Recent Research Directions in U.S. Winter Air Quality:

Progress and Challenges

An overview of research efforts that focus on winter air quality, an important issue in the United States and elsewhere.

Fine particulate matter (particulate matter with dimeter less than 2.5 microns; $PM_{2.5}$) is an important worldwide health and air quality issue.[1] Recent dramatic decreases in emissions of major air pollutants have resulted in significant reductions in $PM_{2.5}$ mass across the United States. Annual emissions of sulfur dioxide (SO_2) have decreased by over 90% since 1990, while nitrogen oxides (NOx = NO + NO_2) and volatile organic compounds (VOCs) have fallen by 60% and over 30%, respectively.[2] At the same time, however, the seasonality of U.S. $PM_{2.5}$ has shifted. Urban areas exhibit both a summer and a winter maximum in $PM_{2.5}$, but since the year 2000 the summer maximum has declined while the winter maximum has remained relatively constant, leading to higher average levels of $PM_{2.5}$ in winter than summer.[3] Chemical speciation of $PM_{2.5}$ shows nationwide declines in sulfate during the summer, but lesser or negligible declines in nitrate and organic carbon that are more prevalent during winter, particularly in the western United States. Similar trends are apparent in other regions with recently declining emissions (e.g., Eastern China).[4]

This summer to winter shift in U.S. $PM_{2.5}$ with declining emissions suggests complex changes in atmospheric chemical mechanisms that convert precursor gases into secondary PM. For example, while SO_2 reductions lead to overall reductions in sulfate, they may also lead to unintended increases in nitrate due to the replacement of sulfate by nitrate at relatively constant ammonia levels, an effect that is pronounced in winter. Emissions changes also lead to responses in the availability of oxidants that may actually increase oxidation rates even as levels of precursor gases decline. For example, recent models suggest that heterogeneous (i.e., gas to particle or droplet reactions) sulfur oxidation has increased during winter. Oxidation of NOx occurs through both gas phase and heterogeneous mechanisms that are known to be strong, non-linear functions of the NOx emissions themselves. Winter oxidation rates of VOCs leading to particulate organic carbon remain poorly defined but may be regionally significant.

Although the national trend in $PM_{2.5}$ emissions and response is clear, winter air quality issues also remain region-specific and responsive to local emission sources. For example, Fairbanks, Alaska is well known for high winter $PM_{2.5}$ likely resulting from a combination of residential wood combustion and heating oil use. Contrasting to Fairbanks is Salt Lake City, Utah, which has high winter ammonium nitrate resulting from a combination of urban and agricultural emissions in confined valleys. Denver, Colorado suffers from winter haze that is similar in composition and origin to that of Salt Lake City, but that does not currently exceed regulatory standards. California's San Joaquin Valley (SJV) has severe winter $PM_{2.5}$ resulting from widespread agricultural emissions and a collection of smaller urban areas. Emissions of VOCs from the oil and gas industries have led to recent, unusual events of high wintertime ozone in sparsely populated mountain basins of Utah and Wyoming. The northeastern United States has winter $PM_{2.5}$ that is generally below regulatory standards but due to a mix of mobile source, home heating, electric power generation and agricultural emissions across a wide region. The midwestern United States suffers from periodic episodes of ammonium nitrate during cold periods.

Common to all of these regional $PM_{2.5}$ issues is winter meteorology, characterized by generally shallow boundary layers that confine surface level emissions in a more concentrated layer near the surface than is typical in summer. This effect is particularly pronounced in the western United States, where meteorological inversions below the confining terrain of mountain basins lead to multi-day stagnation events known as persistent cold air pools (PCAPs). The interaction between boundary layer meteorology, emission sources and atmospheric chemical cycles is extremely complex. Mixing of surface level emissions throughout the boundary layer during daytime is weaker and shorter in duration, and vertical stratification from early evening through mid-morning leads to differences in conversion rates of precursor gases to secondary pollutants as a function of height. These effects are particularly important for ammonium nitrate, a common component of winter $PM_{2.5}$.

Measurement of chemical composition as a function of height above ground are important to understanding these interactions between atmospheric chemistry and meteorology. Field intensives with highly instrumented aircraft provide the most detailed characterization of these processes but are expensive and difficult to carry out. A particular challenge is the requirement for sustained low altitude flights in the range characteristic of winter boundary layers from 150–800 m (~500–2,500 feet) above ground level.

Several recent aircraft-based field intensives have investigated

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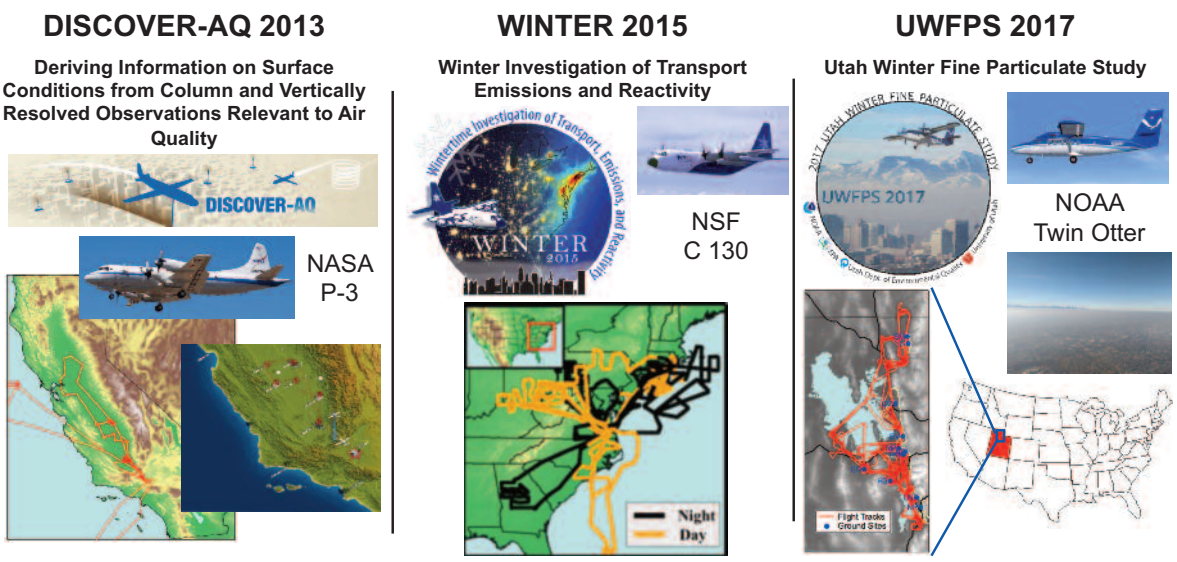

Figure 1. Overview of recent aircraft-based winter field intensives in the United States.

winter atmospheric chemistry and air quality in the United States, as Figure 1 illustrates. The DISCOVER-AQ campaign (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) used the National Aeronautics and Space Administration (NASA) P-3 aircraft to investigate air quality in the SJV region of California during January–February 2013. The sampling strategy included daytime flights with a repeated flight pattern that incorporated vertical profiles from 3,000 m to the surface using low approaches at airfields. Major findings demonstrated the response of $PM_{2.5}$ to decreasing NOx emissions and suggested that declining NOx will shift the mechanism for ammonium nitrate production from nighttime to daytime dominated.[5,6]

The WINTER campaign (Wintertime Investigation of Emissions, Transport and Reactivity) surveyed the eastern United States with the heavily instrumented NCAR/NSF C-130 aircraft during February–March 2015. Flights sampled a wide region, from the Ohio River Valley to the waters off the coast of New England, and as far south as Atlanta, Georgia and coastal Florida. Flights were evenly distributed between nighttime and daytime, with 70% of the ~100 research flight hours in the boundary layer below 1-km altitude. Major findings showed that wintertime emissions inventories, particularly for NOx, appear to be accurate,[7] that chemical mechanisms for nighttime NOx oxidation and halogen activation are not well represented in models[8] and that winter organic aerosol is widespread.[9]

The UWFPS campaign (Utah Winter Fine Particulate Study) investigated the factors governing high levels of $PM_{2.5}$ in the mountain basins of northern Utah during January–February 2015. The study utilized the much smaller National Oceanic and Atmospheric Administration (NOAA) Twin Otter, which executed 23 separate short (~3 hour) research flights during both daytime and nighttime, sampling an altitude range from the surface (~1,300 m at Salt Lake City) through approximately 4,000 m with a limited chemical payload. Major findings showed that the region as a whole is primarily nitrate limited but that ammonia limitation may be important in the urban area of Salt Lake City.[10] A NOx–VOC analysis that is more typically used in the analysis of summertime ozone showed that initial VOC control may be a more effective s trategy for reducing $PM_{2.5}$ than NOx control.[11]

Despite these recent field intensives, multiple scientific questions remain. These include the emission sources most relevant to winter $PM_{2.5}$ as overall precursor emissions continue to decline, the resulting response of chemical mechanisms for production of secondary sulfate, nitrate and organic carbon and improved measurement and modeling of wintertime boundary layer meteorology. Future field intensives are currently in the planning stages to address these issues. The forthcoming ALPACA study (Alaskan Pollution and Chemical Analysis; https://alpaca.community.uaf.edu), currently planned for the winter of 2020–2021, will provide the first chemically detailed investigation of pollution events in that area. The AQUARIUS study (Air Quality Research in the western United States; https://www.atmos.utah.edu/aquarius/index.php) is in early stages of planning to provide both aircraft and ground based measurements of mountain basins subject to cold pool meteorology and air quality issues across the western United States, with a focus on the exceedance areas in the San Joaquin Valley and Salt Lake City regions. The tentative time frame for this study is the winter of 2022–2023.

Winter air quality continues to be an important issue in the

United States and other nations. Domestic and international research efforts focused on this problem are intended to provide improved scientific understanding of the relevant emissions, chemical transformations and boundary layer meteorology that are specific to winter. These efforts are taking place against a backdrop of rapidly changing emissions in the United States, Europe, and Asia, illustrating the global context of these regionally focused field studies. **em**

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