Interim Report of the 2017 Utah Winter Fine Particulate Study (UWFPS 2017)

Executive Summary

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Introduction

The air basins along Utah's Wasatch Range, a region with 2.4 million residents (2010 Census), experience some of the most severe fine particulate matter (particulate matter with aerodynamic diameter less than 2.5 micron, $PM_{2.5}$) air pollution in the United States. High levels of $PM_{2.5}$ degrade visibility and are a significant public health concern associated with increased incidence of respiratory illness such as aggravation of asthma and premature mortality. $PM_{2.5}$ pollution episodes in northern Utah are closely related to the passing of high pressure ridges that favor the formation of persistent cold air pools (PCAPs) in Utah's topographic basins. During these multi-day episodes, vertical mixing is suppressed and pollutants emitted or formed near the surface accumulate in a shallow region known as the atmospheric boundary layer. In the Salt Lake valley, the depth of this boundary layer is 300 - 800 meters (1000 - 2500 feet). Boundary layer depths in other nearby regions, such as the Cache Valley, are typically somewhat shallower. Pollutant accumulation and subsequent atmospheric chemistry leads to elevated levels of primary and secondary pollutants including $PM_{2.5}$. The National Ambient Air Quality Standard (NAAQS) for 24-h $PM_{2.5}$ of 35 μg m^{-3} is exceeded during 6 multi-day events comprising 18 winter days in a typical winter.

The US EPA declared three regions in northern Utah, home to approximately 80% of Utah's population, as non-attainment areas (NAA) for 24-h $PM_{2.5}$ in 2009. Two regions along the Wasatch Front (Salt Lake City Utah NAA, Provo Utah NAA) have been re-designated in December 2016 as "serious" NAA's, requiring more stringent regulations to reduce $PM_{2.5}$. Despite the severity of these pollution episodes, uncertainties remain in the chemical and meteorological processes governing the formation of secondary aerosol. Improved scientific understanding of the emissions and meteorological and chemical processes governing these pollution episodes is needed for the development of effective control strategies to reduce $PM_{2.5}$.

The principal component of $PM_{2.5}$ during winter air pollution events is ammonium nitrate (NH_4NO_3), which is formed in the atmosphere via reversible reactions of ammonia (NH_3) and nitric acid (HNO_3). Ammonia is directly emitted from both agricultural sources, such as animal husbandry, and combustion sources, such as vehicle emissions or residential wood combustion. Nitric acid, by contrast, is a secondary pollutant that forms from chemical reactions of nitrogen oxides (NO_x = NO + NO_2). The latter are primary pollutants associated with combustion sources, such as vehicle emissions, power generation and industrial activity. Particulate ammonium nitrate therefore results from a complex interaction of meteorology, emissions sources and atmospheric chemistry.

Methodology

The wintertime chemistry that leads to ammonium nitrate is expected to be altitude dependent, with more rapid chemistry occurring in the upper parts of the polluted boundary layer compared to the surface level. Previous studies have been limited to surface based measurements, and therefore have not been able to fully characterize the rates and governing factors behind winter $PM_{2.5}$ formation. The 2017 Utah Winter Fine Particulate Study (UWFPS), one of the most comprehensive studies in this region to date, was conducted to address major questions with respect to ammonium nitrate aerosol formation in the stable, polluted boundary layers associated with the valleys of northern Utah. As part of this study, a National Oceanic and Atmospheric Administration's (NOAA) light aircraft, the Twin Otter, flew a suite of scientific instrumentation over the three major valleys of northern Utah, including Cache, Salt Lake, and Utah valleys, from January 15 to February 15, 2017. These flights surveyed the chemical conditions responsible for the formation of fine particulate pollution and probed the horizontal and vertical distributions of the most important trace gases and aerosols.

The use of a heavily instrumented light aircraft is a new approach to the study of air pollution in winter boundary layers. It is possible in part due to recent technological advances that allow for detailed atmospheric chemical measurements with instruments that are light enough to be deployed on small aircraft, which in turn can fly within the required, low altitude range. Application of this approach will improve understanding of winter air pollution in northern Utah and potentially other regions of the United States in the future.

In addition to the aircraft flights, a wide suite of trace gas, particulate and meteorological observations were made at multiple sites located in Cache, Salt Lake, and Utah Valley to augment the aircraft data and provided continuous measurements during both clean and polluted conditions. Detailed meteorological observations were carried out to monitor the meteorological formation, persistence and decay of cold-air pools. Exchange of air between different basins, as well as exchange with the Great Salt Lake (referred to as a "lake breeze"), are additional important meteorological processes that lead to transport and mixing of pollutants from different locations and altitudes that were characterized during UWFPS.

Results

Seven multi-day pollution episodes with elevated $PM_{2.5}$ were observed during 2016 – 2017 winter. Two dominating episodes with multiple NAAQS exceedances occurred during the UWFPS period, providing an opportunity to study the chemical and meteorological conditions during and outside pollution episodes in different environments and examine the temporal, spatial, and vertical variability of chemical conditions. The Twin Otter executed 23 successful research flights across 13 flight days, capturing moderate to severe pollution events and clean periods. Despite the heterogeneity of the sources, the chemical composition of $PM_{2.5}$ was very uniform in all the basins. Consistent with previous ground based studies, $PM_{2.5}$ mass was dominated by ammonium nitrate during high pollution events, with an average contribution based on aircraft data of 75%. Other components of $PM_{2.5}$ that are common in other urban areas, such as organic species (composed of carbon, oxygen and hydrogen) and sulfate (composed of sulfur and oxygen) made up the remaining mass (<20% and <5% on average). The average vertical profile shows an increase of aerosol mass loading with altitude with a maximum at 350 m (1200 feet) above ground level (AGL) and low values above 700 m (2300 feet) in the Salt Lake Valley. In contrast, the vertical distribution of aerosol mass loadings in Utah and Cache Valleys show shallower mixing depth with a maximum near the surface in Utah Valley and a relatively uniform aerosol distribution within the first 300 m (1000 feet) above ground level (AGL) in Cache Valley.

Nitrogen oxide (NO_x) levels show a clear spatial gradient across the northern Utah domain, with higher levels in more populated urban areas closer to the local sources and lower in rural areas such as the Cache Valley. The observed spatial patterns are broadly consistent with those in the emissions inventory used by the Utah Division of Air Quality (DAQ) for air pollution modeling. As a pollution episode progresses, NO_x and ammonium nitrate increase in the three valleys. Ozone, an oxidant that is typically associated with summertime air pollution, is lower during winter pollution episodes in the Wasatch region, an effect that is generally characteristic of urban winter air pollution. The influence of the daily cycle of the mixing within the boundary layer is evident in the observed temporal changes and the vertical distribution of the trace gases and $PM_{2.5}$. The highest NO_x and lowest ozone are often found near the surface at night as the NO_x pollutants are emitted and trapped within a near surface layer that is much shallower than the boundary layer. Nighttime reactions of the concentrated NO_x emissions tend to reduce ambient ozone levels. The shallow nighttime surface layer mixes during the day with the boundary layer due to the effect of sunlight heating the surface, but the boundary layer depth remains relatively constant, with little mixing of pollution to elevations above the cold air pool. These characteristics are manifested in the Twin Otter observations. Aircraft and surface level measurements of NO_x and ozone show clear deviations at night due to the lack of mixing at that time of day, but the same observations agree reasonably well during daylight hours when mixing of surface level and air aloft is more efficient. As the aircraft flies in and out of the boundary layer, a sharp distinction between the polluted air (higher NO_x and $PM_{2.5}$, lower ozone) trapped in the valley and the air above the basin was also seen at all times of day.

Preliminary Findings

1. Limiting Reagents in Ammonium Nitrate Formation

One of the key questions with respect to ammonium nitrate is attribution of the limiting reagent (i.e., primary ammonia or secondary nitrate) within the three basins. The Twin Otter payload was designed to sample the gas and particulate phase chemical composition of the air and provided spatially resolved data of the relative amounts of gases and particles across the region and as a function of height above ground. The ground based observations at Logan in Cache Valley, and at the University of Utah site in the Salt Lake Valley, provided continuous observations of the ammonium nitrate system at fixed locations. Using the combined results, two types of analyses were performed to address the question of the limiting reagent.

The first method examines the ratio of the observed total nitrate to total reduced nitrogen. Twin Otter measurements in the three major air basins (Cache, Utah and eastern Salt Lake Valleys) indicate that across northern Utah, total secondary nitrate, expressed as the sum of both gas and particulate phases, is present in lower concentrations than total primary reduced nitrogen (derived from ammonia), again expressed as the sum of both gas and particulate phases. Thus, secondary nitrate appears to limit the process of ammonium nitrate formation, suggesting that controls leading to reductions in secondary nitrate would be effective at reducing $PM_{2.5}$. However, the extent to which the system is nitrate limited varies from valley to valley and as a function of time and altitude. Of the three major valleys, the Cache Valley was most clearly in the nitrate limited regime although the nitrate limitation may be more severe at the surface than aloft. Both Salt Lake and Utah Valleys are predominantly nitrate limited, but may also have periods of reduced nitrogen limitation later in a pollution episode if the PCAP persists longer. It is possible that as pollution episodes proceed, the system evolves from a more nitrate to a less nitrate limited regime. Of the three major valleys, the Salt Lake Valley is the least nitrate limited during pollution episodes and is often within the ammonium limited regime later in the episodes. This may suggest that the system in Utah Valley and particularly the one in the Salt Lake Valley is close to the equivalence point between nitrate limited and ammonium limited regimes, and reductions in either reagent (nitrate and/or ammonia) may be effective in leading to reductions in ammonium nitrate aerosol. Validation of this result, including a quantitative analysis of the associated uncertainties, will require further analysis of UWFPS data.

A second method for determining limiting reagents relies on complex modeling of the thermodynamics of gas and aerosol phase nitrate and reduced nitrogen. The model is then used to determine the response of total $PM_{2.5}$ to an arbitrary, fixed reduction in either component. Results of this approach may be more definitive than the simpler comparison of total concentrations outlined in the preceding paragraph, but will require further analysis to reach robust conclusions regarding the most effective control strategies.

2. Emissions

During UWFPS, high time resolution real-time ammonia measurements were conducted aboard the Twin Otter and in Logan in the Cache Valley. However, continuous ammonia measurements were limited along the Wasatch Front. The observed ammonia concentrations are typically much higher in the Cache Valley air basin compared to the Wasatch Front, but also more variable, showing significant enhancements near agricultural sources. Similarly, ammonia in Utah valley shows more variability with lower mixing ratios above Utah Lake and larger regions of isolated higher concentrations near Spanish Fork and Lindon. Ammonia levels in the Salt Lake Valley are on average lower than in Utah Valley. The observed high spatial variability of ammonia is in disagreement with the current inventory, which shows comparable NH_3 emissions in the inventories for Cache, Utah and Salt Lake Counties, indicating a potential misrepresentation of the spatial distribution of ammonia sources in the inventory. Since the Wasatch Front appears to be less nitrate limited, better characterization of NH_3 sources in this region will be critical. Multiple independent observations indicate the importance of inter-basin exchange processes that can potentially transport ammonia from Utah Valley to Salt Lake Valley. If the transport of ammonia is a significant contributor to the ambient ammonia in the Salt Lake Valley, a reduction of ammonia in Utah Valley prior to the passages of high pressure systems could potentially reduce $PM_{2.5}$ in the Salt Lake and Utah Valleys during pollution episodes. More work is needed to characterize the ammonia sources and determine the contribution of transport processes on ambient ammonia.

As noted above, the observed distributions of nitrogen oxides, the precursors to secondary nitrate, are similar to those found in the emissions inventories and are concentrated near the largest urban areas in the region. Therefore, although the current analysis of the UWFPS data does not provide a quantitative assessment of the accuracy of nitrogen oxide emission inventories, observations suggest no qualitative disagreement with distribution of these emissions as represented by the inventory.

3. Residential Wood Combustion

Past ground-based work has shown varying contributions of wood smoke emissions to $PM_{2.5}$ in the study region. The major emissions from wood smoke include carbon containing compounds that are thought to contribute mainly to the organic component of $PM_{2.5}$, which as noted above contributed less than 20% to the final particulate mass in the aircraft observations during high pollution episodes. This figure represents a preliminary, upper limit to the contribution of wood combustion to regional fine particulate mass as other sources such as vehicular emissions can also contribute to organic aerosol. The actual contribution of residential wood combustion to total $PM_{2.5}$ is uncertain and may vary spatially depending on the neighborhood and time of the day. Hence, the potential importance of spatial and vertical variability needs to be considered while interpreting surface site observations for determining the residential wood combustion contribution to total $PM_{2.5}$ in a basin. Further analysis of the aircraft data for organic aerosol may serve to provide more quantitative estimates of the contribution of wood smoke to total $PM_{2.5}$ in northern Utah.

Future Research

Although UWFPS was arguably the most comprehensive study of its kind and used a novel methodology for the study of winter air pollution, significant further research will be required to make best use of the data and to reach more definitive findings regarding the sources and processes leading to winter fine particulate matter in northern Utah and elsewhere. Continuing analysis of UWFPS 2017 data, and field studies that build on UWFPS 2017 in the future, should ideally include**,** but not necessarily be limited to, the following.

- Chemical mechanisms for ammonium nitrate and oxidants. The next phase of UWFPS analysis will focus on the atmospheric chemical processes that lead to ammonium nitrate, and the time of day and altitude range in which they are most prevalent, to improve models of ammonium nitrate formation.
- Quantification of key meteorological processes. Data from UWFPS will aid in determining the role of processes such as exchange of air between different valleys in northern Utah, exchange with the Great Salt Lake, and exchange between the polluted boundary layer air and the clear air aloft.
- Variation of nitrate limitations during pollution episode evolution. A preliminary UWFPS finding is that the system is limited by secondary nitrate early in pollution episodes, but that this limitation becomes less severe with time. The finding has important implications for control strategies, and will be more carefully examined during further analysis.
- Uncertainties in ammonia sources. Emissions inventories, their spatial distributions, and the contributions of inter-basin exchange of ammonia are important, unresolved issues that will require further analysis and measurement. Determination of the relative contribution of vehicular and agricultural sources of ammonia is an important issue that will require further examination using UWFPS data and potentially further measurements in the future.
- Uncertainties in wood smoke contributions to fine particulate mass. Measurements during UWFPS can partially, but not fully, quantify this particulate matter source. Future studies that include more comprehensive measurements and spatial distribution of carbon containing gas and aerosol phase species are required.
- Better characterization of volatile organic compounds (VOCs). Measurements during UWFPS targeted the major components of ammonium nitrate aerosol. However, VOCs play important roles in the chemical processes that lead to ammonium nitrate, and are themselves important tracers of emission sources. Future measurements should also incorporate state of the art measurements of these compounds
- Uncertainties in meteorological modeling. Limitations imposed by local topography, Salt Lake water temperature specification, land-use characterization including snow and urban effects, parameterized boundary layer physics, and current computational constraints can all prevent adequate simulations of cold-pool development and evolution. Ongoing meteorological research will make use of UWFPS and future observations to better constrain these processes so that regulatory models best represent the evolution of pollution episodes in northern Utah.