Observational Needs for Four-dimensional Air Quality Characterization

by Richard Scheffe, Russell Philbrick, Clinton MacDonald, Timothy Dye, Mike Gilroy and Anne-Marie Carlton

Surface-based monitoring programs provide the foundation for associating air pollution and causal effects in human health studies, and they support the development of air quality standards and the preparation of emission reduction strategies. While surface oriented networks remain a key tool for addressing traditional single pollutant human exposure challenges, these networks in isolation are not capable of adequately addressing emerging assessment challenges that consider linkages across pollutant categories, spatial scales, and environmental media. This increasing complexity of air quality assessments will gradually accelerate the reliance on environmental modeling systems featuring multi-dimensional descriptions in time and space. Accordingly, our design perspectives on observation systems must adapt complementary observation and modeling approaches. Paralleling this increased importance of models are major concerns about the adequacy of existing observation systems to provide the observations needed to support model evaluations and data assimilations for day-to-day operational assessments of the environment. For example, a model's ability to characterize surface air quality and deposition depends on properly characterizing physical and chemical processes throughout the atmospheric column, as well as the larger scale regional transport into the area. Because most pollutants and precursors for chemical and aerosol production reside above the immediate surface where point sensors provide measurements, it becomes important to examine the spatial and temporal heterogeneity through the atmospheric column. This article provides an overview of observation systems in use today to address comprehensive characterization of the planetary boundary layer, air quality and meteorological variables, and puts forth a series of recommendations to be considered in addressing important observations gaps.

What applications benefit from 4D characterization?

Observations are necessary to constrain calculations and keep the elements of the four-dimensional space-time array within bounds to properly characterize the current and changing state of the environment. Sufficient numbers of adequate quality observations are required to assure representative descriptions of the environment from model calculations and provide a basis for:

1. Linking human health and environmental welfare effects with air quality, which is the basis for developing U.S. National Ambient Air Quality Standards;
2. Determining compliance with relevant standards;
3. Developing emission reduction strategies through source apportionment studies, air quality model evaluations and model applications;
4. Assessing progress in response to implemented emission strategies;
5. Forecasting air quality to inform the public of potential adverse air pollution; and
6. Elucidating atmospheric processes to improve physical and chemical basis for air quality modeling systems.

Ultimately, the value of the environmental policies depends upon the observational data used for developing and constraining the model data products.

To date, these objectives were often approached from a single pollutant perspective. The modeling of physical and chemical processes involved in formation and accumulation of the trace species and aerosols from various emission sources require far more integrated assessments that capture a variety of pollutants over multiple spatial and temporal scales (intercontinental to local/near source environments). They must also consider both air and terrestrial/aquatic media impacts that account for bi-directional interactions between air quality and climate, as well as health relevant issues. Superimposed on these integrated themes is the need to understand the effects pathways from emissions sources to ambient conditions, human exposure, and ecosystem impacts[1]; and finally to the ultimate consequences upon ecosystem and human health. All of this "accountability" suggests characterization requirements both across a range of spatial scales covering the source to effects continuum as well the necessary temporal extension (retrospectively and prospectively) to relate emission changes to consequences. These integration themes, articulated in the 2004 National Academies study on air quality management[2] and elsewhere[3,4], point to the need to comprehensively characterize air quality in multiple dimensions and argue for the complementary use of observations, respecting the model's inherent multi-dimensional simulation capability and the inherent "trust" in the observations.

Improving the treatment of chemical and physical processes used in air quality models is perhaps the most important application of vertically distributed observations of chemical species, aerosols, and meteorological properties. To illustrate, the availability of vertically resolved aerosol organic carbon observations throughout boundary layer enables an assessment of the impact of adding a cloud processing mechanism for secondary carbon formation to CMAQ, EPA's national air quality model. Without access to aloft data, a real world assessment of a process operating above the surface layer would not be possible. Figure 1 shows an example of the benefit resulting from aloft data that extends the ability to improve our understanding of processes occurring aloft; as now, the properly distributed vertical mass results in improved surface predictions by addressing a known under prediction of secondarily formed aerosol carbon

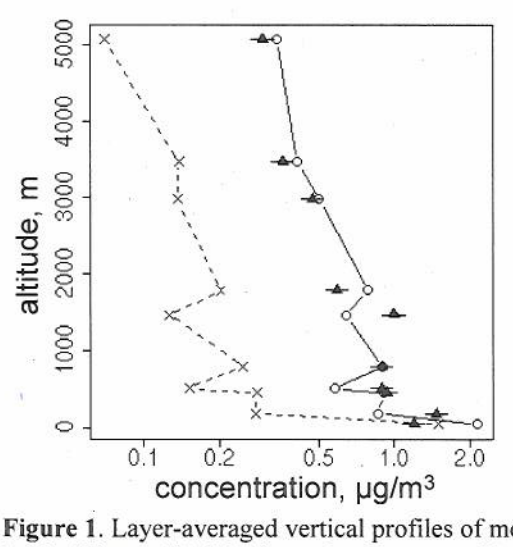

Figure 1. Layer-averaged vertical profiles of modeled and observed organic carbon (OC) on August 14, 2004. Note: Dashed line and "×" indicates layer-averaged base CMAQ OC prediction. Solid line and "O" indicates CMAQ OC prediction with cloud-produced SOA included. OC observations "▲" are based on the International Consortium for

Atmospheric Research on Transport and Transformation (ICARTT) New England Air Quality Study - Intercontinental Transport and Chemical Transformation (from Carlton et al.[5]). The *x*-axis is log scale.

Overview of Existing Observation Systems

A variety of Federal and State agencies, international and private sector organizations support measurement programs underlying air quality assessments. These include routine surface based regulatory and deposition networks, intensive field studies, remote sensing systems, sondes, aircraft campaigns, satellites, and focused fixed site special purpose networks (Figure 2). While the overarching theme of this paper addresses the roles of observational programs complementing air quality models to foster comprehensive air quality characterization, the individual components of the observational infrastructure are driven by separate design needs based on the specific missions of sponsor entities. Accordingly, this system of systems is very loosely organized and the objectives for comprehensive characterization are opportunistic in nature.

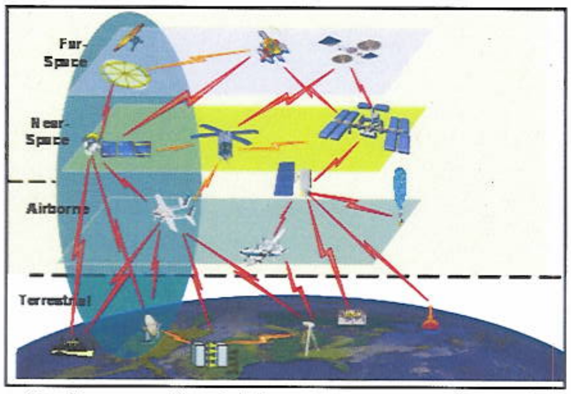

Figure 2. Concept of multiple sensor systems for surface and aloft measurements over local, regional, and global scales.

Routine surface based networks

Routine U.S. air- and deposition-monitoring networks provide data at over 3000 fixed surface sites, including measurements of many gaseous species and suspended particulate (PM) properties (Figure 3). Many U.S. networks were established in response to needs from the 1970 Clean Air Act (CAA), and subsequent 1977 and 1990 amendments. The evolution of U.S. standards, based upon reviews by various stakeholders, such as the U.S. government, the private sector, and advisory groups like the U.S. National Academy of Sciences of the National Research Council, have resulted in periodic reorganization and/or enhancements of the measurement networks.

Many individual monitoring sites are operated by a combination of local, state or tribal government entities, research investigators, contractors, or private parties. The U.S. Environmental Protection Agency (U.S. EPA) provides funding, database support and operational guidance for the majority of routine network operations addressing air quality standards and management practice. Other U.S. federal agencies, including the National Park Service (NPS), National Oceanic and Atmospheric Association (NOAA), Department of Energy (DOE), United States Department of Agriculture (USDA), and U.S. Geological Survey (USGS) also support a variety of observational network operations.

The State and Local Air Monitoring Networks (SLAMs) is an overarching structure for the routine U.S. regulatory networks used primarily for determining compliance with National Ambient Air Quality Standards (NAAQS) and supporting the associated development and testing of emissions reduction strategies. The SLAMS includes the majority of measuring instruments for monitoring ozone (~ 1100), $PM_{2.5}$ gravimetric mass (~ 1100), continuous $PM_{2.5}$ mass (~500), $PM_{2.5}$ chemical speciation (~500), several hundreds of NO_x, CO, and SO_2 sites, and roughly 75-sites with Photochemical Assessment Measurement Stations (PAMS) providing our major source of volatile organic carbon measurements. The national core network[6] (NCore), to be deployed by 2011 is a 75-site multiple pollutant network, which will provide measurements of a variety of key trace gases and aerosols, and is intended as a science support complement to the SLAMs networks. These air toxics measurements will include a variety of particle bound metals and gaseous organic compounds spanning low carbon number volatile organics through semivolatile and persistent halogenated compounds.

Outside the SLAMS umbrella, the Clean Air Status and Trends Network (CASTNET), and the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, both provide a variety of aerosol and gaseous observations which constitute the majority of our routine rural based monitoring operations.

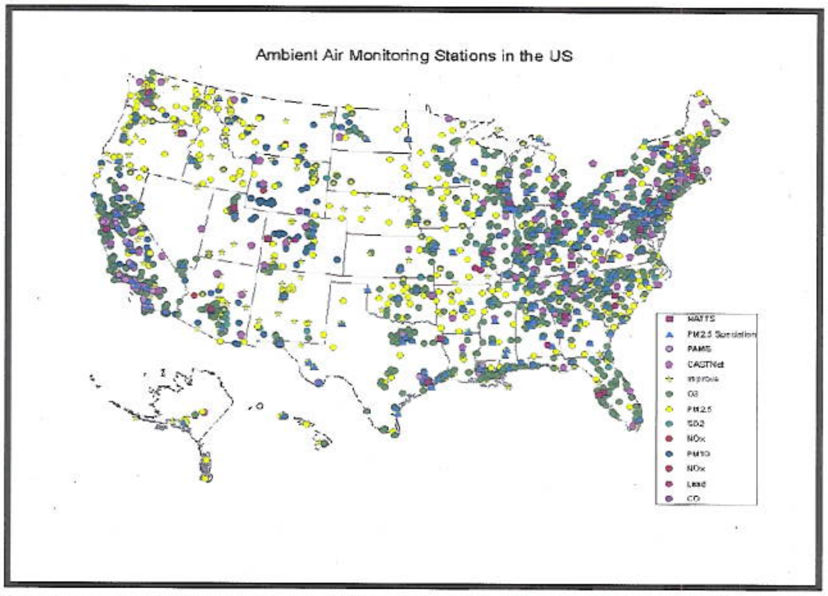

Figure 3. Aggregate map of the majority of routine U.S. monitoring stations illustrating relatively broad coverage across the continental U.S. with noted spatial gaps in low populated areas.

Chemical and meteorological observations characterizing the planetary boundary layer.

Chemical Observations. Aircraft, sondes, tethered balloons, ground based remote sensors and satellite platforms are sources of measurements used to characterize the vertical distribution and total pollutant loading throughout the atmospheric column. In contrast to the routine surface based networks discussed above, chemical measurements aloft are generally conducted as part of intensive field campaigns (see below) or special studies designed to characterize specific atmospheric processes, and focus on efforts to obtain insight on transport, transformation, and removal of air pollutants. However, there are several routine operations providing chemical information aloft supporting regional air quality, climate change, and stratospheric ozone depletion assessments:

- **NOAA operations (http://www.esrl.noaa.gov/gmd/) including**: an ozone sonde network (8 U.S. sites) providing one day per week vertical ozone profiles with approximately 100 m resolution from the surface through the stratosphere; the Dobson ozone spectrometer network (15 sites, 9 U.S. locations) provide near continuous daytime measurements of total atmospheric column ozone data; routine aircraft flights that characterize vertical distribution of air pollutant species (O_3, CO, CH_4, CO_2, N_2O, SF_6) relevant to both climate and air quality assessments; and tall tower sites (9-15) that are part of a larger interagency North American Carbon Program (NACP) characterizing carbon sources, sinks and removal processes; these towers are currently located throughout the continental United States and use television and cell phone transmission towers (100-500 m). These sites provide regionally representative boundary layer measurements of near continuous CO_2, CO, CH_4 and associated fluxes, various trace gases, and meteorological parameters;

- **European based aircraft programs (MOZAIC and CARIBE)** - The measurements of ozone, water vapour, carbon monoxide and nitrogen oxides aboard Airbus in-service aircraft (MOZAIC http://www.fz-juelich.de/icg/icg-ii/mozaic/home) and Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC http://www.caribic-atmospheric.com/) programs provide near continuous air quality measurements conducted in commercial European based airlines, and these data include several trans-Atlantic flights to and from North American cities. These programs, addressed in depth in the companion paper[7] provide the most extensive routinely-collected and vertically-distributed air quality data throughout the PBL.

- **Satellite missions** - NASA and European space agency satellite platforms addressed in the companion paper[8], provide total atmospheric column estimates for aerosols and several trace gases. Although these missions are focused on research efforts typically operating less than a decade, the transition to new missions often incorporates observations compatible with previous missions. Most satellites with air quality sensors are polar orbiting; thus they provide near global spatial coverage with resolution ranging from 10 to 50 km with one or two views per day. NOAA's GOES weather satellites, which provide aerosol information, follow an equatorial orbit allowing near continuous views of the tropical regions. Satellite measurements using remote sensing techniques will play an even more important roll in providing future observations of certain key parameters on global and regional scales. Figure 4 shows an example of the value associated with four-dimensional measurements in depicting the evolution of a Saharan dust storm[9] from the CALIPSO satellite's lidar instrument. During a period of more than a week, the vertical and horizontal distribution optical scattering profiles of the dust were measured. The detail information on the spatial distribution and temporal variations in this type of event cannot be obtained by ground based observations.

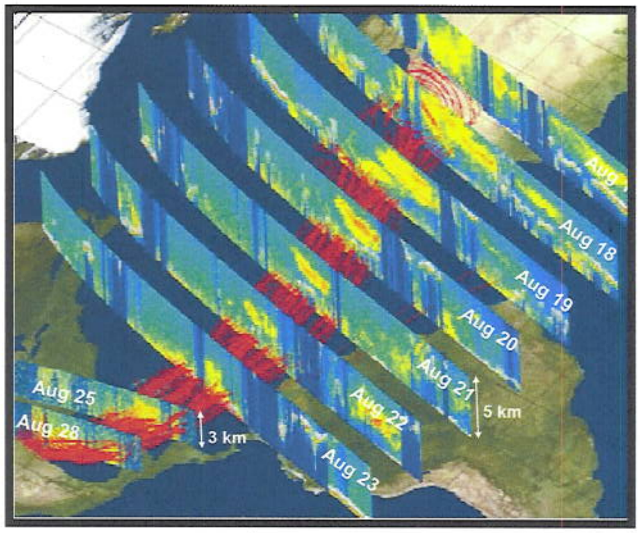

Figure 4. Four-dimensional observations from the CALIPSO satellite lidar measurement sequences of August, 2006 show African dust transport across the Atlantic Ocean (source, Liu et al.[9]).

Intensive field campaigns. The majority of relevant PBL chemical information is provided by specialized field campaigns which often include aircraft, and a variety ground based in-situ and remote sensing platforms. These campaigns, often lead by NOAA and NASA, are designed to shed light on poorly characterized atmospheric processes. Since the characterization of most atmospheric processes requires vertically and highly temporally resolved information, these specialized studies provide the most useful data sets for diagnosing air quality model behavior and improving the details of the chemical and physical processes to yield model improvements.

Ground based remote sensing. Remote sensing techniques provide a primary element of our understanding of the physical and chemical processes of the lower atmosphere because of the continuity and accuracy of key measurement parameters of the atmospheric column that can only be acquired by ground based remote sensors. Many extremely important properties can be measured from aircraft, balloon releases, and tethered balloons, because scaled versions of most laboratory instrument techniques can be carried aloft to measure the properties of a trace species, aerosol, or the values for a meteorological parameter. However, by their very nature of operation and cost-per-data-point, it is not feasible to obtain the continuity and the spatial dimensions for the measurements needed to investigate many of the processes, sources, event evolution, or the multi-parameter meteorological factors that control the local environment. Remote sensing techniques rely on the interaction of an electromagnetic or the acoustical propagation wave disturbance with the atmosphere, and use the scattered signals to determine a property, process, or parameter of a scattering volume.

Radars, lidars, radio beacons, radio acoustic sounders, and sodars make up the families of instruments typically used for remote sensing of atmospheric properties. Radars and lidars make use of a rather large portion of the electromagnetic spectrum. The wavelength compared with the size of the scatterer determines the cross-section, and therefore the sensitivity of the detection. For this reason, lidars at ultraviolet, visible and infrared wavelengths are used to investigate the molecular species and aerosols. The radar and radio wave interactions are used for studies of the ensemble motions and bulk scattering properties, such as wind and turbulence, by measuring signals associated with the refractive scattering of permittivity variations in the medium. The acoustical sounding techniques can directly measure the speed of sound, which provides a direct measure of virtual temperature, and with a small correction to the speed of sound due to the amount of water vapor present, the gas kinetic temperature is obtained. An example of the capability of remote sensing lidar is shown in Figure 5. Vertical profiles of water vapor, ozone, temperature, optical extinction and other parameters can be measured independently and simultaneously using Raman lidar techniques. A sequence[10,11] of 24-hours of profiles measured during the period of a summer air pollution episode at the NARSTO-NEOPS site in Philadelphia are shown in Figure 5. The water vapor measurements show that the air transported in the residual layer (back trajectories show that the air mass came from the Ohio valley) is mixed rapidly to the surface near local noon by convection. In the surface layer, thermal decomposition followed by photochemical processes cause rapid increases in ozone and aerosols to unhealthy levels. Remote sensing with lidar and radar provide a special capability for investigating the physical and chemical processes and closely checking the chemical and dynamical response of models. Eventually these techniques should provide primary data sets of assimilation into operational models of the environment.

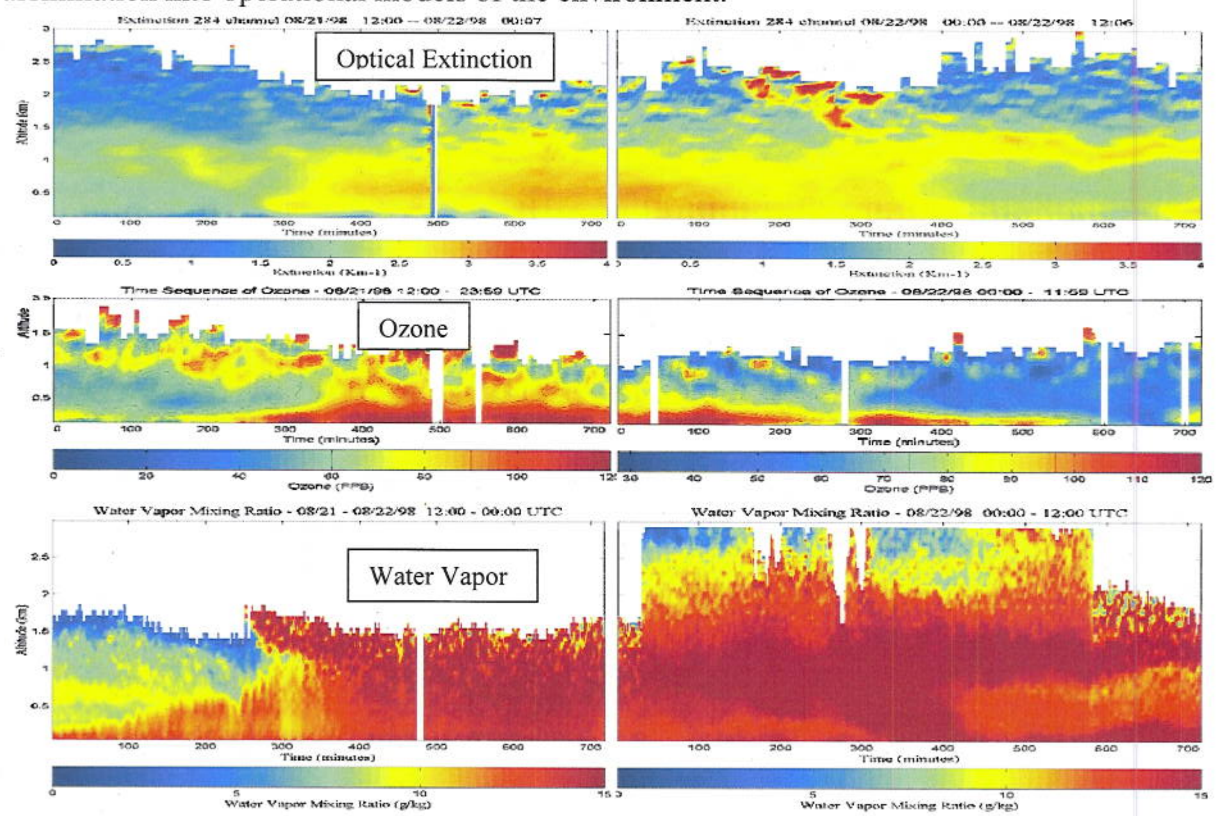

Figure 5. Time sequences of the PBL vertical profiles of water vapor, ozone, and optical extinction measured by a Raman lidar at the NARSTO-NEOPS field site in Philadelphia during August 21-22, 1998[7,8]].

Key Meteorological Observations for PBL Characterization and Model Evaluation

The ability for scientists to accurately model pollution and to conduct useful air quality research requires measurements that capture the spatial (both horizontal and vertical) and temporal characteristics of winds, temperature, and vertical mixing within the atmospheric boundary layer. Historically, boundary layer information has been collected using by twice-daily radiosondes. However, the limited temporal frequency and spatial coverage of these measurements are not adequate to resolve many boundary layer phenomena that have strong influence on air quality such as (1) local flows that are forced by diurnal heating and cooling cycles (e.g. land/seabreeze and mountain/valley circulations); (2) the strength, height, and hour-to-hour and day-to-day variability of temperature inversions, and the nocturnal, marine, and convective boundary layers; and (3) the existence and strength of nocturnal jets[1]. Fortunately, increased routine and episodic deployment of remote sensing instruments (such as sodars, radar wind profilers, lidars, and microwave radiometers) and improvements to instrument data processing algorithms (such as algorithms to automatically detect mixing heights from radar wind profiler and ceilometer data) have greatly improved our ability to discern boundary layer phenomena.

A radar wind profiler (RWP) points vertically in clear air to provide measurements of hourly or sub-hourly vertical profiles of horizontal winds and vertical velocity, and convective boundary layer height. Although there are several types of RWPs, for air quality applications the "Boundary Layer" profiler is most useful. The Boundary Layer profiler operates at 915 MHz and provides sub-hourly data from about 100 to 3,000 m agl with a vertical resolution of 60 to 120 m. Figure 6 presents an example of various boundary layer wind phenomena measured by a Boundary Layer profiler. There are currently about 35 Boundary Layer profilers routinely operating in the United States. Wind data from many of these sites can be obtained from http://madis.noaa.gov/. An RWP can be operated with a Radio Acoustic Sounding System (RASS) to provide hourly virtual temperature (T_v) profiles from about 100 to 1,500 m agl with a vertical resolution of 60 m. In addition, hourly daytime mixing heights can be derived from the RWP signal-to-noise ratio, vertical velocity, and spectral width data. The mixing heights diagnosed using RWP data can range from about 100 m to about 4000 m agl. Although algorithms to derive mixing heights are available, the mixing heights require expert review and editing.

In addition to the Boundary Layer profiles, NOAA operates a network of about 35 tropospheric profilers in the Central United States. In "low" operational mode, the tropospheric profilers measure winds from about 500 m above ground level (agl) up to about 9,000 m agl and have a vertical resolution of 250 m. In "high" mode they have greater altitude coverage up to 16,000 m agl and a vertical resolution of 250 m. For air quality applications, data from the tropospheric profilers are useful for evaluating models ability to capture meso- and synoptic-scale flow patterns that influence long-range pollutant transport.

Sodars operate in a similar fashion as the RWP; however, they use sound instead of electromagnetic radiation to determine wind speed and direction with altitude. There are many

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[1] Low-level jets are a common meso- and synoptic-scale feature that are often not well detected by models, but can have a strong impact on air quality conditions by transporting polluted air masses a great distance at night. Once transported, the pollution can mix down to the surface during the day impacting local pollution concentrations[12,13].

types of sodars with various operating characteristics. In general, sodars provide higher time and vertically resolved data compared to a RWP, but have limited height coverage. For example, "mini-sodars" provide 10-minute averaged profiles of wind speed and wind direction from 15 m above the ground up to 150 m agl, with a 5-m vertical resolution. Whereas, other higher powered sodars can provide data from roughly 50 m agl up to 1000 m agl (depending on atmospheric conditions), with a 15 m vertical resolution. Collocated sodars and RWPs can provide full coverage of boundary layer winds and high resolution data to resolve shallow flows in the nocturnal boundary layer to flows within a deep convective boundary layer. Some air quality agencies, such as the South Coast Air Quality Management District (SCAQMD) have been monitoring boundary layer phenomena.

The ceilometer (Figure 7) works by vertically emitting an eye-safe laser beam and detecting the scattering of the beam by particles. Hourly or sub-hourly boundary layer heights with a vertical resolution of 10 m can be derived from the backscatter data. A spatially extensive network for broad application is available through the NOAA Automated Surface Observing System (ASOS); however, mixing height data are not currently available from the network.

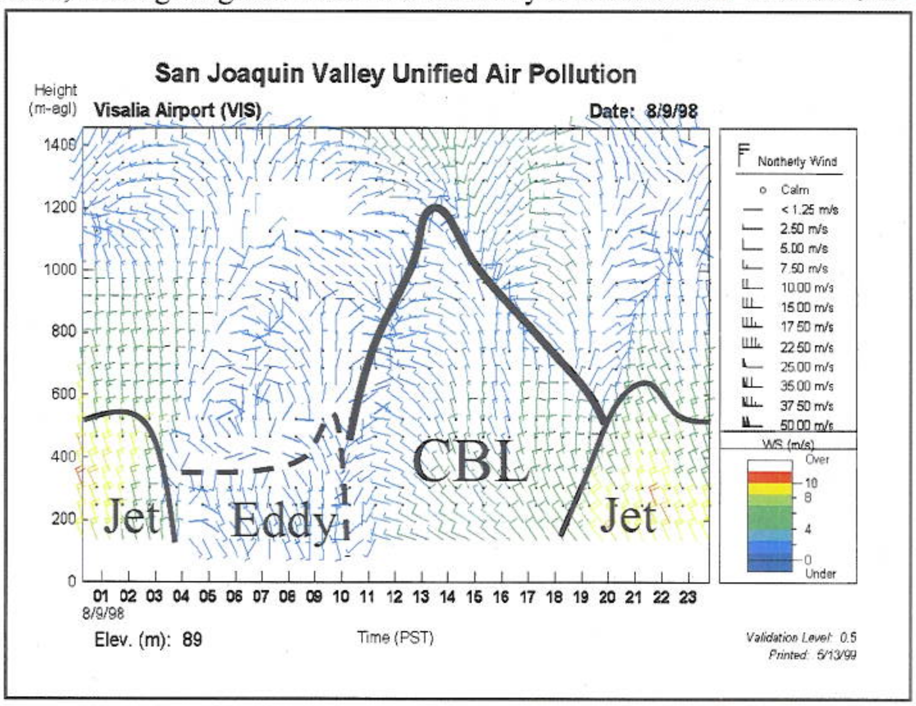

Figure 6. Radar wind profiler data at Visalia on August 9, 1998, showing the nocturnal jet, convective boundary layer (CBL), and eddy flow. This wind pattern was observed on the majority of the episode days.

Since 2004, over 400 commercial aircraft have been collecting meteorological variables (temperature, pressure, RH, winds) as a part of the tropospheric Airborne Meteorological Data Reporting (TAMDAR - http://www.airdat.com/./tamdar/index.php) system. While TAMDAR is designed to provide near real time data for forecasting, the system provides a valuable set of vertical profiles of temperature data (and other variables) during ascents and descents that can potentially be synthesized to fill in temporal and spatial gaps of ground based profilers.

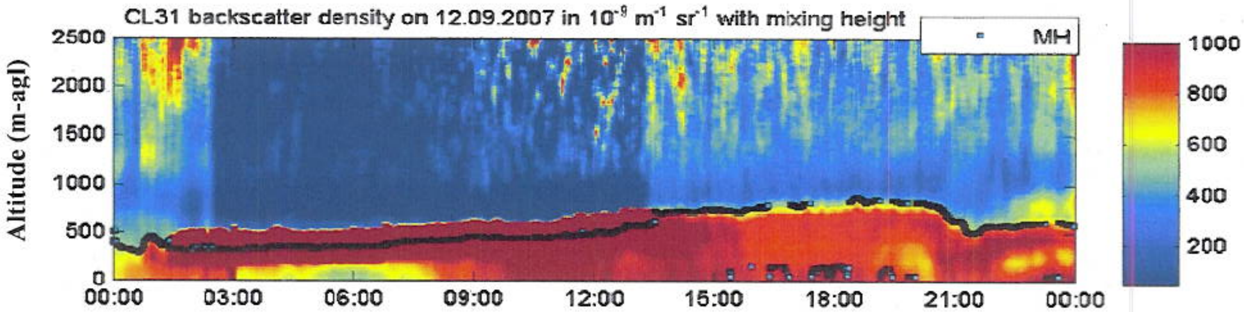

Figure 7. Ceilometer backscatter and mixing height data collected by Puget Sound Clean Air Agency for September 9, 2007.

Enhancements to Improve 4D Characterization

The routinely operating programs discussed above were designed for specific objectives and model evaluation needs. While observations are relevant to virtually any air quality related assessment, the air quality data are not explicitly considered as objectives of routine networks, with certain exceptions noted in both the PAMs and NCore program designs. Consequently, despite the wealth of surface based measurements, many important observational gaps relevant to air quality model evaluation exist; some of the more critical information needs include:

- **Surface based point measurements designed to leverage satellite observations**. The information potential afforded by satellites would be highly leveraged with the addition of key surface based measurements. Specifically, the addition of true nitrogen dioxide and formaldehyde observations in both remote and urban locations would, at a minimum, allow a meaningful assessment of the relevance of space based observations to boundary layer conditions and, more optimistically, would provide the basis for correlating and adjusting space based observations to represent near surface concentration fields. Nitrogen dioxide and formaldehyde are air quality indicators critical to understanding a variety of emissions and atmospheric chemistry processes and have well documented air quality applications using satellite data.

- **Vertically distributed observations of key air quality species**. The United States does not have routinely operating measurement programs dedicated to characterizing the vertical distribution of important trace gases and aerosols through the PBL. While this information is provided by intensive field campaigns, we are left with very limited snapshots of key information to diagnose model behavior. In addition, vertically distributed air quality data enhance the relevance of total column satellite data. A combination of strategically placed surface remote sensing sites and coordinated aircraft flights in combination with satellite imagery could provide a system capable of challenging the model's ability to adequately characterize mass throughout the PBL which in turn would lead to very basic improvements in our emissions estimates and a variety of modeling processes. In addition to key reactive species such ozone, nitrogen oxides and formaldehyde, conservative species such as carbon monoxide, CO, should be given high priority as CO can help delineate meteorological and emissions based influences without the complicating effects of chemistry. Specific examples include:

- Expansion of NOAA's ozone sonde program to provide added coverage in the continental U.S. and addition of key trace gas measurements.

- A sustained U.S. based aircraft campaign (national and international) similar to the MOZAIC/CARIBE European effort that would produce routine vertical profiles of key trace gases and aerosols;

- Deployment of fixed site LIDARS at key locations throughout North America to provide continuous profiles of back-scattered light serving as a direct link between ground-based AIRNOW $PM_{2.5}$ in-situ samplers and MODIS and CALIPSO satellite instruments. Such a network could build on the existing Regional East Atmospheric Lidar Mesonet (REALM) proposal[14] that includes 6 sites in the Northeastern U.S. and Canada.

- **Organized network of PBL observations.** With the exception of California, most of the remote PBL instrumentation is limited to single sites largely disconnected from a network of sensors that allow for broad spatial coverage of the daily evolution of the PBL. This need is particularly important in coastal locations with high populations and noted air quality problems that are impacted by complex land-sea phenomena. An excellent source of boundary layer data is the Meteorological Data Ingest System (MADIS - http://madis.noaa.gov/). MADIS is an integrated system incorporating observations from a variety of surface based, vertical profile and satellite networks that provides a centralized source of observations servicing evaluation efforts. However, a data synthesis system to ingest and process these data for PBL characterization would add value to existing systems and possibly spur the addition of new instruments. The recent National Academies of Sciences Study on mesoscale measurements[15] addresses many of the scientific and infrastructure challenges to metrological observations.

- **Geosynchronous AQ satellite mission.** Polar orbiting satellites[8,16] provide extensive spatial coverage but afford limited temporal resolution as each orbiting platform is limited to two swaths per day. In response to the NRC's decadal survey on Earth Science and Applications from Space, the interagency Geo-CAPE proposal[17] calls for deployment of geostationary platforms focused on North America providing near continuous streams of chemical information comparable to that provided by NOAA's GOES (http://www.oso.noaa.gov/goes/index.htm) platforms, widely used for observing weather systems and meteorological forecasting. Agencies with strong interests in boundary layer characterizations (e.g., EPA) should support satellite missions and other atmospheric column characterization campaigns so that operational designs are matched efficiently with program objectives.

Information technology to enhance data access, integration and analysis. Accessing and manipulating observational and modeling data sets present significant challenges to users requiring familiarity with a variety of systems developed independent of each other with diverse data formatting and meta data standards. Information technology applications to organize, access and integrate data sets can reduce the burden on analysts in understanding and manipulating an increasingly wide range of disparate data sets obtained in different programs, including routine air monitoring. However, accessibility requirements remain a problem because of non-uniform database organization adopted by various investigators. The recently conceived federated data system incorporates both observations and modeling results (DataFed - http://datafedwiki.wustl.edu/index.php/DataFed_Wiki) is an outgrowth of the Global Earth Observation System of Systems (GEOSS). Data Fed is an attempt to coordinate earth observations catalyzed by the Group on Earth Observations (GEO, http://www.earthobservations.org/index.html).

Summary

Challenging multi-dimensional air quality assessments benefit from the complementary use of surface, aircraft and satellite based observation systems and air quality simulation models. The value of vertically resolved and total column measurements of gaseous species and aerosol properties, together with the meteorological conditions enable improvement of key processes and assessment of model performance to extend both the applicability and confidence in environmental simulation models. Unfortunately, there are critical observational gaps that compromise our ability to fully characterize atmospheric behavior across three dimensional space and time as measurements aloft as well as key surface based indicators generally are derived from occasional intensive field campaigns. A more highly coordinated multiple organizational approaches to observational design are recommended to address this important need.

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