**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 well 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 | Aqueous versus dry phase chemistry | Water vapor and clouds increase through anthropogenic and natural processes during a PCAP lifecycle, forming fog and potential wet chemistry processes that are not well-studied. |
| Vertical layering of temperature and moisture  | Vertical layering of pollutants | PCAP vertical structures are highly complex and variable throughout the lifecycle. How chemical aging and particle growth processes couple to vertical meteorological processes is unknown. |
| 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.  |
| Diurnal meteorological forcing  | Photochemical chemistry | Complex feedbacks between vertical layers, mixing of precursors, and actinic flux. |

**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 establish an experimental design that will lead to the meteorological and air chemistry observations that are necessary to improve our understanding of the production, transformation, cycling, and destruction of chemical species during the life cycle of PCAPs. No previous field campaign has provided the breadth and depth of contemporaneous observations at the surface and aloft that could address that goal. 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. 2019).

 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?)***

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?)***

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 chemistry or is it meteorology? 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 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?)***

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). 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.

**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 need to have as much meteorological instrumentation be collocated with air chemistry observations 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)
* Long-path Differential Optical Absorption Spectroscopy LP-DOAS (pollution, mixing)

 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. Long-path DOAS using an array of retro-reflectors at different altitudes (e.g. deployed on a tower) allows for continuous observations of vertical profiles of gases such as NO2, SO2, O3, HONO, HCHO, and NO3. The profiles of these gases probe both vertical mixing processes and chemical transformations.

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

**Section 3a. Introduction**

The AQUARIUS study seeks to collect observations and complete modeling efforts that are designed to collect the data required to improve model physics and emissions inventories and also inform emissions and chemistry research. Considerable work is underway or has been conducted in recent years improving research 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. 2019). 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). As a result, air quality forecasts during PCAPS are frequently inaccurate due to the meteorological model errors.

 The noted errors in wintertime air pollution chemical modelling studies result largely from a combination of three interrelated factors: 1) poor emissions inventories, 2) inadequate model meteorology (mixing and transport) and 3) inadequate model chemistry. In order to improve wintertime air quality models, all three of these subtopics must be directly addressed by AQUARIUS. Weather and air quality forecasters have difficulty providing timely and accurate guidance regarding PCAPS metrics such as their intensity, duration and decay due to the aforementioned errors often noted in meteorological and photochemical models during the PCAP episodes. The AQUARIUS field campaign will seek to incorporate a numerical forecasting component to observational and research modeling efforts.

**Section 3b. Adequate Observations to Evaluate Models**

A key outcome of the AQUARIUS workshop is the identification of a clear need to improve coupled meteorological-chemical observations so that more rigorous evaluation and validation of photochemical models can be conducted, providing the data needed to develop improved turbulence and other physical process parameterizations with which to improve the models. Observations of *coupled* meteorological-chemical processes to validate these models or emission inventories in Western US basins in wintertime are sparse. Generally, field efforts have been focused on either chemistry (e.g., UWFPS, 2017) or meteorology (e.g., PCAPS). The goal of AQUARIUS is to adequately couple these observations such as to provide the needed data to improve the models (and emissions inventories) and also shed new scientific insight into meteorological-chemical coupling. It is critical in collecting the vertical profiles that the meteorological instrumentation be collocated with air chemistry observations at as many sites as possible.

**Section 3c. Improving Emission Inventories**

A key extension of spatially coherent quantification of meteorological-chemical coupling through spatially distributed chemical and meteorological observations will be the concurrent capability to develop strategies to quantify emissions and ultimately improve emissions inventories for modeling and regulatory purposes. The detailed coupled meteorological-chemical observations will allow for quantification of unknown emission sources, improved quantification of currently known sources, and to quantify the temperature and photochemical dependenceof emissions and to differentiate primary vs. secondary sources for various pollutants (e.g., particulates, formaldehyde). Some additional topic areas that need to be addressed with respect to emissions during AQUARIUS include agricultural, mobile, and natural sources. Emissions data need to be determined from spatially distributed surface and aircraft-based emissions observations.

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

A number of coupled meteorological and air pollution chemical modelling studies (both 3-D and box models) have been conducted in stable boundary-layers in California’s Central Valley and several Utah Basins. These have included both 3-D (e.g., Pun et al. 2009; Kelley et al. 2019) and box models (e.g., Womack et al. 2019). These models in most cases struggled to accurately predict wintertime primary and secondary aerosol pollution formation within these basins.

 Some of the key difficulties in modeling the photochemistry and resultant pollution concentrations in stable wintertime boundary layers include inadequate parameterizations of chemical processes, poor representation of photochemistry and surface albedo, and inaccurate 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.

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