Utah Winter Fine Particulate Study (UWFPS)

Research flights of the NOAA Twin Otter to address particulate matter pollution in urban areas of the Great Salt Lake Basin

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Background: The urban air basins along Utah's Wasatch Front, a region with 2.4 million residents (2010 Census), experience some of the most severe particulate matter (PM) air pollution in the Nation. Particulate matter concentrations smaller than 2.5 microns ($PM_{2.5}$) in Salt Lake City exceed the National Ambient Air Quality Standard (NAAQS, 35 µg m^{-3}, 24 hours) on average 18 days out of the year [*Whiteman et al.*, 2014]. The majority of exceedence days occur in December, January and early February during a period when winter meteorology leads to strong, multi-day inversions known as persistent cold air pools (PCAPs) that trap urban emissions within a shallow boundary layer [*Lareau et al.*, 2013]. Particulate matter concentrations increase at rates of 6-10 µg m^{-3} day^{-1} during these events to reach maximum concentrations of 70-80 µg m^{-3} [*Silcox et al.*, 2012; *Whiteman et al.*, 2014]. One report from Logan, UT gave 24 hour average $PM_{2.5}$ at 132.5 µg m^{-3}, and hourly average $PM_{2.5}$ at 182 µg m^{-3} in January of 2004 [*Malek et al.*, 2006]. Particulate matter exceedences in the Salt Lake Valley and the adjacent Cache Valley and Utah Valley (Figure 1) are similar to those seen in urban mountain valleys across the western United States, but Salt Lake City is by far the largest urban area subject to these air quality issues in the intermountain west, and it consequently experiences the highest PM concentrations [*Green et al.*, 2015]. These PM exceedences have been specifically associated with adverse health effects in the region, including a 42% higher rate of emergency room visits for asthma during the latter stages of air pollution events [*Beard et al.*, 2012] and a 4.5% increase in the risk for acute ischemic coronary events per 10 µg m^{-3} particulate concentration [*Pope et al.*, 2006]. The EPA declared three regions in northern Utah as non-attainment areas in 2009 [*Silcox et al.*, 2012]. The Utah Division of Air Quality (UDAQ) is currently developing a State Implementation Plan (SIP) to address the NAAQS $PM_{2.5}$ exceedences in this region.

Meteorological conditions associated with PM buildup in the Wasatch Front have been a focus of recent studies in the region [*Green et al.*, 2015; *Lareau et al.*, 2013; *Whiteman et al.*, 2014]. Persistent cold air pools are complex events that occur when a topographic depression fills with cold air for a period longer than a diurnal cycle. They can have a variety of causes and can be particularly challenging to model correctly [*Lareau et al.*, 2013], with implications for accurate forecasting of air quality. In the case of the Salt Lake Valley, elevation 1300 m ASL, the confining terrain of the Wasatch Mountains to the east and Oquirrh Mountains to the west is 1700 and 1200 m above the valley floor, respectively. Increases in PM mass are associated with large "heat deficits", defined as

the energy required to adiabatically mix a layer of air from the valley floor to the adjacent ridge height [*Green et al.*, 2015; *Whiteman et al.*, 2014]. Potential temperature differences between the valley floor and the terrain height can be as large as 25 K, with 8 K thought to be a figure of merit that defines the onset of $PM_{2.5}$ buildup during a PCAP [*Lareau et al.*, 2013]. Snow cover is an important factor in driving large heat deficits and / or potential temperature differences, but snow is not required to induce PM exceedence events. For example, EPA monitoring data for Salt Lake City from 2000 – 2013 shows average $PM_{2.5}$ of 24±2 and 15±1 µg m^{-3} on snow covered and non-snow covered days in November – February, respectively. NAAQS exceedences occur on 25% of snow covered days and 8% of non-snow covered days [*Green et al.*, 2015]. The highest $PM_{2.5}$ levels tend to occur during the 2nd and 3rd weeks of January, and the probability of high $PM_{2.5}$ events falls dramatically after mid February [*Whiteman et al.*, 2014].

Figure 1. Map of northern Utah showing major valleys and mountain ranges

The chemical composition of $PM_{2.5}$ in northern Utah, and the mechanisms by which it forms, have received considerably less attention than in other regions of the Nation despite the severity of the problem in Utah. Chemically speciated $PM_{2.5}$ measurements come from EPA and UDAQ monitoring sites in the Cache, Salt Lake and Utah Valleys [*Hansen et al.*, 2010; *Kelly et al.*, 2013; *Kuprov et al.*, 2014; *Mangelson et al.*, 1997; *Silcox et al.*, 2012; *Whiteman et al.*, 2014]. Much of the database is based on 24 hour filter measurements, but is augmented by intensive operating periods in different years that produce data at hourly time resolution for major primary trace gases, inorganic acids and bases, inorganic $PM_{2.5}$ ions, and organic matter.

The principal component of regional PM is ammonium nitrate, which is responsible for up to 70% of PM mass during inversion periods and 40% outside of inversions (Figure 2) [*Kuprov et al.*, 2014]. Ammonium chloride may also

contribute up to 15% of $PM_{2.5}$ mass during periods of high concentrations, although the source of the aerosol chloride is unclear [*Kelly et al.*, 2013]. The proximity to the Great Salt Lake is an obvious explanation for chloride aerosol, but reactions that lead to ammonium chloride require acid displacement from salt aerosol, which has been deemed unlikely. Nearly all studies within the last 20 years find sulfate to be a minor contributor to total $PM_{2.5}$ during winter inversions [*Hansen et al.*, 2010; *Kelly et al.*, 2013; *Kuprov et al.*, 2014; *Long et al.*, 2003; *Long et al.*, 2005a; *Long et al.*, 2005b; *Mangelson et al.*, 1997; *Silcox et al.*, 2012; *Whiteman et al.*, 2014], consistent with the data shown in Figure 2. The contribution of organics is more difficult to estimate. Residential wood combustion (RWC) is in general thought to be an important source of organic aerosol during wintertime in western mountain basins [*Green et al.*, 2015]. Several studies in the Salt Lake City area have inferred organic mass from the difference between total mass and the sum of all inorganic ions, as shown in Figure 2. Other studies have determined organic mass from filter samples and a total organic carbon measurement following IMPROVE protocols [*Green et al.*, 2015]. Some studies find a dominant contribution from ammonium nitrate, while others find roughly equal contributions from ammonium nitrate and organics.
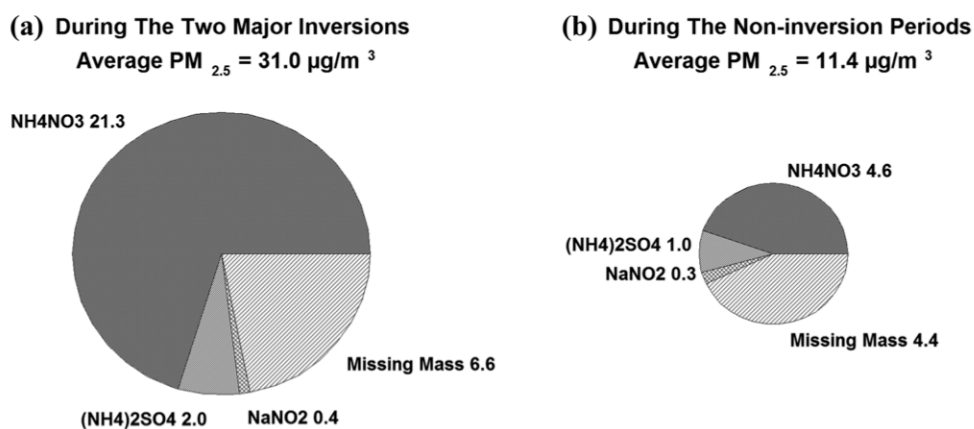

(a) During The Two Major Inversions
Average $PM_{2.5}$ = 31.0 µg/m³

NH4NO3 21.3

Missing Mass 6.6

(NH4)2SO4 2.0   NaNO2 0.4

(b) During The Non-inversion Periods
Average $PM_{2.5}$ = 11.4 µg/m³

NH4NO3 4.6

(NH4)2SO4 1.0
NaNO2 0.3

Missing Mass 4.4

Figure 2. Average composition and total $PM_{2.5}$ mass during inversion and non-inversion events in January and February 2009 in Salt Lake City. From [*Kuprov et al.*, 2014].

Factors affecting Ammonium Nitrate: Because ammonium nitrate is generally the single largest component of $PM_{2.5}$ mass, it is subject to the greatest scrutiny with respect to its sources and potential control strategies. A key question is the limiting reagent (i.e., agricultural emissions of NH_3 or urban NO_x emissions that produce HNO_3 through secondary reactions). Nearly every study that has considered this question indicates that HNO_3 is the limiting reagent, though via varying lines of evidence. Measurement of total inorganic ions in PM2.5 suggests that NH_4^+ is balanced by the sum of sulfate and nitrate, providing evidence for excess NH_3 since a cation deficit would be expected in the NH_3 limited case [*Kelly et al.*, 2013]. Surface level data from a site in the center of Salt Lake City also suggested that ammonium nitrate aerosol formation is limited by the availability of nitrate based on total reduced nitrogen ($NH_3(g) + NH_4^+(p)$) compared to total nitrate ($HNO_3(g) + NO_3^-(p)$), although levels of total reduced nitrogen only slightly exceed those of total nitrate [*Kuprov et al.*, 2014]. Attribution of the limiting reagent in this study may

lie within the experimental uncertainty of the oxidized and reduced nitrogen instrumentation. As discussed further below, attribution of the limiting reagent in the ammonium nitrate equilibrium may be a complicated question, since there may be considerable horizontal and vertical variation in the distribution of each reagent that has not been adequately probed from surface level measurements.

A second key question is the role of residual layer atmospheric chemistry and transport in determining surface level $PM_{2.5}$ levels. Figure 3 shows a diurnal cycle in $PM_{2.5}$, NO_x and O_3 at the UDAQ Hawthorne site in January of 2013. Aerosol mass exhibits a sharp rise between the hours of 9 AM – 12 PM local time, well ahead of the peak in O_3. Levels of NO_x (though they may have a substantial contribution from oxidized nitrogen due to the measurement method) are easily sufficient to fully titrate O_3 at the surface during nighttime hours. The time dependences in this figure are consistent with mixing of aerosol from the residual layer to the surface during morning breakup of the nocturnal boundary layer. However, the degree of stratification within the inversion layer and the depth and intensity to which the surface level O_3 titration persists are essentially unknown. The figure likely does not indicate a strong photochemical nitrate source, although there is likely a photochemical contribution. Further, long term monitoring in Salt Lake City suggests that primary pollutants, such as CO and NO_x, undergo characteristic weekday-weekend cycles seen in other locations, but that $PM_{2.5}$ does not have a strong weekday-weekend cycle [*Whiteman et al.*, 2014]. This observation is also consistent with a residual layer $PM_{2.5}$ reservoir with considerable time lag from the primary NO_x emissions. Thus, the key to understanding the mechanism for winter $PM_{2.5}$ formation in Salt Lake City and adjacent areas is to understand the composition and transport within the residual layer.
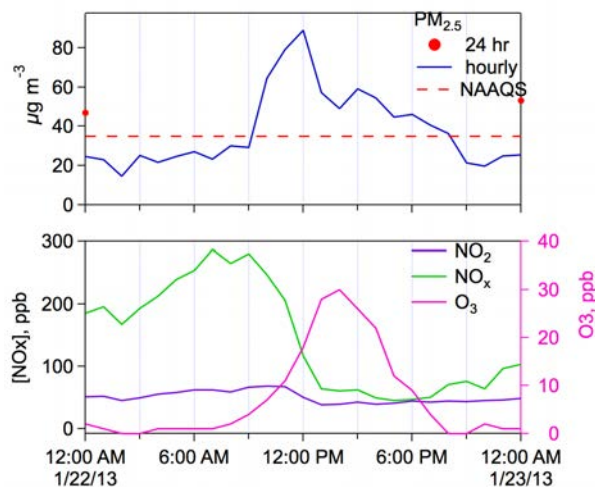

Figure 3. Diurnal cycle of $PM_{2.5}$, NO_x and O_3 at the Hawthorn site in Salt Lake City in January 2013.

This conceptual model is similar to the interpretation of the behavior of winter ammonium nitrate aerosol in California's southern San Joaquin Valley. Watson and Chow observed similar characteristics at a supersite in Fresno, CA in winter 2000, and further observed that although NO_x emissions were highly concentrated in urban areas such as Fresno, ammonium nitrate aerosol was widespread throughout rural areas [*Watson and Chow*, 2002]. They proposed a model with efficient residual layer chemistry converting NO_x to soluble nitrate, together with higher residual layer wind speeds that could transport soluble nitrate over a wider geographic area. More recent analysis of data from the San Joaquin Valley has confirmed this model, showing very different diurnal patterns of NO_x, O_3 and $PM_{2.5}$ from atop a 90 m tower at a rural site relative to

surface level. Residual layer $PM_{2.5}$ increased steadily during nighttime at 90 m, giving rise to a mid-morning surface level maximum during residual layer breakup [*Brown et al.*, 2006; *Chow et al.*, 2006; *Lurmann et al.*, 2006].

A third question is the limiting reagent for the oxidation of NO_x emissions to produce HNO_3 and nitrate. Because NO_x is primarily emitted as nitric oxide (NO), approximately two molecules of oxidant are required to convert NO_x to HNO_3 by daytime photochemical reactions.

$$NO + HO_2 \rightarrow NO_2 + OH, \text{ or } NO + O_3 \rightarrow NO_2 + O_2$$
$$NO_2 + OH \rightarrow HNO_3$$
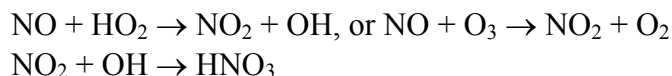
Alternatively, approximately 1.5 molecules of oxidant are required in nighttime heterogeneous chemistry:

$$2NO + 3O_3 \rightarrow N_2O_5 + 3O_2$$
$$N_2O_5 + H_2O \text{ (het)} \rightarrow 2HNO_3$$
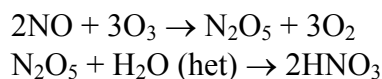
Sources of oxidants include background ozone or photochemical reactions of VOC and NO_x that produce oxidants including OH and O_3. If entrainment of background O_3 is the major source, then the oxidant limitation depends on the dynamics of the PCAP and the mixing efficiency with background O_3 from higher up in the inversion or the free troposphere. If photochemistry is the major oxidant source, then its response to both VOC and NO_x emissions becomes important, as is the case for summertime urban boundary layers.

A fourth question that has not been previously considered in the analysis of winter $PM_{2.5}$ in either California or Utah is the interaction between the reactive nitrogen and the chloride cycle. Nighttime conversion of NO_x to soluble nitrate is now known to occur simultaneously with substantial activation of photolabile halogen in the form of nitryl chloride ($ClNO_2$) in regions rich in aerosol chloride [*Osthoff et al.*, 2008; *Thornton et al.*, 2010].

$$N_2O_5 + Cl^- \text{ (het)} \rightarrow NO_3^- + ClNO_2$$
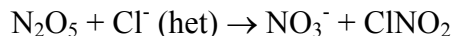
The recent analysis from Salt Lake City showing several $\mu g \ m^{-3}$ attributable to ammonium chloride aerosol, accounting for 15% of $PM_{2.5}$ mass, suggests that this chemistry may be particularly active in that region. If so, the atmospheric chemistry of winter $PM_{2.5}$ in northern Utah may be extremely complex, with both a nighttime residual layer component and a daytime photochemical component that results in part from atomic chlorine. This chemistry is likely to exhibit considerable vertical variation as a function of time of day [*Young et al.*, 2012].

Aircraft Measurements: Probing the residual layer composition and chemistry can be accomplished most effectively with a light aircraft. Measurements well above the elevation of the valley floor are essential due to the likely strong altitude dependence of

the relevant atmospheric chemistry. Regular vertical profiling with some level of chemical detail would be especially informative. The NOAA twin otter is capable of flying level legs as low as 150 m AGL during daytime and 300 m AGL during nighttime over populated areas. Furthermore it can execute missed approached to airfields throughout the Wasatch Front region to obtain vertical profiles to near surface level. Figure 4 shows an example of the residual layer structure and aerosol distribution as measured with a laser ceilometer. Aerosol mass during this event is distributed through 400 – 600 m AGL, with low clouds late in the time series that obscure the aerosol signal above them [*Lareau et al.*, 2013]. Measurements taken with aerosol samplers placed along a mountain transect confirm the presence of aerosol through at least 450 m above the valley floor, but with vertical variation [*Silcox et al.*, 2012].
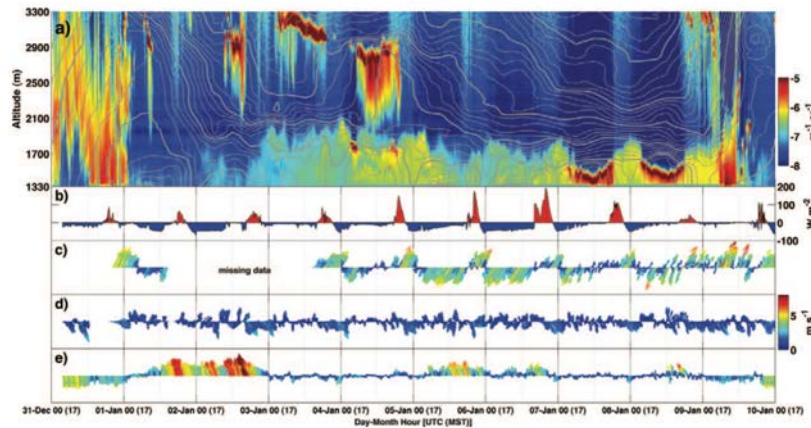

Figure 4. Laser ceilometer measurements of a 2011 winter PM$_{2.5}$ event. From [*Lareau et al.*, 2013].

The NOAA twin otter carries a sufficient payload to measure the relevant gas and aerosol phase species. The total useful payload for instruments is approximately 2000 lbs, and total power is approximately 5 kVA. Table 1 below lists a set of instruments that would fit within the limits of the twin otter's capabilities and also address the major scientific objectives of a winter study in northern Utah. Preliminary discussions with engineers at the NOAA aircraft operations center (AOC) indicates that this payload can be distributed across the available floor space and sampling ports of the twin otter.

Table 1. Proposed NOAA twin otter instrumentation

Instrument	Species Measured	PI	Power (kVA)	Weight (lbs)
Aerosol Mass Spectrometer	Speciated PM 1	Ann Middlebrook (NOAA)	1.1	546
Iodide Time of Flight Chemical Ionization Mass Spectrometer	HONO, HNO_3, N_2O_5, $ClNO_2$, other species	Joel Thornton (University of Washington	1.7	380
Nitrogen Oxide CRDS	NO, NO_2, NO_y, O_3	Steve Brown (NOAA)	0.5	257
Ammonia QCL Spectrometer	NH_3	Jenifer Murphy (University of Toronto)	1.0	200
Picarro CRDS	CO, CO_2, CH_4	Colm Sweeney (NOAA)	0.3	70
Total			4.6	1453

 A sampling strategy similar to that of the recent NASA DISCOVER-AQ program, which employed repeated flights with the same pattern over a period of weeks, would provide a dataset with sufficient statistics to characterize trace gas and aerosol vertical and horizontal distributions throughout the Salt Lake City area and adjoining valleys. The twin otter has an endurance of approximately 4.5 hours at a cruising speed of approximately 120 knots, for a range of approximately 500 miles per flight. This range is sufficient to sample all three major valleys (Cache, Salt Lake, Utah) together with the major bodies of water (Great Salt Lake, Utah Lake). Figure 5 shows the airspace in northern Utah. Flight planning will be developed in close collaboration with investigators at NOAA and UDAQ, together with NOAA AOC and local air traffic control to meet scientific objectives within the context of a safe and easily executable flight plan.

The single flight plan would be repeated at different times of day to sample periods of morning boundary layer evolution (8 AM takeoff), late afternoon / evening boundary layer establishment (4 PM takeoff), and residual layer in darkness (12 AM takeoff). We estimate approximately 15 research flights within the study area, or 5 of each type. The flights would principally target inversion periods, but would also sample during at least one non-inversion period for contrast.
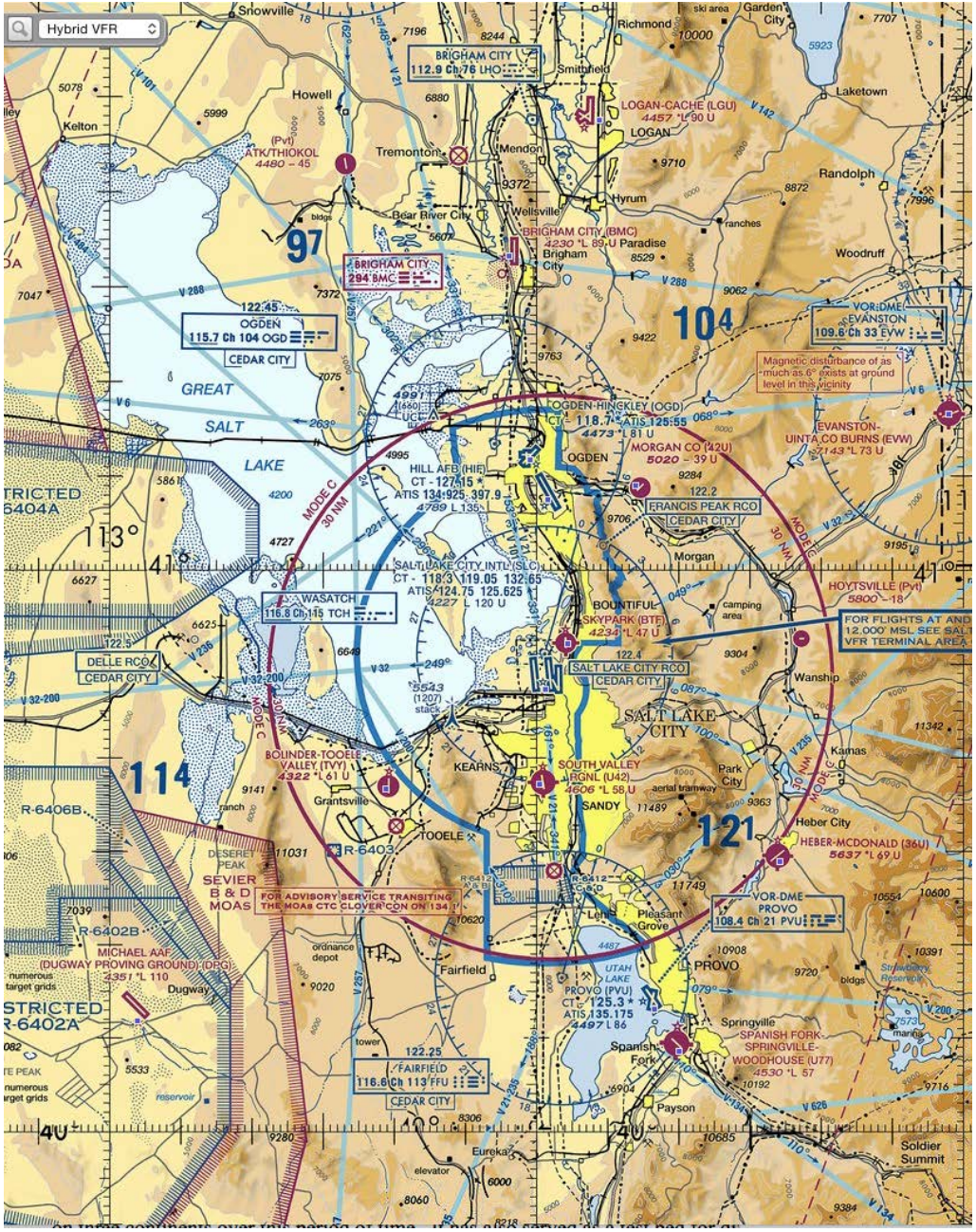
Figure 5. VFR map of the Salt Lake City air space.

The major concerns for execution of a safe flight plan that can meet scientific objectives are 1) the depth and structure of the boundary layer relative to the safe operating altitude range of the twin otter: 2) the presence of low level clouds and / or fog during inversions that would limit visibility and flight operations. As noted above, available data indicates that the boundary layer structure is deep enough that level flight legs of the twin otter will sample it. Nevertheless, vertical profiling over the airfields shown in Figure 5 will be essential to characterize vertical boundary layer composition and structure at different times of day. Data from the Salt Lake City airport indicates that there should be sufficiently clear conditions, even during inversion periods, to fly in reasonable visibility

environments. Figure 6 shows visibility, cloud ceiling and temperature data for Salt Lake City International airport for the most recent seven Januaries. Visibility and cloud ceiling are most often unlimited (>10 miles and >1500 m AGL, respectively), with distributions that are not significantly different for temperatures below -5°C (taken here to be more characteristic of inversions).
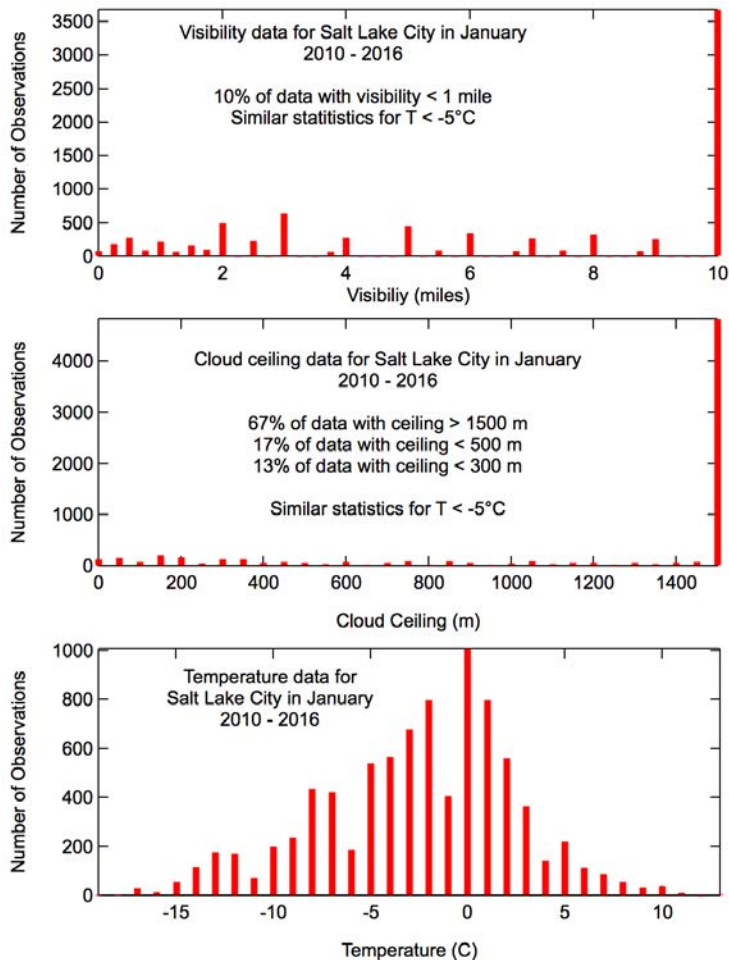

Figure 6. January visibility, cloud ceiling and temperature statistics at Salt Lake City International Airport (KSLC) for 2010-2016.

Scientific Questions and Deliverables:

1. What is the spatial distribution, both vertically and horizontally, of key trace gases and aerosols related to PM formation? What are the limiting and excess reagents in ammonium nitrate formation, and what are the key source regions? What are the limiting and excess reagents in oxidant and nitric acid formation? Do these limitations and / or sources vary significantly across the region?

> Emissions within the three major valleys of the Wasatch Front are expected to be heterogeneous, with the largest urban emissions in the Salt Lake Valley and the largest agricultural emissions in the Cache Valley. Aircraft flights across all 3 valleys at different times of day will characterize the spatial distribution of NH_3,

HNO_3 and NO_x, together with the key intermediates in NO_x oxidation. Comparison of these data to thermodynamic aerosol models will help to determine which reagents are limiting and where. Further, the aircraft measurements at different times of day will characterize transport of trace gases and aerosols, to assess the efficiency of residual layer transport in the distribution of pollutants and / or inter-valley coupling.

2. How do these distributions and the associated chemistry vary as a function of time of day? What are the most important chemical mechanisms for ammonium nitrate aerosol?

Morning, afternoon and night flights will characterize the chemical and meteorological evolution of boundary layer breakup, re-establishment, and overnight residual layer chemistry and transport. Rates of chemical reactions may be a strong function of altitude at all times of day. Vertical profiles will probe this chemical composition for an integrated analysis of boundary layer structure and chemistry.

3. What is the role of the Great Salt Lake and Utah Lake, both chemically and meteorologically, in regional air quality? Are they significant sources of aerosolized chloride that perturbs nitrogen oxide chemistry? Do they play a role in transport and or storage of polluted air masses?

Flights over large bodies of water at various altitude ranges should be feasible. These flights will characterize the vertical structure of winds over the lakes the transport pollutants, as well as the chemical composition at different altitudes.

4. Are there significant aerosol sources other than ammonium nitrate? What is the role of, for example, residential wood combustion as a source for organic aerosol?

Deployment of the AMS will provide unprecedented detail in the measurement of organic aerosol composition to the northern Utah valleys. Furthermore, the iodide CIMS instrument will provide measurements of oxygenated organic and nitrogen containing compounds that are characteristic of residential wood combustion sources. The spatial distribution of these organic aerosol and oxygenated VOCs, together with the daily time evolution, will provide accurate data on the primary and secondary organic aerosol source from residential wood combustion.

5. What are the key emission sectors for aerosol precursors? What is the role of agricultural, industrial, urban, home heating, and natural emissions?

Aircraft measurements can be compared to emissions inventories developed by UDAQ to assess the emission sources responsible for $PM_{2.5}$. This comparison will benefit from local 3D air quality modeling capabilities of investigators in Utah, at the EPA or at NOAA. Identification of a modeling component to this study will be a critical need going forward.

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