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Raman Lidar Contributions to Understanding Air Quality Issues

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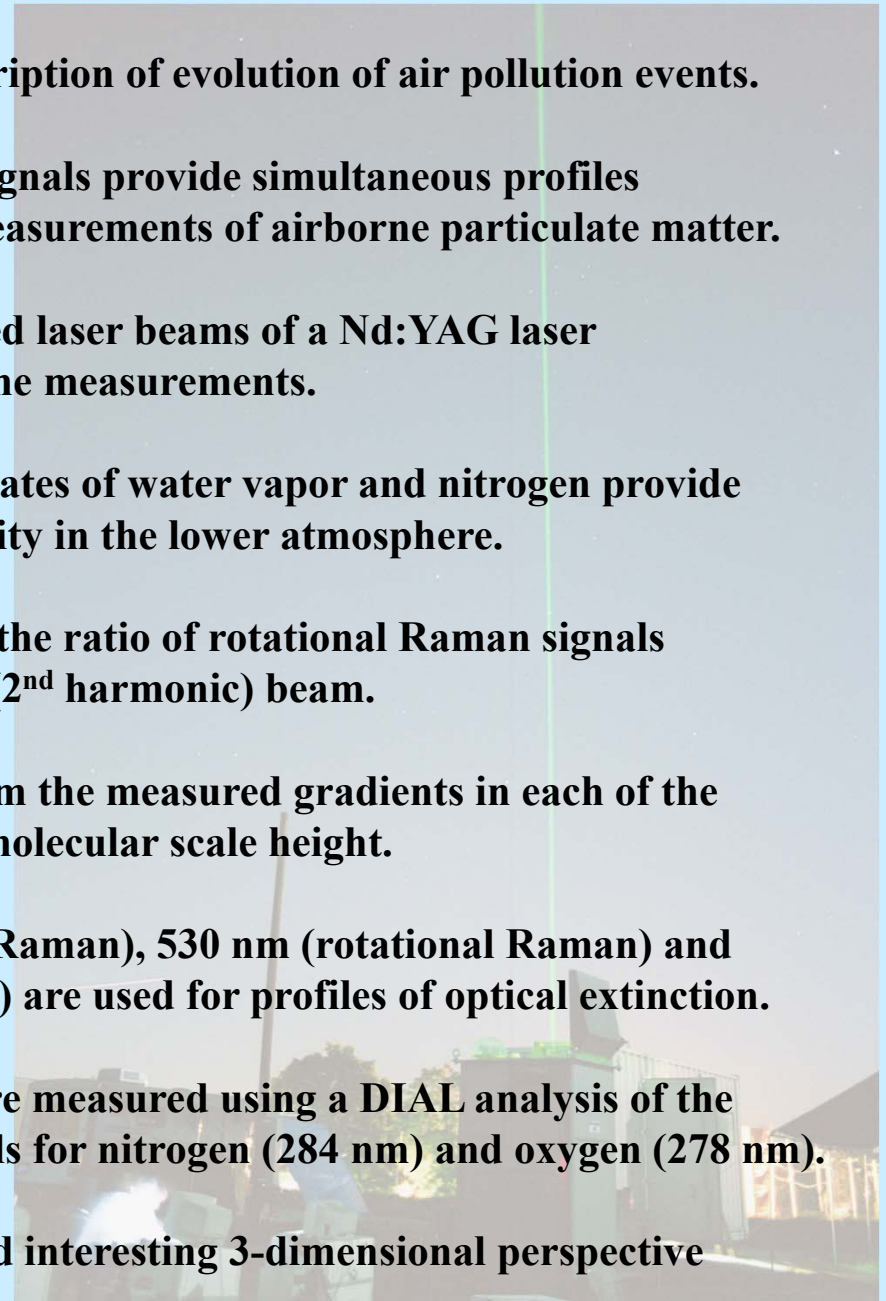
Air Quality Research Goals

- Investigate, understand and model the physical and chemical processes important in evolution of air pollution events
- Identify the local and transport sources that contribute to increased concentrations of ozone and PM_{2.5}
- Connect the sources of air pollution with population exposure and health effects
- Develop and test models which fully predict the distribution of air pollutants to predict and test regulatory measures
- Develop and improve the measuring techniques needed for process monitoring

Raman LIDAR Techniques –

Raman lidar techniques provide valuable description of evolution of air pollution events.

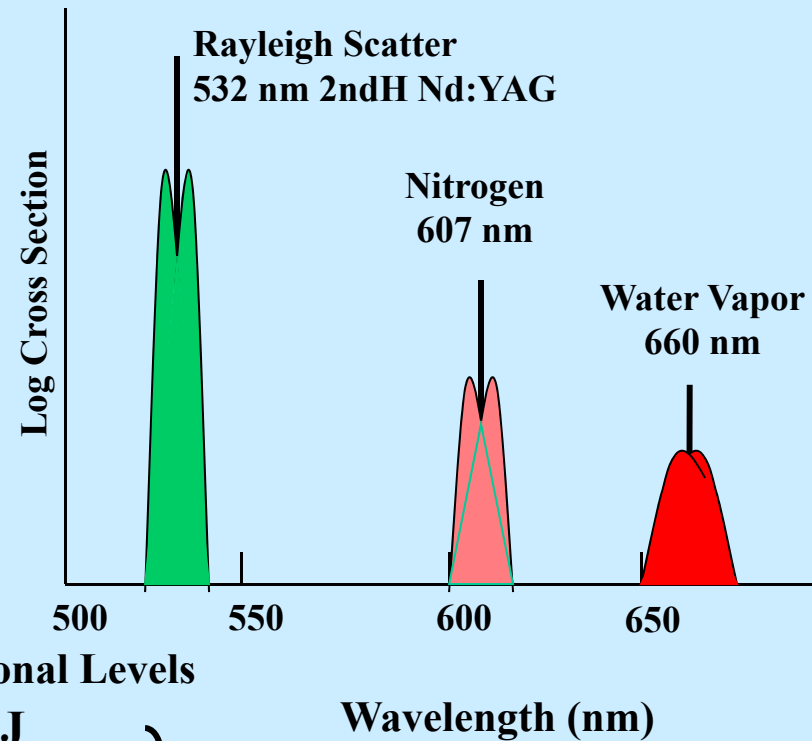
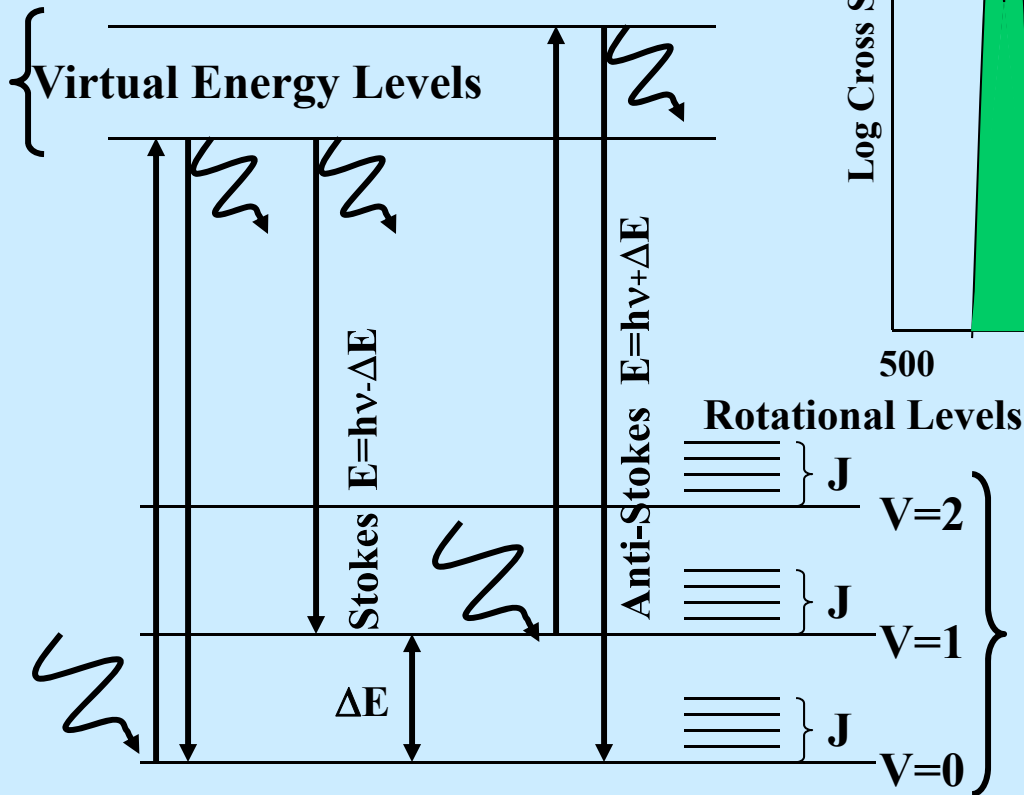
- The vibrational and rotational Raman lidar signals provide simultaneous profiles of meteorological data, ozone and measurements of airborne particulate matter.**
- We make use of 2nd and 4th harmonic generated laser beams of a Nd:YAG laser to provide both daytime and nighttime measurements.**
- The Raman scatter signals from vibrational states of water vapor and nitrogen provide robust profiles of the specific humidity in the lower atmosphere.**
- The temperature profiles are measured using the ratio of rotational Raman signals at 530 and 528 nm from the 532 nm (2nd harmonic) beam.**
- Optical extinction profiles are determined from the measured gradients in each of the molecular profiles compared to the molecular scale height.**
- Wavelengths of 284 nm (nitrogen vibrational Raman), 530 nm (rotational Raman) and 607 nm (nitrogen vibrational Raman) are used for profiles of optical extinction.**
- The ozone profiles in the lower troposphere are measured using a DIAL analysis of the ratio of the vibrational Raman signals for nitrogen (284 nm) and oxygen (278 nm).**
- Several campaigns have provided this new and interesting 3-dimensional perspective for air pollution events.**





Raman Scatter

Excited Electronic States



Vibration Energy Levels



Measurements by LAPS Lidar

<u>Property</u>	<u>Measurement</u>	<u>Altitude</u>	<u>Time Resolution</u>
Water Vapor	660nm/607nm	0 to 10 km	Night – 1 min
	294nm/285nm	0 to 3 km	Day & Night – 1 min
Temperature	528nm/530nm	0 to 10 km	Night – 5 min
Ozone	276nm/285nm	0 to 3 km	Day & Night – 10 min
Optical Extinction	285nm	0 to 5 km	Day & Night – 10 min
	530nm	0 to 10 km	Night – 5 min
	607nm	0 to 10 km	Night – 5 min

Ozone Measurement Technique

- DIAL (Differential Absorption Lidar) analysis of the Raman shift of N₂ (284 nm) and O₂ (277 nm), which occur on the steep side of the Hartley absorption band of ozone
- Integrated ozone density is obtained by taking the ratio of the O₂ and N₂ Raman-shifted signals, and correcting for molecular scattering and absorption
- The number density of ozone is calculated by differentiating the integrated ozone density, given by:

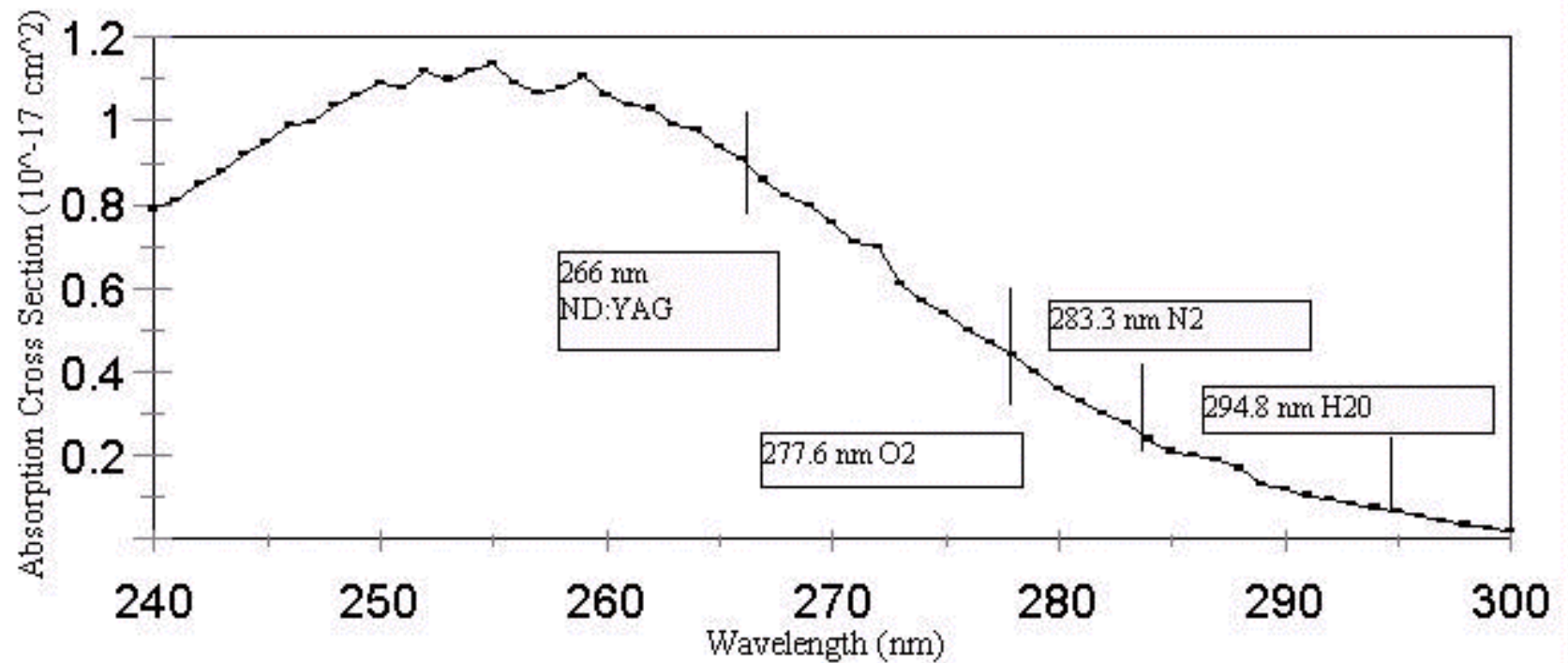
$$[O_3(z)] = \frac{d}{dz} \left[\ln \left(\frac{P_{O_2}(z)}{P_{N_2}(z)} \frac{1}{k_{system}} \right) \left(\frac{1}{(\sigma_{N_2} - \sigma_{O_2})} \right) + \frac{(\sigma_{O_2} - \sigma_{N_2})}{(\sigma_{N_2} - \sigma_{O_2})} K(z) \right]$$



Ozone Lidar

Ratio of O₂/N₂ Raman Signals

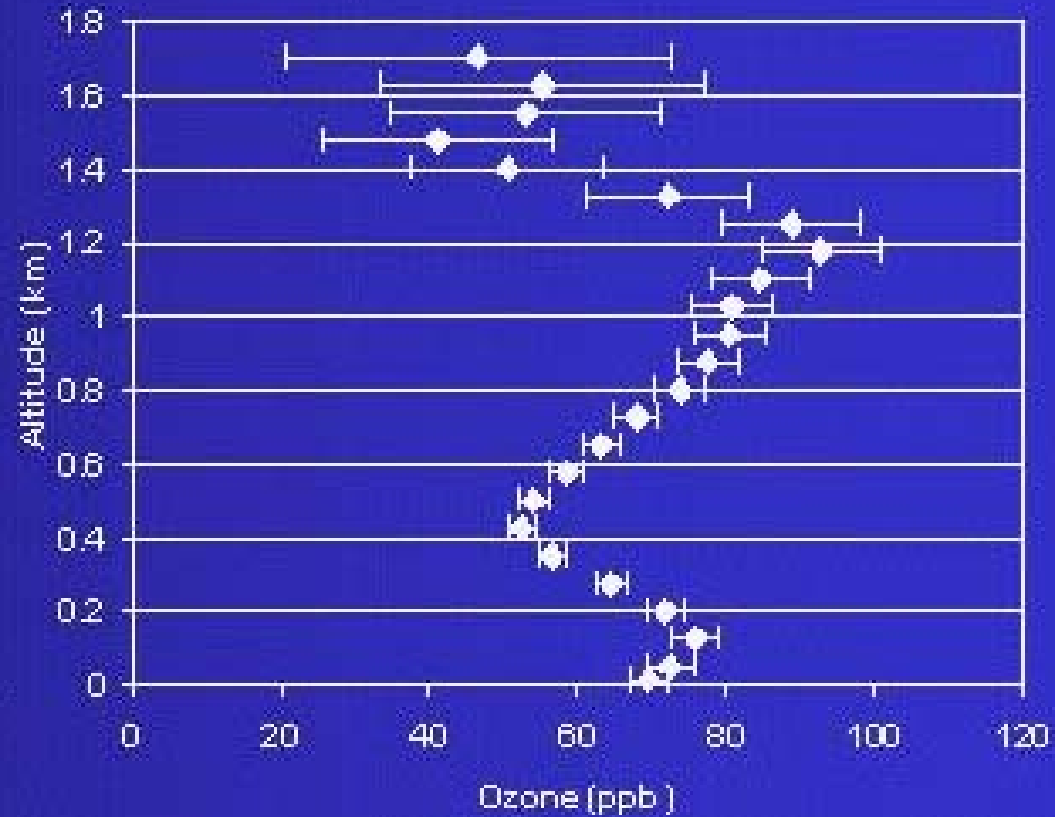
Ozone Absorption Cross Section
Hartley Band



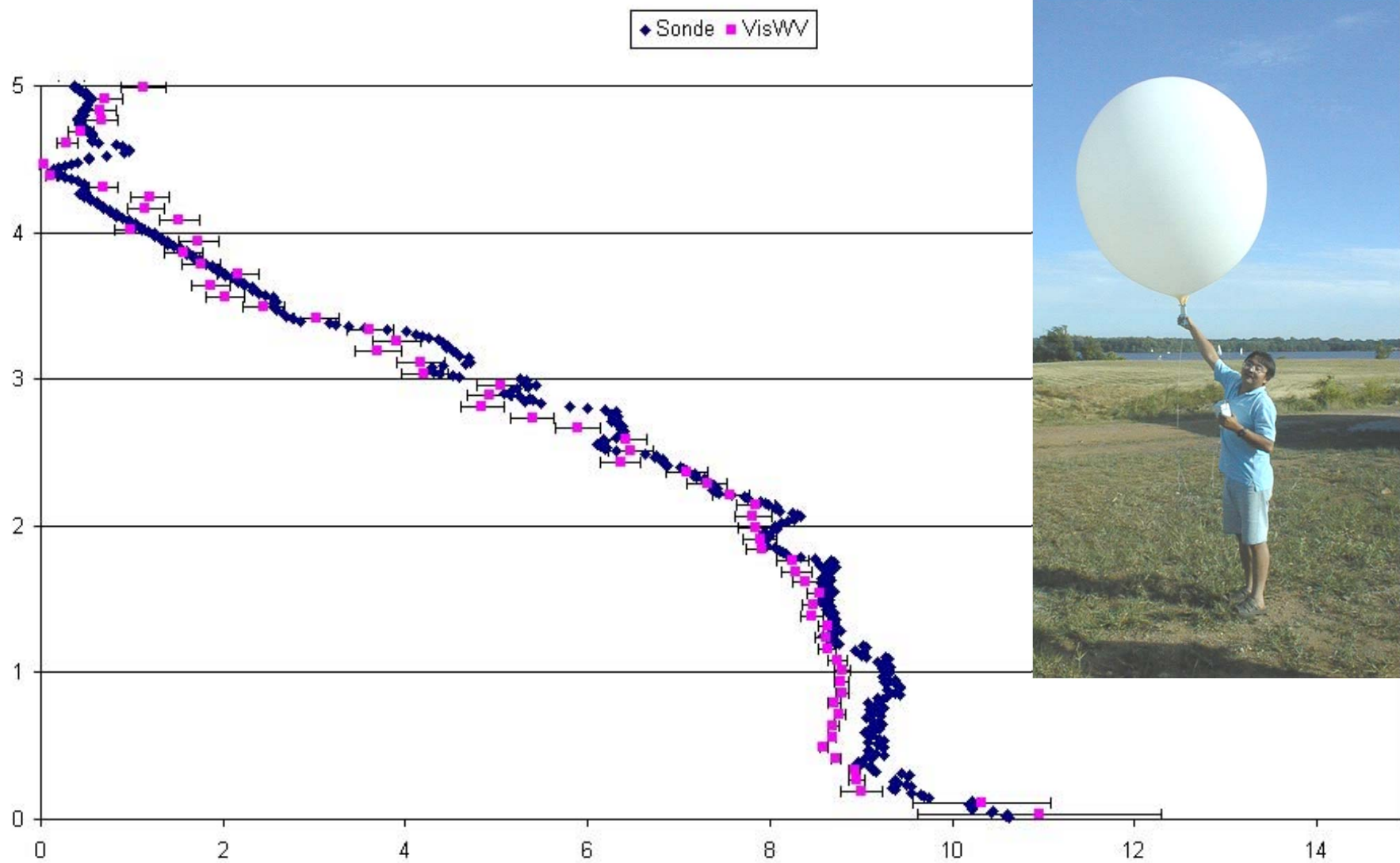


Ozone Profile

Ozone 08/21/98 15:00-15:59 UTC
Philadelphia, PA

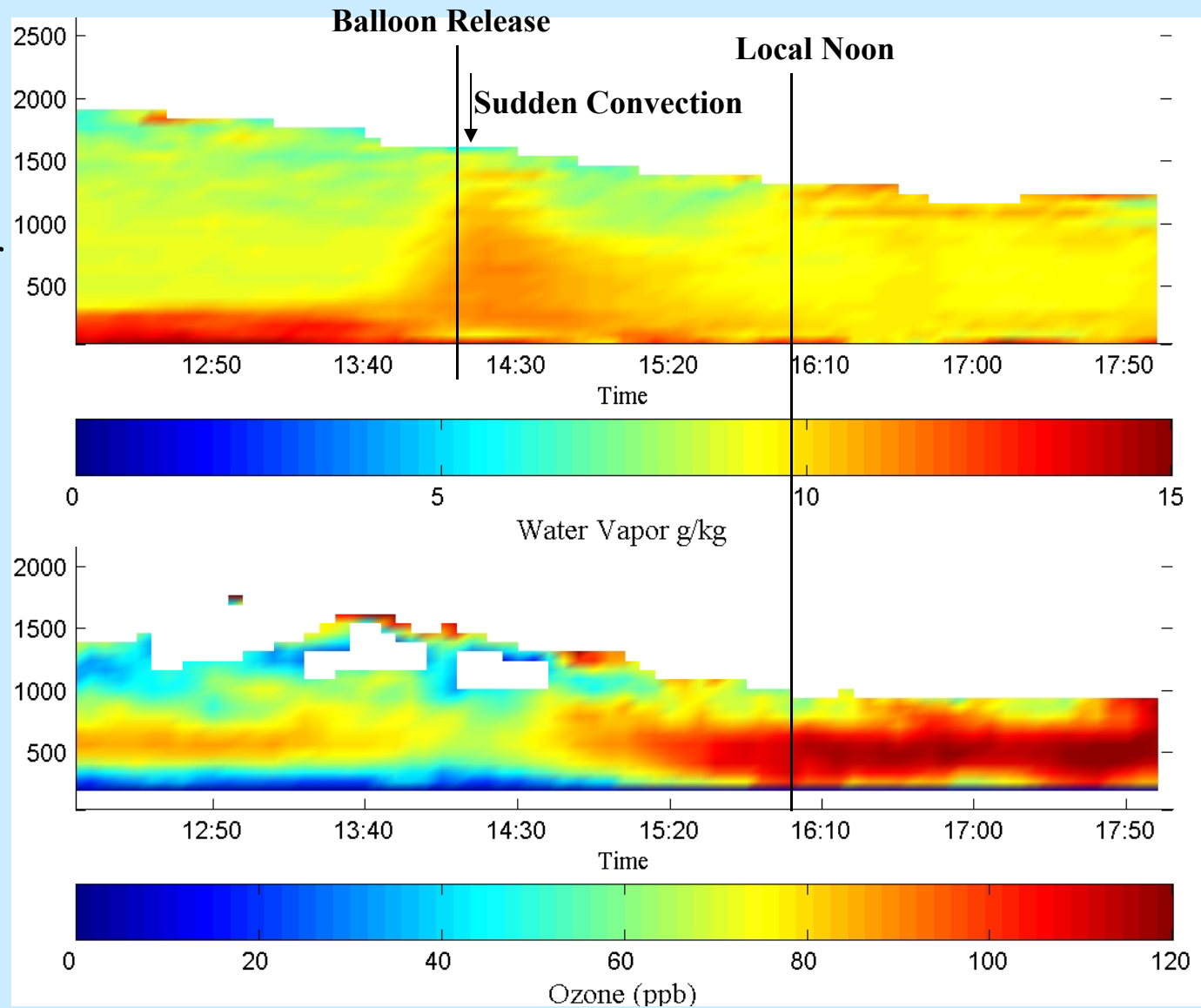


Comparison of Lidar and Sonde Water Vapor Mixing Ratio for 07/10/01
Sonde - 02:10 UTC ; Lidar 01:54 - 02:24 UTC

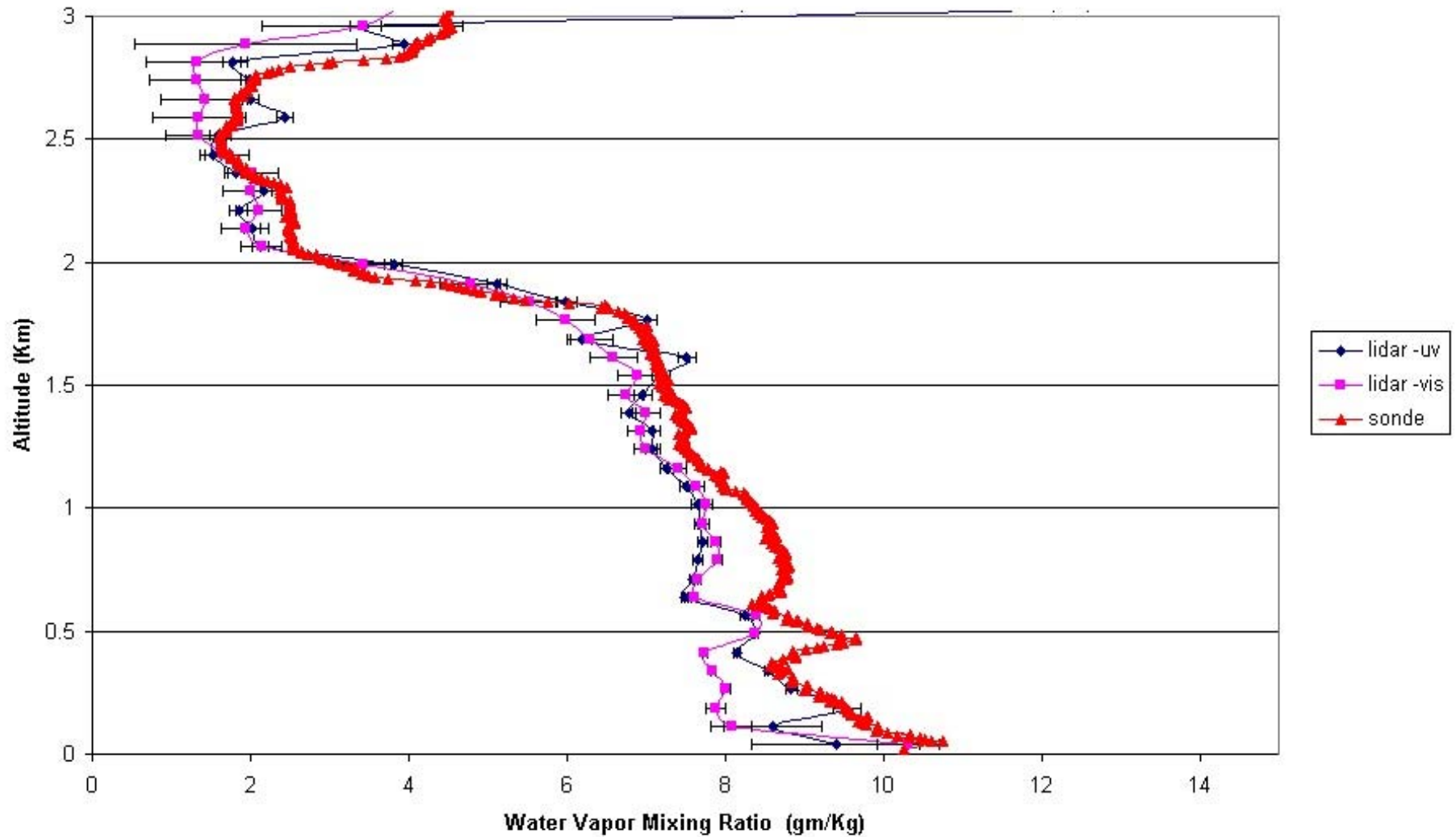


Water Vapor and Ozone – 10 July 2001 – 1200-1800 GMT (0800-1400 EDT)

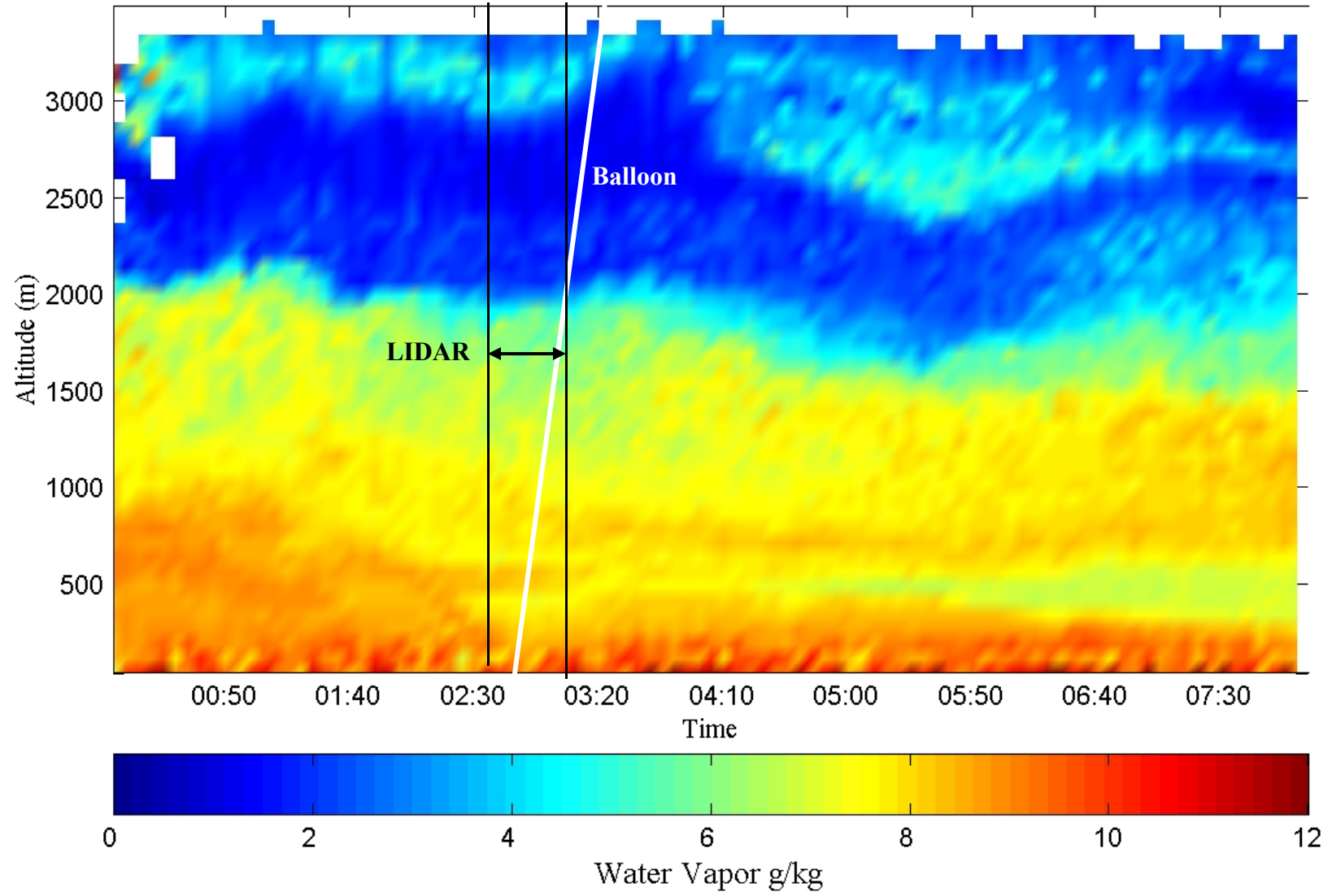
PSU LAPS
Lidar profiles
of water vapor
and ozone
during small
brief episode
on 10 July.

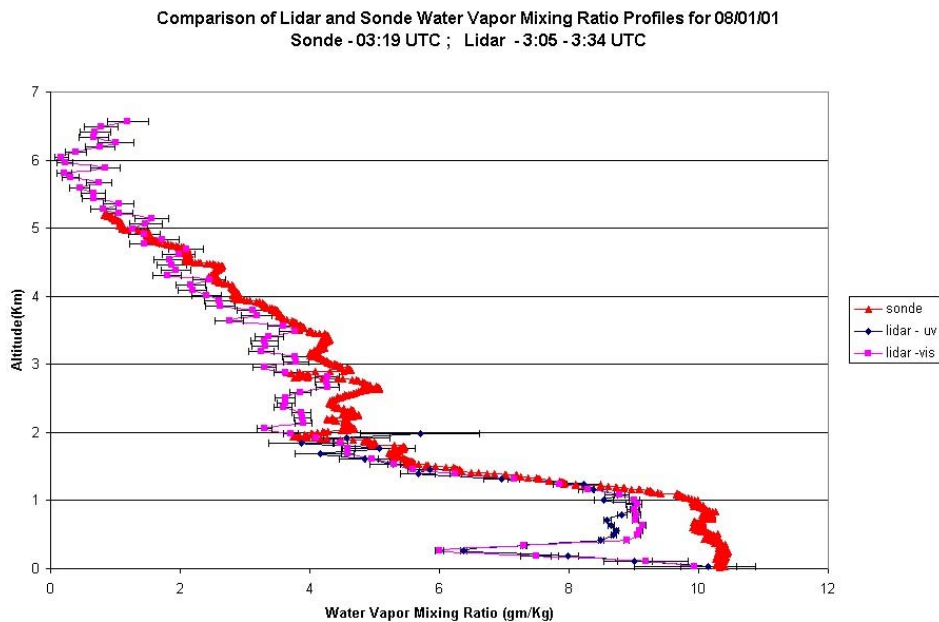
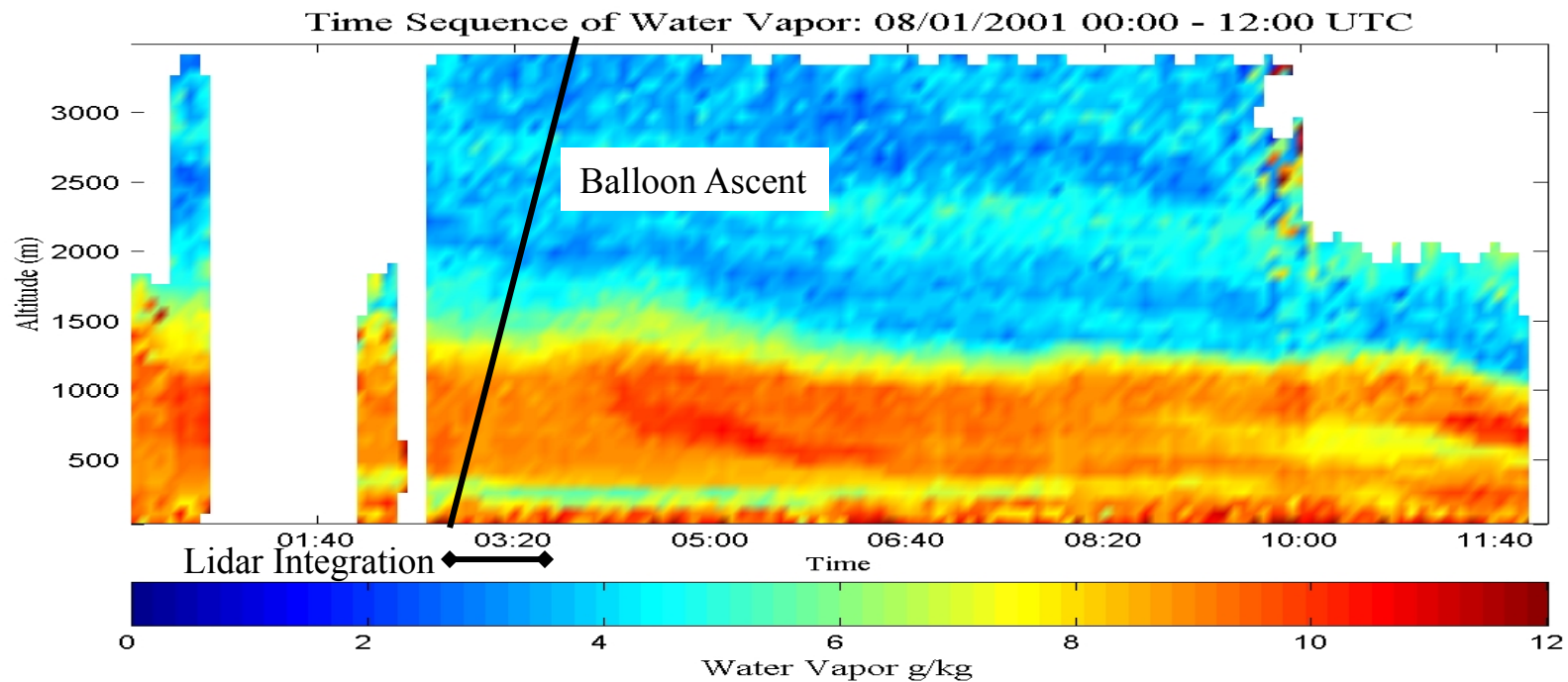


Comparison of Lidar and Sonde Water Vapor Mixing Ratio for July 31st
Sonde - 02:51 UTC ; Lidar 02:39 - 03:09 UTC



Time Sequence of Water Vapor: 07/31/2001 00:00 - 08:00 UTC

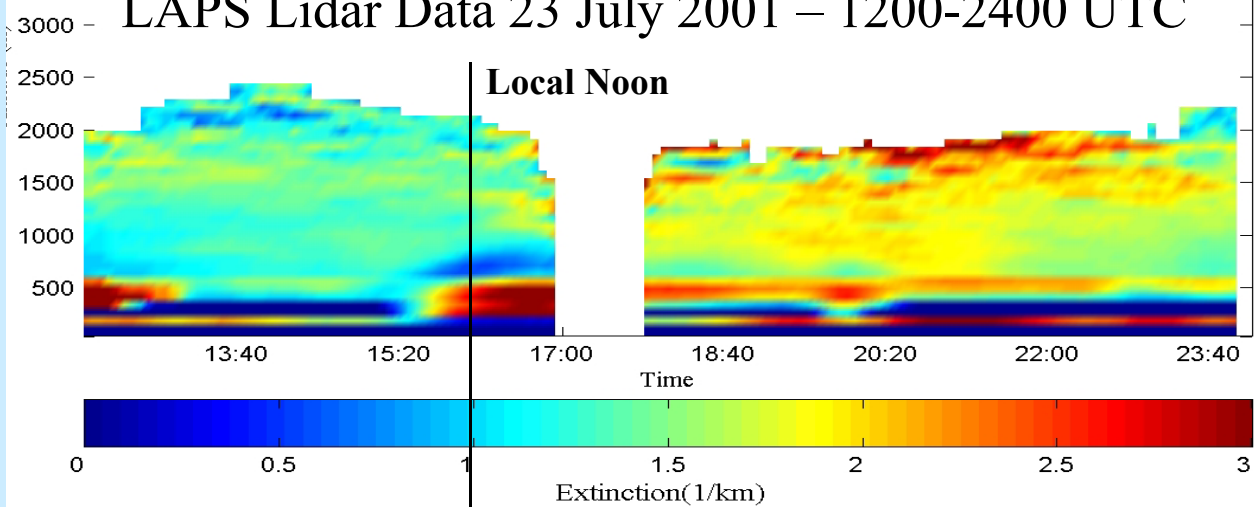




Lidar profile provides more accurate picture of atmospheric structure. Example is taken from night of 31 July – 1 August 2001 during NARSTO-NE-OPS in Philadelphia.

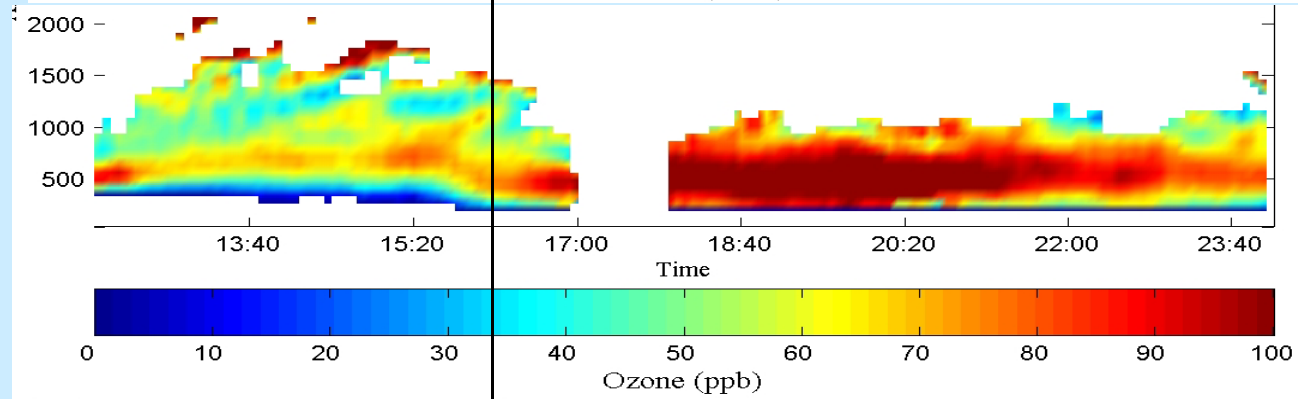
LAPS Lidar Data 23 July 2001 – 1200-2400 UTC

**Optical Extinction
284 nm**

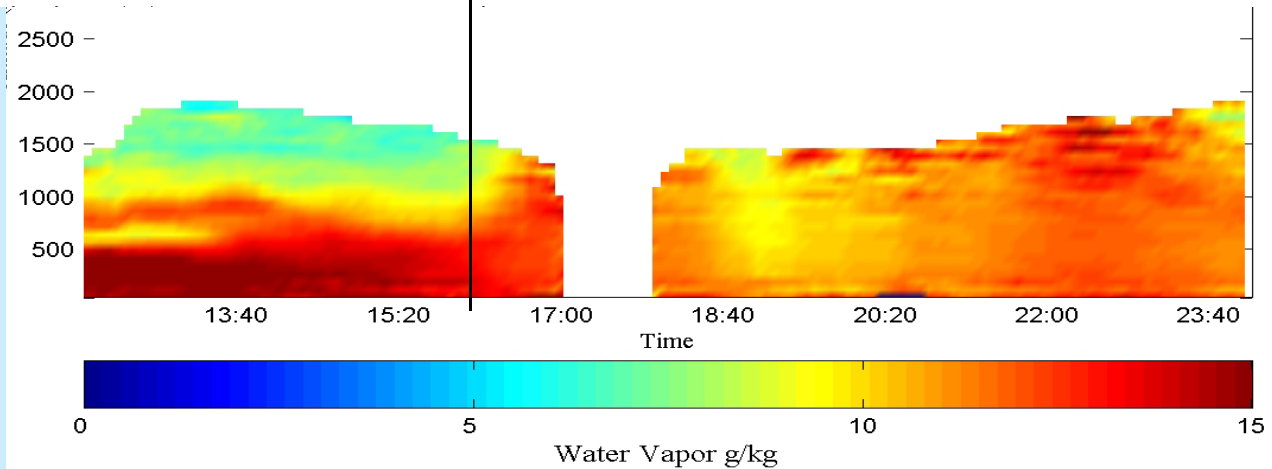


Ozone

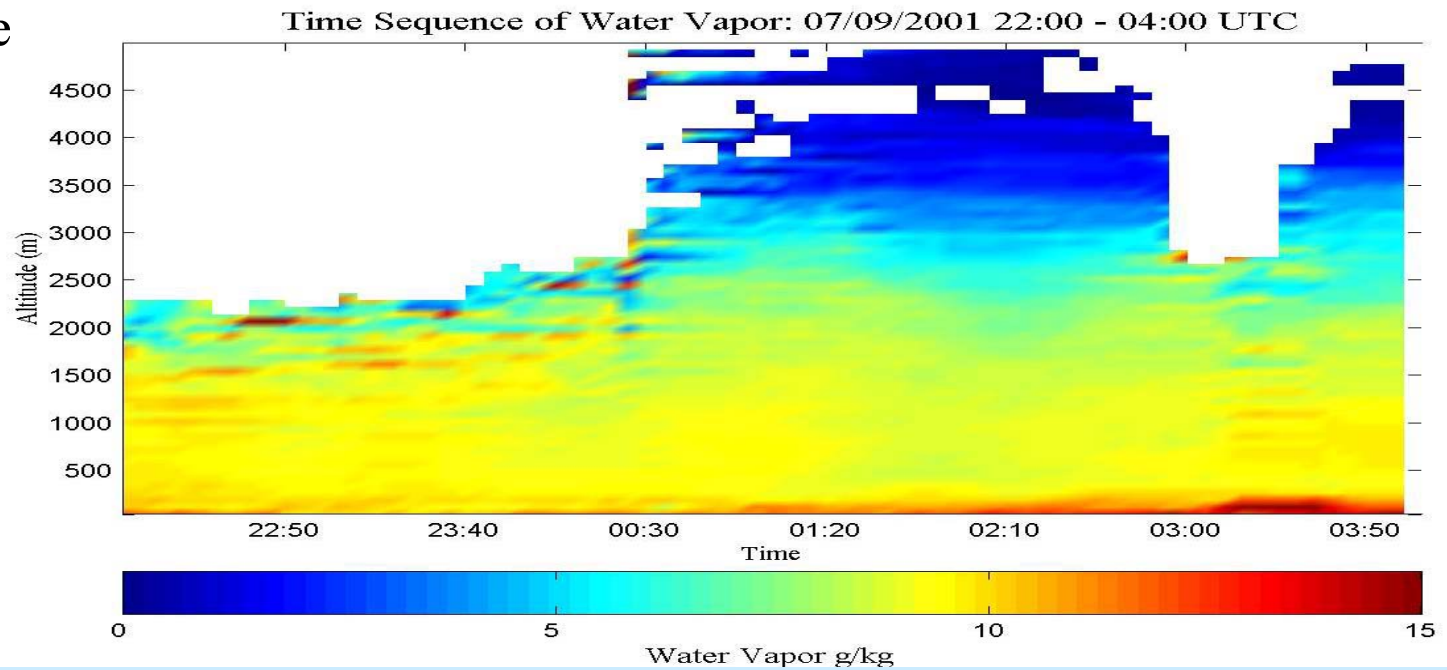
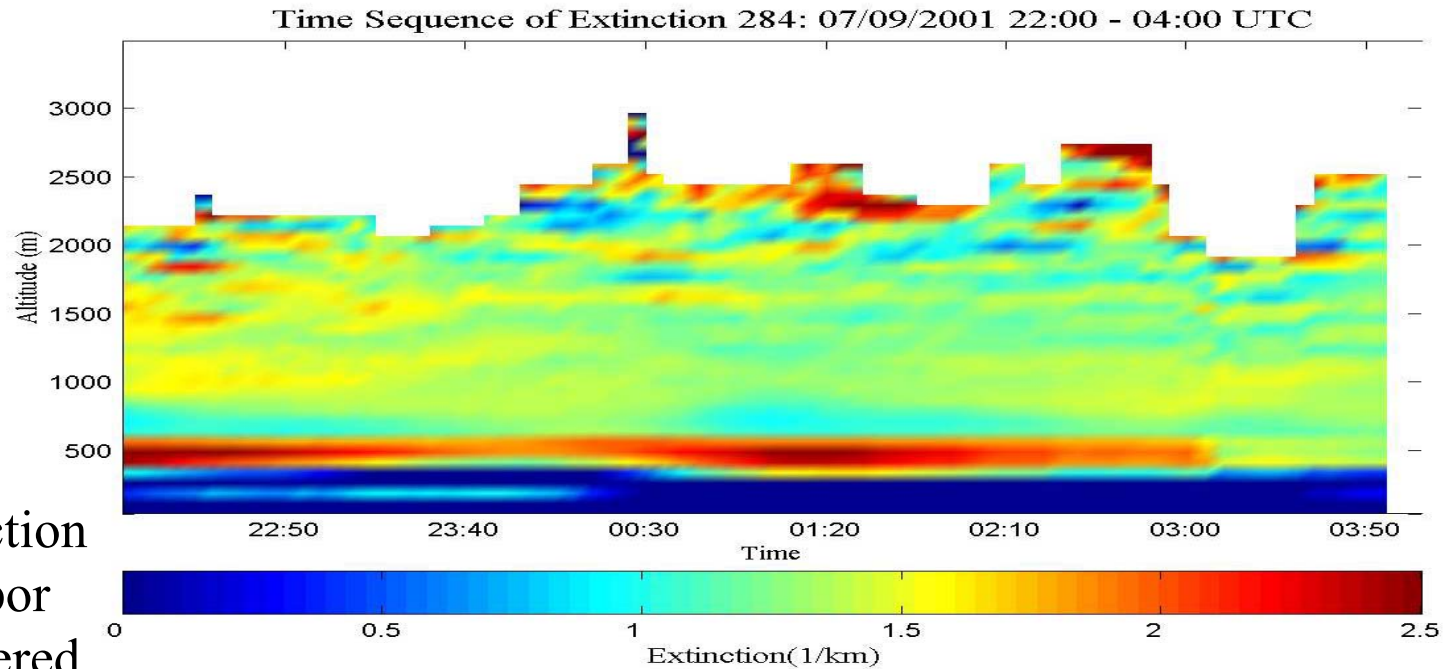
Altitude (km)



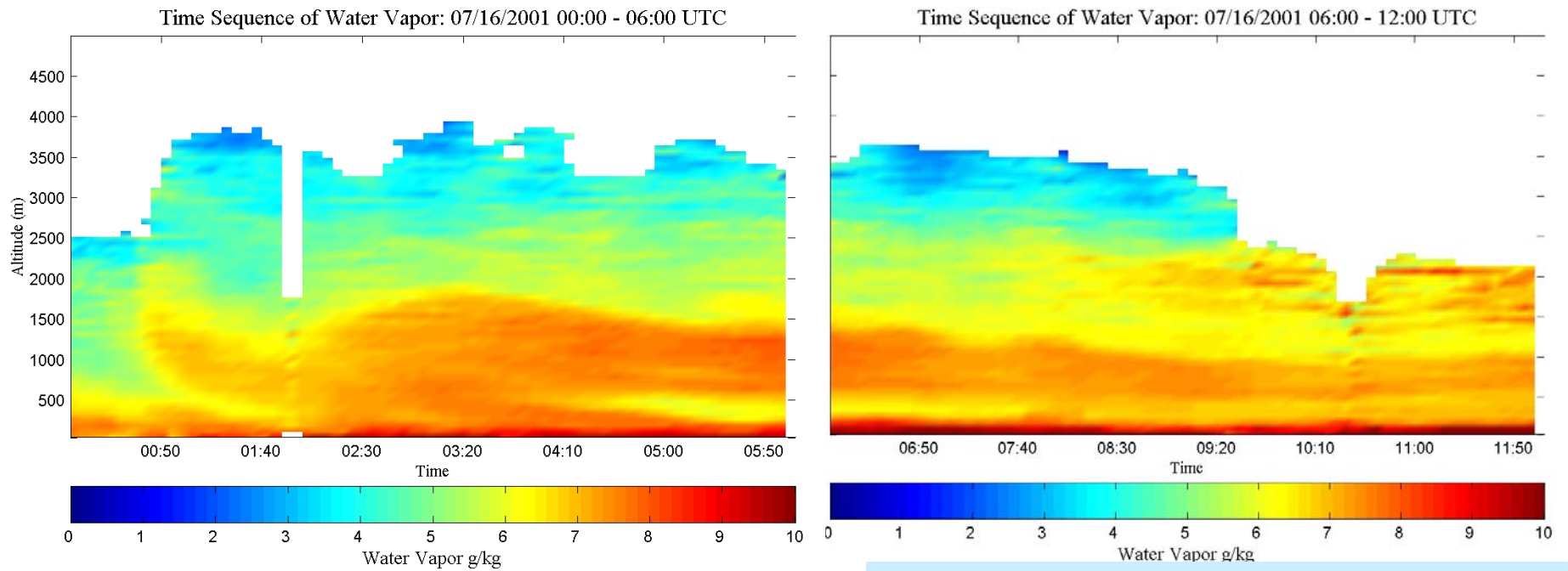
**Water Vapor
Specific Humidity**



Optical Extinction and Water Vapor data show layered structure at the same time reported by UMD aircraft.



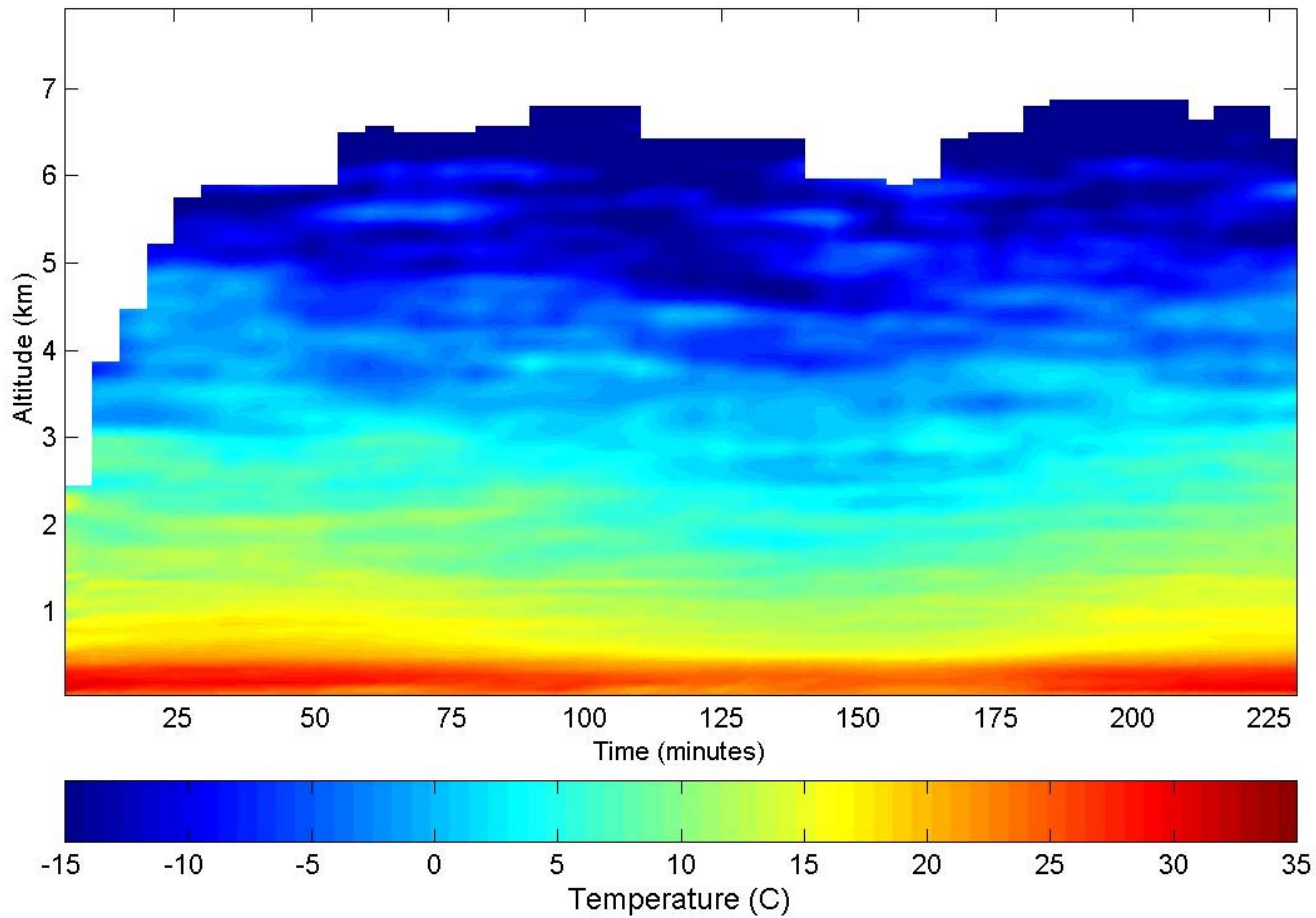
Variations of the nighttime boundary layer between 8PM and 8AM





Rotational Raman Temperature

Time Sequence of Temperature 07/02/99 00:56 - 04:42 UTC



plotted on: 07/26/00
 signal clipped at error: $\pm 5^\circ\text{C}$

step size: 5m
 integration period: 30m

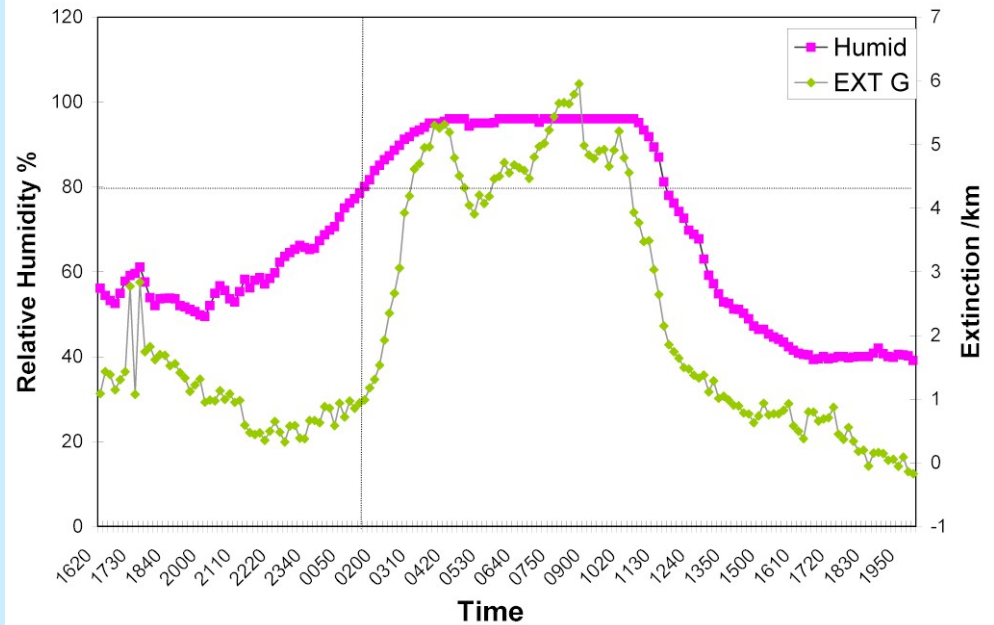
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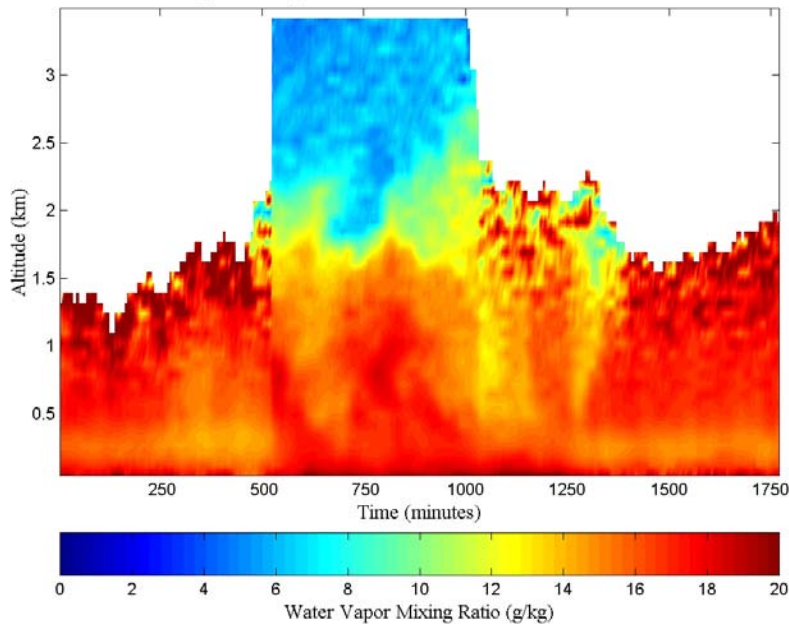
Humidity control of extinction

>80% relative humidity
causes striking increase
in extinction

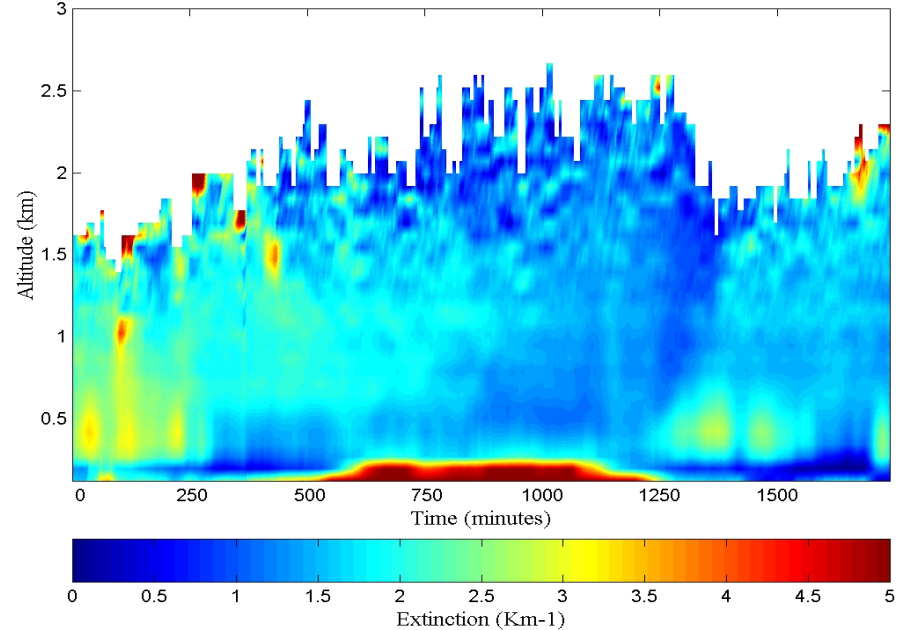
Ground Level Extinction and Relative Humidity
July 3 16:20 - July 4 22:10 UTC



Water Vapor Mixing Ratio - 07/03 - 07/04/99 16:09 - 21:50 UTC



Extinction 284 channel 07/03/99 16:35 -- 07/04/99 21:47UTC

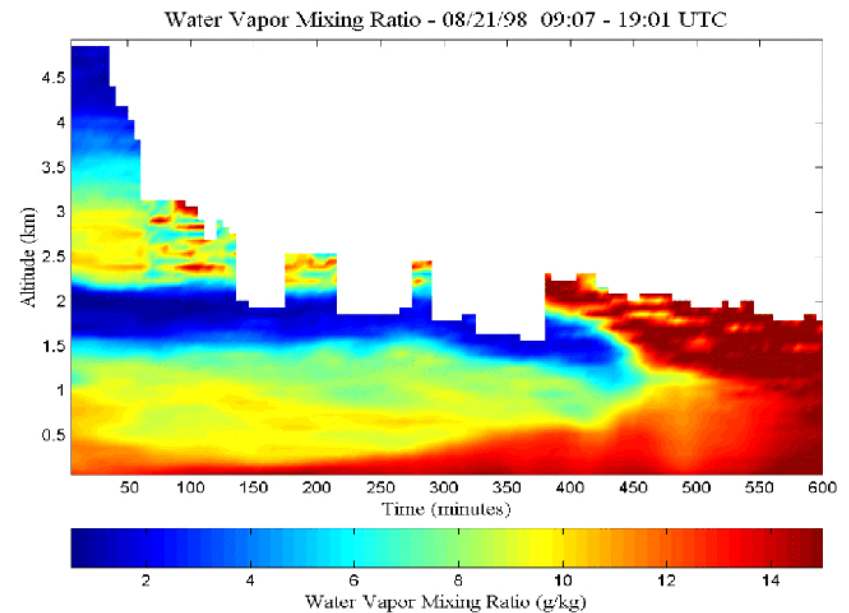
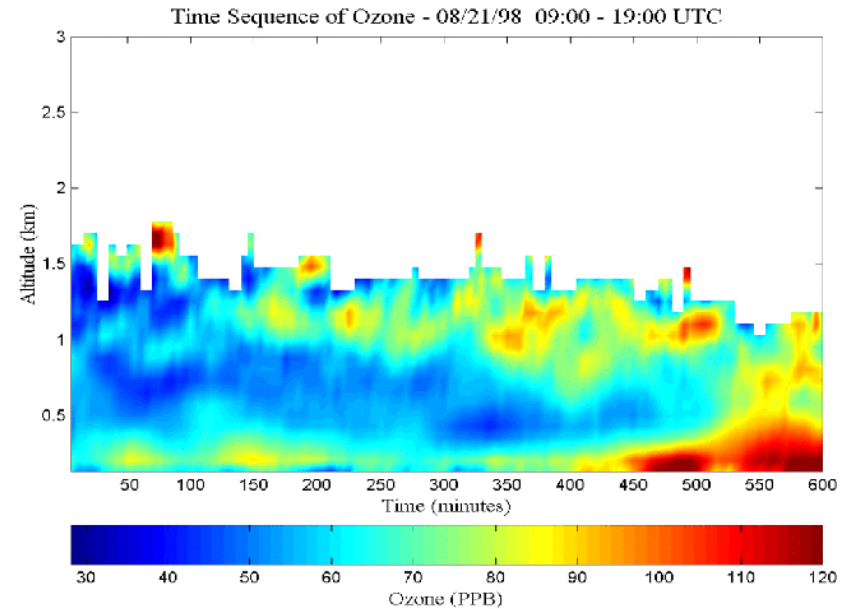


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Precursor Transport

NARSTO-NE-OPS – Ozone suddenly increases when an elevated layer carrying precursor chemicals from upper mid-west region mixes with rising PBL at about 1PM local time. The water vapor provides a tracer of the elevated layer. In this case, the air pollution event appears to be triggered by the precursor materials that are temperature sensitive such as PAN (peroxyacetyl nitrate).

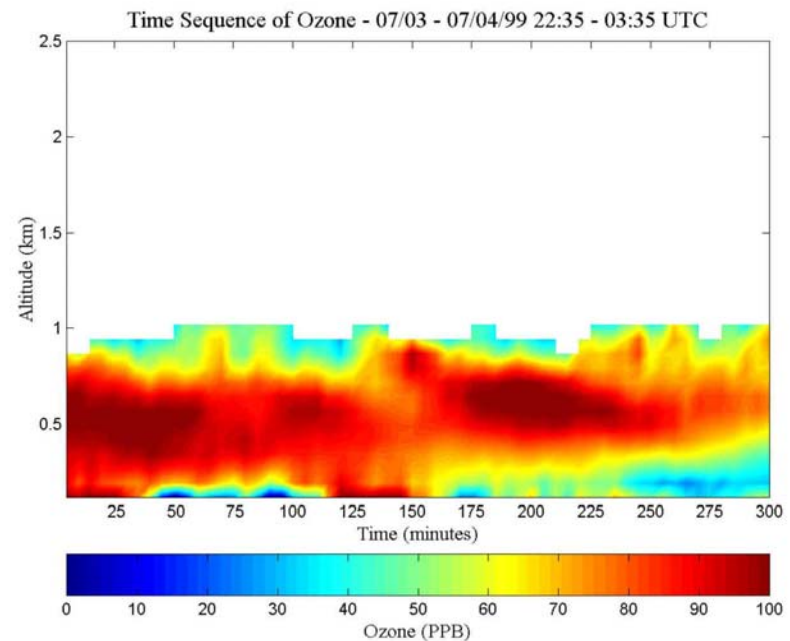
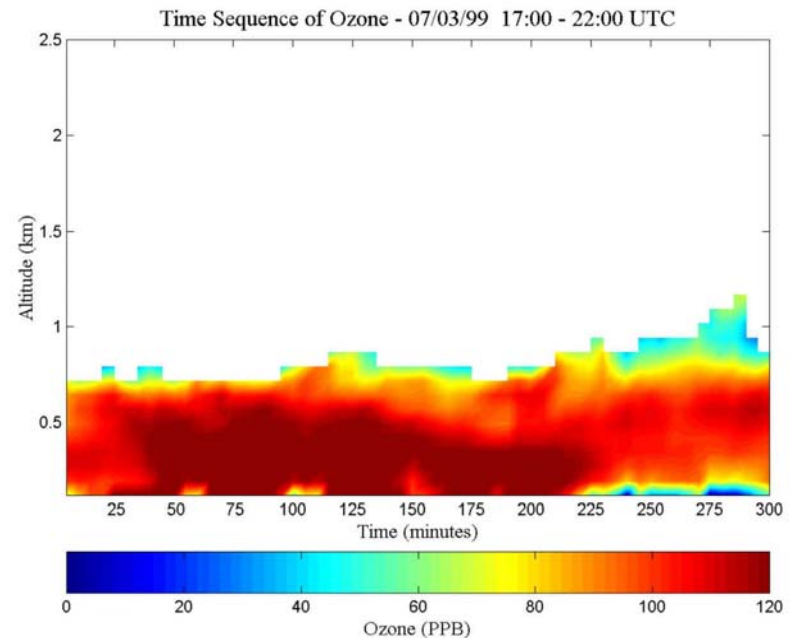


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10 Hour Sequence of Ozone

Afternoon rush hour provides sufficient NO_x to reduce ozone concentration, then formation of nocturnal inversion cuts off supply of ozone transport to replace surface losses – results in storage aloft at night.





Relationship between Ozone and Particulate Matter

- Tropospheric ozone is photochemically formed from chemical emissions of nitrogen oxides and volatile organic compounds
- Key ingredient in smog and contributes to detrimental health effects
- Ozone and PM are related through their formation processes and transportation
- Thermally dependent transport reservoirs of ozone precursors remain stable in cool temperatures can be transported over large distances
- When the temperature increases, the precursors will be released and can readily react to form ozone and other pollutant species, thus increasing both ozone and PM levels

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RAMAN LIDAR

- Raman lidar uses signal ratios and provides robust technique
- Several important properties can be routinely measured -
 - water vapor
 - temperature
 - ozone
 - optical extinction - 530 nm, 607 nm, 285 nm
 - optical backscatter - 532 nm, 266 nm
- Time sequences provide description of the dynamics (1 min step and 5 min smooth for water vapor and extinction, 10 min step and 30 min smooth for ozone and temperature)
- Lidar measurements are capable of providing the data needed to test and validate models and replace balloon sondes



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Summary

Raman Lidar has been demonstrated to provide important information of the 3-D characteristics of the meteorological and air quality properties: Ozone, Water Vapor, Optical Extinction, Temperature

Combining the Raman Lidar data with Doppler radar provides a complete set of results for testing model predictions, evaluating dynamical processes (vertical and horizontal) and describing the meteorology of the lower atmosphere.

Examples of preliminary results from the Raman Lidar data obtained as part of the NARSTO-NE-OPS project are shown. These results are expected to provide an important input to the 3-D picture of the local and regional processes controlling air pollution events.