decreases in incoming shortwave radiation into the
253 PCAP due to reflection of the shortwave solar energy back to space at cloud top) - on the vertical
254 stratification and thus vertical mixing processes within the PCAPs.
255 The complexity of interactions between topographical and other physical characteristics
256 of basins, meteorological, and chemical processes introduced in Fig. 1 leads to pollution with
257 characteristics, severity, and duration differing both in time and space and across different
258 basins. For example, variations in the vertical temperature profile and magnitude of turbulence
259 and mixing from solar heating or wind shear can dramatically impact the amount of
260 entrainment/dilution of pollutants and vertical layering of pollutant precursors within the basin
261 atmosphere.
262 The workshop identified four critical couplings processes between meteorology and
263 chemistry requiring investigation as part of the proposed field program : 1) surface fluxes of
264 energy and momentum and chemistry; 2) moisture and fog, and heterogeneous, multi-phase and
265 aqueous-phase chemistry; 3) PCAP vertical thermodynamic profiles and vertical chemical
266 profiles, and 4) meteorology and chemistry associated with thermally and dynamically-forced
267 exchange processes. No prior wintertime field campaign has provided the breadth of
268 observations necessary to analyze the meteorological-chemical linkages outlined here or to
269 validate complex processes within coupled atmosphere-chemistry models.
270 A critical tool for both basic research and air pollution control strategies are
271 coupled meteorological and chemical models. Development of useful guidance for the research
272 and regulatory communities requires improvements to both meteorological and chemical
273 components of these models (Giovanni et al. 2021). To improve meteorological model
274 simulations of PCAPs, considerable work is underway or has been conducted in recent years

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

(e.g., Saide et al. 2011, Lareau and Horel 2015b, Ahmadov et al. 2015, Saide et al. 2016, Tran et al. 2018, Sun and Holmes 2019; Kelly et al., 2018; Sun et al. 2020). Some of the key meteorological processes that are very difficult to model in stable wintertime boundary layers include vertical temperature and humidity structure, cloudiness, turbulent mixing and boundary-layer flows (Baklanov et al., 2011, Holmes et al., 2015). Improvements in and testing of the representations of heterogeneous chemical processes (Holmes et al., 2019; Brown et al., 2006b), links between aerosol phase and gas-particle equilibration and partitioning (Shiraiwa et al., 2013b, Zaveri et al., 2018), secondary organic aerosol lifecycles (Cappa et al., 2016) and organic compound oxidation pathways (Bianchi et al., 2019), especially as they occur within colder wintertime environments, is needed to accurately understand and predict both aerosol and gas-phase abundance and composition, discussed further later on.

The coupled meteorological and chemical models parameterize vertical mixing using relationships between turbulent fluxes and mean thermodynamic profiles that have significant impacts on predicted atmospheric composition. It is therefore paramount that meteorological (e.g., temperature and wind speeds to quantify mixing and transport, relative humidity and downwelling radiance to estimate liquid water path and boundary-layer moisture) and chemical vertical profile measurements be collocated with observations of the radiative and turbulent components of the surface energy balance. This approach will allow a detailed quantification of mass, moisture, heat, and chemical budgets within basins. It is also important that variations in the surface energy balance across the basins be well captured using flux sites and satellite data. Important land surface parameters to observe include the depth and age of snow (which can impact the amount of reflected solar radiation and rates of photochemical reactions), and soil

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297 temperature and moisture (which can impact fog formation and vertical mixing processes, both
298 which impact chemistry).

299 Sufficient data, both temporally and spatially, are needed to ensure that spatial gradients
300 are captured across the basins of interest to form a three-dimensional representation of the
301 atmospheric state for both chemical and meteorological properties. Chemical processes driven by
302 mixing also need to be resolved alongside the meteorological measurements for turbulence,
303 mixing, and transport, to allow linking of these two processes. A holistic, interdisciplinary, and
304 multi-agency approach will be used in this study where existing infrastructure such as National
305 Weather Service daily rawinsonde launches, weather stations from public and private sectors
306 available from MesoWest (Horel et al. 2002), wind sodars, lidar profilers, and ceilometers will
307 be supplemented with instrumentation dedicated to this field study.

308 One study design approach is a process-focused deployment, targeting regions of interest
309 within a basin to investigate and quantify coupled chemical-meteorological processes. Three
310 examples of targeted meteorological processes and their impacts on the chemistry include: 1)
311 inter-basin exchange, 2) sidewall- and canyon-flow transport, and 3) vertical layering and
312 exchange (Fig. 1).

313 In addition to using frequent rawinsonde launches and vertical-profiling wind lidars, sodars,
314 ceilometers, and acoustic-sounding systems at fixed locations, use of mobile vertical profilers
315 and tall buildings as instrument platforms can help resolve the vertical structure of the basin
316 atmosphere. Mobile ground-based systems measuring both meteorology and chemistry, such as
317 the California Air Resources Board (CARB) Mobile Measurement Platform (Park et al. 2011), or
318 the TRAX air quality light-rail train (Mitchell et al. 2018; Mendoza et al. 2019), will fill gaps in
319 the fixed-instrumentation networks. Mobile ground-based systems driven up canyons and slopes

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

can provide measurements along topographic sidewalls, and drones can obtain shallow but highly resolved vertical profiles in areas with limited topography. Coupled meteorological data (including turbulence and the surface energy balance) collected on towers and high buildings at multiple locations within the basins would also provide valuable observations in the lowest 100 m of the PCAP atmosphere. A potential exists for deploying airborne Doppler wind lidar to provide similar vertical wind-flow information along a 2-D transect, during times and in locations where fog and low stratus do not impede operations. Lidar-based vertical wind profiles at basin boundaries would greatly assist in quantifying background pollution and meteorological parameters as well as transport processes within a single basin and between adjacent basins.

The field deployment will also include modeling teams, using tools that span the scales from large eddy simulations (meters resolution) to regional coupled meteorological and chemical models (kilometers resolution), to forecast PCAP events and contribute to flight planning; this will help ensure the measurements address the modelers' needs. Complementing research-grade models and forecasts, operational air quality forecasts used to warn the public about pollution events will be evaluated to identify deficiencies and accelerate the transition of any improvements from research to operations.

4. Emissions of Relevant Short-Lived Pollutants

Within basins that experience adverse air quality in winter, direct emissions of aerosols are of interest, as well as emissions of short-lived gases, including reactive nitrogen species (e.g., NO_x and NH_3) and intermediate-volatility/volatile organic compounds (I/VOCs; e.g., hydrocarbons, amines, oxidized and reduced sulfur compounds, and oxygenated organic

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

compounds). Such emissions are largely from urban, agriculture, and biomass burning sources. Here, residential wood combustion is considered an urban emissions source, and biomass burning is limited to wildland fires (wildfires and prescribed burns). Different sources emit at different elevations above the surface, with consequent implications for the fate of emitted compounds and their impacts on surface air quality. For example, emissions from vehicles occur at the surface, whereas emissions from some industrial sources or the power sector may occur at elevations 10's to 100's of meters above the surface.

Mobile sources and residential wood combustion contribute significantly to primary and secondary aerosols in Salt Lake City in wintertime (e.g., Kelly et al. 2013). Similar sources contribute to elevated aerosol concentrations in the SJV; in addition to local transportation and residential wood combustion, cooking was identified as significant source of aerosols, particularly in the evening (Young et al. 2016). While emissions reductions from passenger vehicles have led to a continuing decline in on-road vehicle contributions to NO_x and aerosols (Dallman and Harley 2010; Bishop et al. 2012; Utah DAQ 2019), the contributions of older and off-road vehicles to NOx and aerosols are not well quantified. In addition, the temperature dependence of mobile source emissions is not well-constrained for real-world conditions. Previous in-use laboratory testing has shown that lower ambient temperatures can lead to higher gaseous and particulate mass emissions from gasoline and diesel vehicles, and to changes in chemical composition (Zielinska et al. 2004). In addition, vehicle operation specific to winter conditions, such as changes in the frequency of hot vs. cold starts and an increase in idling, can affect mobile source emissions and ambient concentrations. Finally, NH_3 emissions from selective catalytic reduction systems may increase in winter, given the increase in NH_3-forming deposits with decreases in ambient temperature (Strots et al. 2010).

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

Reductions in mobile source I/VOC emissions spanning several decades (e.g., Warneke et al. 2012) have led to an increase in the relative importance of other, under-studied sources of I/VOCs including cooking and volatile chemical products (VCPs) (McDonald et al. 2018). A large fraction of VCP emissions occur inside buildings. More restrictive air exchange between buildings and ambient air during the winter to conserve energy may affect VCP emissions (Pagonis et al. 2019); currently, there are few constraints regarding the seasonality in VCP emissions. Evaporative emissions of transportation fuels generally decrease as ambient temperature decreases (Harley et al. 1992; Rubin et al. 2006), which also may be observed for VCPs.

Regarding residential wood burning, in areas affected by poor wintertime air quality, efforts have been made to reduce solid fuel combustion (wood, wood pellets, coal, etc.) through implementation of burn restrictions when poor air quality is forecast, and incentives to exchange wood-burning devices for natural gas heating options. Such strategies appear to be reducing the prevalence of solid-fuel burning, including wood (Kotchenruther, 2020); however it has been demonstrated that primary and secondary aerosols originating from residential wood burning persist, even when solid-fuel burning is prohibited (Glisson et al. 2019; Kotchenruther, 2020). In addition, solid fuels used in wood stoves are poor in nitrogen, which affects the emissions of nitrogen-containing VOCs, NOx, and NH_3 (Coggon et al. 2016).

Agricultural emissions have been directly linked to atmospheric aerosol formation (Bauer et al. 2016), mainly via the dominant contribution of agriculture to NH_3 emissions both globally and in the study region (Bouwman et al., 1997; Paulot et al., 2014). Uncertainties in quantifying the contribution of NH_3 to aerosols in winter are largely associated with parameterizations of emissions as a function of temperature. NH_3 emissions from manure vary

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

non-linearly with temperature, with higher temperatures increasing volatility (e.g., Hempel et. al 2016). For animals in heated housing, there is concern that process-based NH_3 predictions for ambient temperature may underpredict winter emissions (Moravek et al. 2019). In addition to NH_3, several recent studies have pointed to the importance of fertilized agricultural soils as a source of NO_x (Almaraz et al. 2018; Trousdell et al. 2019), which could have a particularly large impact in California where crop fertilization occurs in the winter months.

A number of individual agricultural VOCs have been identified, and include aliphatic and aromatic hydrocarbons, phenols, alcohols, aldehydes, ketones, esters, carboxylic acids, terpenes, heterocyclic compounds, amines, and reduced sulfur compounds (e.g., Blunden et. al. 2005; Filipy et al. 2006; Shaw et al. 2007; Turan et al. 2007; Trabue et al. 2008; Rumsey et al. 2012; Rumsey and Aneja, 2014). While past studies suggested that, relative to other anthropogenic sources, agricultural VOCs do not contribute significantly to O_3 production, recent studies indicate their role has been overlooked (Pusede et al., 2014; Parrish et al., 2017; Trousdell et al., 2019). Several of these identified compound classes are known aerosol precursors, particularly amines and reduced sulfur compounds. Amines have been reported as a significant component of the carbonaceous fraction of aerosol observed during cold wintertime inversion conditions in western agricultural valleys, in Utah (Silva et. al 2008), and Washington (Bottenus, et. al 2018). Reduced sulfur compounds (e.g., dimethylsulfide, dimethyldisulfide, and methanethiol) have received less attention (Trabue et al. 2008; Rumsey et al. 2014), but agricultural contributions of such compounds can be significant (Shaw et al. 2007).

In certain states and regions, there may be unique agricultural sectors unaccounted for in emissions inventories and air quality models. For example, the state of Utah is the number two producer of mink pelts in the U.S., but such emissions are lacking in the National Emissions

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

Inventory because there are no data available. Also, rapid changes in agricultural source emissions may occur owing to the recent expansion of anaerobic digesters. California alone has dozens of digesters under construction for the dairy industry and they are being installed in other states as well. Few studies have been conducted to see how the digester processes might impact non-GHG emissions.

Wildland fire emissions likely contribute to the elevated $PM_{2.5}$ concentrations observed during the winter months. Fire activity in winter has become more prominent in the western U.S. For example, the Thomas Fire in California, which started in early December, became one of the largest wildfires recorded in California (Kolden and Abatzoglou, 2018). Higher summer temperatures and decreased precipitation in the fall could potentially continue to extend the western United States fire season into the winter (Guzman-Morales and Gershunov, 2019). Transported emissions from these late fall/early winter wildfires in the western US could then contribute to enhanced $PM_{2.5}$ concentrations in the SLC region.

5. Winter Atmospheric Chemical Cycles

Chemical transformations and subsequent phase partitioning of primary air pollutants such as NO_x, VOCs, and reduced nitrogen are responsible for the conversion of these primary emissions into aerosols. While the daily integrated solar radiation available to drive winter photochemistry is generally lower than summer, multiphase chemical reactions that are prevalent in winter can lead to photolabile radical sources that drive aerosol formation. The nature and magnitude of radical sources impacts the sensitivity of chemical oxidation cycles to primary emissions of NO_x and VOCs, leading to fundamental shifts in the prevalent chemical regime

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

435 during winter. The shallow, stable boundary layer associated with PCAPs tends to concentrate
436 primary emissions, further shifting the sensitivities of oxidation cycles. This section examines
437 some of the features of winter chemical oxidation cycles in polluted boundary layers and
438 identifies major uncertainties.

439 Snow cover is conducive to the formation and intensification of PCAP conditions due to
440 enhanced radiative cooling at the snow surface that strengthens the low-level wintertime stable
441 layer. In the SLV, for example, $PM_{2.5}$ exceedances are observed four times as frequently on
442 snow covered days compared to those without snow cover (Whiteman et al. 2014). The effect of
443 snow cover is complex, since it changes both the boundary layer dynamics and atmospheric
444 chemistry. Reflection of solar radiation by snow increases actinic flux and photochemical
445 reaction rates. Surface snowpacks also directly impact atmospheric composition by serving as a
446 sink/reservoir for atmospheric trace gases and particles upon deposition and as a source from
447 reactions on and within the snow grain surface (Grannas et al. 2007). For example, nitrate
448 deposited on the snowpack undergoes photolysis to produce NO_x and HONO released to the air
449 above (Chen et al. 2019; Honrath et al. 2000; Michoud et al. 2015; Zatko et al. 2016). Snowpack
450 photochemistry also contributes to near-surface OH through the production and subsequent
451 photolysis of H_2O_2 and carbonyls, including formaldehyde, acetaldehyde, and acetone (Couch et
452 al. 2000). Dinitrogen pentoxide (N_2O_5) reactions on saline snow grains can result in $ClNO_2$
453 formation, with the snowpack serving as a net source or sink of $ClNO_2$ depending on temperature
454 (Wang et al. 2020).

455 The presence of large surface area in the particulate phase and in fogs also increases
456 heterogeneous reaction rates during winter, altering levels of several criteria air pollutants
457 including ozone (O_3), nitrogen dioxide (NO_2), SO_2 and particulate matter (Sarwar et al. 2012;

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

458 Sarwar et al. 2014). One important heterogeneous reaction known to contribute to accumulation
459 of aerosol mass is that of N_2O_5 on deliquesced aerosol particles. N_2O_5 chemistry occurs primarily
460 during darkness, and is favored at lower temperatures and elevated NO_x levels, which are
461 characteristic of urban areas during winter (Chang et al. 2011). This reaction regulates the
462 wintertime NO_x lifetime and leads to the formation of nitric acid (HNO_3) (Chang et al. 2011),
463 which contributes to particulate nitrate levels in the presence of ammonia (McDuffie et al. 2019).
464 Nitryl chloride ($ClNO_2$), a photolytic source of chlorine radicals ($Cl^{\cdot}$) and NO_2, is produced from
465 N_2O_5 where chloride-containing particles are present (Osthoff et al, 2008). Atomic chlorine is
466 highly reactive and initiates VOC oxidation cycles and SOA formation (Wang and Hildebrandt
467 Ruiz, 2018). The fate of the reactive uptake of N_2O_5 and subsequent impacts on oxidant and PM
468 concentrations depend on individual particle surface composition (Gaston and Thornton 2016;
469 McNamara et al. 2020), which remains poorly constrained.
470 A second important wintertime heterogeneous and multiphase reaction is the production
471 of nitrous acid (HONO), a radical reservoir whose photolysis may be a large OH source that
472 affects urban air quality (e.g., Fu et al. 2019). Numerous heterogeneous and multiphase HONO
473 sources have been proposed, such as photochemical generation from snowpacks (Chen et al.
474 2019) and photolysis of nitrate aerosol (Ye et al. 2017), although the latter remains controversial
475 (Romer et al. 2018). Given the variety of potential HONO sources and associated uncertainties,
476 future measurements are needed to ascertain dominant formation pathways.
477 Dissolution of trace gases and subsequent aqueous-phase reactions within fog droplets
478 and deliquesced aerosol can also lead to secondary aerosol formation, including sulfate and high
479 molecular weight organic compounds (Ervens et al. 2011). Previous SJV wintertime studies have
480 shown aqueous-phase formation of high molecular weight organic compounds (Qin and Prather

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

2006), as well as hydroxymethanesulfoante (Whiteaker and Prather 2003). This compound has been suggested as a significant winter sulfate source in highly polluted regions such as China (Song et al. 2019). Aqueous phase sulfur oxidation driven by NO_2 at high aerosol pH has recently been proposed to explain rapid sulfate formation in China (Wang et al., 2011). Fog pH has been increasing in recent years within the SJV, where ammonium nitrate is a major component in the north and sulfate is prevalent in the south (Herckes et al. 2015), providing a potential test of this and other known and proposed aqueous sulfur oxidation mechanisms (Alexander et al., 2009; Calvert et al., 1985; Green et al., 2019). Studies of the multiphase processes remain too sparse to accurately parameterize their contribution to winter air quality.

O_3 and NO_2 are often collectively termed odd oxygen (Ox) and tracked together to determine the growth of O_3 beyond its rapid interconversion with NO_2 (Wood et al. 2009). Odd oxygen is frequently defined more broadly to include other nighttime reservoirs such as NO_3, N_2O_5 and gas phase HNO_3 (Brown et al. 2006c; Liu, 1977). Recent work has expanded the definition of O_x further (referred to as $O_{x,total}$) to include additional reactive nitrogen compounds and particulate nitrate (Womack et al. 2019). Tracking $O_{x,total}$ allows for the investigation of the role that oxidation plays in the buildup of pollutants in boundary layers. During the Utah Winter Fine Particulate Study (UWFPS) (Baasandorj et al. 2018), $O_{x,total}$ increased dramatically above the 45 ppbv O_3 background during pollution episodes and rapidly became dominated by NO_2 and particulate nitrate under conditions of depleted O_3. Box modeling demonstrated that the high ratio of NO_x to VOCs in the SLV altered the traditional O_3 photochemical cycle (Kleinman 1994; Lin et al. 1988) to primarily form particulate nitrate (Womack et al. 2019). This finding contrasts with the adjacent Uintah Basin, UT, where high concentrations of VOCs from oil and gas extraction with relatively lower NO_x levels have been shown to force the photochemical

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

cycle to produce $O_{x,total}$ in the form of wintertime O_3 (Ahmadov et al. 2015; Edwards et al. 2013; Edwards et al. 2014). These two analyses of air pollution in adjacent basins, SLV and Uintah, demonstrated the important role of photochemistry during winter and the utility of $O_{x,total}$ as a parameter for evaluating its role in the generation of both O_3 and particulate nitrate pollution. The contrast between the two basins shows that particulate nitrate and O_3 are closely coupled and can be regarded as chemically identical (Meng et al. 1997). Modeling techniques that have long been used for evaluating mitigation strategies for ozone can also be used for particulate nitrate (Nguyen and Dabdub 2002; Pun et al. 2009). In the wintertime SLV, a NO_x-VOC sensitivity isopleth demonstrated that the formation of $O_{x,total}$ is most sensitive to reductions in VOCs, and is NO_x-saturated, meaning that reductions in NO_x emissions would initially increase aerosol pollution (Womack et al. 2019), a result that is counterintuitive since NO_x is also the precursor to particulate nitrate, the dominant component of $PM_{2.5}$ in SLV. However, the NO_x-VOC relationship for particulate nitrate must be understood in detail in the context of the competing processes that govern particulate nitrate formation and chemical and meteorologically driven loss, which remain incompletely understood.

6. Processes Governing Particulate Matter Formation and Loss

The formation and loss of atmospheric aerosols is tightly linked to the partitioning of the chemical constituents between the particle and gas phases, and the multiphase reactions described in Section 5. Observations demonstrate that particulate nitrate and OA dominate wintertime composition in a variety of urban areas within valleys (e.g., Fresno, CA; Salt Lake City, UT; Beijing, China; Po Valley, Italy) (Bressi et al. 2016; Franchin et al. 2018; Lu et al.

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

2019; Young et al. 2016). Analysis of the OA composition indicates oxygenated OA, likely synonymous with SOA, often comprises a large fraction of the OA, along with OA derived from major primary sources, including biomass combustion, vehicles, and cooking (Bressi et al. 2016; Crippa et al. 2013; Lu et al. 2019; Young et al. 2016; Zhang et al. 2007; Paglione et al. 2020). Yet, substantial challenges remain regarding the ability of models to quantitatively predict ambient aerosol concentrations in these wintertime environments, especially of the secondary components (Fountoukis et al. 2016; Kleeman et al. 2019; Schroder et al. 2018). These challenges arise from the incomplete understanding of the physical, chemical, and thermodynamic processes that govern aerosol formation and partitioning, and how these are impacted by wintertime meteorological conditions. In addition, differences in the chemical environment and processes occurring within the nocturnal residual layer versus the surface layers are of particular importance in wintertime owing to shallower nocturnal boundary layers coupled with less available sunlight compared to other seasons (Baasandorj et al. 2017; Prabhakar et al. 2017; Pusede et al. 2016; Wang et al. 2018).

Dry deposition may influence ozone and particle loss rates, but particle deposition velocities in the key submicron particle range are generally small and thus dry deposition will likely only play a considerable role when the nocturnal boundary layer is particularly shallow (Emerson et al., 2020) and the aerodynamic resistance is low (i.e., vigorous turbulent mixing.) Dry deposition of gases such as HNO_3 or oxygenated VOCs can drive evaporation of particulate nitrate and organic aerosol and thus indirectly contribute to loss of $PM_{2.5}$ (Pusede et al., 2016; Knote et al., 2015). For HNO_3, such indirect loss processes are most important in warmer, ammonia-deficient conditions, when HNO_3 comprises a large fraction of the total nitrate, and when the boundary layer is particularly shallow (Prabhakar et al, 2017) and loss of OA will be

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

similarly enhanced in warmer conditions. Direct measurements of dry deposition of such gases can help constrain understanding of the importance of these loss processes (e.g., Nguyen et al., 2015), and further examined through the use of process-level models (e.g., Prabhakar et al, 2017). Ozone dry deposition is highly variable based on the surface characteristics and if not measured directly, it can presumably be parameterized. However, particle loss via scavenging by fog may be important in valley regions when the fog penetrates to the surface (Gilardoni et al., 2014); chemical processing within fog may also play a role in transforming the ambient aerosol composition (Gilardoni et al., 2016). Improved treatment of scavenging by wet deposition that accounts for variability in in-cloud water has been shown to improve model skill at predicting nitric acid and particle phase nitrate under wintertime conditions (Luo et al., 2019). Strong, multi-day pollution events are often ended by a change in synoptic conditions that brings increased ventilation and, potentially, precipitation (Largeron and Staquet, 2016).

One consideration is cold conditions with low absolute humidity but variable relative humidity characteristic of many wintertime valley environments. The limited availability of thermodynamic data for electrolyte solutions—especially mixtures containing organics—at low temperatures means that predictions of gas-particle partitioning and aerosol pH using existing thermodynamic models are extrapolations and may not be accurate. Direct measurements of the gas-particle distribution of inorganic and some organic species (e.g. organic acids) can serve to evaluate model predictions of equilibrium phase partitioning and aerosol pH (Murphy et al. 2017; Guo et al., 2016; Nah et al., 2018). However, the low temperatures may give rise to long equilibration timescales, making assumptions of thermodynamic equilibrium on typical model timescales questionable. Comparison between sufficiently detailed observations and model predictions will allow for investigation of sensitivities to temperature and RH (Evanoski-Cole et

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

al. 2017). The viscosity of particles varies over many orders of magnitude, dependent on the particle composition, ambient temperature, and relative humidity (Renbaum-Wolff et al. 2013). Consequently, particles exhibit material properties, e.g., diffusivities, ranging from liquid-like to solid (Koop et al. 2011), which affects gas-particle partitioning and equilibration timescales (Shiraiwa and Seinfeld 2012; Shiraiwa et al. 2013a), heterogeneous reactions (Gaston et al., 2014; Li et al. 2018; Liu et al. 2018), and particle growth dynamics (Shiraiwa et al. 2013b; Zaveri et al. 2014; Zaveri et al. 2018). Additionally, components within particles can undergo phase-phase separation into an organic-rich and inorganic/aqueous-rich phase, leading to unexpected internal morphologies within individual particles (Song et al. 2012; You et al. 2012). It is unclear how such phenomena impact rates of mass transfer between particles and the rates of aerosol-forming chemical processes, such as N_2O_5 reactive uptake, that occur on and within particles in the cold wintertime conditions.

Observations of N_2O_5 reactive uptake coefficients exhibit complex dependencies on particle OA mass fraction (Bertram et al. 2009), along with the OA chemical composition (e.g., the O:C atomic ratio) and potential for liquid-liquid phase separation (Gaston et al. 2014). The N_2O_5 reactive uptake coefficient may increase with decreasing temperature, perhaps owing to increased solubility of N_2O_5 (Abbatt et al. 2012; Hallquist et al. 2003; Schweitzer et al. 1998; Wagner et al. 2013), but requires further characterization for particle compositions reflective of wintertime valleys.

Particle composition and abundance vary with size, with especially large differences between submicron and supermicron particles. Compositional differences consequently engender size-varying differences in aerosol water content—critical to N_2O_5 uptake and other heterogeneous processes. The importance of considering and characterizing refractory and

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

596 supermicron particles varies between regions. For example, supermicron particle contributions in
597 the wintertime SJV are often small (Parworth et al. 2017), while in the colder SLV it can be
598 substantial as a result of dust, road salt and lake salt (Hrdina et al. 2020). Compared to submicron
599 particles, field observations of supermicron particle concentration, size, and composition are
600 often limited, challenging model-measurement comparisons. Particle composition affects both
601 N_2O_5 uptake and resulting products (HNO_3 versus $ClNO_2$), and often there are differences
602 between laboratory parameterizations and field observations of $ClNO_2$ yields (McDuffie et al.
603 2018). McNamara et al. (2020) recently showed that $ClNO_2$ production, driven by the surface
604 reaction of N_2O_5 (Gaston and Thornton 2016), is explained by surface area-weighted single-
605 particle composition and developed a new parametrization that requires testing in other
606 environments. Notably, the SLV has two unique chloride sources – playa dust and road salt –
607 that likely contribute to $ClNO_2$ (Mitroo et al. 2019; McNamara et al. 2020). Measurements
608 completely characterizing the time and size-varying particle composition in wintertime valley
609 environments are needed to accurately simulate nighttime production of HNO_3 via N_2O_5 uptake.
610 Gas-particle partitioning of HNO_3 and other acids depends on size-varying particle and
611 gas composition. In submicron particles, acid-anion phase partitioning is strongly controlled by
612 NH_3 availability, alongside temperature, RH, and aerosol pH, the latter of which depends on the
613 identity and abundance of other aerosol constituents (e.g., particulate sulfate or organic aerosol)
614 (Murphy et al. 2017). In contrast, HNO_3 can react effectively irreversibly with supermicron dust
615 or sea salt particles, displacing CO_2 (from $CaCO_3$) (Usher et al., 2003) or HCl (from NaCl) (Gard
616 et al. 1998). As dry deposition is much faster for supermicron particles than accumulation mode,
617 uptake to supermicron particles can thus suppress submicron nitrate formation. An

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

618 observationally constrained understanding of the dynamic links between HNO_3, NH_3, and sub-
619 versus supermicron reactive uptake and partitioning is needed.

620 Formation of SOA in valley environments during winter has received relatively little
621 consideration compared to summer, although SOA can comprise a substantial fraction of aerosol
622 dependent upon the conditions. Some observations indicate that the [SOA]/[NO_3^-] ratio, or the
623 [OA]/[NO_3^-] ratio when the former is not available, decreases as the total aerosol loading
624 increases, although the absolute SOA concentration tends to increase with aerosol loading
625 (Bressi et al. 2016; Franchin et al. 2018; Lu et al. 2019; Young et al. 2016). This suggests some
626 link between SOA and nitrate formation, potentially related to the dependence of aerosol liquid
627 water content on particulate nitrate; higher aerosol water content can lead to enhanced uptake
628 and reaction of water-soluble organic compounds--including at night (Hodas et al. 2014; Sullivan
629 et al. 2016). Aqueous processing, especially of VOCs from residential wood combustion, lead to
630 production of high molecular weight (Qin and Prather 2006) and absorbing organic aerosol (i.e.,
631 brown carbon, BrC) (Laskin et al., 2015; Gilardoni et al. 2016), which can influence the local
632 and regional radiative budget (Mohr et al. 2013). Understanding of SOA and BrC formation in
633 wintertime environments is lacking, both within and outside of valleys, with large model-
634 measurement differences in some environments and a strong sensitivity to the particular SOA
635 parameterization used (Chrit et al. 2018; Fountoukis et al. 2016; Meroni et al. 2017; Russell et al.
636 2018; Schroder et al. 2018; Shah et al. 2019). It may be that current parameterizations, developed
637 almost entirely using room-temperature experimental data, are not properly capturing T-
638 dependent changes in both gas- and particle-phase chemical pathways and in compound
639 volatility (Bianchi et al. 2019; Stolzenburg et al. 2018; Ye et al. 2019). Winter SOA formation
640 via reaction of NO_3 radicals with VOCs, especially from sources such as VCPs and residential

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

wood combustion in poorly mixed nighttime boundary layer structures has received relatively little attention (Mohr 2013, Yuan 2016), even though NO_3 chemistry has been proposed as a dominant SOA source during summer in the SJV (Rollins, 2012).

Overall, the identity and contribution of major submicron aerosol constituents in wintertime valleys is well-known, with less known about the supermicron aerosol. Still, gaps in process-level understanding of the formation pathways and diurnally varying partitioning confront the development of clear control strategies and major improvements in wintertime air quality in such regions. Further, particulate matter loss processes, including wet and dry deposition, are poorly constrained by observations, as are the loss processes of PM precursors Targeted, comprehensive measurements that can be compared with models are necessary to close these gaps.

7. Greenhouse Gas (GHG) Emissions

The proposed field study will yield unique insights into wintertime GHG emissions and how they relate to air pollutant and associated precursor emissions in western U.S. basins. Wintertime emissions likely differ from other seasons owing to the presence of heating needs (Gurney et al. 2012; Mitchell et al. 2018). Specifically, this field campaign will link GHG to emission sectors to understand how shifts in these emissions are associated with changes in short-lived pollutants and use GHG emissions as transport tracers to elucidate meteorological processes.

Three GHG emissions sectors will be carefully considered: transportation, agriculture, and oil/gas and energy distribution. These are most relevant in the Western U.S. The on-road

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

transportation sector is one of the largest emitters in urban settings, often accounting for nearly 50% of CO_2 and other pollutants, but is generally the least well-constrained sector within carbon emission inventories, with large uncertainties (Gately and Hutyra, 2017; Mendoza et al. 2013). Agriculture is an important source of GHG and air pollutants in the western U.S., in particular methane (CH_4) and nitrous oxide (N_2O), respectively comprising nearly 40% and 80% of total emissions (U.S. Environmental Protection Agency, 2019). Several studies have now shown that inventories underestimate methane emissions from oil and gas production (Alvarez et al. 2018; Robertson et al. 2017; Lyon et al. 2015). To constrain the contribution of all of these (and other) source types to degradation in air quality, will require monitoring a variety of trace gases.

Changes in vehicle fleets and improvements in technology have reduced mobile-sector emissions of both CO_2 (Gately et al. 2015) and air pollutants (McDonald et al. 2018). For example, NO_x and carbon monoxide (CO) emissions in cities have fallen (Parrish et al. 2011; Hassler et al. 2016), although the decline has slowed recently (Jiang et al. 2018). Also, changes in the U.S. fuel mix (U.S. Energy Information Administration, 2019), may affect emissions of both GHGs and pollutants. These trends point to changes in the urban atmospheric composition that might manifest themselves in observed concentrations. Work in Salt Lake City (Lin et al. 2018), with the longest running urban CO_2 network, indicates that CO_2 enhancements have remained flat in the urban core. This is despite population growth, implying energy efficiency improvements, and increased CO_2 enhancements in the suburbs due to new anthropogenic emissions occurring where previously there were none (Mitchell et al. 2018a). These long-running data sets in Salt Lake City are complemented by a novel platform on the public transit light-rail trains that measure GHGs and air pollutants as the trains traverse the city (Mitchell et al. 2018b; Mendoza et al. 2019). Concentration ratios from this platform have been used to

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

687 fingerprint NO_x emission sources (Mitchell et al. 2018b) and characterize how fine scale
688 meteorological phenomena, such as canyon flows and inter-basin transport, affect air pollution
689 distributions (Baasandorj et al. 2018). The proposed field campaign will utilize GHG and
690 pollutant datasets from aircraft, mobile laboratories and ground sites.
691 The chemical inertness of GHGs such as CO_2 and CH_4 allows for their use as passive
692 tracers of atmospheric transport (Pataki et al. 2005). This is particularly true in the wintertime,
693 when photosynthetic uptake of CO_2 is minimal (Pataki et al. 2003; Strong et al. 2011).
694 Measurements of CO_2 in the SLV have revealed strong relationships between levels of CO_2 and
695 atmospheric stability, with the presence of PCAPs associated with significant enhancements in
696 CO_2 concentrations (Pataki et al. 2005; Bares et al. 2018; Lin et al. 2018). Therefore, if GHG
697 emissions are constrained, levels of GHG can potentially be used to quantify the effects of
698 atmospheric transport processes and separate the contributions of such transport effects from
699 atmospheric chemistry.
700
701 **8. Campaign Design**
702
703 The recent 2017 Utah Winter Fine Particulate Study (UWFPS) in SLV was an aircraft and
704 ground-based campaign utilizing the NOAA Twin Otter, a medium sized research aircraft
705 (Bassandorj et al. 2018). The study demonstrated the capability to conduct in-situ aircraft
706 measurements in shallow winter boundary layers within complex terrain during periodic
707 episodes of low visibility. A clear lesson learned from the UWFPS study was that the
708 complexity of the science governing wintertime air quality is better addressed through a broader
709 suite of instrumentation and focal area than could be accommodated by the payload and range of

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

a Twin Otter. A larger aircraft (e.g., NSF C-130, NOAA P-3) would allow for a more extensive payload providing detailed in situ and remote sensing instrumentation, as well as the ability to sample multiple basins within a domain that could encompass most or all of the western U.S. (see Figure 1).

The workshop considered the optimal combination of both ground-based and airborne measurements and platforms required to address the science questions identified. A recurring theme was the importance of combining co-located, comprehensive meteorological and chemical measurements. The workshop attendees stressed the need for large aircraft to carry the payload required to characterize both the organic and inorganic gases and particle composition and understand their relationships. Participants also expressed the need for airborne and surface measurements of coupled meteorological and chemical processes to bring insight to not only the formation and dissipation of the PCAPs that allow for the build-up of these extreme air quality events, but especially the vertical and horizontal transport and mixing processes modulating the concentrations of pollutants and precursors and constraining chemical processes. Based on the climatology of PCAPs in these regions (Whiteman et al., 2014), the optimum period for an airborne field campaign is from approximately in mid-December early through mid-February. Measurements may not be continuous during this period and would depend on logistical considerations, such as total aircraft hour allocation to the project.

Key measurements that will be needed onboard the aircraft includes standard meteorological parameters along with fast-response measurements of VOCs, most likely using chemical ionization mass spectrometry, the various NO_x and NO_y species (e.g., NO, NO_2, HNO_3, N_2O_5, HONO, NO_3) and other important trace gases (e.g., O_3, SO_2, HCHO, CO) and greenhouse gases (CO_2, CH_4, N_2O), and submicron and supermicron aerosol composition and size

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

733 distributions. A suite of ground-based measurements (in situ and remote sensing) will
734 complement aircraft data to characterize the coupled meteorological-chemical system in three
735 dimensions during the field campaign. These include wind and aerosol lidars, radio acoustic
736 sounders, rawinsonde, ceilometers, and long-path Differential Optical Absorption Spectroscopy
737 (DOAS) to provide continuous observations of vertical profiles (e.g., temperature, wind speed
738 and, trace gases such as NO_2, SO_2, O_3, HONO, HCHO, and NO_3). Specifically, a western U.S.
739 winter air quality study should include comprehensive measurements of major radical sources,
740 including CH_2O and other aldehydes, HONO, $ClNO_2$, and O_3, together with measurements of
741 actinic flux to define photolysis rates and radical sources. Measurements of major radical
742 species, including OH, HO_2 and preferably RO_2, together with nighttime measurements of NO_3
743 radicals, must also be available to constrain diel radical cycles and concentrations and to
744 compare with the process-level predictions. Speciated measurements of these radicals and their
745 precursors are most easily carried out from ground sites. However, measuring a subset of these
746 compounds from a research aircraft allows for extension of the conclusions regionally and as a
747 function of altitude. Comparison of continuous measurements at a single location to vertical and
748 horizontal distributions of radicals and/or radical precursors provides a more complete view of
749 the radical budget.

750 Complete characterization requires fully instrumented aircraft measuring $O_{x,total}$ and its
751 partitioning between nitrogen and oxygen species (O_3, NO_2, NO_3, HNO_3, N_2O_5, $ClNO_2$, etc.)
752 within and above the boundary layer during daytime and nighttime. In general, $O_{x,total}$ budgets
753 have not been fully quantified in valleys in the western U.S. However, it is clear that odd-oxygen
754 species play a substantial role in the buildup of wintertime pollutants. Owing to the significance
755 and uncertainty in HONO sources, nitrous acid measurement via multiple techniques and from

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

ground and aircraft platforms will be an essential component of a future western U.S. winter air quality study.

High temporal and spatial resolution measurements of NO_x, VOCs from multiple sources (transportation, VCPS, cooking, wood combustion, biogenic, industrial, agriculture), NH_3 and amines are required. Multiple tracers will enable quantitative attribution of GHG emissions to different sectors through their enhancement ratios. These include enhancement ratios of CO_2 and CH_4 relative to tracers such as CO and NO_x. For instance, using the CH_4:CO ratio measured from aircraft, a recent study revealed that CH_4 emissions from urban areas in the East Coast corridor were drastically underestimated by inventories (Plant et al. 2019). Similarly, observed VOC:GHG correlations would allow attribution to oil/gas emissions (Petron et al. 2012). In addition, novel tracers like atmospheric O_2 will be considered for source attribution as part of the planned field campaign.

9. Summary and Conclusions

The interactions between complex wintertime chemistry and numerous meteorological processes regulating the occurrence and rates of chemical processes in PCAPs has not yet been observed in a detail needed to provide satisfactory understanding of the evolution of these complex pollution episodes. This paper provides an outline of the comprehensive chemical and meteorological datasets needed from airborne platforms with extensive range, coupled to a variety of surface-based measurements with high vertical resolution at numerous representative sites to provide enhance understanding of these pollution episodes. The workshop has outlined the design and requirements for a field campaign investigating wintertime basins to provide the

Accepted for publication in *Bulletin of the American Meteorological Society.* DOI 10.1175/BAMS-D-20-0017.1.

779 breadth of observations necessary to characterize the meteorological-chemical linkages to
780 validate complex processes within coupled atmosphere-chemistry models.
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782
783 **Acknowledgements:**
784 NSF Atmospheric Chemistry Program (Award Number 1912664) and the NOAA Atmospheric
785 Chemistry, Climate and Carbon Cycle Program (Award Number NA19OAR4310078) supported
786 the workshop. We appreciate the assistance of Chase Brandt Hodges-Heilmann with citation
787 management for this article. We appreciate the assistance of Chris Rapp in website development
788 for the workshop. We also appreciate the administrative support from the Department of
789 Atmospheric Sciences at the University of Utah in hosting the workshop, specifically the help
790 from Holly Moreno and Nola Lucke. The views expressed in this manuscript are those of the
791 authors alone and do not necessarily reflect the views and policies of the U.S. Environmental
792 Protection Agency.
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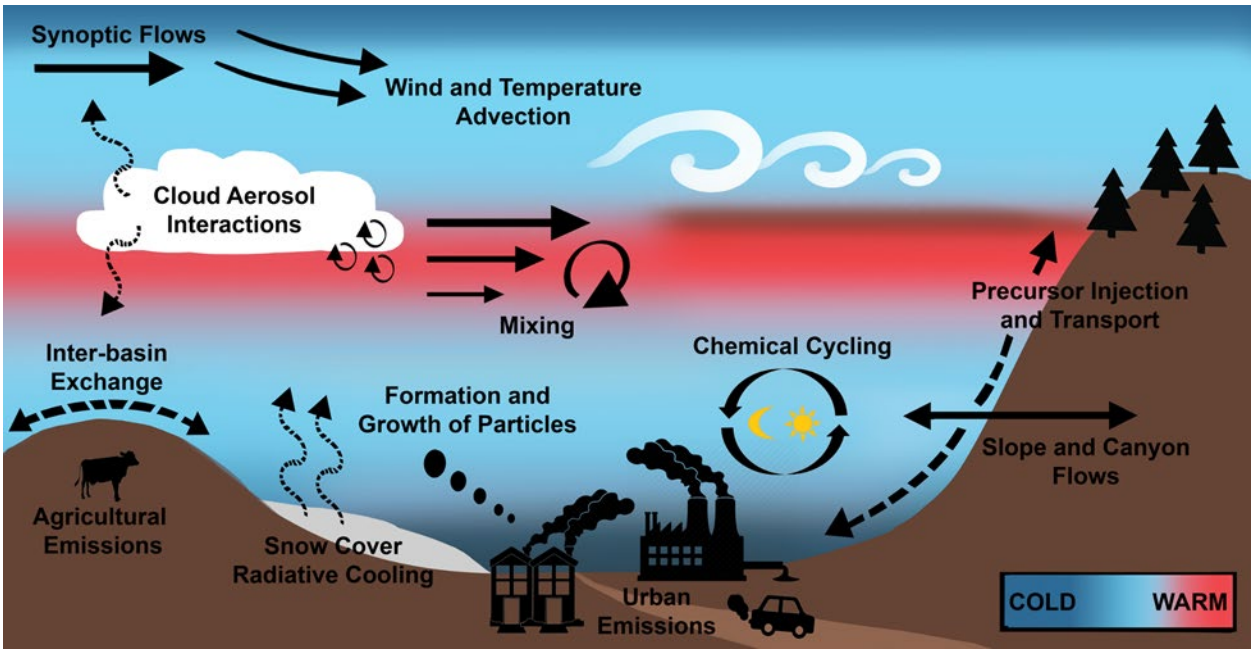

Figure 1. Schematic of various coupled meteorological and chemical processes within wintertime PCAPs. Figure is not to scale. Importantly, stable nocturnal inversions can be extremely shallow (10's of meters) and elevated inversions can also be present depending on the large-scale synoptic flow.

Accepted for publication in *Bulletin of the American Meteorological Society*. DOI 10.1175/BAMS-D-20-0017.1.

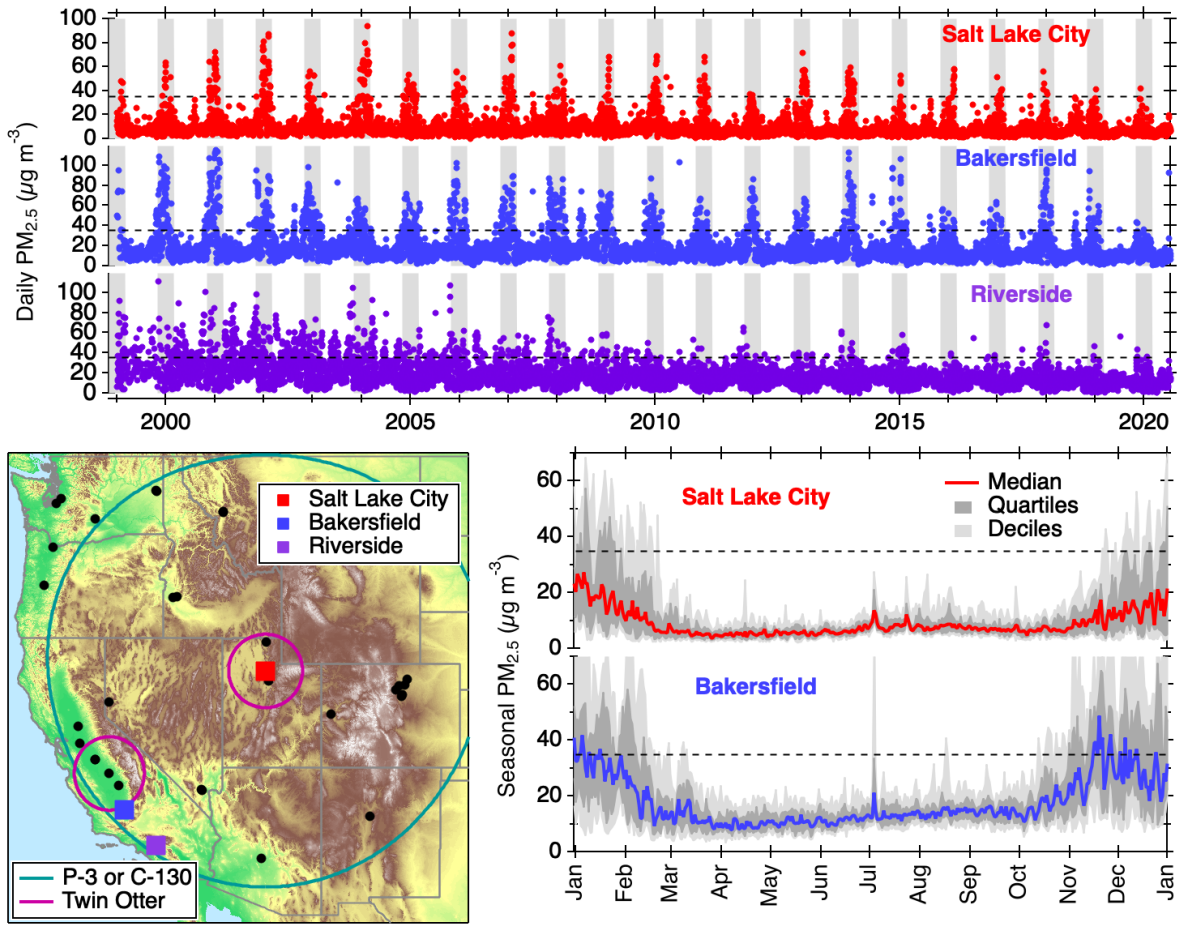

Figure 2: *Lower left:* Elevation map of the Western U.S. showing selected air quality monitoring sites (black dots) that exhibit winter maxima in $PM_{2.5}$. Colored markers show three sites that are highlighted in the top figure. Rings show the approximate ranges of a large research aircraft (P-3 or C-130) based out of Salt Lake City or smaller aircraft (Twin Otter) based out of either Salt Lake City, UT or Fresno, CA. *Top*: Daily $PM_{2.5}$ at Salt Lake City, UT and Bakersfield, CA from 1999-2020. The dashed lines are the 24-hour U.S. NAAQS for $PM_{2.5}$ of 35 μg m^{-3}. Grey shaded areas indicate November - February. *Lower Right*: Median, 25th and 75th percentile, and 10th and 90th percentile $PM_{2.5}$ at Salt Lake City, UT and Bakersfield, CA for each day of year from the record in the top figure. Dashed lines indicate the NAAQS, as in the top figure.

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