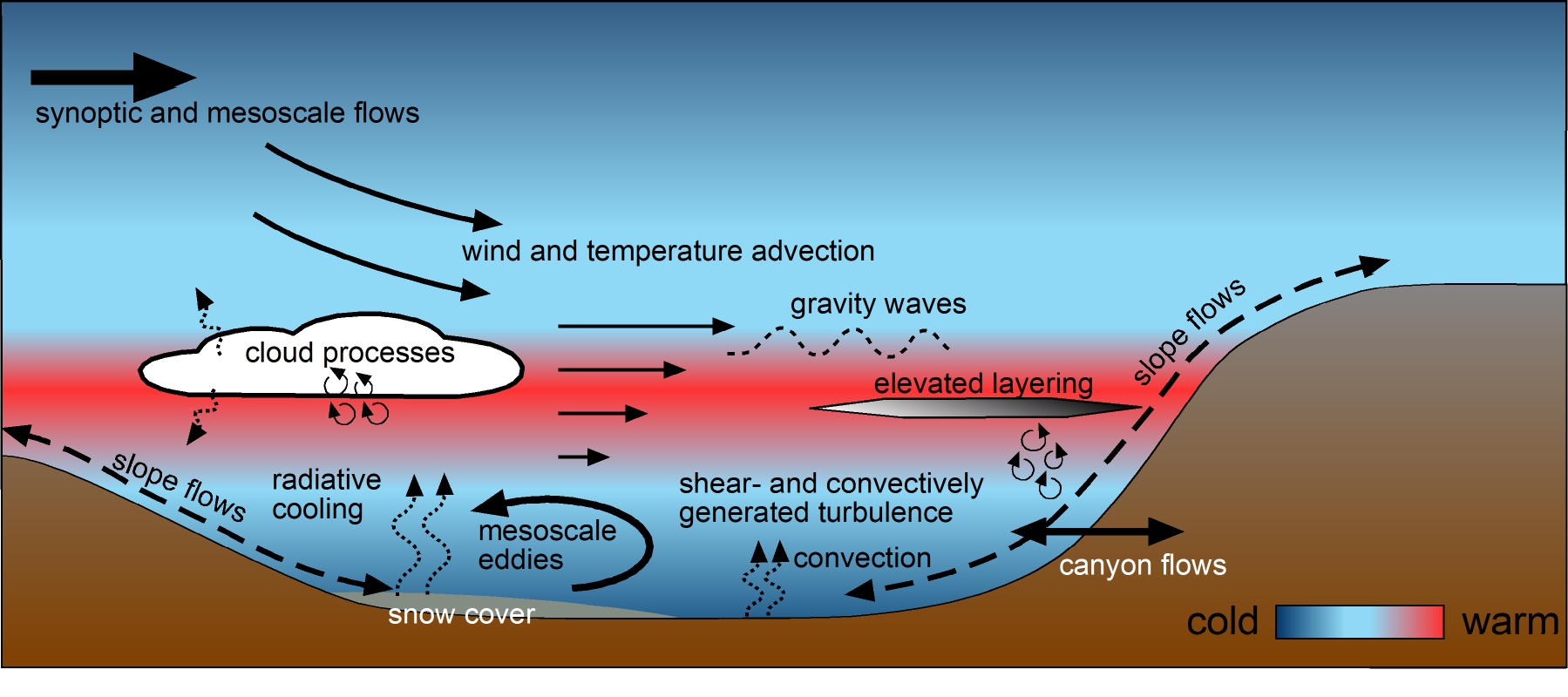
**Air QUAlity Research In the western United States (AQUARIUS)**

**Content Draft for White Paper**

**Section 1. Overview of the coupling between meteorological and chemical processes controlling severe wintertime air pollution episodes in the Western US**

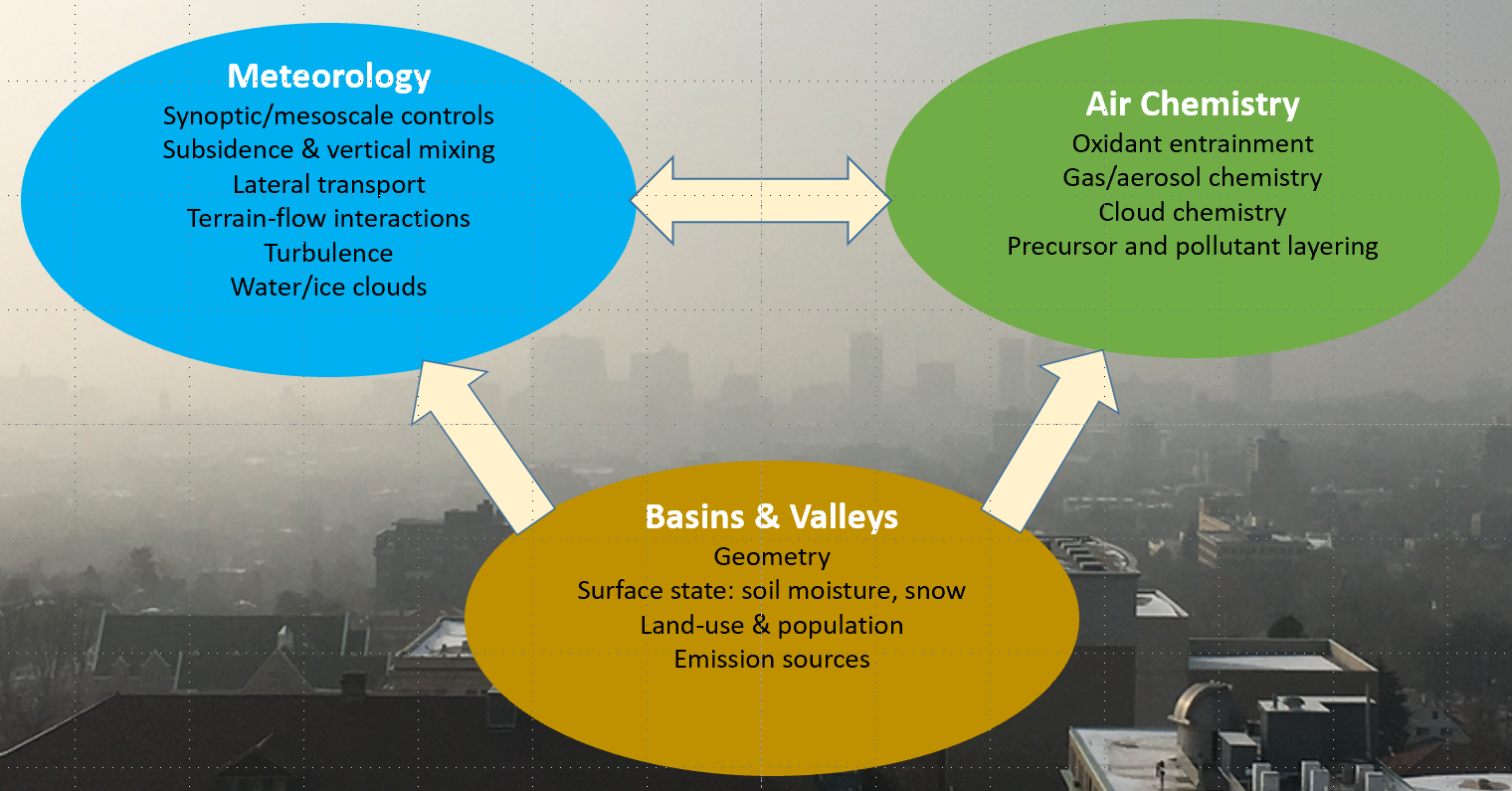
Winter episodes of high particulate concentrations occur frequently in urban and agricultural basins and valleys in the western US (Whiteman et al., 2001, Whiteman et al. 2014, VanReken et al., 2017, Franchin et al. 2018). These episodes often arise due to the development of persistent cold-air pools (PCAPs), which are stably-stratiﬁed boundary-layer airmasses sheltered from lateral and vertical mixing by surrounding topography (Lareau et al. 2013). PCAPs may last from days to weeks when a combination of warming aloft and cooling near the surface lead to stable stratification and air mass stagnation ([Dorninger et al. 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878" \l "bb0070), [Reeves et al., 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878#bb0230), [Sheridan et al. 2014](https://www.sciencedirect.com/science/article/pii/S0169809516303878" \l "bb0240), Holmes et al. 2015, McCaffrey et al. 2019, Sun and Holmes, 2019, Ivy et al., 2019).

Fair weather arising from subsidence above the mountain ranges of the Western US due to high pressure and regional-scale warm air advection aloft are a key factor for the formation of the stable stratification that traps cold air in basins and valleys. The depth, intensity, and duration of PCAPs depend on the characteristics of the underlying surfaces within the basins (e.g., dry or wet soils or the presence of snow cover), the surface energy and radiation budget, and complex time- and space-varying interactions between regional- and local-scale flows with the surrounding terrain (Neeman et al. 2015, Lareau and Horel, 2015a, Foster et al. 2017). Long-duration PCAPs are more common in deep, enclosed basins that tend to be sheltered from passing weather disturbances than shallow or partially enclosed ones (Clements et al. 2003, Vosper et al. 2008, Hoch et al., 2010, Yu et al. 2017, Sheridan 2019). Once PCAPs are established, many other factors that affect radiative, sensible, and latent heat fluxes at the surface and within the entire depth of the PCAPs become important, e.g., time of year, presence of low clouds within PCAPs and mid- and high-level clouds above them (VanReken et al. 2017). Figure 1 illustrates many of these meteorological processes.



**Figure 1**. Processes affecting PCAPs.

The meteorological factors dominating the setup, maintenance, and demise of PCAPs in the western US and elsewhere are well understood. **However, the complex thermodynamic, radiative, and dynamical processes evolving in space and time during PCAP episodes that strongly affect pollutant concentrations are not well documented, understood, analyzed, or forecasted.** As illustrated in Fig. 2, the meteorological and chemical processes are highly intertwined and need to be studied as a coupled system within valleys and basins that exhibit different underlying surface states and emission profiles (Baasandorj et al. 2017, Womack et al. 2019).



**Figure 2.** Schematic of the coupling between the characteristics of basin and valleys and meteorological and chemical processes during wintertime PCAPs.

The complexity of the coupling between the characteristics of basins and valley and meteorological and chemical processes introduced in Figs 1 and 2 lead to pollutant episodes in the western US of varying severity and duration. Although the setup and evolution of episodes in the large Central Valley of California differ in many respects from those in basins and valleys of the Intermountain West, the AQUARIUS program is focused on understanding how these highly-coupled meteorological and chemical processes combined with local conditions contribute in different ways through improved understanding of the production, transformation, cycling, and destruction of chemical species during the life cycle of PCAPs.

The following questions were posed to the participants in the AQUARIUS Workshop to focus discussion on PCAP-related pollution episodes:

* How do meteorological “cold-air pool” conditions contribute to wintertime basin air quality, and how can meteorological observations and modeling efforts be designed to most effectively inform chemical and emissions research?
* What are the key meteorological factors that contribute to the production, transformation, cycling, and destruction of chemical species during the life cycle of persistent cold-air pools?
* To what extent are chemical processes controlled by vertical mixing and entrainment between layers of differing stability relative to horizontal transport (interbasin and intrabasin)?
* How are chemical pathways affected by differences between thermodynamic and dynamical processes in the core of basins relative to those processes near their edges?
* Do chemical processes affect the structure of the boundary layer, for example, through contributing to the formation of clouds or reducing solar insolation received at the surface?
* How do variations in the fluxes of heat, moisture, and momentum arising from heterogeneous surfaces within and between basins (e.g., rural or urban; vegetated, snow or water covered) lead to variations in the production and destruction of pollutants?
* What are the appropriate modeling approaches to use within wintertime basins and how may that differ as a function of factors such as basin dimensions, surface characteristics, exposure to differing mesoscale and synoptic conditions, etc.?
* What meteorological observations are required to help validate photochemical models?

Based on feedback obtained during the Workshop, Table 1 summarizes examples of linkages between meteorological and chemical processes that are not well understood. Further refinement and discussion led to the four critical topics described in the following subsections that are recommended to be examined as part of the AQUARIUS program. No prior field campaign has provided the breadth and depth of contemporaneous observations at the surface and aloft necessary to analyze the meteorological-chemical linkages outlined in Table 1 nor serve as input to or validation for atmosphere-chemistry models. Establishing an experimental design to provide those meteorological and air chemistry observations and requisite modeling studies to address these topics are described in Sections 2 and 3.

**Table 1.** Coupled meteorological and chemical processes for which their impacts are incompletely understood along with the subsections that discuss these processes.

|  |  |  |
| --- | --- | --- |
| **Meteorology** | **Chemistry** | **Impacts** |
| * Effects of land use, soil moisture, surface state | * Photochemical chemistry | * Modifies surface albedo, latent and sensible heat fluxes, radiative budget * Photochemical production |
| * PCAP depth and volume | * Concentration and photochemical gradients | * Feedbacks between PCAP lifetime and lateral and vertical pollutant gradients |
| * Moisture and fog/clouds | * Cloud chemistry versus particle- and gas-phase chemistry * Photolysis * Gas-particle partitioning | * Alters surface energy balance and turbulent production * Enhances the importance of aqueous and heterogenous chemical pathways * High moisture content promotes uptake of semi-volatile gases by particles * Reduces actinic flux and photolysis reactions |
| * Vertical temperature and moisture structure | * Vertical layering of precursors, primary and secondary pollutants | * Affects vertical mixing, chemical aging and particle growth * Leads to coupling/decoupling from surface emissions * Complicates feedback of emission controls on pollution levels |
| * Lateral exchange and transport | * Oxidant, pollutant, and precursor transport and concentration | * Pollutant, precursor, and oxidant concentrations vary spatially due to boundary-layer transport, mixing and terrain-driven flows |
| * Temporal variations in meteorological conditions | * Actinic fluxes and photochemistry * Reaction rates, chemical vs. transport/mixing time scales | * Formation of secondary pollutants follow different pathways during day and night * Turbulent mixing during day often replaced at night by decoupled surface and residual layers |

*Section 1.1: Surface fluxes and radiative processes*

The atmospheric boundary layer is coupled to the underlying surface through surface turbulent fluxes with reduced surface sensible and latent heat fluxes observed during PCAP episodes in the Salt Lake Valley (Sun and Holmes 2019). However, surface turbulent heat fluxes calculated using Monin–Obukhov similarity theory relied upon by most atmospheric models generally do not agree well with those observed within stable wintertime boundary layers (Karsisto et al. 2016, Massey et al 2017). The variability of turbulent surface fluxes and incoming and outgoing radiation as a function of land use and varying surface state conditions within the urban and agricultural valleys of the western US is poorly understood.

Formation of secondary pollutants requires that precursor emissions are oxidized to their secondary products. Under wintertime conditions, daylight hours are limited, temperatures are cold, and, as a result, daytime photochemistry that produces secondary pollutants may be limited. Since many important reactions that drive secondary pollutant formation are photolytic, characterizing the distribution of actinic flux is important to understand the influence of photochemical processes during PCAPs episodes. This can be challenging in wintertime due to variations in land use and snow cover on albedo as well as the presence of fog and cloud. These variations subsequently impact boundary-layer temperature structure, vertical and horizontal transport and cycling of pollutants, temperature-dependent chemical processes, and photochemical reaction rates.

While boundary layer depth is often assumed to depend strongly on sensible heat flux, the intensity of downwelling shortwave radiation is also important (Pal and Haeffelin, 2 015; Trousdell et al., 2016). Possible coupling between cloud and/or aerosol shading and turbulent mixing will be important to examine during AQUARIUS field campaigns. Feedbacks between surface state (e.g., snow cover vs. no snow cover) albedo and photochemical rates have been shown to be important (Neeman et al. 2015; Foster et al. 2017), but more comprehensive evaluation of the coupling between surface shortwave, sensible/latent heat and turbulence fluxes and surface state, and their subsequent impacts on the production and destruction of pollutants, is needed. Detailed observations of photolysis rates and actinic fluxes are important to be able to quantify photochemical-meteorological coupling.

*Section 1.2: Moisture content and clouds*

Atmospheric moisture and presence of fog and clouds influence the formation of secondary particulate matter. The relative humidity in the surrounding environment and the hygroscopicity of the aerosol chemical constituents determine the water content of the aerosols found within PCAPs. Under cool, humid conditions, condensation of semi-volatile species onto particles is favorable, whereas such formation is relatively unfavorable under warm, dry conditions and the other characteristics of aerosols also vary strongly with humidity (Winkler 1988) Urban areas enhance boundary-layer water vapor during wintertime through anthropogenic emissions (Salmon et al. 2017). Complex feedbacks exist between natural and anthropogenic water vapor and particulate aerosol emissions within a PCAP, and the subsequent development of aqueous particulate pollution, low clouds and fog. Aqueous chemistry follows very different pathways in cloud and fog than in aerosol particles since cloud and fog droplets are highly dilute, whereas particles have high ionic strengths that lead to highly nonideal chemistry (Pye et al., 2019). Intense fog not only reduces aerosol loading through wet removal but can also provide a medium for aqueous-phase chemical reactions by altering particle size distribution by selectively removing water-soluble species. As a result, primary aerosols may be transformed into secondary aerosols (Dall’Osto et al. 2009 ).

Prior work on fog chemistry (Collett et al., 1999; Ge et al., 2012; Herckes et al., 2015) provides a framework for a more comprehensive investigation of the role of clouds and fog in PCAPs. Nucleation scavenging and removal of aerosol particles dominates in dense fog (Seinfeld and Pandis 2016) although exceptions in highly polluted environments have been observed (Bisht et al. 2016). Convective layer growth and turbulence during the afternoon helps to decrease particulate concentrations and evaporate fog and stratus, which leads to new hygroscopic aerosol particles and haze (Pandis et al. 1990; Zhu et al. 2019). The transition from cloud-free to cloudy PCAPs often coincides with a plateauing of particulate concentrations (VanReken et al 2017; Baasandorj et al. 2017).

Greater understanding of these feedbacks between aerosols, water vapor, clouds and fog, and atmospheric boundary-layer structure and evolution is needed. Does aerosol chemistry differ during high and low humidity conditions? How does the chemistry vary between wet (cloudy) and dry (non-cloudy) PCAPs? Does fog impact sulfur or organic oxidation? Does different chemistry occur during shallow near-surface fog versus an elevated stratus layer? What factors contribute to the tendency for the rate of particulate concentration increase flatten when fog and clouds form within PCAPs?

*Section 1.3: Vertical structure*

A key research area for AQUARIUS is to identify and understand the common, as well as unique, features for the coupling between vertical profiles of meteorological properties, chemical precursors, and particulate pollution during the lifetime of PCAPs episodes in different basins and valleys in the western US. Since poor air quality at the surface often involves mixing of pollutants formed aloft to the ground, the effects of intermittent vertical stratification on chemistry must be thoroughly characterized to improve the models used in air quality management and health assessments. Evaluating those processes under the distinctly different vertical profiles commonly present in California’s Central Valley relative to those in much smaller basins and valleys in Utah will be critical. However, even defining adequately the depth of PCAPs is notoriously difficult when one or more stable layers at the surface or aloft are present (Collaud et al. 2014). Developing approaches to improve what is considered the boundary layer depth will be crucial to help improve air chemistry model simulations for example of dry deposition rates for critical compounds such as O3, NOx, NOy, and PM.

Pollutant formation may be more favorable within elevated layers exposed to oxidant abundance that are less affected by NOx titration near the surface (Baasandorf et al. 2017). Assessing the extent and factors that control such elevated oxidant-rich layers throughout the western US will be highly relevant for AQUARIUS. The time scales affecting vertical mixing between such layers and determining how sensitive they are to temperature, moisture, and clouds needs to be addressed. Greater attention is also needed to understand the impacts of aerosols within PCAPs through their absorption and scattering of solar radiation that affects the surface energy budget and vertical profiles of temperature and moisture (add refs).

Turbulence from the surface through the entire depth of PCAPs and above into the free atmosphere aloft is intrinsically linked to vertical mixing processes for heat, moisture, and precursor and pollutant concentrations within PCAPs (refs to be inserted). The time evolution and intermittency of vertical wind shear, gravity wave production, and entrainment across the interface between PCAPs and the free atmosphere are recognized as being critical, but their impacts on pollutant concentrations are poorly understood (refs to be inserted). Although characterizing the spatial and temporal variations in turbulence within PCAPs in different basins will be difficult, that will be a critical task to understand vertical transport processes and meteorological coupling with chemical processes.

*Section 1.4: Thermodynamical and dynamical processes within basins and valleys*

The chemical processes that lead to secondary pollutant formation in basins and valleys are sensitive to the interplay between regional pollutant advection along with lateral and vertical mixing within them of fresh emissions and pollutants that have undergone varying degrees of local chemical aging. The characteristics of the basins and valleys (size, depth, surrounding terrain, land use, emission sources) help define the thermodynamical and dynamical processes shown schematically in Fig. 1 that affect these chemical processes. For example, the broad expanse of California’s Central Valley and the Snake River Basin allow substantial advection into those regions along with considerable lateral transport and cycling of pollutants within their large volumes. On the other hand, PCAP episodes in Intermountain West basins and valleys are affected strongly by the interaction with slope, tributary-canyon, and other dynamically-driven flows. Hence, the scales of basins have a large impact on the relative importance of inter- (between) and intra- (within) basin exchange and transport processes. Prior field studies have lacked sufficient simultaneous meteorological and chemical measurements across basins and valleys to adequately address all of the intertwined factors outlined in Fig. 2.

Replenishment of oxidants through entrainment from aloft or vertical mixing resulting from thermally-driven slope flows promotes sustained production of secondary pollutants throughout a PCAPs episode. Although VOC emission controls may help for episodes that terminate through oxidant depletion, episodes that experience oxidant replenishment might be insensitive to such controls. Despite the importance of entrainment processes for air quality management during PCAPs, the extent of oxidant entrainment is not well quantified in western basins. Previous studies have hypothesized the importance of horizontal transport processes on the chemical processes within PCAPs (e.g., Baasandorj et al. 2017). Oxidant injection from tributary canyons, or agricultural ammonia from inter-basin transport have been explored in Utah. The entrainment of nitrate PM produced overnight in the residual layer within the San Joaquin Valley was found to be responsible for approximately 80% of the near surface levels in the morning hours (Prabhakar et al., 2017).

Origins of the nocturnal air aloft may dramatically influence PM levels during the next day. Hence, observing the time-evolving three-dimensional flow and thermodynamic gradients within basins is crucial. Particular attention is requires in basins of smaller size to sample adequately the canyon and slope regions (Oldroyd et al. 2016). Sensible surface heat fluxes along the surface and basin/valley slopes, as well as in the adjacent canyons are important for driving the wind systems important for inter/intra basin, and slope/canyon exchanges. Spatial variability in these surface heat fluxes can be caused by many factors, including patchy snow cover, sidewall orientation, land-cover variability (e.g. urban vs rural), and elevation-related factors. Even during stable wintertime conditions, shallow layers with upslope flows can be present along the valley and canyon sidewalls. The subsequent venting processes transport pollutants away from the lower reaches of the cold air pool, but can be recirculated aloft leading to elevated pollution layers. Comparison of vertical profiles away from basin walls (e.g. from towers or via remote sensing) to pseudo profiles taken with sensors vertically deployed up a sidewall can probe the role of canyon wall exchange relative to daytime PBL growth in the interior of the basin (add refs from Hoch). Lagrangian-based approaches to track the evolution of the flows entering, within, and exiting basins are needed to arrive at a better understanding of the impact of nonstationary boundary layers on chemical processes..

**Section 2. AQUARIUS Study Design: Vertical Profiles**

**Section 2a. Introduction**

Ground-based platforms will be the cornerstone of a fully integrated suite of vertical measurements used to characterize the coupled meteorological-chemical system in three dimensions during the AQUARIUS campaign. The AQUARIUS study design for measuring meteorological and chemical coupling, three-dimensional transport, and precursor chemical emissions must take into account a number of important points, including spatial representation (density and number) of observations, spatial collocation of meteorological and chemical measurement, and effectively utilizing both *in situ* and remote sensing methods.

Because vertical mixing in photochemical models is parameterized using relationships between turbulent fluxes and mean profiles, it is paramount that meteorological and chemical profile measurements be collocated with surface energy balance stations and sensors to measure atmospheric turbulence and resulting meteorological and chemical exchange processes. These vertical profiles need to be adequately resolved in both time and space such that the relationships between meteorological and chemical processes can be determined. This approach will allow mass, moisture, heat, and chemical budgets of basin air masses to be quantified in unprecedented detail. For example, collocated vertical profiles of vertical velocity, water vapor, and chemical composition would enable computation of flux and flux divergence profiles needed to estimate the boundary budget of these quantities (Lareau 2019). Numerical modeling analyses may be useful in helping inform difficult choices about sensor distribution to ensure column observations that best resolve inter- and intra-basin exchange and mixing processes, and to inform location, scope, and size of the study domains in smaller (e.g., Utah) or larger (e.g., San Joaquin Valley) basins targeted during AQUARIUS.

The regions of interest to be most heavily instrumented in individual basins will depend on the basin size, topography, emission sources, and important transport processes. Which of the coupled processes discussed in Section 1 will be most targeted through observation will depend on the basin of interest. The various capabilities of proposed AQUARIUS vertical profiling instrumentation to address these goals will be discussed in more detail in the following sections.

**Section 2b. Basin Vertical Profiling Instrumentation**

An overview of proposed observational platforms to be deployed and their purposes are illustrated graphically in Figure **7**. Many types of vertical profiling instrumentation, both *in situ* and remote sensing, will need to be utilized during AQUARIUS to address the core science questions. The most effective spatial representation and colocation of instruments is not shown in Figure 7 as the geographical distribution of sensors will depend on the unique geography, meteorology, and emission sources in the basin of interest. Technical specifications and advantages and limitations of the various proposed vertical profiling sensors systems are given in Table 3.

Surface observation sites, particularly those with chemical and turbulence measurements (not shown) will also need to be situated in a way that their observations can be easily blended to vertical profile data.



**Figure 7.** *Schematic of proposed vertical profiling instruments during AQUARIUS.*

A careful analysis of the conditions under which some profiling platforms cannot be utilized due to flight restrictions or weather conditions, especially fog will need to be conducted. Surface-based instruments deployed on high towers, tall buildings, or terrain slopes on the edges of basins will also need to be utilized for obtaining vertical and “psuedo-vertical” profiles of some atmospheric and chemical measurements (Fig. 7).

Many turbulence measurements (both vertically and horizontally) will be needed in order to capture the impact of varying meteorological processes (e.g., canyon or slope flows) on the turbulence budget. The turbulence budget can then be coupled with chemical process observations. Doppler lidar observations will, for example, provide an unprecedented view of mixing processes over the PCAP depth, including near the top interface of PCAPS where shear and buoyancy interactions can change the PCAP structure. To date these processes have not been measured, but are known to strongly impact PCAP evolution via top-down fluxes (e.g., Lareau and Horel 2015b). Similarly there is a need to better quantify near-surface fluxes that drive mixing from the bottom up and depend on the surface state and terrain heterogeneity. These surface data are also critical in contextualized lidar-observed variations of mixing aloft

Table 3. AQUARIUS Vertical Profiling Platforms for Coupled Meteorology-Chemistry Measurements

|  |  |  |  |
| --- | --- | --- | --- |
| **Vertical Profiling Instrumentation** | **Key Parameters Measured** | **Vertical and Horizontal Resolution.** | **Limitations** |
| Airborne wind LiDAR | Vertical and horizontal wind speed, turbulence | Horizontal ~2-3 km, vertical ~25 m. | No data in thick clouds, very clean (low aerosol) air |
| Surface-based wind LiDAR | Vertical and horizontal wind speed, turbulence | Horizontal ~2-3 km, vertical ~25 m. | No data in thick clouds, very clean (low aerosol) air |
| Surface-based Raman LiDAR | Water-vapor mixing ration, temperature, aerosol, and cloud optical properties. | 100 m vertical resolution, 3000 range | Discontinuities in ground-based time height cross-sections |
| Surface-based Dial LiDAR | Atmospheric concentrations of gases such as water vapor and ozone. | 150 m vertical resolution | Retrievals are sensitive to clouds, leading to larger errors in the temperature and WV profiles. |
| Airborne cloud RADAR | Cloud and precipitation structure, vertical winds |  |  |
| Airborne in situ meteorological probes | Cloud microphysics, thermodynamics, turbulence, dynamics |  |  |
| Differential Optical Absorption Spectroscopy (DOAS) Radiometers | Estimate of the vertical temperature profile, static stability, relative humidity, and cloud liquid water path and gases NO2, SO2, O3, HONO, HCHO, and NO3. |  |  |
| Scanning Aerosol Backscatter  Lidar | Pollution layers, aerosols, cloud top and base |  |  |
| Aerosol ceilometers | Air pollution and meteorology, pollution layers |  |  |
| Rawinsonde systems | Boundary-layer and tropospheric thermodynamic and wind profiles and pressure | Vertical resolution of 10-30 m. | Slantwise profile. Limited frequency. |
| Pseudo-vertical profiles and building profiles | Wide array of meteorological and chemical species | Variable. | Surface fluxes and flows sometimes make these observations unrepresentative of free atmosphere |
| Mobile vertical profilers | Ceilometers, wind LiDARS commonly deployed on mobile ground-based units. |  |  |
| DIAL LiDAR |  |  |  |

Vertical remote sensing by the suite of sensors listed in Table 3 will be a critical component of adequately observing the vertical, temporal, and spatial evolution of boundary-layer meteorology and the depth and layering of particulate pollutants. Ground-based wind and aerosol LiDARs, ceilometers, sodars, and other remote sensing instruments will be carefully deployed in order to get a full picture of meteorological and chemical coupling. Aerosol lidars and ceilometers will be used to monitor the temporal and spatial variations in vertical profiles of aerosol backscatter. Different scanning strategies of the various platforms will be used. A graphical example of the different scanning approaches for a wind LiDAR to determine three-dimensional flow structure in the basin atmosphere as shown in Figure 8.



***Figure 8.***Schematic illustrating commonly used scanning approaches for wind liDARS. The liDAR scans can target the horizontal winds (conical PPI scans, VAD retrieval), the vertical structure of turbulence (sigma\_w, vertical stares), and mass transport through a tributary canyon (RHI scans).

In addition to fixed location vertical profiling, there is the potential for mobile vertical profilers to map the spatial structure of the basin atmosphere. Any available mobile ground-based systems measuring both meteorology and chemistry will be utilized, such as the CARB Mobile Measurement Platform (MMB)(Park et al. 2016) or the TRAX air quality light-rail train (Mendoza et al. 2019). These ground-based systems can be driven up canyons and slopes for pseudo-vertical profile measurements similar to tall buildings and instruments placed along mountain slopes. These vertical profiles at basin boundaries will assist in quantifying background pollution and meteorological parameters as well as intra-basin transport processes. Recent field campaigns in these basins highlight the importance of capturing the meteorological variability and air-pollution coupling (Baasandorj et al. 2017, Faloona et al. 2020). Truck or trailer mounted Doppler lidars, for example, can be used to probe variations in the vertical velocity, boundary layer mixing height, and elevated aerosol layers. While these data are not as easily collocated with other measurements, they can provide important spatial context for time-series measurements at fixed site sensors. Figure. 9 shows and example of mobile Doppler lidar profiling in complex terrain, revealing spatial differences in aerosol backscatter, vertical velocity, and boundary layer height. Similar sampling could be used during AQUARIUS.

A close up of a logo

Description automatically generated

**Figure 9.** Example of mobile-lidar observations of variation in boundary layer vertical velocity (top) and aerosol backscatter (bottom) in complex terrain.

Vertical *in-situ* sensing will also be utilized in AQUARIUS observational design (Table 3). Multiple rawinsonde systems, tethersondes, and small drones are all recommended and would help supplement the remote sensing systems. These systems will be used as much as possible in areas where they are allowed and below federal aviation regulation height limits. The types of sensors also include weather stations deployed along basin slopes (meteorology, pseudo-vertical approach).

**Section 2c. Basin Vertical Profiling Study Design**

The size of the basins studied will determine the vertical sampling approach that is utilized during AQUARIUS. For a small basin such as Utah’s Cache Valley, a single central vertical profiling site may be adequate. For the medium-sized Salt Lake Valley and expansive California Central Valley, multiple vertical profiling sites, combined with mobile observation platforms and aircraft measurements are likely needed to capture important spatial variations in meteorology and chemistry. Alternatively, airborne or mobile ground-based systems equipped with instruments such as Doppler and Raman lidars can be deployed to capture spatial variability of meteorology (including turbulence) and chemistry. Any available mobile ground-based systems measuring both meteorology and chemistry will be utilized during AQUARIUS.

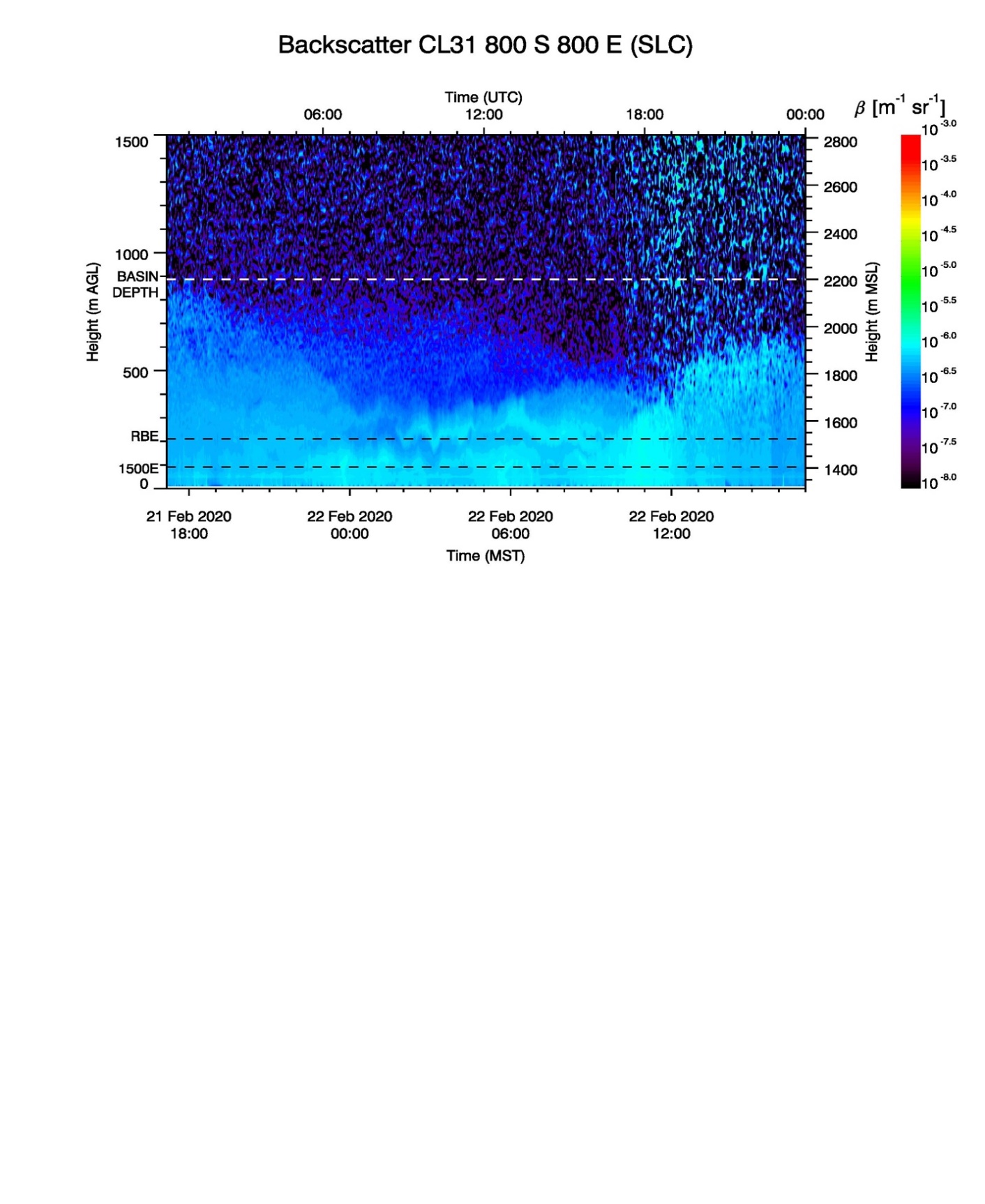
Sufficient data collection to ensure spatial gradients are captured across the basin of interest is of great importance. There must be adequate observation locations spatially (and with adequate temporal consistency) that a three-dimensional representation of the atmospheric state for both chemical and meteorological properties is obtained. The spatial representation aspect will be easier for small basins and more difficult for larger basins. A holistic and interdisciplinary and multi-agency approach is needed, where existing infrastructure such as National Weather Service daily rawinsonde launches, weather stations from public and private sectors available on MesoWest (Horel et al. 2002), wind sodars, lidar profilers, and ceilometers will be supplemented with instrumentation dedicated to AQUARIUS.

One AQUARIUS study design approach is a process-focused deployment, selecting regions of interest within a basin to investigate the details of known coupled chemical-meteorological processes, for example, mobile lidar observations might help fill in important data gaps in large basins. For example, different sampling strategies could focus on capturing the coupled meteorology and chemistry along a sidewall, a tributary, a topographic gap, etc. This would allow a deeper study of known processes and could lead to the quantification of the individual processes. Three brief examples of such targeted sampling strategies are provided below:

*Sidewall ventilation:* a pseudo-vertical transect of chemistry from basin floor to PCAP top could be combined with slope-normal wind and aerosol lidar scans, to quantify the timing, strength and importance of this process during the life cycle of a PCAP event.

*Inter-Basin exchange:* Using vertical profiles of wind and chemistry to estimate pollution and/or precursor transport under the changing conditions of a PCAP life cycle. For example, the transport of chemcial precursors into the PCAP could be analyzed by looking at flow structures and aersol backscatter from a laser ceilometer (Figure 10).

*Cold-air pool layering and vertical exchanges*: In this case, coupling, decoupling and re-coupling of these different layers (i.e. reservoirs) in the PCAP may occur. Vertical chemistry profiles could be combined with wind LiDAR observations (vertical stares) and other vertical profiles such as aerosol backscatter from ceilometers to investigate the importance of different vertically separated reservoirs where different chemical may dominate. Understanding and resolving the timing of the de-coupling (night) and re-coupling via a surface based convective BL may be crucial to fully replicate and correctly model chemical processes.



**Figure 10.** *Example of (aerosol) ceilometer resolving low-PM air being injected into the Salt Lake Basin nocturnal CAP. Air is injected through a smaller tributary canyon with its mouth at the elevation marked “RBE”.*

Chemical processes driven by mixing also need to be resolved alongside the turbulence, mixing and transport meteorological measurements so that these two processes can be linked together in the study analysis. Coupled turbulence, meteorological (including turbulence and the surface energy balance) data on towers and high buildings at multiple locations within the basins are needed. For example, locations such as radio towers or high building could be used to sample a vertical profile of data using ground-based sensors. Many of these stations should be collocated with long-path Differential Optical Absorption Spectroscopy DOAS using an array of retro-reflectors at different altitudes (e.g. deployed on a ground based tower) allows for continuous observations of vertical profiler (for a minimum of temperature, wind speed and, profiles of gases such as NO2, SO2, O3, HONO, HCHO, and NO3. The profiles of these gases probe both vertical mixing processes and humidity) to collect data that can be used to investigate the validity of Monin-Obuhkov Similarity Theory (MOST) during PCAPS chemical transformations.

**Section 3. AQUARIUS Meteorology-Chemistry Modelling**

**Section 3a. Introduction**

Comprehensive atmospheric chemical transport models (CTMs) driven by output from mesoscale meteorological models are the preferred tools for air quality management and forecasting. Obtaining CTMs for PCAP conditions requires that complex feedbacks between model meteorology (mixing and transport), emission inventories, and atmospheric chemistry processes all be adequately simulated. CTM parameterizations for atmospheric mixing and other processes are developed for broad application and generally perform well in characterizing summertime ozone and PM2.5 pollution. However, achieving good model performance is more challenging for simulations of the meteorology and air quality conditions during stable wintertime PCAPs episodes in Western air basins. For instance, model performance issues have been identified in CTM studies for the San Joaquin Valley (SJV) and suggest that key processes may not be fully resolved, although modeling for SJV has achieved reasonable results overall in some cases (Ying et al., 2008; Chen et al., 2014; Kelly et al., 2018). For smaller basins such as Salt Lake City, CTM modeling is more challenging and relatively limited in the literature, in part because inadequate simulations of meteorology often preclude meaningful scientific application of CTMs.

To improve meteorological model simulations of PCAPs, considerable work is underway or has been conducted in recent years (e.g., Saide et al. 2011, Lareau and Horel 2015b, Ahmadov et al. 2015, Saide et al. 2016, Foster et al. 2017, Tran et al. 2018, Sun and Holmes 2019; Kelly et al., Sun et al. 2019, 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](https://www.sciencedirect.com/topics/earth-and-planetary-sciences/boundary-layer-flow) ([Baklanov et al., 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878" \l "bb0025), [Price et al., 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878#bb0220), Holmes et al. 2015).

Deficiencies in emissions inventories and model treatment of complex chemical mechanisms in CTM modeling also limit the ability to simulate air pollution processes in western basins. However, meteorological modeling issues are especially important to address in these cases because meaningful investigations of emissions and chemistry are challenging without reliable meteorological modeling. Prior work has used constrained box-modeling to provide insights on chemical processes in the absence of reliable CTM modeling (e.g., Womack et al., 2019), and over-reliance on box-modeling is recognized an outstanding issue in characterizing chemical processes during PCAPs episodes.

Finally, weather and air quality forecasters have difficulty providing timely and accurate guidance to the public and policymakers regarding PCAP metrics such as their intensity, duration and decay due to the many challenges associated with CTM and meteorological modeling of PCAPs. A careful study design will be employed during AQUARIUS to address the needs for data to validate current models and improve model parameterizations of complex physical processes. By integrating a comprehensive suite of meteorological and chemical measurements with numerical forecast modeling, AQUARIUS will provide an unprecedented characterization of the key physical processes to inform research to improve model emission inventories and chemistry and physics parameterizations.

**Section 3b. Measurements for Model Evaluation**

The coupled three-dimensional meteorological and chemical measurements collected during the AQUARIUS workshop will be used to evaluate, validate, and improve physical parameterizations of CTMs. Typically, only routine monitoring is available to characterize air quality and validate meteorological models in Western basins. Such monitoring is useful in identifying model performance issues but not sufficient for diagnosing the causes of problems at a process level. In limited cases, field studies have been performed to provide measurements for diagnostic model evaluation. However, these efforts have focused on either chemistry (e.g., UWFPS, 2017) or meteorology (e.g., PCAPS) and were not designed to address the effects of the *coupled* meteorological-chemical processes that dominate secondary pollutant formation processes. The goal of AQUARIUS is to couple these observations, and to do so at high resolution, characterize and provide new scientific insight on meteorological-chemical coupling during PCAPs episodes through an integrated campaign of meteorological and chemical measurements. The lack of such measurements limits the detailed model evaluation necessary to begin developing improved model parameterizations.

Characterizing collocated vertical profiles of key meteorology and chemistry variables at many sites will be critical to this effort to evaluate model performance of transport and mass balances within basins and to provide observations to be used to improve the parameterization of processes in models, and ultimately air quality forecasts. Additionally, many of the chemical transport models use the flux-profile relationship for heat or moisture to simulate the pollutant mixing. Therefore, by collecting data for the vertical profiles of pollutant species and turbulent fluxes of the species, model improvements to simulate the pollutant mixing can also be investigated.

To this end, AQUARIUS will focus on process level data observations that can be compared directly high resolution simulations (e.g., LES) and thus facilitate identification of critical shortcomings in subgrid parameterizations for these processes in coarse resolution models. Turbulence measurements have generally been few and far between, despite the importance of turbulence (both within the stable boundary-layer and at the interface with the free atmosphere above the PCAP). Having collocated vertical profile instruments and turbulence sensors will be crucial to obtain the datasets necessary to improve the meteorological parameterization. Detailed turbulence measurements coupled with other meteorological observations will be a focal point during AQUARIUS. For example, Doppler lidar vertical profiles acquired at 1 second temporal resolution and 18 m vertical resolution will provide highly resolved data to characterize turbulence quantification (w’,w’w’, w’w’w’) over the depth the PCAP. Collocated high resolution aerosol backscatter and water vapor mixing ratio observations can then be used to compute covariance terms associated with boundary layer physical processes (e.g., w’q’) and that strongly impact boundary layer development. These data coupled with surface flux observations will then inform model parameterization improvement efforts. Collecting such measurements in multiple Western basins during different large-scale meteorological regimes during AQUARIUS will provide valuable data to aid in the aforementioned process level understanding, since the important processes and types of meteorological-chemical coupling observed are expected to differ among basins due to differences in basin geometry, temperature and moisture profiles, vertical profiles of chemical composition, etc. These observations will yield unprecedented representation of the coupled atmosphere-chemistry system and emission sources in PCAPS.

**Section 3c. Emission Inventories**

Gridded fields of pollutant and precursor emissions are key inputs to the photochemical models used in air quality management. Adequate characterization of emissions can be challenging for the multitude of pollutants and sources present in a typical air basin. An important use of the spatially distributed chemical and meteorological observations will be to provide top-down constraints on the emissions from key sectors in the air basins. The detailed coupled meteorological-chemical observations will be used to improve quantification of known emission sources, potentially identify currently unknown emissions sources, better characterize the temperature and activity dependenceof emissions, and differentiate primary vs. secondary sources for various pollutants (e.g., particulates, formaldehyde). Some specific emission-related sectors to be considered using AQUARIUS data include agricultural, mobile, and natural sources (e.g., soil NO). Spatially distributed surface and aircraft-based measurements of pollutant concentrations will be the primary data source for investigating pollutants emissions. To relate emissions to these ambient concentration measurements, photochemical grid models and other models (e.g., back trajectories) may be used. Also, emissions will be evaluated by using them to drive air quality models and evaluate them during periods outside PCAPs, where models are expected to have less meteorological uncertainties. This will allow for quantification of the uncertainties in the suggested emissions inventories for chemical species such NOx and NH3, which can lead to underestimates of the ammonium nitrate formation in chemical transport models. Thus, it is important to for measurements to be continuous and over an extended period of time capture these conditions improve emissions characterization and make study findings directly relevant to modeling for typical regulatory assessments.

**Section 3c. Photochemical Modeling for Research and Forecasting**

Several coupled meteorology and air pollution CTM modeling studies have been conducted for conditions of stable boundary-layers in California’s Central Valley and several Utah Basins. Studies in California’s Central Valley have included both 3D Eulerian grid models (e.g., Kleeman et al., 2005; Ying et al., 2008a, 2008b, 2009; Zhang et al., 2010; Ying, 2011; Chen et al., 2014; Kelly et al. 2018; Chen et al., 2020) and box models (e.g., Pun et al. 2009). High resolution CTM modeling for the Central Valley appeared to capture the main features and limiting precursor for ammonium nitrate formation during winter PM2.5 episodes. However, several model performance limitations were identified, and many questions remain related to emissions, radical sources, transport, and vertical oxidant entrainment due to the lack of comprehensive coupled and meteorological measurements in previous campaigns.

A major focus of the AQUARIUS photochemical modeling effort will be to evaluate turbulence properties of high-resolution models and carefully validate against high-resolution (spatially and temporally) coupled meteorological-chemical observations (including turbulence quantification as discussed earlier). It is currently not well understood whether using a high-resolution WRF-LES (large-eddy simulation) model during PCAPS would better simulate the turbulence and vertical profiles in the lowest part of the ABL (i.e., <2km) compared to the standard WRF simulations. Having the observational datasets for a rigorous model evaluation of numerical weather prediction models, turbulence models, and chemical transport models will allow for detailed numerical studies to be performed. Not all of the turbulence models and numerical weather prediction models are capable of being coupled to an atmospheric chemistry model, therefore this evaluation will become necessary when assessing which NWP parameterizations work with coupled meteorology-chemistry simulations and which parameterizations would be best to improve.

Another challenge is that model performance varies widely across basin and synoptic weather regime. For the smaller valleys in Utah, accurate modeling with 3-D chemical transport models is more challenging than for the Central Valley of California. As a result, key studies of atmospheric chemistry in Utah valleys have relied on box modeling (e.g., Womack et al., 2019). Many questions remain on how to configure models to adequately simulate meteorology and air quality conditions in Utah valleys during PCAPs episodes. PM2.5 nitrate is typically under-predicted in western basins, including in Utah and California, during winter PM2.5 episodes using operational 12-km national modeling (e.g., Kelly et al., 2019). To improve operational and research modeling for PCAPs, the AQUARIUS campaign seeks to address the full range of challenges in simulating the atmospheric chemistry of pollutant formation in stable wintertime boundary layers, including limitations in parameterizations of chemical processes, representations of photochemistry and radical sources, characterizations of surface albedo and snow cover, and emissions inventories (Zhu et al. 2019). It is difficult to isolate these processes to determine which one (i.e., emissions, chemistry, or meteorology) causes the greatest uncertainty in the air quality modeling for PCAPS. Future studies, with observations from meteorology-chemistry coupled field campaigns would allow for these different processes to be evaluated in a systematic manner to better isolate the uncertainties in the chemical transport modeling.

A number of state and federal agencies provide forecasts which are used for warning the population in advance about poor air quality events. These forecasts are important in terms of forecasting PCAPS with enough advance notice to provide public awareness and subsequent air pollution action advisories, and for appropriate health guidelines such as for school recess. Thus, besides evaluating research grade models specifically configured for this project, it is also necessary to evaluate the operational forecasts systems to provide guidance on their errors and possible ways to improve them. Since these forecasts are available in near-real time, they can be used during the field experiment to have a preliminary assessment of their performance and help in campaign planning. Additionally, modeling groups participating in the field campaign will deploy their own forecasts specifically tailored to the campaign needs. These will include higher resolution in regions with intensive sampling, model configuration that has found to perform better for PCASPs, and preliminary assumptions on how emissions vary with weather conditions. Additionally, the forecasts can be combined into a PCAP “ensemble” that could also be evaluated to assess potential improvements on forecasting these events based on multiple models.

In summary, an integrated observational and modeling approach is needed during AQUARIUS to improve CTM models of PCAPS. High-resolution meteorological modeling should be carefully validated against measurements. First, in the model evaluation will be to determine how accurately the CTMs simulate the formation of secondary pollutants during the study period. If evidence exists that CTM performance issues are related to limitations in model emissions, than attempt to better resolve emissions will be conducted using emission sensitivity simulations or by updating bottom-up emission estimates as appropriate. If evidence exists that performance issues are related to limitations in model chemistry, attempt to resolve those issues by updating model chemical mechanisms using process-based empirical parameterizations or other approaches. Once model simulations of secondary pollutant formation are found to be adequate based on high-resolution modeling, then attempts to parameterize processes such that they can be represented using typical air quality model configurations that might be applied at coarser scales can be developed. The goal is to incorporate understanding developed through the field study into every component of downstream modeling applications for air quality management and health assessments.

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