Absorbing aerosol radiative effects in the limb-scatter viewing geometry

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Outline

- Absorbing aerosols in the upper troposphere
- Absorbing aerosol detection in the upper troposphere
- OSIRIS instrument
- SASKTRAN forward model
- Model-calculated OSIRIS sensitivity to upper tropospheric aerosols
    - Absorbing / Non-absorbing
    - Single / Multiple / Total Scattering
- Conclusions

Absorbing aerosols in the upper troposphere

- **Solid** ice nuclei (IN) concentrations in the upper troposphere (UT, > 7 km) influence cirrus cloud properties and produce a climate forcing via the indirect effect

- **Dust** is a very efficient IN, however, it has been shown that it is most likely to reach the UT only if it survives extensive cloud processing [Wiacek and Peter, 2009; Wiacek et al., 2010], which is difficult to model accurately

- **Soot** is thought to be a much less efficient IN (ongoing research into this), however, as it is deposited directly in the UT by commercial aircraft, it may have a role to play

- "Cirrus clouds form via heterogeneous freezing on mineral dust and metallic aerosol, not homogeneously or on elemental carbon or biological particles." – Cziczo et al., 2013, Science

Absorbing aerosol detection in the UT

- In the upper troposphere what we know about the **chemical composition** and spatial distribution of absorbing aerosols comes from infrequent **aircraft campaigns** employing filter as well as single-particle collection and analysis methods

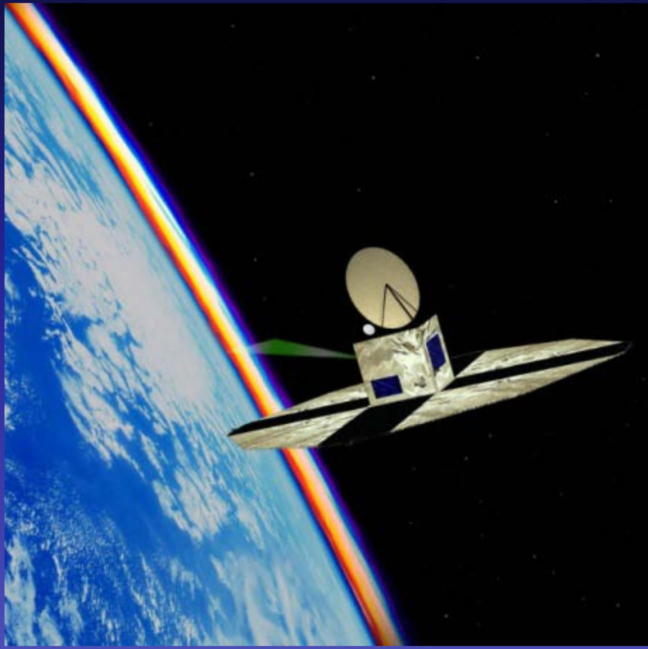
- Currently **nadir-viewing** passive satellite sensors can detect **major** dust and soot plumes in the near-UV (320-400 nm), however, these are confined to the **lower troposphere** (0-7 km)

- While the **limb-scatter viewing** geometry provides good **sensitivity** (~50-100 km observation path length), high vertical **resolution** (~1-2 km), and good spatial **coverage...**

Absorbing aerosol detection in the UT

- …UTLS extinction retrievals **may** be complicated by the need of *a priori* information on aerosol **single scattering albedo**
- Little attention has been devoted to the retrieval of aerosol extinction in the upper troposphere
- None have reported attempts to discriminate aerosol composition

OSIRIS: Optical Spectrograph and Infra-Red Imager System



Optical Spectrograph

Grating spectrometer

1-2 km vertical resolution

280 – 810 nm spectral coverage

10 – 100 km tangent altitudes

Infrared Imager

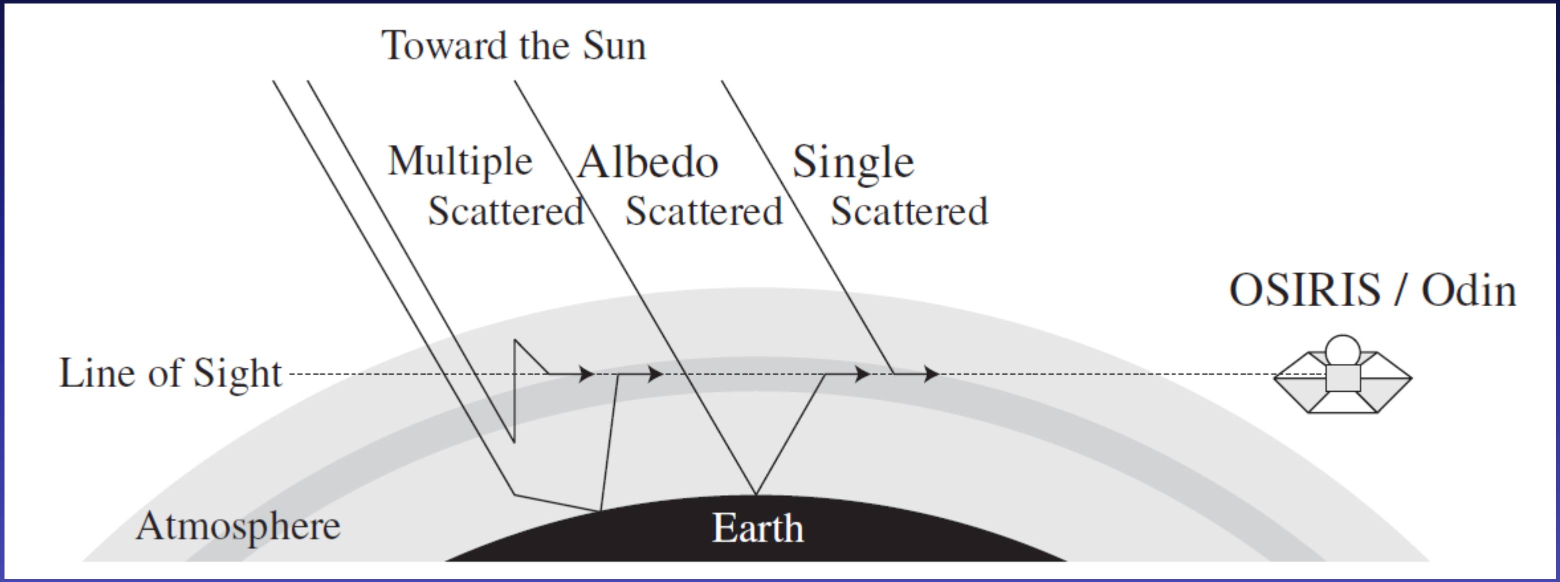
Linear array detector spans ~ 100 km

1.26 μm, 1.27 μm, 1.53 μm

2001 – 2007: 50% aeronomy, 50% astronomy

2007 – Present: 100% aeronomy

Limb-scatter viewing geometry



[Bourassa et al., 2008]

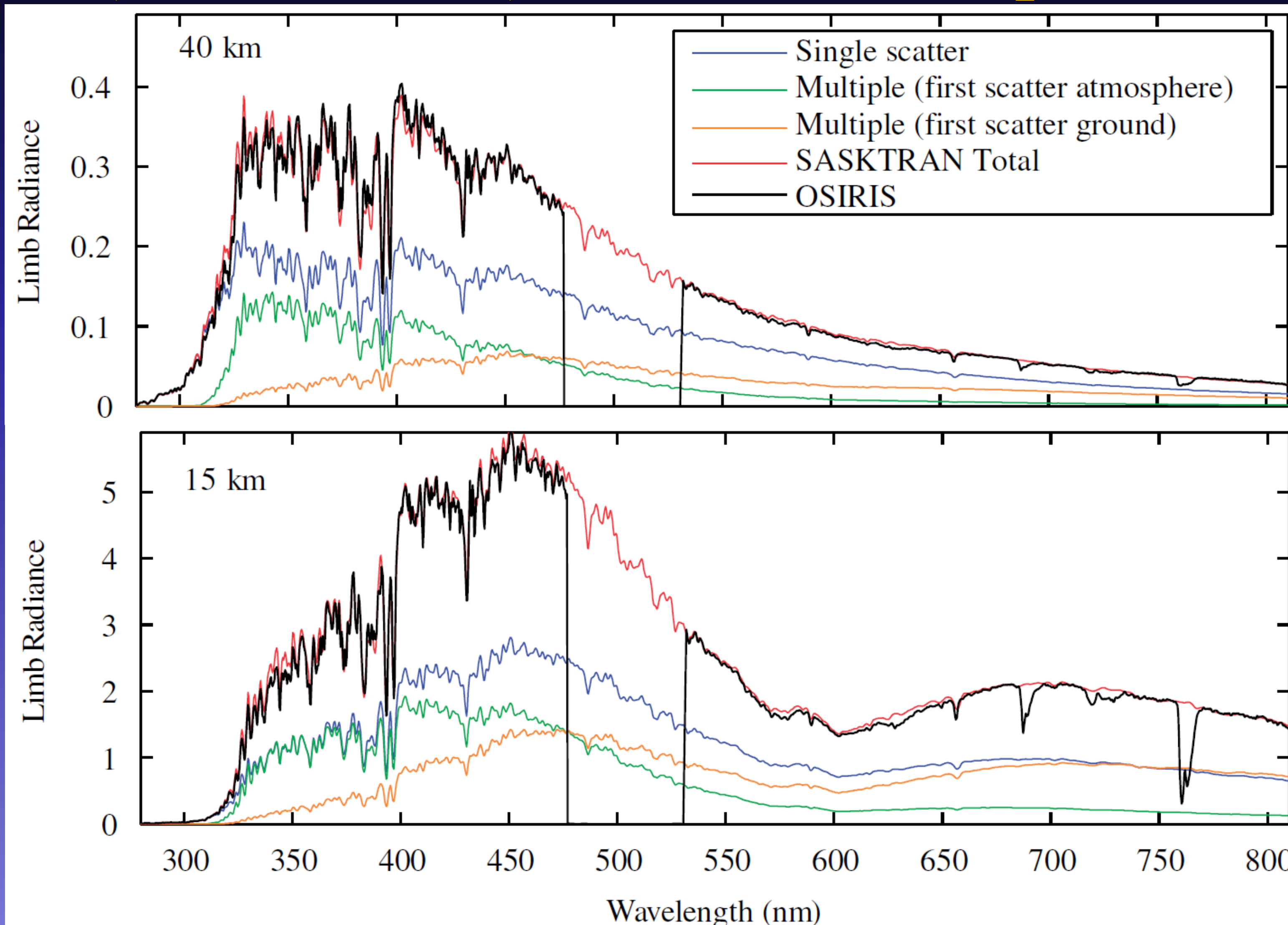
- Contribution from light that has scattered multiple times in the atmosphere and/or has been reflected from the ground is significant

SASKTRAN Radiative Transfer Code

- C++ code optimized to run efficiently on PC / Linux
- Scalar radiative transfer equation with multiple scattering evaluated in a fully 3-D, spherical shell model of the atmosphere
  - "Successive orders of scattering" approach used to trace 1st and 2nd order scattering events exactly
  - Higher order scattering term contributions estimated by integrating previous order scattering source term at local zenith
  - Lambertian reflection at the Earth's surface
- Scattering by air molecules and aerosols
  - Altitude-dependent cross-sections and phase functions
  - Can be calculated (Mie) or specified directly (tabular)
- Absorption by trace gases and aerosols
  - Temperature-dependent cross-sections for trace gases
  - Complex index of refraction for aerosols

Measured (OSIRIS) and modelled (SASKTRAN) limb radiance spectra

(10^{13} photons/s/cm^2/nm/sterad)



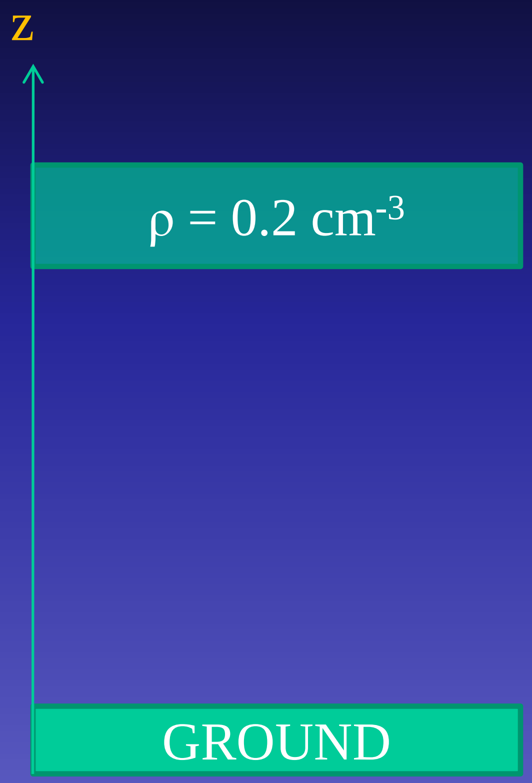
[Bourassa et al., 2008]

SASKTRAN Simulation Setup

- Model 4 wavelengths = [337 nm, 377 nm, 452 nm, 813 nm]
- Model 2 albedos = [0, 1]
- Model 2 solar zenith angles = [~65°, ~87°]
- Model one order of scattering (single-scattering)
- Model 50 orders of scattering (total scattering)
- Calculate multiple scattering (50 orders – 1 order)

SASKTRAN Simulation Setup

- Add species one-by-one
  - Air only (0-100 km)
  - Aerosol only (5-km between 10-15 km)
  - Air + aerosol
- Isolate dust (and other species') absorption effects via **index of refraction**
- Always same set of OSIRIS tangent heights



Simulated Aerosol Properties

- ρ = number concentration
- r,σ = lognormal mode radius and width
- n_r, n_i = real and complex index of refraction

Aerosol	ρ	r, σ	n_r			
	[cm^{-3}]	[μm]	337 nm	377 nm	452 nm	813 nm
Absorbing dust (D1)	0.2	1.0, 1.6	1.55	1.55	1.55	1.55
Non-absorbing dust (D2)	0.2	1.0, 1.6	1.55	1.55	1.55	1.55
Pure soot (PS)	20	0.0125, 1.6	1.95	1.95	1.95	1.95
Smoke (SM)	20	0.075, 1.6	1.51	1.51	1.51	1.53
Sulfate aerosol (SO4)	30	0.08, 1.6	1.47	1.45	1.43	1.43
Ice (ICE)	0.2	1.0, 1.6	1.33	1.32	1.32	1.30

	n_i			
	337 nm	377 nm	452 nm	813 nm
D1	1.953×10^{-2}	1.380×10^{-2}	6.928×10^{-3}	6.441×10^{-4}
D2	0	0	0	0
PS	0.79	0.79	0.79	0.79
SM	2.677×10^{-2}	2.677×10^{-2}	2.677×10^{-2}	1.653×10^{-2}
SO4	1.0×10^{-10}	2.1×10^{-10}	2.5×10^{-9}	8.8×10^{-8}
ICE	5.5×10^{-9}	2.7×10^{-9}	1.7×10^{-9}	1.5×10^{-7}

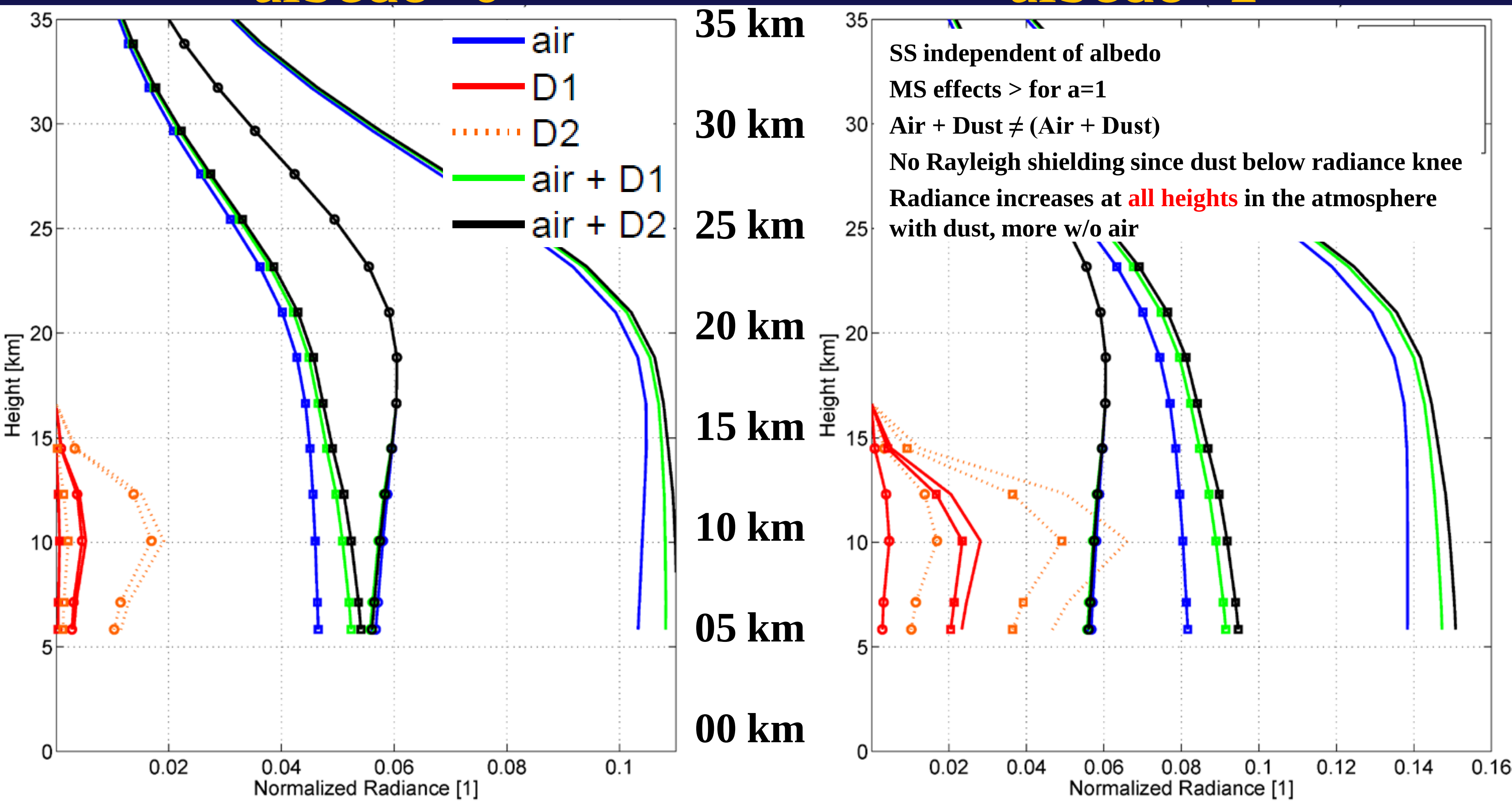
Single
Multiple
Total

~340 nm

SZA~66°, SAA~70°

D1 = absorbing
D2 = non-abs.

albedo=0 | albedo=1



Normalized Radiance [0→1] | Normalized Radiance [0→1]

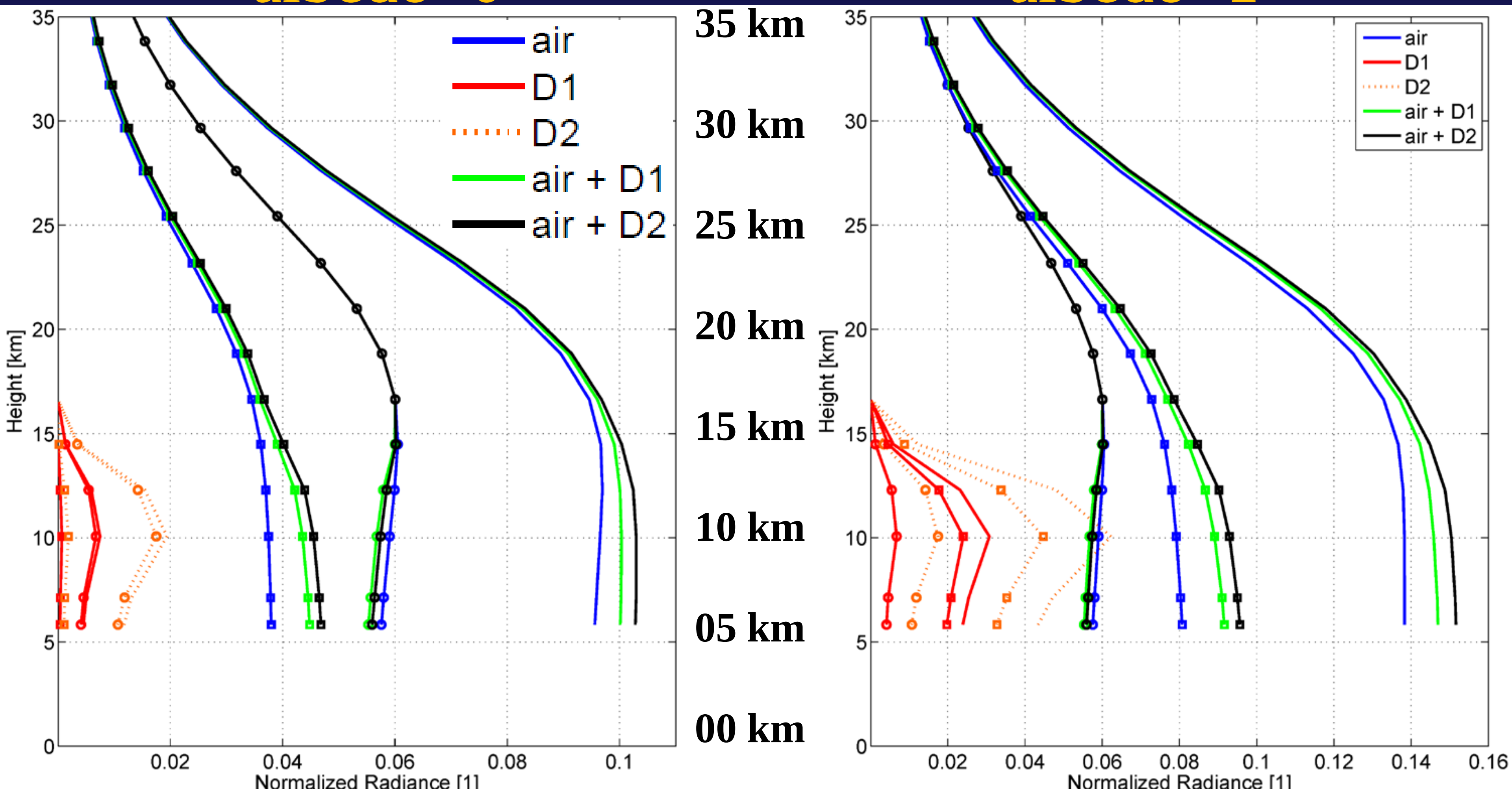
Single
Multiple
Total

~380 nm

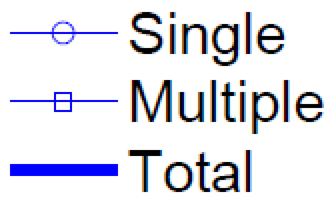
SZA~66°, SAA~70°

D1 = absorbing
D2 = non-abs.

albedo=0 albedo=1



Normalized Radiance [0→1] **Normalized Radiance [0→1]**

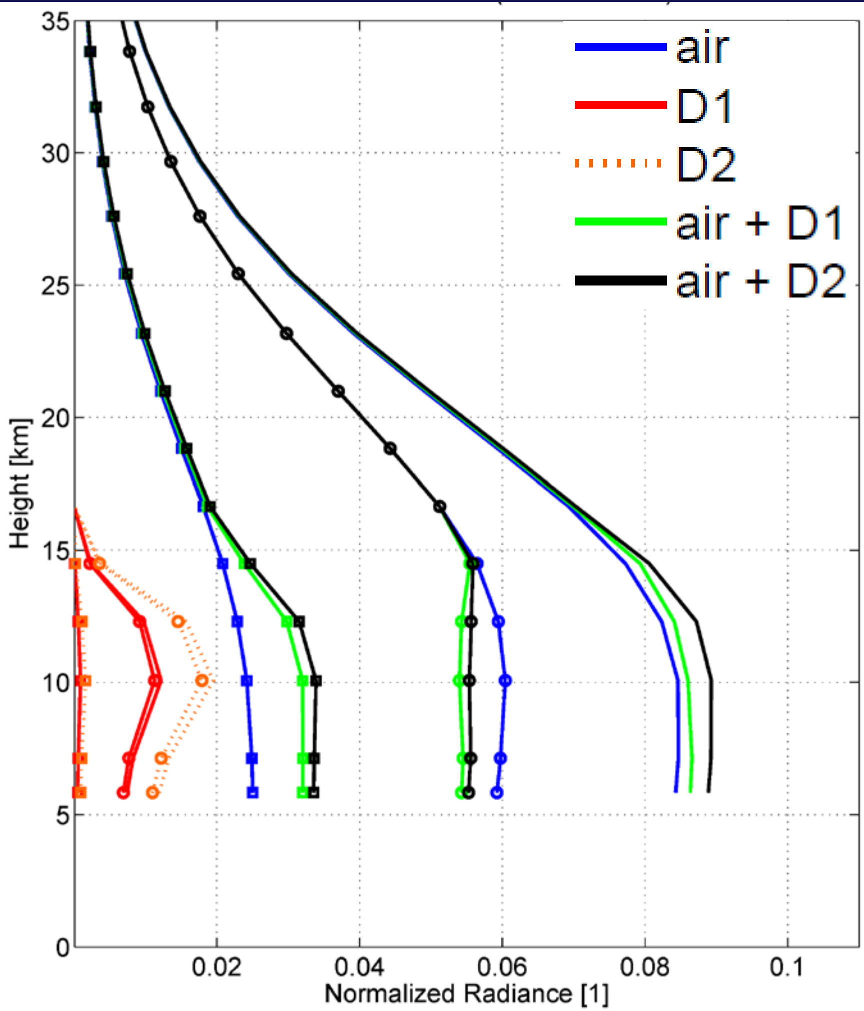


~450 nm

SZA~66°, SAA~70°

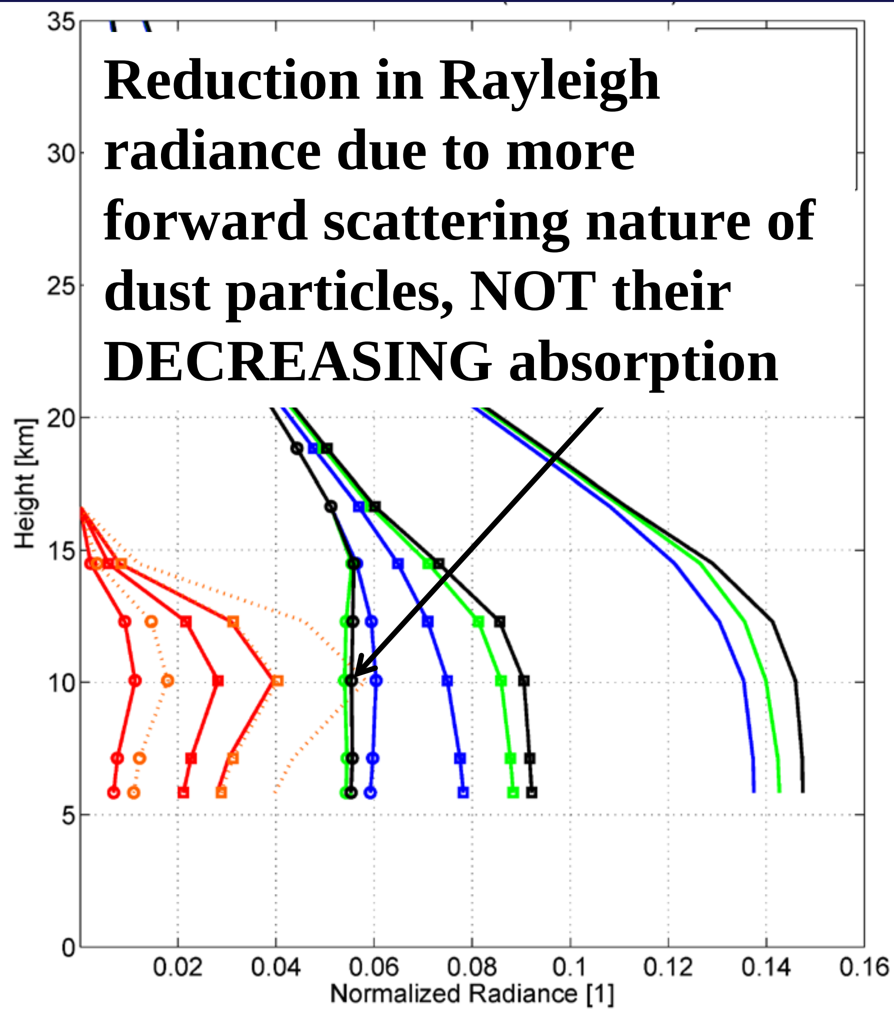
D1 = absorbing
D2 = non-abs.

albedo=0



35 km
30 km
25 km
20 km
15 km
10 km
05 km
00 km

albedo=1



Reduction in Rayleigh radiance due to more forward scattering nature of dust particles, NOT their DECREASING absorption

Normalized Radiance [0→1]

Normalized Radiance [0→1]

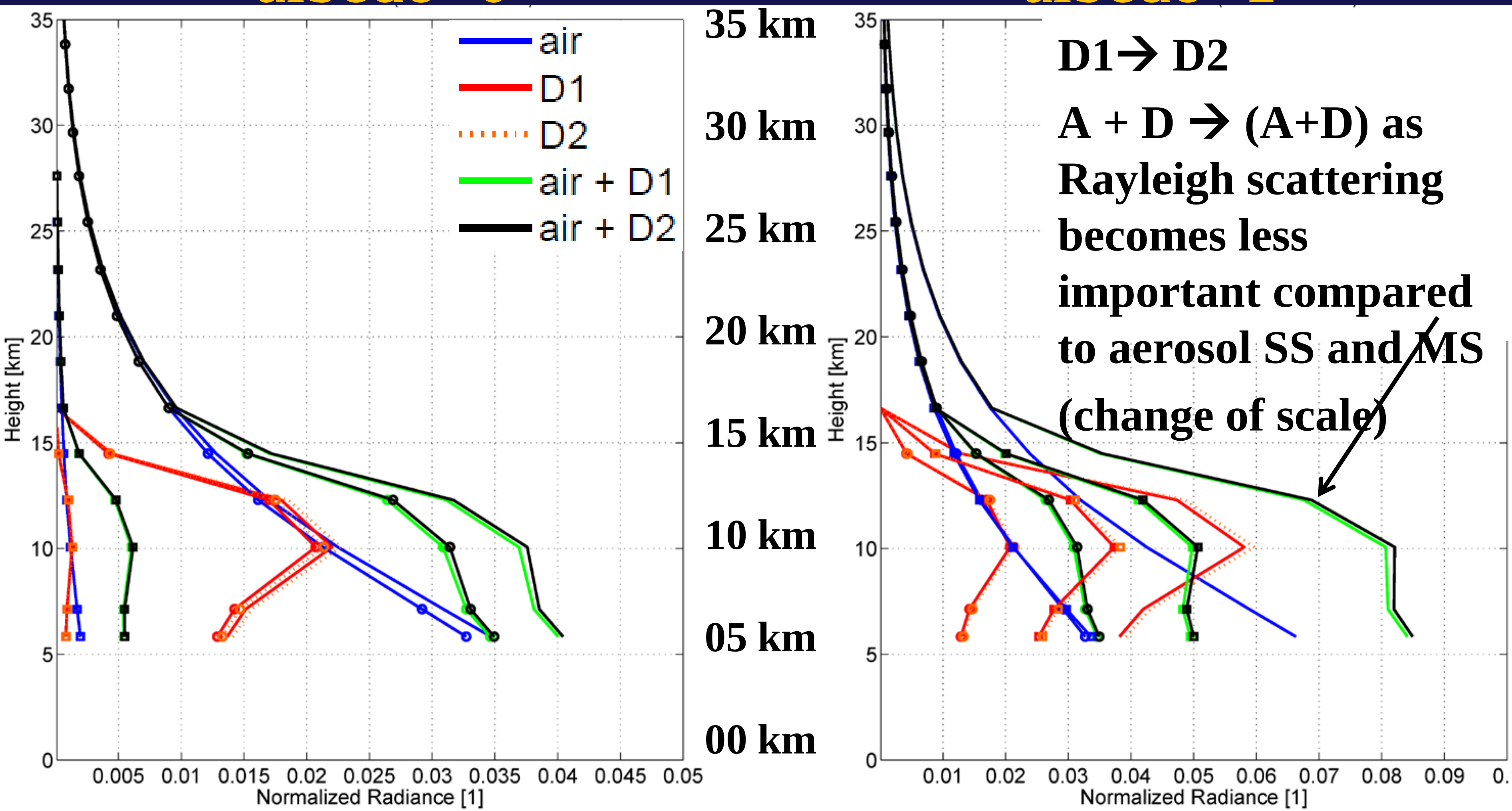
~800 nm

SZA~66°, SAA~70°

- Single
- Multiple
- Total

D1 = absorbing
D2 = non-abs.

albedo=0 | albedo=1



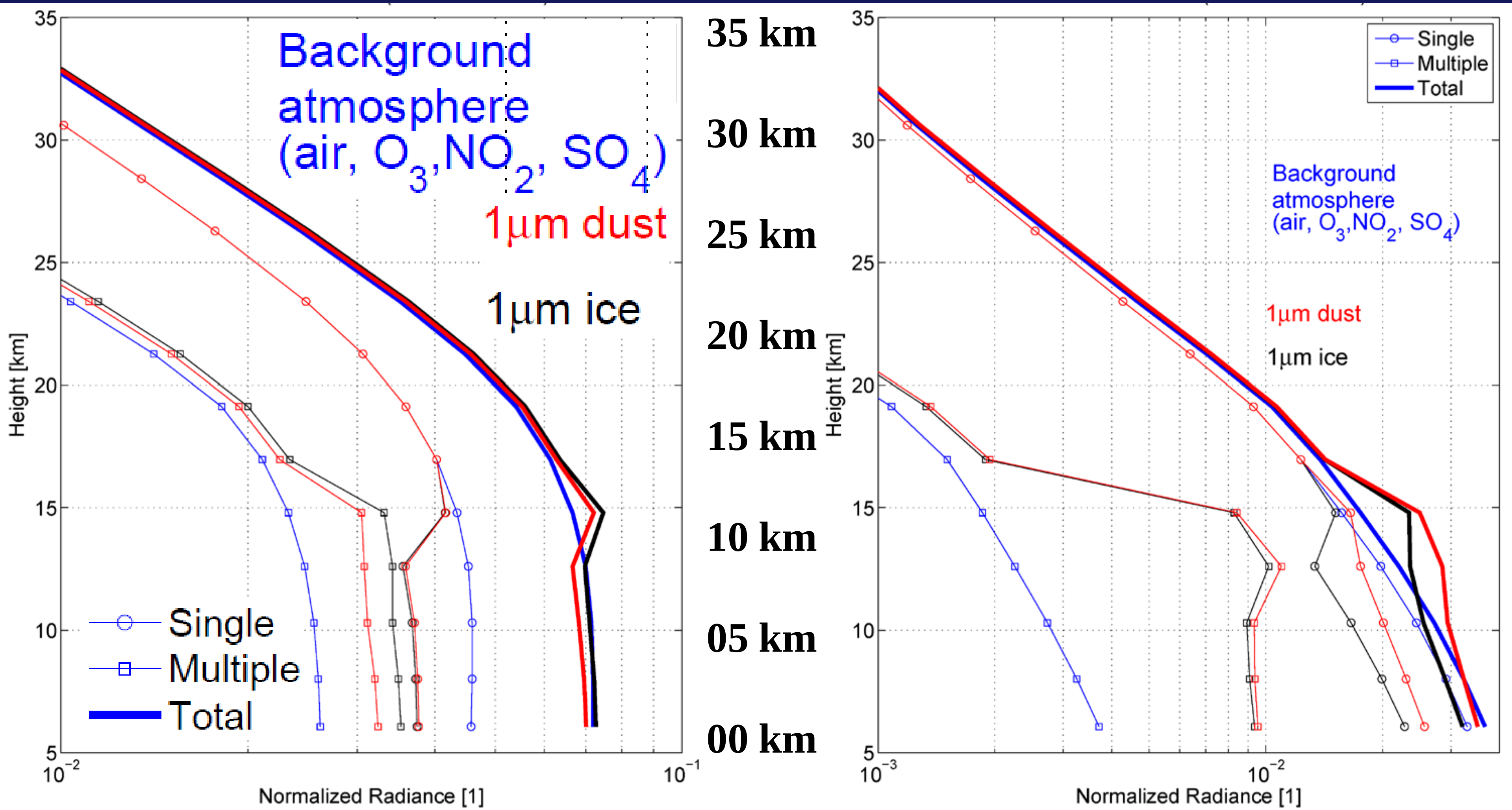
Normalized Radiance [0→1]

Normalized Radiance [0→1]

Absorbing Dust vs. Ice (14-15 km, ρ=20cm^{-3})

SZA~66°, SSA~105°, albedo=0

450 nm   800 nm



Normalized Radiance [0→1]   **Normalized Radiance [0→1]**

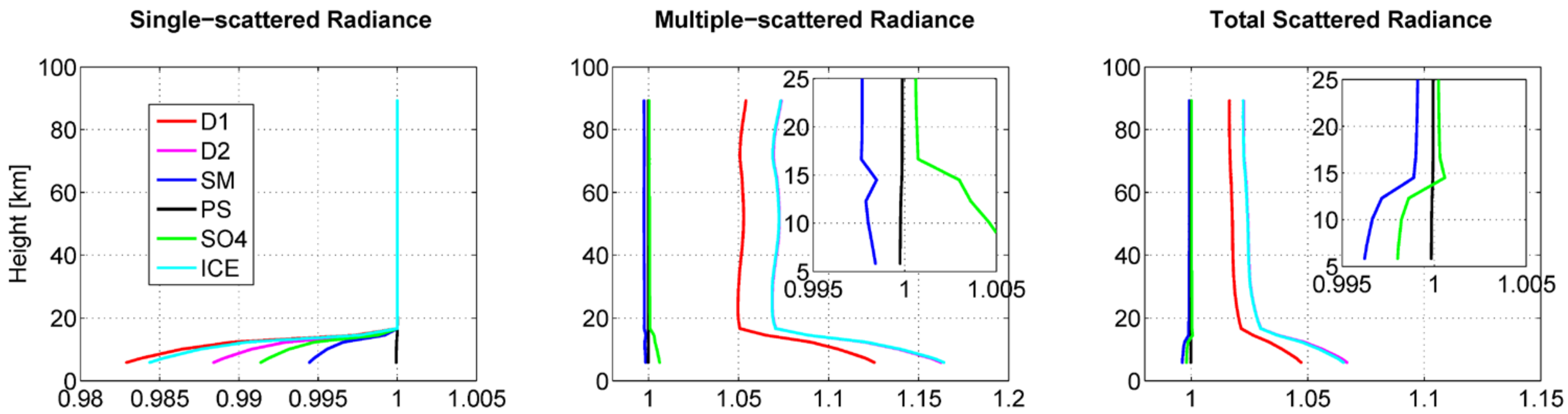
Summary (1)

- Optically thick dust well below the radiance knee has a negligible effect on the SS Rayleigh radiance field
- Optically thick dust near the radiance knee shields the SS Rayleigh radiance field at and below the dust
- Optically thick dust well above the radiance knee compares with or dominates the SS Rayleigh radiance field
- As we move from SS Rayleigh shielding to SS Rayleigh increases, we move from non-linear to linearly additive radiance effects of the air + dust combination
- The effect is very similar for negligibly absorbing ice
- Dust absorption plays a very small role in limb-scatter obs.

Other aerosols at ~340 nm

albedo=0, layer between 10-15 km

SZA~66°, SAA~70°



Radiance Ratio =

$$\frac{\text{Radiance}_{(\text{air + aerosol})}}{\text{Radiance}_{(\text{air})}}$$

Aerosol	ρ [cm^{-3}]	r, σ [μm]
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Non-absorbing dust (D2)	0.2	1.0, 1.6
Pure soot (PS)	20	0.0125, 1.6
Smoke (SM)	20	0.075, 1.6
Sulfate aerosol (SO4)	30	0.08, 1.6
Ice (ICE)	0.2	1.0, 1.6

Percent radiance reduction at 35 km altitude

aerosol layer still between 10-15 km

SZA~66°, SAA~70°

	$a = 0$					
	Pure soot		Smoke		Hybrid	
ρ [cm^{-3}]	337 nm	813 nm	337 nm	813 nm	337 nm	813 nm
0.2	−0.0001	0.0000	−0.0009	0.0012	−0.0054	0.0014
20	−0.0050	−0.0003	−0.0845	0.1160	−0.5291	0.1366
200	−0.0500	−0.0027	−0.7252	1.1466	−4.4902	1.1967

	$a = 1$					
	Pure soot		Smoke		Hybrid	
ρ [cm^{-3}]	337 nm	813 nm	337 nm	813 nm	337 nm	813 nm
0.2	−0.0001	−0.0001	−0.0017	−0.0004	−0.0096	−0.0087
20	−0.0089	−0.0035	−0.1704	−0.0402	−0.9430	−0.8604
200	−0.0884	−0.0350	−1.6438	−0.3778	−8.1904	−7.7199

Summary (2)

- Any aerosol species we modeled decreases SS radiance towards OSIRIS within the aerosol layer
  - Absorption effects are secondary to scattering phase function, size and number concentration effects
- MS radiance either increases (dust, ice, sulphate) or decreases (soot, smoke) at all heights **except**
- For medium-sized and absorbing particles (smoke) it can do both
- Total radiance decreases at 35 km are a unique signature of absorbing aerosols, but it is a small signature (<0.2%)
- Total radiance signatures within, below and above an aerosol layer mixed with air are complex to interpret
- **Limb-scatter retrievals of aerosol extinction are insensitive to external information on aerosol absorption**

Details in [Wiacek et al., 2013, AMTD]