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

**Content Draft for White Paper**

**Section 1. Overview of the impact of meteorological “cold-air pool” conditions on wintertime air quality in the Western US**

A persistent cold-air pool (PCAP) as deﬁned by Lareau et al. (2013) is a stably-stratiﬁed boundary-layer airmass sheltered from lateral and vertical mixing by the surrounding topography that lasts from days to weeks. PCAPs form most frequently during winter within urbanized and rural basins worldwide when a combination of warming aloft and cooling near the surface lead to stable stratification in the boundary layer ([Dorninger et al. 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878%22%20%5Cl%20%22bb0070), [Reeves et al., 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878#bb0230), Lareau et al., 2013, [Sheridan et al. 2014](https://www.sciencedirect.com/science/article/pii/S0169809516303878#bb0240), Holmes et al. 2015, McCaffrey et al. 2019, Sun and Holmes, 2019, Ivy et al., 2019). PCAPs often lead to elevated levels of particulate air pollution in combination with low clouds, fog, and hazardous ground and air travel (Whiteman et al., 2001, Whiteman et al. 2014, Vanreken et al., 2017, Franchin et al. 2018). Forecasting and numerical modeling of PCAPS are difficult due to many complex and coupled processes, such as the surface heat budget, snow cover, cloud cover, topography, turbulence, temperature advection aloft, and the pressure gradient force (Holtslag,et al. 2011, Lareau et al. 2013, Smith, 2019).

 Generally fair weather arising from high pressure and regional-scale descending motion above the mountain ranges of the Western US contribute to 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) and complex 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 most common in deep, enclosed basins that tend to be sheltered from passing weather disturbances more than shallow or partially enclosed ones (Clements et al. 2003, Vosper et al. 2008, Hoch et al., 2010, Sheridan 2019). Once PCAPs are established, many other factors that affect radiative and 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. Figure 1 illustrates many of these meteorological processes. PCAPS are weakened or destroyed by weather systems accompanied by strong winds, cold temperatures at mountaintop level, or precipitation (Lareau et al. 2013).



**Figure 1**. Placeholder Figure from **DeWekker et al. 2018 for PCAP physical processes.** Do we have a **volunteer** willing to make a summary schematic of the physical processes impacting PCAPS?

 While the meteorological factors that dominate the setup, maintenance, and demise of PCAPs in the western US are broadly known, the complex thermodynamic, radiative, and dynamical processes underway in specific basins and valleys are not well understood, analyzed, or forecasted during PCAP episodes. As illustrated in Fig. 2, the local environment, meteorological boundary-layer processes, and chemical processes are highly intertwined and need to be studied as a coupled system (Baasandorj et al. 2017, Womack et al. 2019; Faloona et al. 2020). The lifecycles of the meteorological and chemical characteristics and processes have been shown to vary considerably during PCAPS. Table 1 illustrates some of the key coupled meteorological and chemical processes that vary during the PCAP lifecycle. **Volunteer** – please work on fleshing out Table 1 and adding references. The AQUARIUS workshop identified several key focus areas where targeted meteorological-chemical coupling observations should be conducted: Large-scale forcing, terrain flows, radiation processes (clouds, albedo, solar angle), vertical and horizontal transport and mixing processes, boundary-layer structure and layering, and clouds (wet vs dry PCAPS) (Table 1 and Figure 2).



**Figure 2.** Schematic of the various coupled meteorological, chemical, and topographical processes that occur in wintertime PCAPs. Please help by coming up with something better!

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| --- | --- | --- |
| **Coupled PCAPS Meteorological Characteristic** | **Coupled PCAPS Chemical Characteristic** | **Changes noted during the lifecycle of the PCAP** |
| Moisture and low clouds/fog | * Cloud/fog chemistry versus particle- and gas-phase chemistry
* Photolysis
* Gas-particle partitioning
 | Water vapor and clouds increase through anthropogenic and natural processes during a PCAP lifecycle. Cloud and fog formation likely enhances the importance of aqueous and heterogenous chemical pathways that are not well-studied during winter PM episodes. Humidity generally promotes uptake of semi-volatile gases by particles, and cloud/fog cover reduces actinic flux and photolysis reactions. |
| Vertical temperature structure and layering of moisture  | Vertical layering of pollutants | PCAP vertical structures are highly complex and variable throughout the lifecycle. Vertical layering of air masses leads to distinct pollutant layers that may be coupled or decoupled from surface emissions and other pollutant layers at different times. The overall effect of vertical meteorological layering on chemical aging and particle growth processes and the response of pollution to emission controls is poorly understood.  |
| Exchange and transport processes | Oxidant and other pollutant and precursor transport | The boundary-layer winds and terrain-driven transport of pollutants, precursor species, and background oxidants has been shown to vary vertically, spatially and temporally. Secondary pollutant formation has been found to be sensitive to oxidant abundance during winter PM episodes. Oxidants may be depleted in isolated atmospheric layers, but entrainment of oxidants from aloft (e.g., the regional background) or from sidewall flows could sustain chemistry that would otherwise be limited by lack of oxidants.  |
| Diurnal meteorological forcing  | Photochemistry | Complex feedbacks between vertical layers, mixing of precursors, and actinic flux. Secondary pollutant formation cycles follow different formation pathways during nighttime and daytime conditions. Vertical mixing may be relatively homogeneous in certain basins during daytime, but the surface layer may be decoupled from residual layer(s) at night. |

**Table 1.** Examples of **coupled meteorological and chemical processes** and noted variations in characteristics over PCAP lifecycle. **Volunteers** – please add to the table.

 A key goal of the AQUARIUS study is to improve our understanding of the production, transformation, cycling, and destruction of chemical species during the life cycle of PCAPs. Establishing an experimental design to provide the meteorological and air chemistry observations to meet this goal is essential. No previous field campaign has provided the breadth and depth of contemporaneous observations at the surface and aloft that could address the complex coupled meteorological and chemistry processes of importance. As we will elaborate in future sections, observing the complexity of coupled chemistry and meteorological processes in topographic basins requires a holistic and novel field study design approach to take advantage of extensive existing sensor networks that will be available at minimal cost to the program as well as deploy diverse sensor types to fill in the gaps using innovative deployment strategies (e.g., plane, in situ and surface-based remote, mobile, drones, IOTs). The science plan factors in the strength and weaknesses of these diverse sensor types for the complex and evolving boundary layer conditions observed during PCAPs. The AQUARIUS field campaign also presents an opportunity to collect data and evaluate forecast model capabilities during the study to improve research and operational simulations of PCAPs. Considerable work is underway or has been conducted in recent years improving model simulations of PCAPs (e.g., Lareau and Horel 2015b, Ahmadov et al. 2015, Foster et al. 2017, Tran et al. 2018, Sun and Holmes 2019; Kelly et al. 2018).

 The coupling between meteorology and chemistry is highly complex. All of the meteorological forcing variables shown in Figure 1 interact with a wide range of chemical processes. PCAPS can be long-lived, lasting one to two weeks, during which a complex and varied array of coupled meteorological and chemical processes occur. The evolution of PCAPS can be characterized by an onset or development phase, mature or steady state phase, and a breakup or decay phase. The meteorological and chemical characteristics of each of these phases vary, thus it is expected that meteorological and chemical process coupling varies similarly through the lifecycle of PCAPS. AQUARIUS observational design will target the entire evolution of PCAPS so that the importance of various processes throughout the lifecycle of PCAPS can be quantified. The development/onset phase of PCAPS is often characterized in colder regions by fresh snow cover, high albedo, cold temperatures, and clear skies and ample solar radiation. As PCAPS mature, fog and stratus may develop along with buildup in secondary particulates and water vapor in the boundary-layer, as well as the lowering of a strong subsidence inversion that typically constrains the vertical depth of the polluted layer. Distinct elevated layers of higher concentrations of particulates and associated chemical precursors often develop during the mature phase, as well as complex inter- and intra-basin transport that may contribute to partial and temporary destruction of the PCAPS in geographically-favored subdomains within them. During the mature phase, many meteorological factors that affect pollutant concentrations are prominent including fluctuations in the depth of the PCAPS, terrain and thermally driven flows, and turbulent mixing laterally and in the vertical. During the breakup or decay phase, the depth of the PCAP typically continues to decrease while concentrations of criteria pollutants may increase.

Additional discussion of the state-of-scientific understanding related to key meteorological-chemical coupling during PCAPs and potential new scientific understanding that would result from coupled meteorological and chemical measurements during the AQUARIUS field campaign for three major topic areas are described below:

*Topic area #1: Solar-radiation-atmospheric boundary-layer-chemistry coupling (****Volunteers please contribute; Hoch, Kelly, Oldroyd, Holmes?)***

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 the influence of snow cover on albedo as well as the presence of fog and cloud during parts of the episode. Variations in actinic flux in the boundary-layer during the winter season are modulated by solar zenith angle, length of daylight, snow cover and other surface properties. These variations subsequently impact boundary-layer temperature structure, vertical and horizontal transport and cycling of pollutants, temperature-dependent chemical processes, and photochemical reaction rates. Greater understanding of radiative feedback processes with respect to coupled meteorology and chemistry is needed during PCAPS. Feedbacks between surface state (e.g., snow cover vs. no snow cover) albedo and photochemical rates have been shown to be important, but more comprehensive evaluation of the coupling between surface 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.

*Topic area #2: Dry-and moist chemistry-atmospheric coupling*. *(****Volunteers please contribute; Lareau, Kelly, Karle?)***

Atmospheric moisture has an important influence on the formation of secondary particulate matter. Under cool, humid conditions, condensation of semi-volatile species onto particles is favorable, whereas such formation is relatively unfavorable under warm, dry conditions. Moreover, the formation of cloud and fog dramatically alters the formation pathways of secondary PM. Aqueous chemistry follows very different pathways in cloud and fog than in aerosol particles, because cloud and fog droplets are highly dilute, whereas particles have high ionic strengths that lead to highly nonideal chemistry (Pye et al., 2019). Urban areas are known to 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. For example, fog in some Utah Valleys is associated with the cessation of PM2.5 growth. Why? Is this related to a change in chemistry, cloud scavenging, or meteorology? Although some work on chemical processes in fogs has been conducted in the past (Collett et al., 1999; Ge et al., 2012; Herckes et al., 2015), a comprehensive understanding of the role of cloud and fog in wintertime PM episodes in western remains elusive and requires an integrated measurement campaign to address. Greater understanding of these feedbacks between aerosols, water vapor, clouds and fog, and atmospheric boundary-layer structure and evolution is needed. How does the chemistry vary between wet (cloudy) and dry (non-cloudy) PCAPS? Does fog impact sulfur or organic oxidation? Does aerosol chemistry differ during high humidity conditions? Does different chemistry occur during shallow near-surface fog versus an elevated stratus layer?

*Topic area #3: Boundary-layer-transport processes-chemistry coupling*. *(****Volunteers please contribute; Hoch, Kelly, DeWekker, Karle, Faloona, Holmes, Oldroyd?)***

The chemical processes that lead to secondary pollutant formation are sensitive to the mixing of diverse air masses that may have undergone varing degrees of local chemical aging or are primarily composed of fresh emissions or regional background air. Pollutant formation chemistry may be favorable only in certain parts of the vertical column due to titration effects of NOx emissions in the shallow surface layer or the limited oxidant abundance in certain vertical layers. 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. The coupling between vertical meteorological properties within PCAPS and vertical profiles of chemical precursors and particulate pollution are a key research area for AQUARIUS. Quantifying vertical and horizontal airmass trajectories and their coupling with chemical processes is needed. For example, what are the relative importance of sidewall ventilation, daytime PBL growth and vertical mixing, and horizontal advection processes for transporting chemical precursors within multi-layered PCAPs? How many distinct chemical-meteorological layers exist and what are the time scales at play for transport processes within these layers? How much do these processes vary between basins, over time, with height and depth of PCAP, etc? Turbulence is a key meteorological process that is intrinsically linked to the vertical mixing processes within PCAP. Characterization of spatial and temporal variations in turbulence within PCAPS is a difficult but critical task for understanding vertical transport processes and meteorological coupling.

*Topic area #4: Inter-basin, intra-basin, and sidewall/canyon exchange-chemical coupling. (****Volunteers please contribute; Stutz, Simpson, Hoch, Kelly, Oldroyd, DeWekker, Faloona?)***

Secondary pollutant formation depends on the availability of oxidants to drive atmospheric oxidation mechanisms. In isolated air masses, pollutant formation processes may terminate when the key oxidants become depleted. Replenishment of oxidants through entrainment from aloft or sidewall mixing can promote sustained production of secondary pollutants throughout a PCAPs episode. Episodes that terminate through oxidant depletion may respond favorably to VOC emission controls, whereas episodes that experience oxidant replenishment might be insensitive to VOC 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 meteorological horizontal transport processes on the chemical processes within PCAPS (e.g., Baasandorj et al. 2017). In Utah basins oxidant injection from sidewall canyons or agricultural ammonia from inter-basin transport have been two recent topics of interest. The scales of basins have a large impact on the relative importance of inter (between) and intra (within) basin. Workshop participants agreed that many previous field studies have not have the spatial data collected of both meteorological and chemical measurements to answer the above questions. Different regions within basins will have different emissions and chemistry, as well as variations in meteorology. Better understanding of spatial variations in thermodynamic/dynamic and chemical emissions and processes are all needed in order to tackle these important questions. Care as to how to sample the canyon and slope regions is needed in order to design experiments of exchange processes along the valley slopes (Oldroyd et al. 2016).

**Section 2. Recommended Study Design for Ground-based Vertical Profiles**

**Section 2a. Introduction (*Volunteers please contribute)***

Ground-based vertical profiles of atmospheric and chemical species will be a cornerstone observational platform for understanding meteorological and chemical coupling during the AQUARIUS field campaign. In order to address the AQUARIUS science questions pertaining to coupled meteorology and chemistry, the study design for ground-based vertical profiles for meteorological-chemical coupling must take into account a number of important points, including spatial representation, *in situ* and remote sensing methods, and spatial collocation of meteorological and chemical measurements. An overview of potential observational platforms to be deployed and their purposes are illustrated graphically in Figure. 3.



Figure 3. Schematic of recommended ground-based vertical profiling instruments to be used during AQUARIUS. (**Hoch** can you make one for us?)

**Section 2b. Co-located Atmospheric and Chemical Measurements (*Volunteers*** please contribute).

The ground-based vertical profiles within basins studied during AQUARIUS must adequately resolve both the intra-basin and inter-basin chemical and meteorological properties and transport processes. It is therefore critical that the vertical profiles collected during the AQUARIUS field study include collocated meteorological and atmospheric chemistry instrumentation as much as possible. The coupled atmospheric and chemical variability, meteorological and chemical exchanges, vertical mixing processes, and vertical profiles of meteorological and chemical species must be adequately resolved in both time and space such that the detailed relationships between meteorological and chemical processes can be determined. This approach will also allow mass budgets of basin air masses to be accurately determined. Obviously, some difficult choices about which regions get sampled and which are not sampled by both meteorological and air chemistry instrumentation will need to be made as the location, scope, and size of the AQUARIUS study is finalized.

**Section 2c. Basin Vertical Profiling Instrumentation (*Volunteers*** please contribute and list and describe your vertical profiling instruments **Oldroyd, Holmes, DeWekker, Hoch, Stutz, Simpson**)

All available types of vertical profiling instrumentation, both *in situ* and remote sensing, will need to be utilized during AQUARIUS to address the science questions. Surface observation sites will also need to be situated in a way that their observations can be related to vertical profile data.

 A wide array of vertical profiling instrumentation will need to be utilized, keeping carefully in mind the conditions under which some profiling platforms cannot be utilized due to flight restrictions or weather conditions. High towers, tall buildings, or terrain slopes on the edges of basins will also need to be utilized for obtaining vertical profiles of some atmospheric and chemical measurements.

 Many turbulence measurements (both vertically and horizontally) will be needed in order to capture the impact of various meteorological processes (e.g., canyon or slope flows) on the turbulence budget to then couple with chemical process observations

 Vertical remote sensing will be a critical component of adequately observing the vertical and spatial evolution of boundary-layer meteorology and the depth and layering of particulate pollutants. Ground-based lidars, ceilometers, sodars, and other remote sensing instruments will be carefully deployed in order to get a full picture of meteorological and chemical coupling. The types of sensors include:

* Aerosol lidar (air pollution)
* Aerosol ceilometer (air pollution and meteorology)
* Wind lidar (meteorology)
* Wind sodar (meteorology)
* Radiometer (meteorology)
* DOAS

 Vertical *in-situ* sensing will also be utilized in AQUARIUS observational design. Multiple rawinsonde systems, tethersondes, and small drones are all recommended and would help supplement the remote sensing systems. However, in some basins federal aviation regulations may make most drones and tethersondes unusable. The types of sensors include:

* Rawinsonde vertical profiles (meteorology)
* Weather stations deployed along basin slopes (meteorology)

**Section 2d. Basin Vertical Profiling Deployments (*Volunteers, Stutz, Simpson, Hoch, Oldroyd, Holmes, DeWekker…)*** please contribute.

Should we do a comparison of Cache, SLV, and CA Central Valley?)

The size of the basins studied will likely 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, a number of vertical profiling ground based sites are likely needed to capture spatial variations in meteorology and chemistry adequately. Vertical profiles at the boundaries of the basins will assist in quantifying the aforementioned 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)

 Sufficient data collection to ensure spatial gradients are captured across the basin of interest is important. There must be adequate observation locations spatially (and with adequate temporal consistency) that a 3-D 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, and ceilometers will be supplemented with instrumentation dedicated to AQUARIUS.

 Chemical processes driven by mixing 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) data on towers and high buildings at multiple locations within the basins are needed. For example, in the Salt Lake Valley, locations such as the KSL radio towers near the Great Salt Lake, the LDS building downtown, and the University of Utah WBB building that houses the Department of Atmospheric Sciences have been identified as potential sites.

**Section 3. AQUARIUS Model Improvement Goals (*Volunteers*** please contribute to this section. Particularly the modelers **Saide, Kelly, Holmes, Mallia, Oldroyd**).

**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. 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 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 model simulations of PCAPs, considerable work is underway or has been conducted in recent years (e.g., Lareau and Horel 2015b, Ahmadov et al. 2015, Foster et al. 2017, Tran et al. 2018, Sun and Holmes 2019; Kelly et al. 2018). 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%22%20%5Cl%20%22bb0025), [Price et al., 2011](https://www.sciencedirect.com/science/article/pii/S0169809516303878#bb0220), Holmes et al. 2015). Issues with emission inventories and chemical mechanisms also limit the ability to simulate air pollution processes in western basins. However, meteorological modeling issues are especially important to address 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 experts at the AQUARIUS workshop indicated that reliance on box-modeling is an outstanding issue in characterizing chemical processes during PCAPs episodes. Moreover, wsissueswith CTM modeling of s

address the three inter-related factors of importance for air quality simulation for challenging PCAPs conditions: (1) model meteorology (mixing and transport), (2) emission inventories and modeling, and (3) atmospheric chemistry. A careful study design will be used to address these factors by integrating a comprehensive suite of meteorological and chemistry measurementswithnumerical forecast .This novel approach willfacilitate a thorough characterization of the key PCAPs processes to inform research emission inventories and chemistry and parameterizations

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

The AQUARIUS workshop identified a clear need to perform comprehensive measurements of coupled meteorological-chemical processes to improve air quality characterization and inform rigorous evaluation and validation of photochemical models. Typically, only routine is monitoring is available to characterize air quality and meteorological 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 casesstudies have been performed to provide measurements for diagnostic model evaluation. However, these 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 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. Collectingcharacterizeinform d parameterizations will also be especially important Ideally, such measurements will be collected in multiple Western basins because processes are expected to differ among basins due to differences in basin geometry and emission sources and other factors.

**Section 3c. Improving 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. Such integration of modeling and measurements will s characterization and make study findings directly relevanttofor typical regulatory assessments

**Section 3c. Needed Photochemical Model Improvements**

Several coupled meteorology and air pollution 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 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. For the smaller valleys in Utah, accurate modeling with 3-D chemical transport models is more challenging than for the Central Valley. 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 underpredicted 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).

**Section 3d. AQUARIUS Modeling Goals**

The recommended model improvement goals for AQUARIUS to are as follows:

* Collect detailed and spatially comprehensive meteorological observations, including 3-D turbulence, that are sufficient to validate the model and determine model weaknesses and assist in future turbulence parameterization development and evaluation. 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).
* Build a real-time suite (array) of forecast models that can be blended into a PCAP “ensemble” that can be validated and evaluated and utilized as forecast support for the science team while addressing forecasting and model improvement goals.
* Evaluate turbulence properties of high-resolution models and carefully validate against unprecedented coupled meteorological-chemical observations collected during AQUARIUS.
* Using high-resolution meteorological modeling that has been carefully validated against measurements, simulate the formation of secondary pollutants during the study period. If evidence exists that performance issues are related to limitations in model emissions, attempt to resolve those issues 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 adequate based on high-resolution modeling, attempt to parameterize processes such that they can be represented using typical air quality model configurations that might be applied at coarser scales. The goal is to incorporate understanding developed through the field study into downstream modeling applications for air quality management and health assessments.

**REFERENCES**

Ahmadov, R., et al. (2015), Understanding high wintertime ozone pollution events in an oil‐ andnatural gas‐producing region of the western US, *Atmos. Chem. Phys.*, **15**( 1), 411– 429, doi:[10.5194/acp-15-411-2015](https://doi.org/10.5194/acp-15-411-2015).

Baasandorj, M., Hoch, S. W., Bares, R., Lin, J. C., Brown, S. S., Millet, D. B., Martin, R., Kelly, K., Zarzana, K. J., Whiteman, C. D., Dube, W. P., Tonnesen, G., Jaramillo, I. C., and Sohl, J.: Coupling between Chemical and Meteorological Processes under Persistent Cold-Air Pool Conditions: Evolution of Wintertime PM2.5 Pollution Events and N2O5 Observations in Utah's Salt Lake Valley, Environ. Sci. Technol.,(2017) 51, 5941–5950, <https://doi.org/10.1021/acs.est.6b06603>.

Baklanov, A. A., and Coauthors, 2011: The nature, theory, and modeling of atmospheric planetary boundary layers. *Bull. Amer. Meteor. Soc.*, **92**, 123–128.

Clements, C.B., Whiteman, C.D. and Horel, J.D. (2003) Cold‐air‐pool structure and evolution in a mountain basin: Peter Sinks. Journal of Applied Meteorology and Climatology, **42**, 752– 768

DeWekker, S.F.J.; Giovannini, L.; Gutmann, E.; Knievel, J.C.; Kossmann, M.; Zardi, D. Meteorological applications benefiting from an improved understanding of atmospheric exchange processes over mountains. Atmosphere **2018**.

M. Dorninger, C.D. Whiteman, B. Bica, S. Eisenbach, B. Pospichal, R. Steinacker. **Meteorological events affecting cold-air pools in a small basin**. J. Appl. Meteorol. Climatol., 50 (2011), pp. 2223-2234.

Faloona, I.C., S. Chiao, A.J. Eiserloh, R.J. Alvarez, G. Kirgis, A.O. Langford, C.J. Senff, D. Caputi, A. Hu, L.T. Iraci, E.L. Yates, J.E. Marrero, J. Ryoo, S. Conley, S. Tanrikulu, J. Xu, and T. Kuwayama, 0: [The California Baseline Ozone Transport Study (CABOTS).](https://journals.ametsoc.org/doi/abs/10.1175/BAMS-D-18-0302.1) *Bull. Amer. Meteor. Soc.,* **0**, <https://doi.org/10.1175/BAMS-D-18-0302.1>

C. Foster, E. Crosman, J. Horel. **Simulations of a cold-air pool in Utah's Salt Lake Valley: sensitivity to land use and snow cover**. Boundary-Layer Meteorol. (2017) **164**, pages63–87(2017)

A. Franchin, D.L. Fibiger, L. Goldberger, E.E. McDuffie, A. Moravek, C.C. Womack, E.T. Crosman, K.S. Docherty, W.P. Dube, S.W. Hoch, B.H. Lee, R. Long, J.G. Murphy, J.A. Thornton, S.S. Brown, M. Baasandorj, A.M. Middlebrook. **Airborne and ground-based observations of ammonium-nitrate-dominated aerosols in a shallow boundary layer during intense winter pollution episodes in northern Utah.**

Hoch, S.W. and Whiteman, C.D. (2010) Topographic effects on the surface radiation balance in and around Arizona's Meteor Crater. Journal of Applied Meteorology and Climatology, **49**, 1114– 1128

Holmes, H. A., J. K. Sriramasamudram, E. R. Pardyjak, and C. D. Whiteman, 2015: Turbulent fluxes and pollutant mixing during wintertime air pollution episodes in complex terrain. Environ. Sci. Technol., **49**, 13 206–13 214, <https://doi.org/10.1021/acs.est.5b02616>.

Horel, J., and Coauthors, 2002: MesoWest: Cooperative mesonets in the western United States. Bull. Amer. Meteor. Soc., **83**, 211–225.

Holtslag, A.A.M., Svensson, G., Baas, P., Basu, S., Beare, B., Beljaars, A.C.M., Bosveld, F.C., Cuxart, J., Lindvall, J., Steeneveld, G.J., Tjernström, M., Van De Wiel, B.J.H., Holtslag, A.A.M., Svensson, G., Baas, P., Basu, S., Beare, B., Beljaars, A.C.M., Bosveld, F.C., Cuxart, J., Lindvall, J., Steeneveld, G.J., Tjernström, M. and Van De Wiel, B.J.H. (2013) Stable atmospheric boundary layers and diurnal cycles: challenges for weather and climate models. Bulletin of the American Meteorological Society, **94**(11), 1691–1706. <https://doi.org/10.1175/BAMS-D-11-00187.1>.

C.E. Ivey, S. Balachandran, S. Colgan, Y. Hu, H.A. Holmes. **Investigating fine particulate matter sources in Salt Lake City during persistent cold air pool events**. Atmos. Environ., 213 (2019), pp. 568-578

James T. Kelly, Shannon N. Koplitz, Kirk R. Baker, Amara L. Holder, Havala O.T. Pye, Benjamin N. Murphy, Jesse O. Bash, Barron H. Henderson, Norman C. Possiel, Heather Simon, Alison M. Eyth, Carey Jang, Sharon Phillips, Brian Timin, 2019. Assessing PM2.5 model performance for the conterminous U.S. with comparison to model performance statistics from 2007-2015, Atmospheric Environment, 214, 116872, ISSN 1352-2310,

<https://doi.org/10.1016/j.atmosenv.2019.116872>.

Kelly, J. T., Parworth, C. L., Zhang, Q., Miller, D. J., Sun, K., Zondlo, M. A., Baker, K. R., Wisthaler, A., Nowak, J. B., Pusede, S. E., Cohen, R. C., Weinheimer, A. J., Beyersdorf, A. J., Tonnesen, G. S., Bash, J. O., Valin, L. C., Crawford, J. H., Fried, A., & Walega, J. G. (2018). Modeling NH4NO3 over the San Joaquin Valley during the 2013 DISCOVER‐AQ Campaign. Journal of Geophysical Research: Atmospheres, 123, 4727– 4745. <https://doi.org/10.1029/2018JD028290>

Lareau, N. P., E. Crosman, C. D. Whiteman, J. D. Horel, S. W. Hoch, W. O. J. Brown, and T. W. Horst (2013), The persistent cold‐air pool study, Bull. Am. Meteorol. Soc., 94( 1), 51– 63, doi:10.1175/Bams‐D‐11‐00255.1

Lareau, N.P. and Horel, J.D. (2015)a  Dynamically induced displacements of a persistent cold‐air pool. Boundary‐Layer Meteorology, **154**(2), 291–316. <https://doi.org/10.1007/s10546-014-9968-5>.

Lareau, N.P. and J.D. Horel, 2015b: [Turbulent Erosion of Persistent Cold-Air Pools: Numerical Simulations.](https://journals.ametsoc.org/doi/abs/10.1175/JAS-D-14-0173.1) J. Atmos. Sci., **72**, 1409–1427, <https://doi.org/10.1175/JAS-D-14-0173.1>

McCaffrey, K., J.M. Wilczak, L. Bianco, E. Grimit, J. Sharp, R. Banta, K. Friedrich, H.J. Fernando, R. Krishnamurthy, L.S. Leo, and P. Muradyan, 2019: [Identification and Characterization of Persistent Cold Pool Events from Temperature and Wind Profilers in the Columbia River Basin.](https://journals.ametsoc.org/doi/abs/10.1175/JAMC-D-19-0046.1) J. Appl. Meteor. Climatol., **58**, 2533–2551, <https://doi.org/10.1175/JAMC-D-19-0046.1>

E.M. Neemann, E.T. Crosman, J.D. Horel, L. Avey. **Simulations of a cold-air pool associated with elevated wintertime ozone in the Uintah Basin, Utah**. Atmos. Chem. Phys., 15 (2015), pp. 135-151

Oldroyd, H.J., Pardyjak, E.R., Higgins, C.W. et al. Buoyant Turbulent Kinetic Energy Production in Steep-Slope Katabatic Flow. Boundary-Layer Meteorol **161,**405–416 (2016). <https://doi.org/10.1007/s10546-016-0184-3>

Pun, B., R. Balmori, and C. Seigneur (2009), Modeling wintertime particulate matter formation in central California, Atmos. Environ., 43( 2), 402– 409, doi:[10.1016/j.atmosenv.2008.08.040](https://doi.org/10.1016/j.atmosenv.2008.08.040).

H.D. Reeves, K.L. Elmore, G.S. Manikin, D.J. Stensrud**. Assessment of forecasts during persistent valley cold pools in the Bonneville Basin by the North American Mesoscale Model**. Wea. Forecasting, 26 (2011), pp. 447-467, [10.1175/WAF-D-10-05014.1](https://doi.org/10.1175/WAF-D-10-05014.1)

Smith, R. B., 2019: 100 years of progress in mountain meteorology research. A Century of Progress in Atmospheric and Related Sciences: Celebrating the American Meteorological Society Centennial, Meteor. Monogr., No. 59, Amer. Meteor. Soc., <https://doi.org/10.1175/AMSMONOGRAPHS-D-18-0022.1>

Salmon, O. E., Shepson, P. B., Ren, X., Marquardt Collow, A. B., Miller, M. A., Carlton, A. G., Cambaliza, M. O., Heimburger, A., Morgan, K. L., Fuentes, J. D., & Stirm, B. H. (2017). Urban emissions of water vapor in winter. *Journal of Geophysical Research: Atmospheres*, 122, 9467– 9484. <https://doi.org/10.1002/2016JD026074>

Sheridan PF (2019) Synoptic-flow interaction with valley cold-air pools and effects on cold-air pool persistence: influence of valley size and atmospheric stability. Q J R Meteorol Soc 145:1636–1659. <https://doi.org/10.1002/qj.3517>

P.F. Sheridan, S.B. Vosper, A.R. Brown **Characteristics of cold pools observed in narrow valleys and dependence on external conditions**. Q. J. R. Meteorol. Soc., 140 (2014), pp. 715-728, [10.1002/qj.2159](https://doi.org/10.1002/qj.2159)

Sun, X. and H.A. Holmes, 2019: [Surface Turbulent Fluxes during Persistent Cold-Air Pool Events in the Salt Lake Valley, Utah. Part I: Observations.](https://journals.ametsoc.org/doi/abs/10.1175/JAMC-D-19-0053.1) J. Appl. Meteor. Climatol., **58**, 2553–2568, <https://doi.org/10.1175/JAMC-D-19-0053.1>

T. Tran, H. Tran, M. Mansfield, S. Lyman, E. Crosman. **Four dimensional data assimilation (FDDA) impacts on WRF performance in simulating inversion layer structure and distributions of CMAQ-simulated winter ozone concentrations in Uintah Basin**

VanReken, T. M., and Coauthors, 2017: Role of persistent low-level clouds in mitigating air quality impacts of wintertime cold pool conditions. Atmos. Environ., **154**, 236–246, [https://doi.org/10.1016/j.atmosenv.2017.](https://doi.org/10.1016/j.atmosenv.2017.01.043)

Vosper, S.B. and Brown, A.R. (2008) Numerical simulations of sheltering in valleys: the formation of nighttime cold‐air pools. Boundary‐Layer Meteorology, **127**(3), 429–448. <https://doi.org/10.1007/s10546-008-9272-3>.

Whiteman, C. D., S. W. Hoch, J. D. Horel, and A. Charland, 2014: Relationship between particulate air pollution and meteorological variables in Utah’s Salt Lake Valley. Atmos. Environ., **94**, 742–753, <https://doi.org/10.1016/j.atmosenv.2014.06.012>.

Womack, C.C., E.E . McDuffie, P.M. Edwards, R. Bares, J.A. de Gouw, K.S. Docherty, W.P. Dubé, D.L. Fibiger, A. Franchin, J.B. Gilman, L. Goldberger, B.H. Lee, J.C. Lin, R. Long, A.M. Middlebrook, D.B. Millet, A. Moravek, J.G. Murphy, P.K. Quinn, T.P. Riedel, J.M. Roberts, J.A. Thornton, L.C. Valin, P.R. Veres, A.R. Whitehill, R.J. Wild, C. Warneke, B. Yuan, M. Baasandorj, S.S. Brown (2019). **An odd oxygen framework for wintertime ammonium nitrate aerosol pollution in urban areas: NOx and VOC control as mitigation strategies** (2019). 46, 4971–4979, Geophys. Res. Lett, [10.1029/2019GL082028](https://doi.org/10.1029/2019GL082028)

Whiteman, C. D., S. Zhong, W. J. Shaw, J. M. Hubbe, X. Bian, and J. Mittelstad 2001: Cold pools in the Columbia basin. Wea. Forecasting, **16**, 432–447.

Yu, L., Zhong, S. and Bian, X. (2017) Multi‐day valley cold‐air pools in the western United States as derived from NARR. International Journal of Climatology, **37**(5), 2466–2476. <https://doi.org/10.1002/joc.4858>.

Zhu, S, Mac Kinnon, M, Shaffer, BP, Samuelsen, G, Brouwer, J and Dabdub, D. 2019. An uncertainty for clean air: Air quality modeling implications of underestimating VOC emissions in urban inventories. Atmos Environ 211: 256–267. DOI: [10.1016/j.atmosenv.2019.05.019](https://doi.org/10.1016/j.atmosenv.2019.05.019)