Lecture 26. Aerosol Lidar

- Motivations to study aerosols and clouds
- Lidar detection of aerosol/cloud properties
- Single-channel vs multi-channel lidar

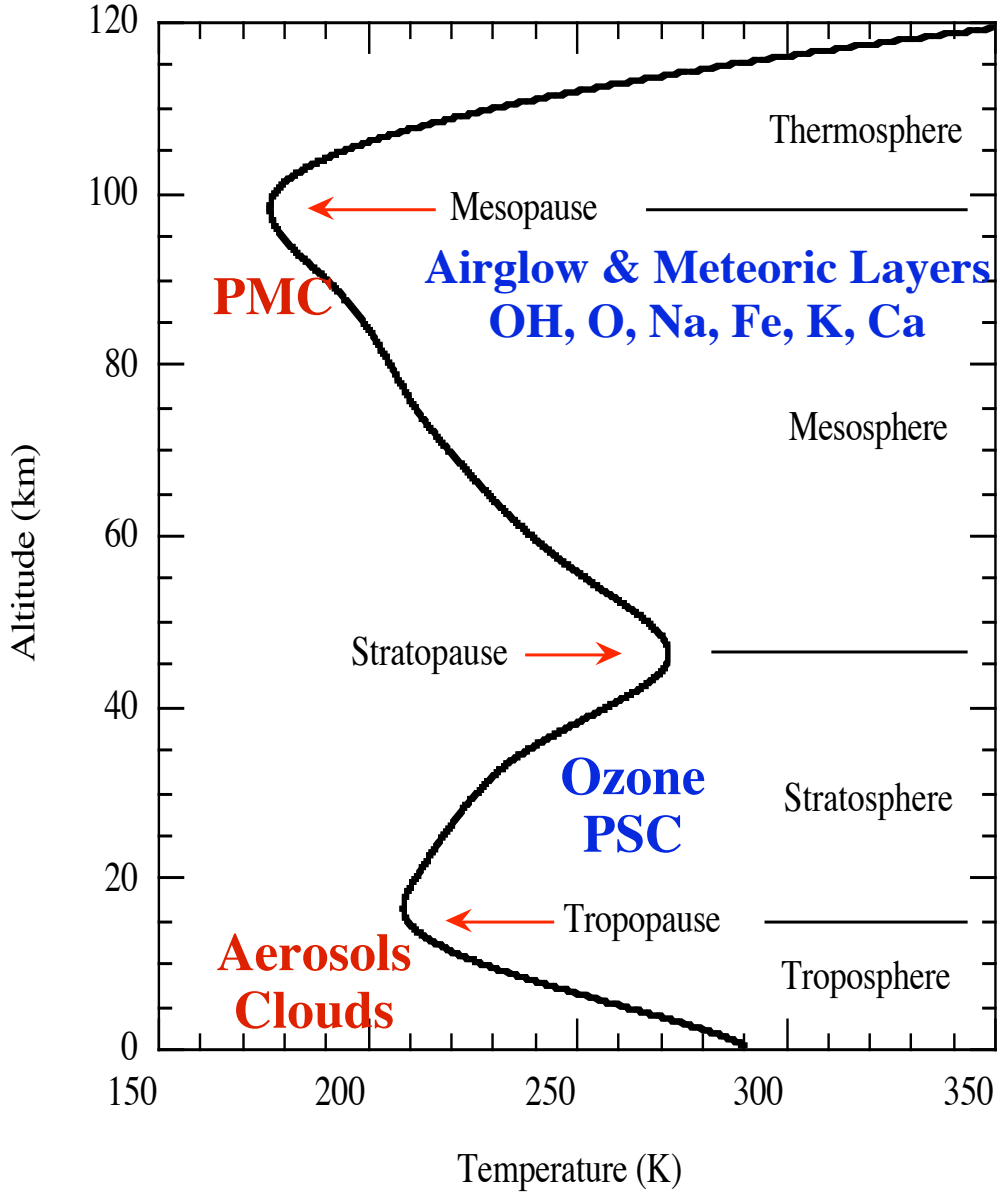
(Elastic-scattering lidar, Raman lidar and HSRL)

- Polarization detection
- Multi-wavelength detection
- Lidar detection of PMC and PSC
- Summary

Motivations to Study Aerosols

- Atmospheric aerosols play an important role in many atmospheric processes. Although only a minor constituent of the atmosphere, they have appreciable influence on the Earth's radiation budget, air quality and visibility, clouds, precipitation, and chemical processes in the troposphere and stratosphere.
- The occurrence, residence time, physical properties, chemical composition, and corresponding complex-refractive-index characteristics of the particles , as well as the resulting climate-relevant optical properties are subject to large diversity especially in the troposphere because of widely different sources and meteorological processes.
- Therefore, vertically resolved measurements of physical and optical properties of particles such as the particle surface-area concentration, volume and mass concentrations, mean particle size, and the volume extinction coefficient are of great interest.
- Routine (long-term), range-resolved observations of these parameters can only be carried out with lidar.

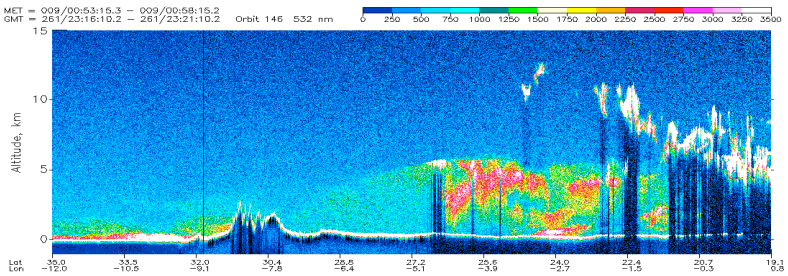
Aerosols and Clouds in Atmosphere



Polar mesospheric clouds (PMC) usually occur in polar summer

Polar stratospheric clouds (PSC) occur in polar winter and spring

Aerosols always present in troposphere with highly variable concentration and composition due to natural and anthropogenic sources



Nucleation mode: r < 0.1 μm
accumulation mode: 0.1 < r < 1 μm,
coarse mode: r > 1 μm

More Motivations

- Aerosol Properties – optical, microphysical, and chemical
- Cloud Properties – optical, microphysical, radiative for ice, water and mixed phase
- Aerosol-Cloud Interactions – aerosol act to seed clouds
- Air Quality and Pollutant Transport – Study long range transport, anthropogenic vs. natural sources, gas/aerosol interactions
- Direct and Indirect Aerosol Radiative Forcing – improve models
- Surface and Atmospheric Radiative effects
- Cloud – Radiation Dynamical Feedback Processes
- Properties of Mixed Phase Clouds
- Polar Stratospheric Clouds – distribution, properties and lifecycle
- Polar Mesospheric Clouds - indication of global climate change
- Aerosol influence is one of the major uncertainties in atmospheric models that are used to predict global climate change.
- All aerosols and clouds are also good tracers of atmosphere environment, so excellent natural indicator or laboratory.

Aerosol Properties vs Lidar Detection

- Physical Properties: Occurrence, Height, Residence Time, Vertical structure.
  - for any scattering lidar

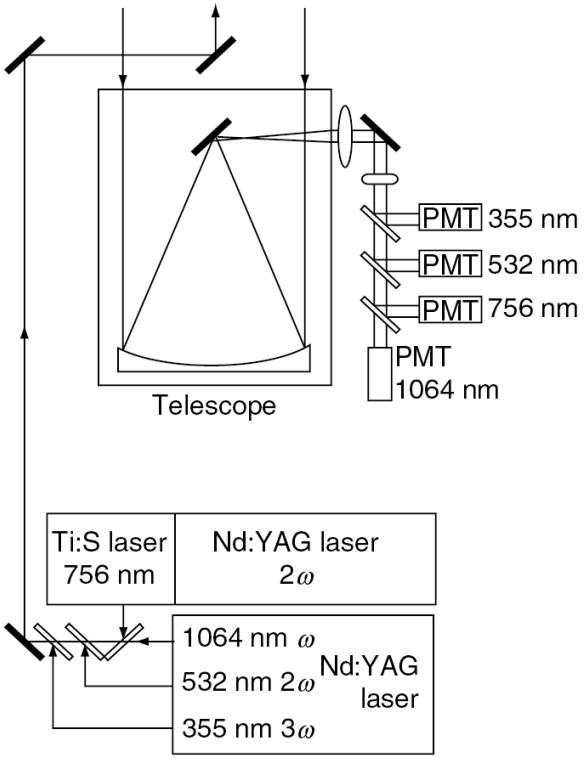
- Optical Properties: light backscatter, absorption, extinction, or albedo, complex-refraction-index
  - single-channel lidar versus multi-channel lidar

- Microphysical Properties: particle size, particle shape, number density, mass density, size distribution
  - multi-wavelength lidar and polarization detection lidar

- Chemical Composition and Process in the Atmosphere
  - laser-induced-breakdown with spectroscopic lidar

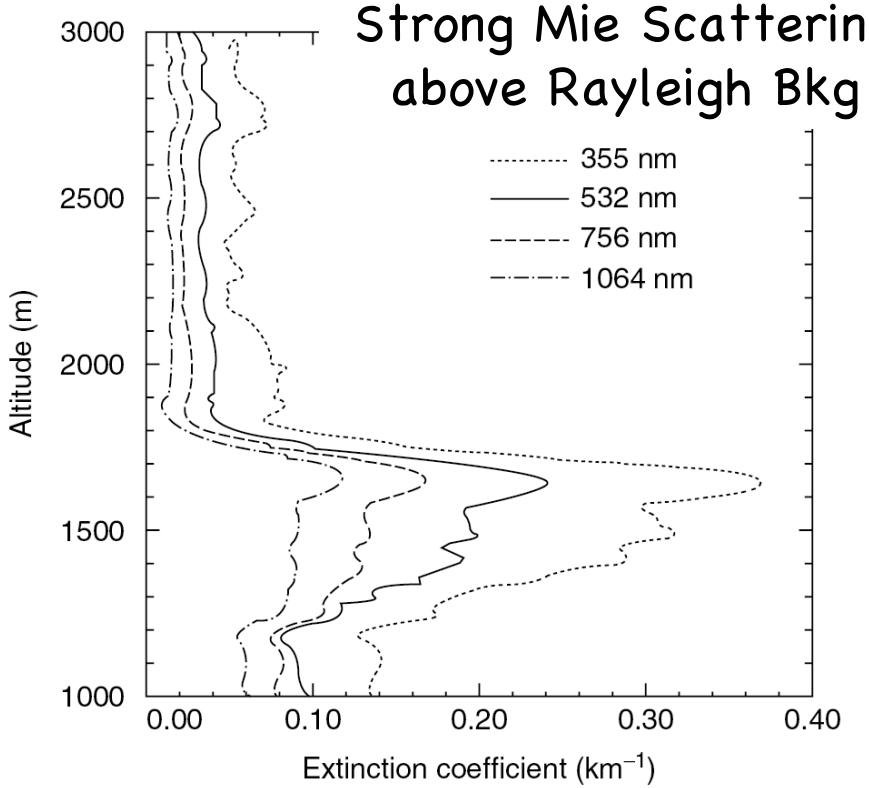
Lidar Detection of Aerosols

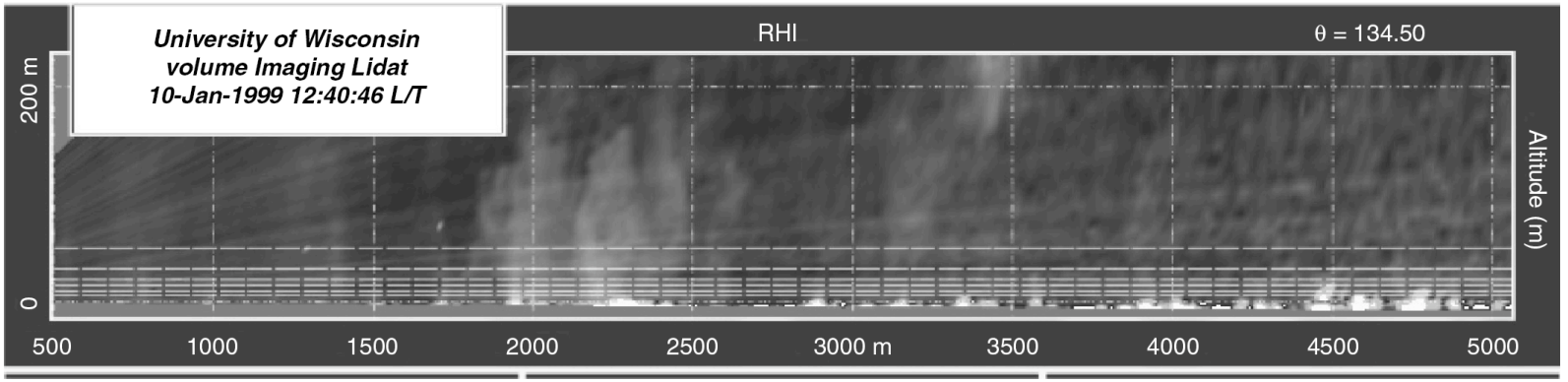
Co-axial Lidar



Block diagram of four-wavelength lidar.

Strong Mie Scattering above Rayleigh Bkg





Elastic-Scattering for Lower Atmos

- Lidar equation for elastic backscatter by air molecules and aerosols

$$P(R) = \frac{E_o \eta}{R^2} O(R) \beta(R) \exp\left[-2\int_0^R \alpha(r)dr\right]$$

O(R) - overlap between laser and FOV
β(R) - backscatter coefficient ($km^{-1}sr^{-1}$)
α(R) - extinction coefficient (km^{-1})

- Backscatter and extinction are contributed by aerosols and molecules

$$\beta(R) = \beta_{aer}(R) + \beta_{mol}(R)$$
$$\alpha(R) = \alpha_{aer}(R) + \alpha_{mol}(R)$$

Index aer refer to aerosol particles
Index mol refer to air molecules

- Combine above equation, we obtain

$$S(R) = P(R)R^2 = E_0 \eta_L \left[\beta_{aer}(R) + \beta_{mol}(R)\right] \exp\left[-2\int_o^R \left[\alpha_{aer}(r) + \alpha_{mol}(r)\right]dr\right]$$

Note: backscatter is local effect while extinction is integrated effect

- Recall the Rayleigh scattering from air molecules $\beta_{mol}(R)$ can be determined from atmosphere temperature and pressure

$$\beta_{mol}(\lambda, z_R) = 2.938 \times 10^{-32} \frac{P(z_R)}{T(z_R)} \cdot \frac{1}{\lambda^{4.0117}}$$

z_R is altitude for range R

Elastic-Scattering Cont'd

- Ignore the molecular absorption, then the extinction by air molecules is the integration of molecular angular scattering coefficient through the entire 4π solid angles. Rayleigh scattering is anisotropic

$$\beta(\theta) = \frac{\beta_T}{4\pi} P(\theta) = \frac{\beta_T}{4\pi} \times 0.7629 \times (1 + 0.9324\cos^2\theta)$$

$$\alpha_{mol}(R) = \beta_T = \frac{8\pi}{3} \cdot \beta(R,\pi)$$

- Define molecule extinction-to-backscatter ratio (lidar ratio) as

$$L_{mol} = \frac{\alpha_{mol}(R)}{\beta_{mol}(R)} = \frac{8\pi}{3} sr$$

The lidar ratio for molecule is range independent

- Thus, among the four parameters $\beta_{mol}(R)$, $\beta_{aer}(R)$, $\alpha_{mol}(R)$, $\alpha_{aer}(R)$, two are known, and the other two $\beta_{aer}(R)$ and $\alpha_{aer}(R)$ are unknown.

- Define the lidar ratio for aerosols (extinction-to-backscatter ratio)

$$L_{aer}(R) = \frac{\alpha_{aer}(R)}{\beta_{aer}(R)}$$

$$Y(R) = L_{aer}(R)\left[\beta_{aer}(R) + \beta_{mol}(R)\right]$$

Derive Aerosol Backscatter and Extinction in Lower Atmosphere

- Substitute lidar ratios and Y(R) into the lidar equation for S(R),

$$S(R)L_{aer}(R)\exp\left\{-2\int_o^R\left[L_{aer}(r)-L_{mol}\right]\beta_{mol}(r)dr\right\}=E_0\eta_L Y(R)\exp\left[-2\int_0^R Y(r)dr\right]$$

- Taking the logarithms of both sides of above equation and differentiating them with respect to R, we obtain

$$\frac{d\ln\left(S(R)L_{aer}(R)\exp\left\{-2\int_o^R\left[L_{aer}(r)-L_{mol}\right]\beta_{mol}(r)dr\right\}\right)}{dR}=\frac{1}{Y(R)}\frac{dY(R)}{dR}-2Y(R)$$

- This is a Bernoulli equation, and can be solved for the following boundary condition

$$Y(R_0)=L_{aer}(R_0)\left[\beta_{aer}(R_0)+\beta_{mol}(R_0)\right]$$

Klett Method

- By solving the Bernoulli equation, we obtain the backscatter coefficient

$$\beta_{aer}(R)+\beta_{mol}(R)=\frac{S(R)\exp\left\{-2\int_{R_0}^{R}\left[L_{aer}(r)-L_{mol}\right]\beta_{mol}(r)dr\right\}}{\frac{S(R_0)}{\beta_{aer}(R_0)+\beta_{mol}(R_0)}-2\int_{R_0}^{R}L_{aer}(r)S(r)T(r,R_0)dr}$$

where $$T(r,R_0)=\exp\left\{-2\int_{R_0}^{r}\left[L_{aer}(r')-L_{mol}\right]\beta_{mol}(r')dr'\right\}$$

- The aerosol extinction coefficient can be estimated by

$$\alpha_{aer}(R)=L_{aer}(R)\beta_{aer}(R)$$

- The solution for the Bernoulli equation can be integrated by starting from reference range R_0, which may be either the near end ($R > R_0$, forward integration), or the remote end ($R < R_0$, backward integration).
- Numerical stability is only given by the backward integration, which is called Klett method [1981].

How Reliable is Single-Channel Lidar?

- The above method is basically using one lidar equation to determine two unknown parameters. Its accuracy critically relies on the input parameter $L_{aer}(R)$ - lidar ratio for aerosol.
- However, this quantity depends on the microphysical, chemical and morphological properties of the particles, and varies strongly with height, especially when marine, anthropogenic (urban, biomass burning), and desert dust particles or mixtures of these are present in layers above each other. Even in the well-mixed layer, the lidar ratio is not constant with height because relative humidity increases with height.

Aerosol type	Lidar ratio
Marine particles	20-35 sr
Saharan dust	50-80 sr
Less absorbing urban particles	35-70 sr
Absorbing particles from biomass burning	70-100 sr

- Variations between 20-100 sr make it practically impossible to estimate trustworthy extinction profiles from single-channel data in lower atmos.

Raman Lidar and HSRL

- To infer aerosol extinction more trustworthy, the key is to add additional channel to provide addition information. At least, two channels of lidar profiles are needed. Raman lidar and HSRL are two major solutions to this problem.

- One method is the measurement of two lidar profiles in one of which the aerosol scattering is zero $\beta_{aer} = 0$. This is the case in Raman lidar. Only molecules, not aerosols, contribute to the inelastic Raman backscatter profile produced by molecular nitrogen or oxygen.

(1) The elastic lidar return is affected by both aerosol extinction α_{aer} and aerosol backscatter β_{aer}.

(2) But Raman lidar return is only affected by aerosol extinction α_{aer} alone, as aerosol scattering β_{aer} is at a different wavelength than the Raman return wavelength, so won't be received by the Raman channel.

- Another method is the high-spectral-resolution lidar (HSRL) that has two channels - one to measure pure molecular scattering, and another to measure the combination of aerosol and molecular scattering.

Raman Scattering Lidar

- Raman lidar measures

(1) elastic scattering from aerosols and molecules, and

(2) inelastic (Raman) scattering by N_2 or O_2 molecules.

For example, if 532 nm laser is Raman shifted by N_2, the return Raman signal is at 607 nm.

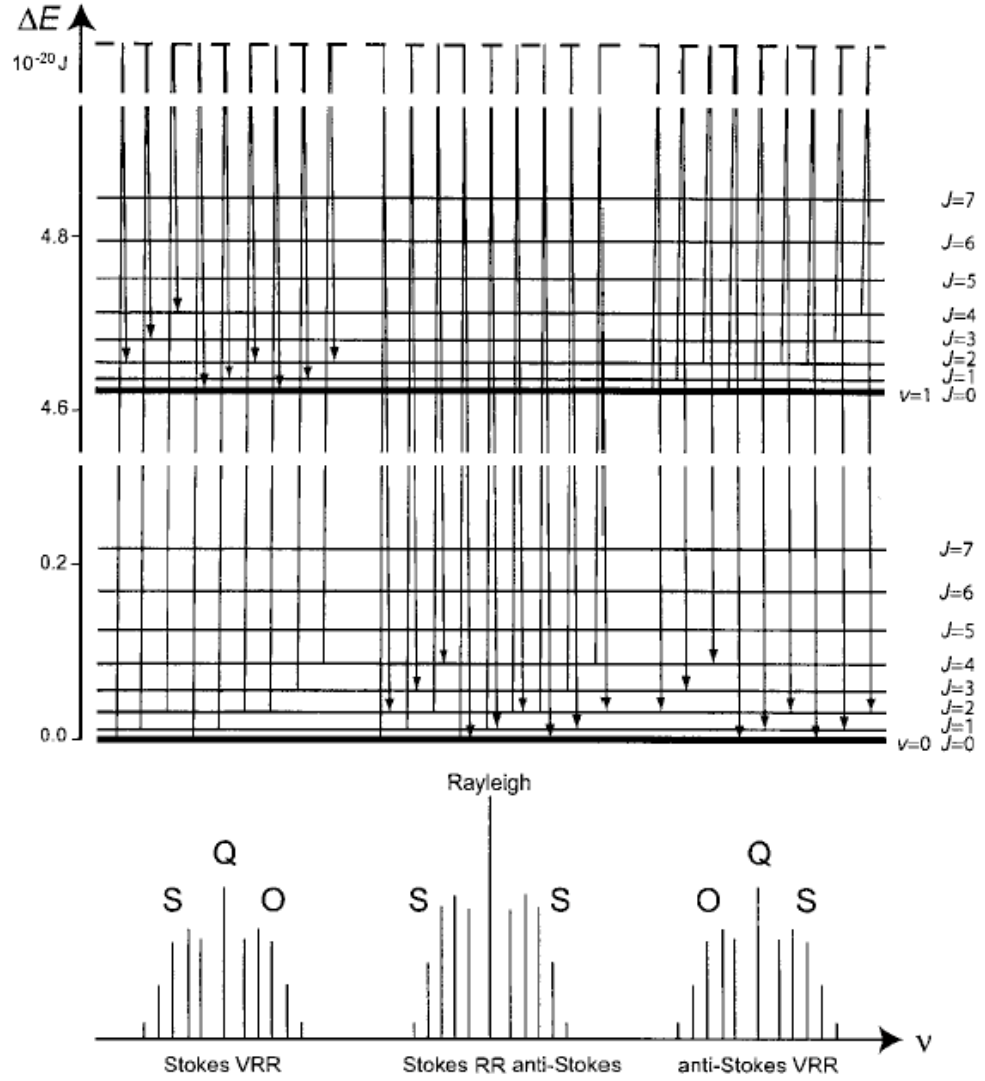


Fig. 9.1. Vibration–rotation energy levels of the N_2 molecule, Raman transitions, and resulting spectrum.

HSRL

- High-Spectral-Resolution-Lidar (HSRL) is to measure the molecule scattering separately from the aerosol scattering, utilizing the different spectral distribution of the Rayleigh and Mie scattering.

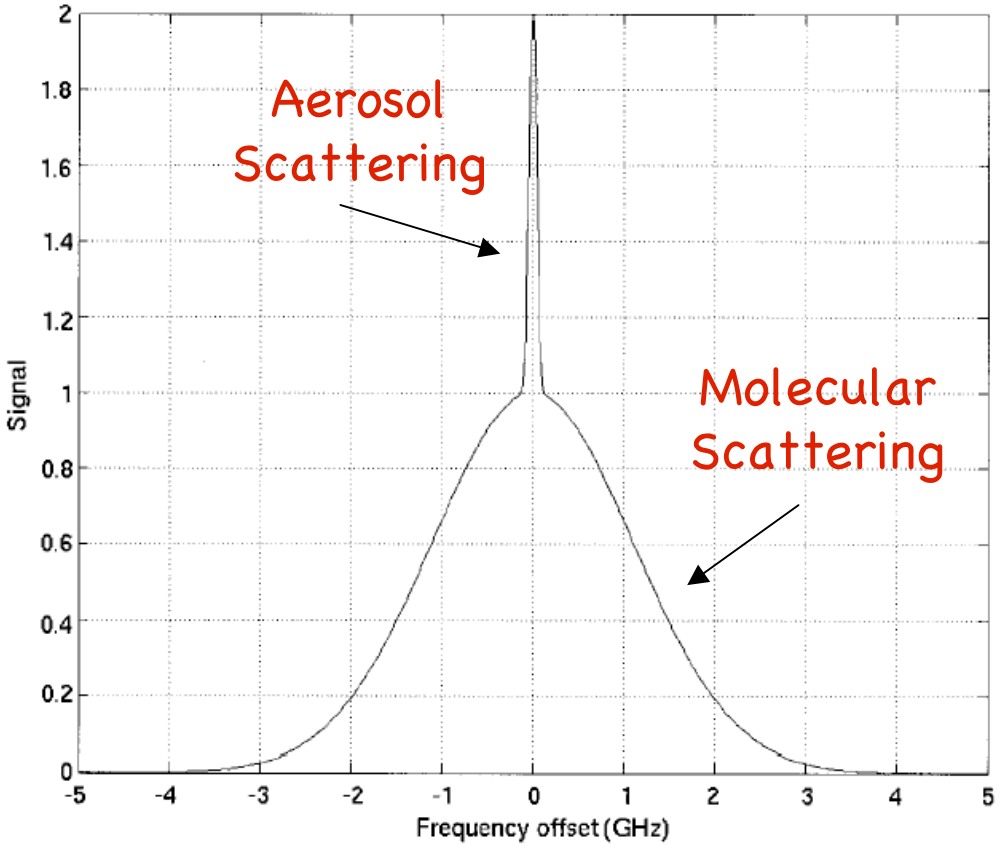


Fig. 5.1. Spectral profile of backscattering from a mixture of molecules and aerosols for a temperature of 300 K. The spectral width of the narrow aerosol return is normally determined by the line width of the transmitting laser.

Derive Extinction from Raman/HSRL

- For the pure Rayleigh channel in HSRL or the Raman channel

$$P(R,\lambda_{Ra}) = \frac{E_o \eta_{\lambda_{Ra}}}{R^2} O(R,\lambda_{Ra}) \beta_{Ra}(R,\lambda_0) \exp\left\{-\int_0^R \left[\alpha(r,\lambda_0) + \alpha(r,\lambda_{Ra})\right] dr\right\}$$

For HSRL, $\lambda_0 = \lambda_{Ra}$;
For Raman, $\lambda_0 \neq \lambda_{Ra}$

On the way up

On the way down

- Rayleigh and Raman scattering from air molecules can be calculated

$$\beta_{Ra}(R,\lambda_0) = N_{Ra}(R) \frac{d\sigma_{Ra}}{d\Omega}(\pi,\lambda_0)$$

- From above two equations, we have

$$\alpha(R,\lambda_0) + \alpha(R,\lambda_{Ra}) = \frac{d}{dR} \ln \frac{N_{Ra}(R)}{S(R,\lambda_{Ra})} + \frac{d}{dR} \ln O(R,\lambda_{Ra})$$

- In the optimum case O(R) ≡ 1, i.e., good overlap

$$\alpha_{aer}(R,\lambda_0) + \alpha_{aer}(R,\lambda_{Ra}) = \frac{d}{dR} \ln \frac{N_{Ra}(R)}{S(R,\lambda_{Ra})} - \alpha_{mol}(R,\lambda_0) - \alpha_{mol}(R,\lambda_{Ra})$$

Derive Extinction Cont'd

- To obtain the extinction coefficient at the transmitted wavelength λ_0, we have to introduce the Angstrom exponent, which describes the wavelength dependence of the particle extinction coefficient.

$$\frac{\alpha_{aer}(\lambda_0)}{\alpha_{aer}(\lambda_{Ra})} = \left(\frac{\lambda_{Ra}}{\lambda_0}\right)^{a(R)}$$

- Thus, the final solution of aerosol extinction is given by

$$\alpha_{aer}(R,\lambda_0) = \frac{\frac{d}{dR}\ln\frac{N_{Ra}(R)}{S(R,\lambda_{Ra})} - \alpha_{mol}(R,\lambda_0) - \alpha_{mol}(R,\lambda_{Ra})}{1+\left(\frac{\lambda_0}{\lambda_{Ra}}\right)^{a(R)}}$$

- For HSRL, the denominator of the above equation is 2, because $\lambda_0 = \lambda_{Ra}$

Derive Backscatter from Raman/HSRL

- Aerosol backscatter coefficient can be calculated from the ratio of the total backscatter signal (aerosol + molecule) to the molecular backscatter signal as below.

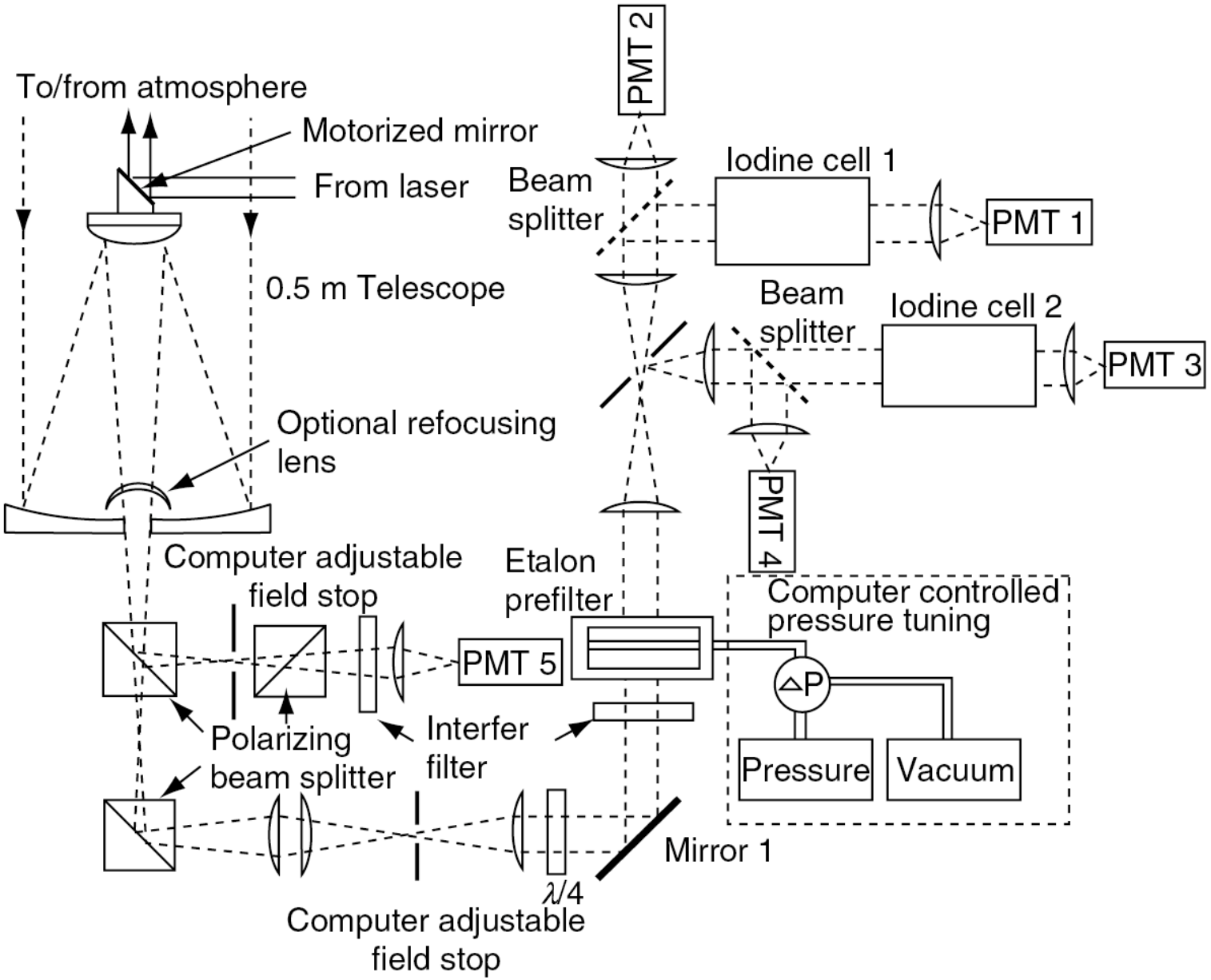
$$\beta_{aer}(R,\lambda_0)+\beta_{mol}(R,\lambda_0)=\left[\beta_{aer}(R_0,\lambda_0)+\beta_{mol}(R_0,\lambda_0)\right]\frac{P(R_0,\lambda_{Ra})P(R,\lambda_0)}{P(R_0,\lambda_0)P(R,\lambda_{Ra})}\frac{N_{Ra}(R)}{N_{Ra}(R_0)}$$
$$\times\frac{\exp\left\{-\int_{R_0}^{R}\left[\alpha_{aer}(r,\lambda_{Ra})+\alpha_{mol}(r,\lambda_{Ra})\right]dr\right\}}{\exp\left\{-\int_{R_0}^{R}\left[\alpha_{aer}(r,\lambda_0)+\alpha_{mol}(r,\lambda_0)\right]dr\right\}}$$

Here, R_0 is a reference point, which is usually chosen in upper troposphere or lower stratosphere so that it is free of aerosols and the molecular backscatter coefficient can be reliably estimated from atmos conditions.

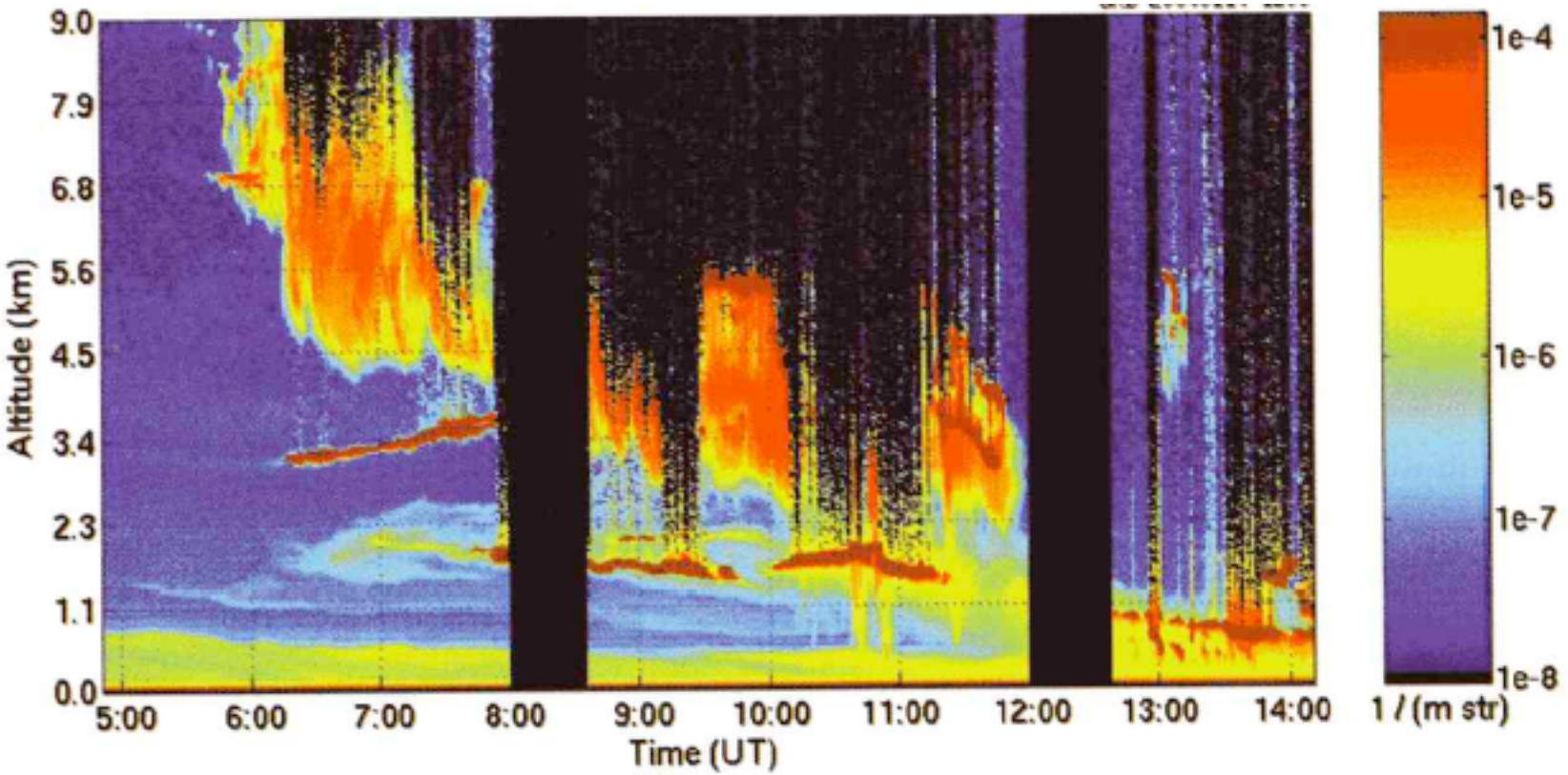
- The aerosol lidar ratio is then given by

$$L_{aer}(R,\lambda_0)=\frac{\alpha_{aer}(R,\lambda_0)}{\beta_{aer}(R,\lambda_0)}$$

Wisconsin HSRL



HSRL Results

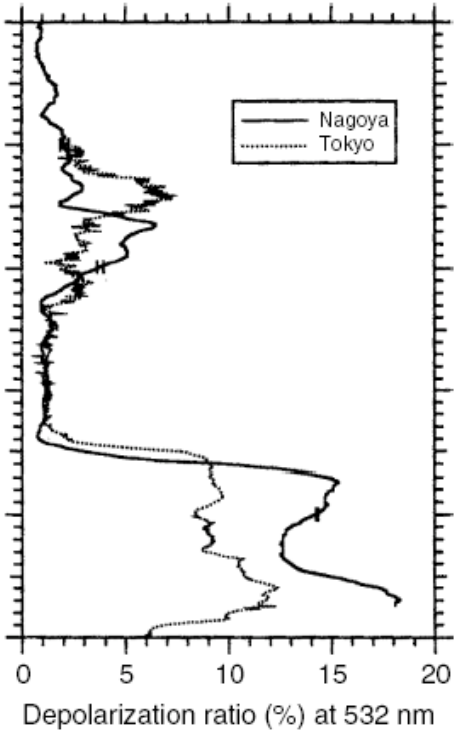


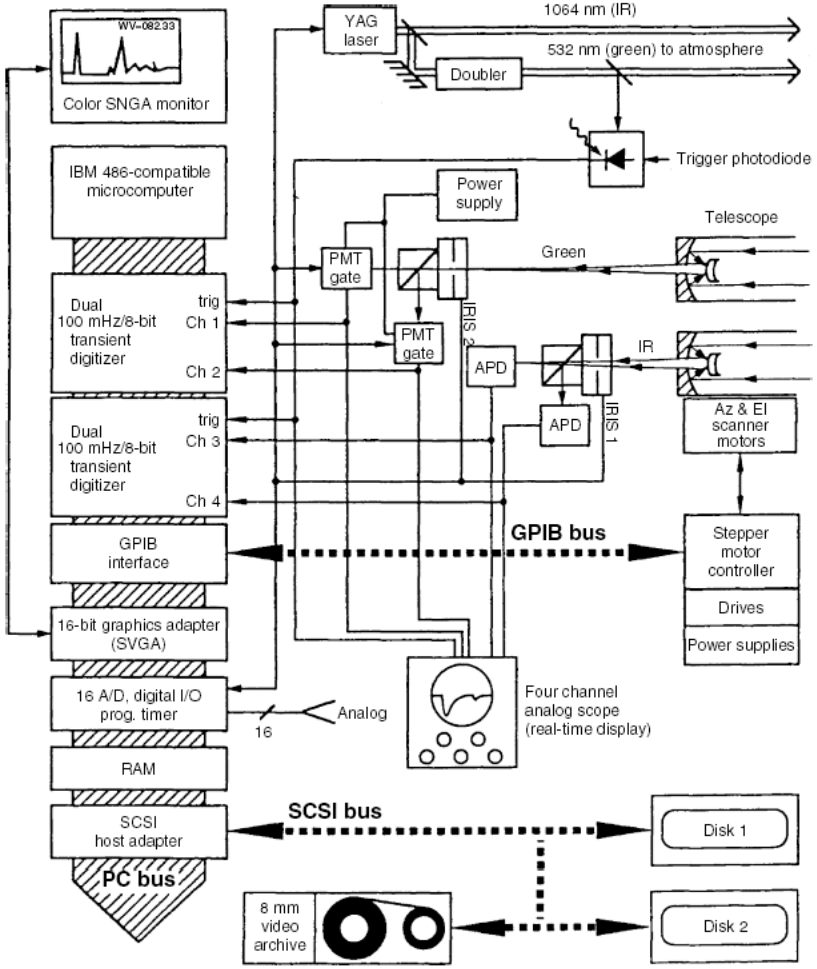
Polarization Lidar Detection

- Backscattering from a spherical particle does not change the polarization, but the non-spherical particle changes the polarization. Thus, by monitoring the polarization status of the scattered light, information on the shape of the aerosol particles can be obtained.

- Depolarization is defined by

$$\delta = I_{\perp} / I_{\parallel}$$



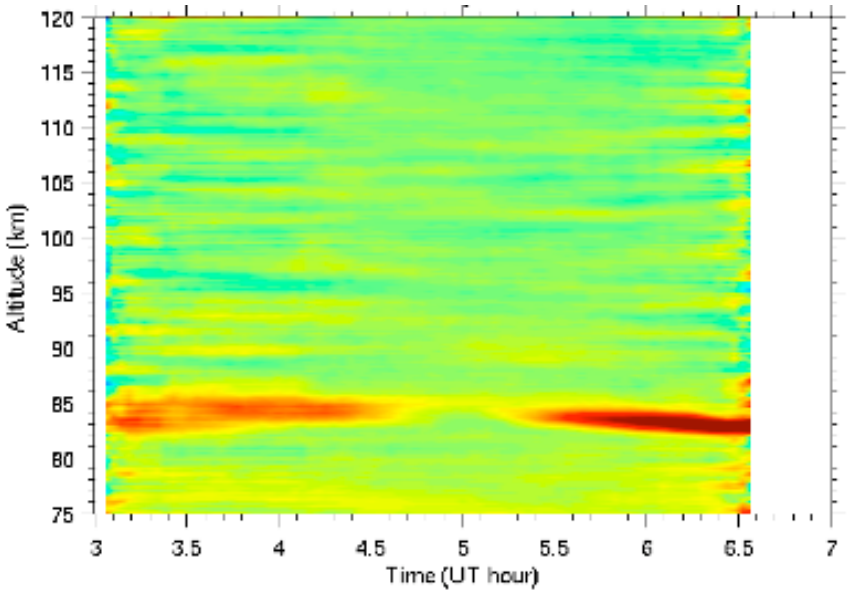


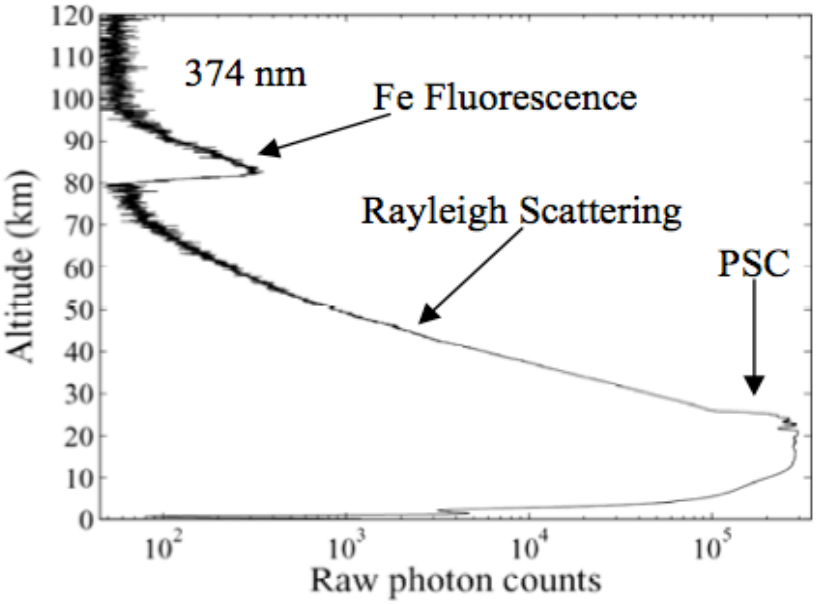
Multi-Wavelength Lidar Detection

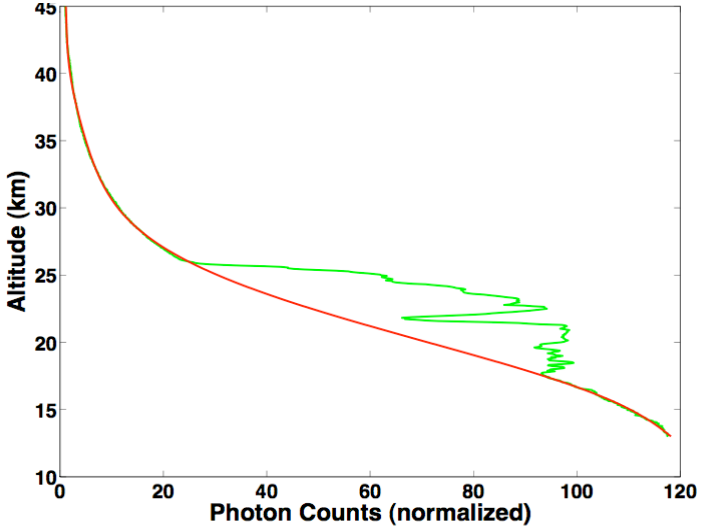
- By taking the color ratio of aerosol scattering, plus some assumption of particle size distribution, e.g., lognormal distribution, the multi-wavelength lidar measurements of aerosol can be used to determine the particle size (e.g., radius, width) and particle number density.
- We will give more explanation in the next lecture.

Aerosols in Middle and Upper Atmos

Polar Mesospheric Clouds

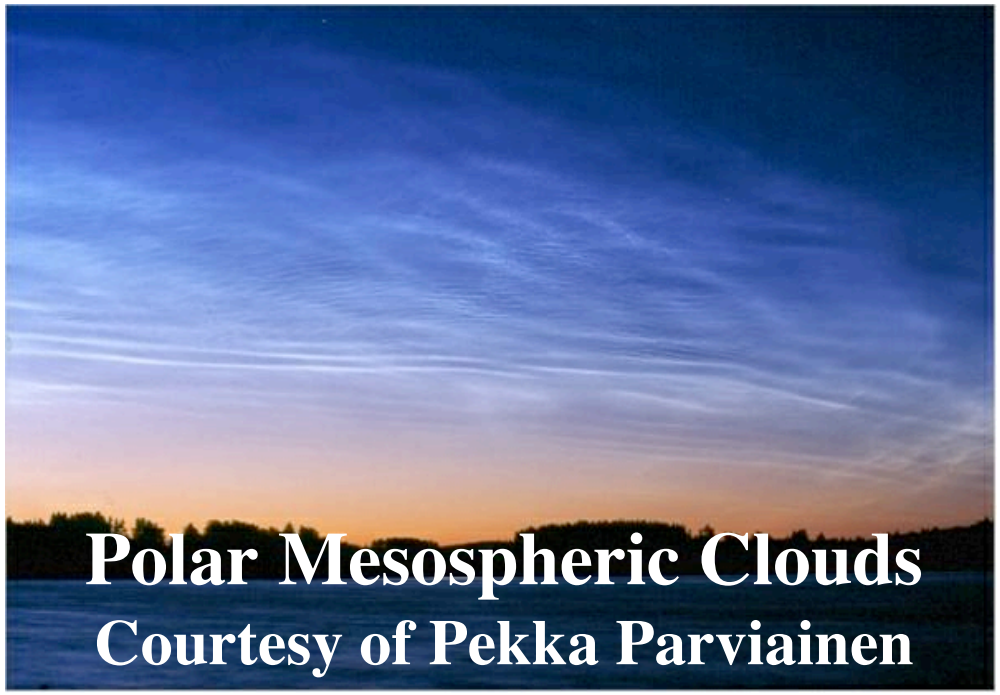






Polar Stratospheric Clouds
- Key role in ozone depletion

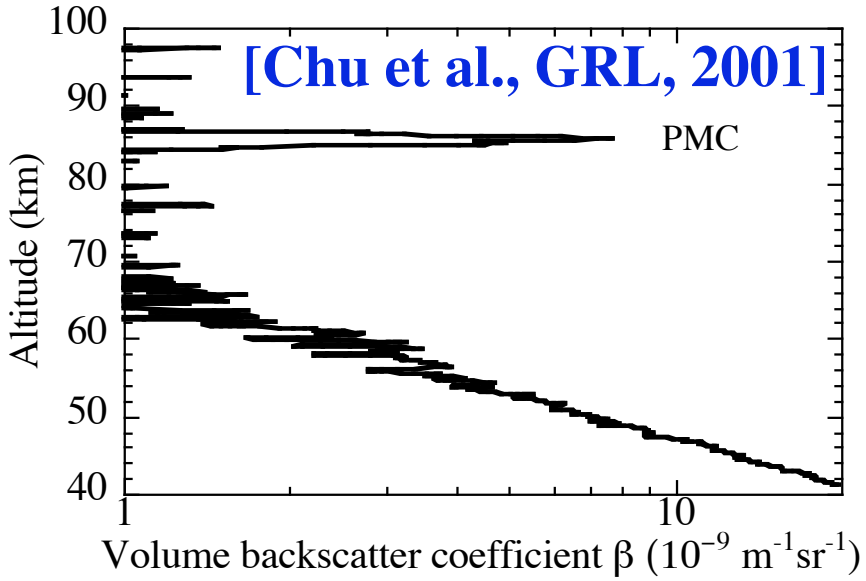
PMC Detection in Polar Region



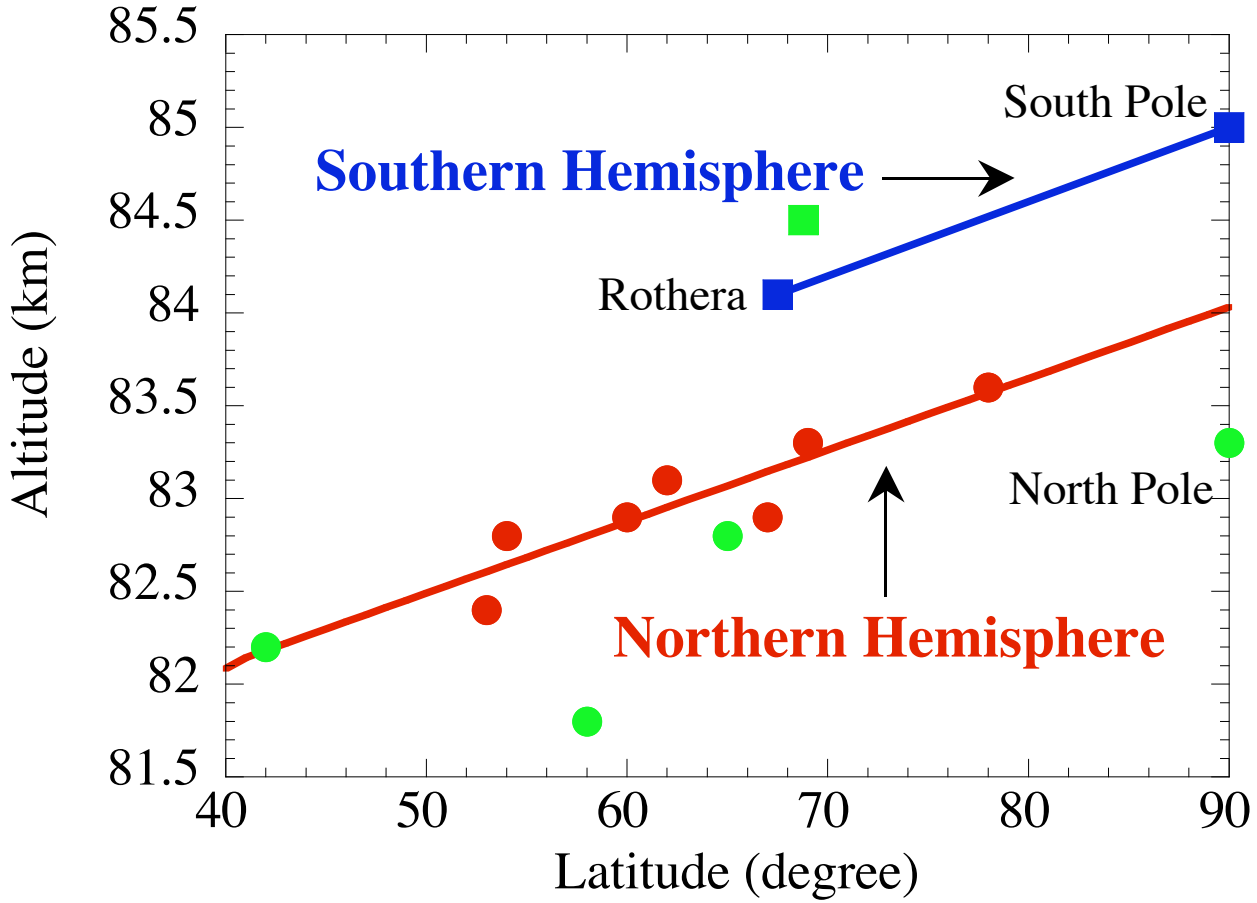
PMC are the thin scattering layers (consisting of water ice particles) ~ 85 km, >50° latitudes, in summer

PMC are regarded as a possible indicator of global climate change Natural laboratory for testing atmosphere models and dynamics

Fe Boltzmann lidar is highly capable of PMC detection
1) Short wavelength (374, 372 nm)
2) High laser power
3) Daytime capability



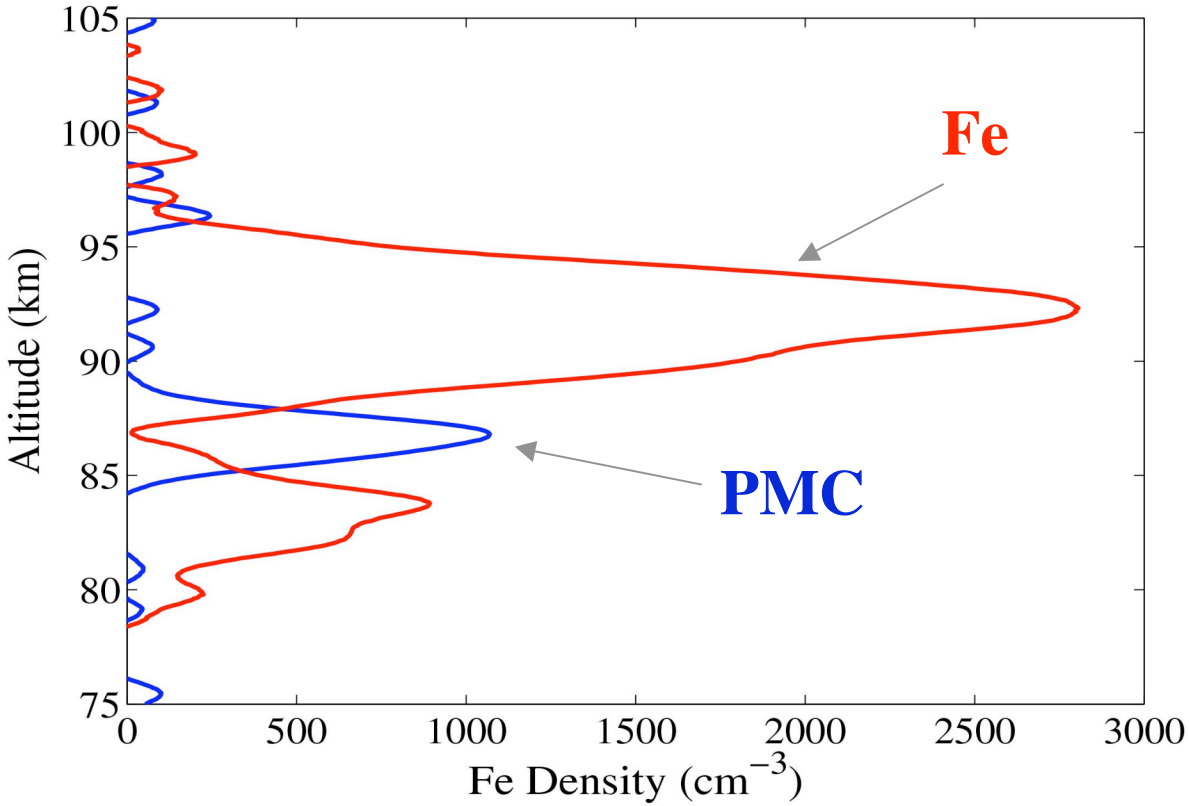
Hemispheric Difference in PMC Altitude



Southern PMC are ~ 1 km higher than Northern PMC

[Chu et al., JGR, 2003; GRL, 2004]

Heterogeneous Removal of Fe by PMC



Plane, Murray, Chu, and Gardner, *Science*, *304*, 426-428, 2004

Fe ablation flux = 1.1 x 10^4 atoms cm^{-2} s^{-1}

Uptake coefficients of Fe and Fe species on ice = 1

Summary

- Aerosol is an important topic in atmospheric science and environmental research. It can be measured/monitored by hot lidar technologies.
- Conventional single-channel elastic-scattering lidar can beautifully address PMC and PSC backscatter problems, and monitor the occurrence, height, vertical structure, etc. However, it is unreliable to derive aerosol extinction.
- Raman lidar and HSRL provide addition information by adding Raman channel or separate molecular scattering from aerosol scattering. Both can measure aerosol backscatter and extinction nicely. However, HSRL is more desirable as Rayleigh scattering is much stronger than Raman scattering.
- Polarization detection and multi-wavelength detection can help identify aerosol shape, size, distribution, and number density.
- Aerosol study is growing, and awaiting for more smart lidar ideas.