Indices of Refraction of Absorptive Aerosol Their Importance and Complexity

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Outline

Absorptive aerosol

- **Why it is important**
- **Discuss the complexity of absorptive aerosol**

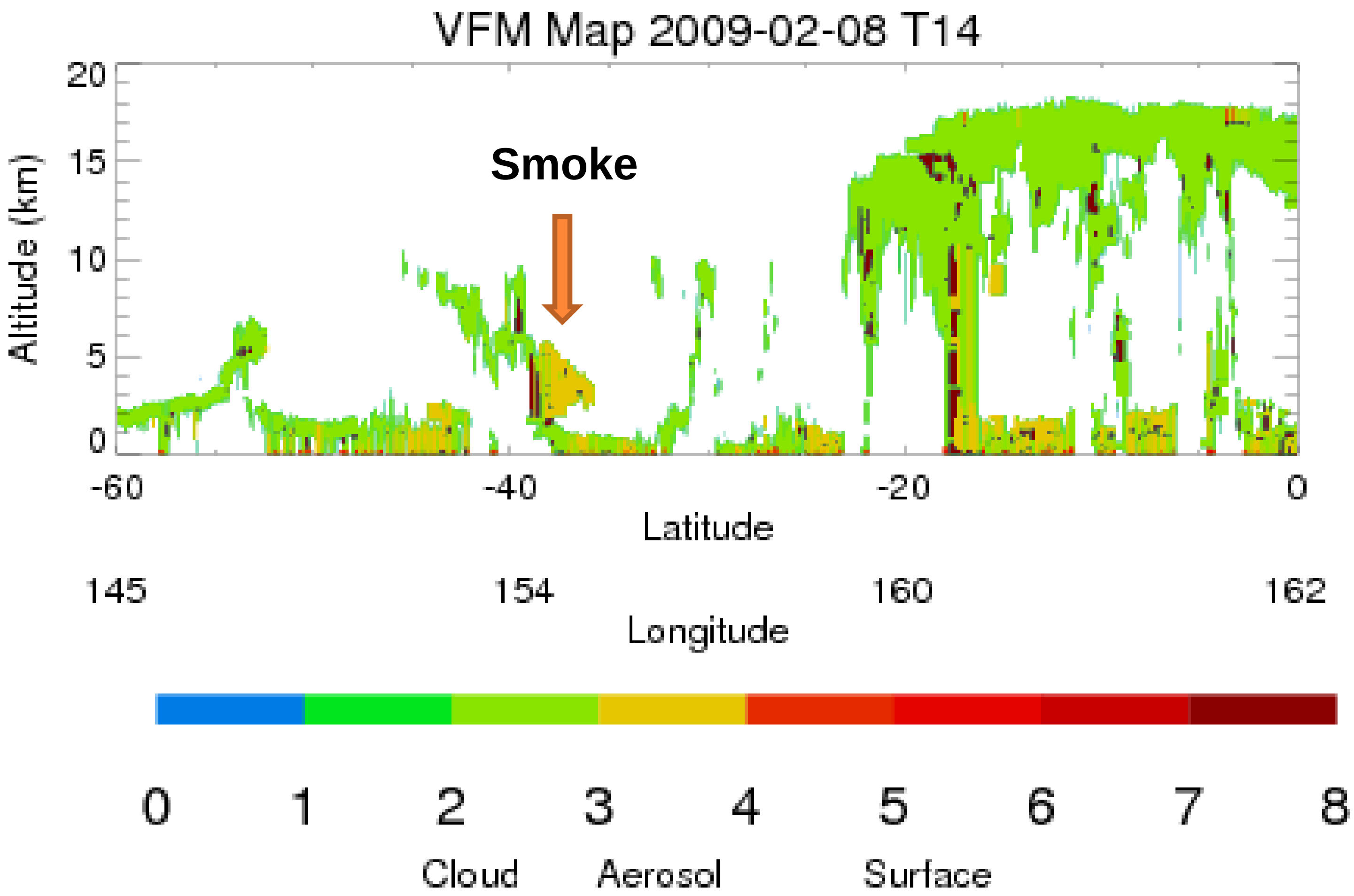
Discuss

- **Sokolik and Toon mineralogical indices**
- **Chang and Charalampopouls flame soot indices**
- **Lund Myhre and Nielsen organic acid indices**

Conclusions and Recommendations

Black Saturday Bushfires - February 7, 2009
MODIS Visible Image of Australia

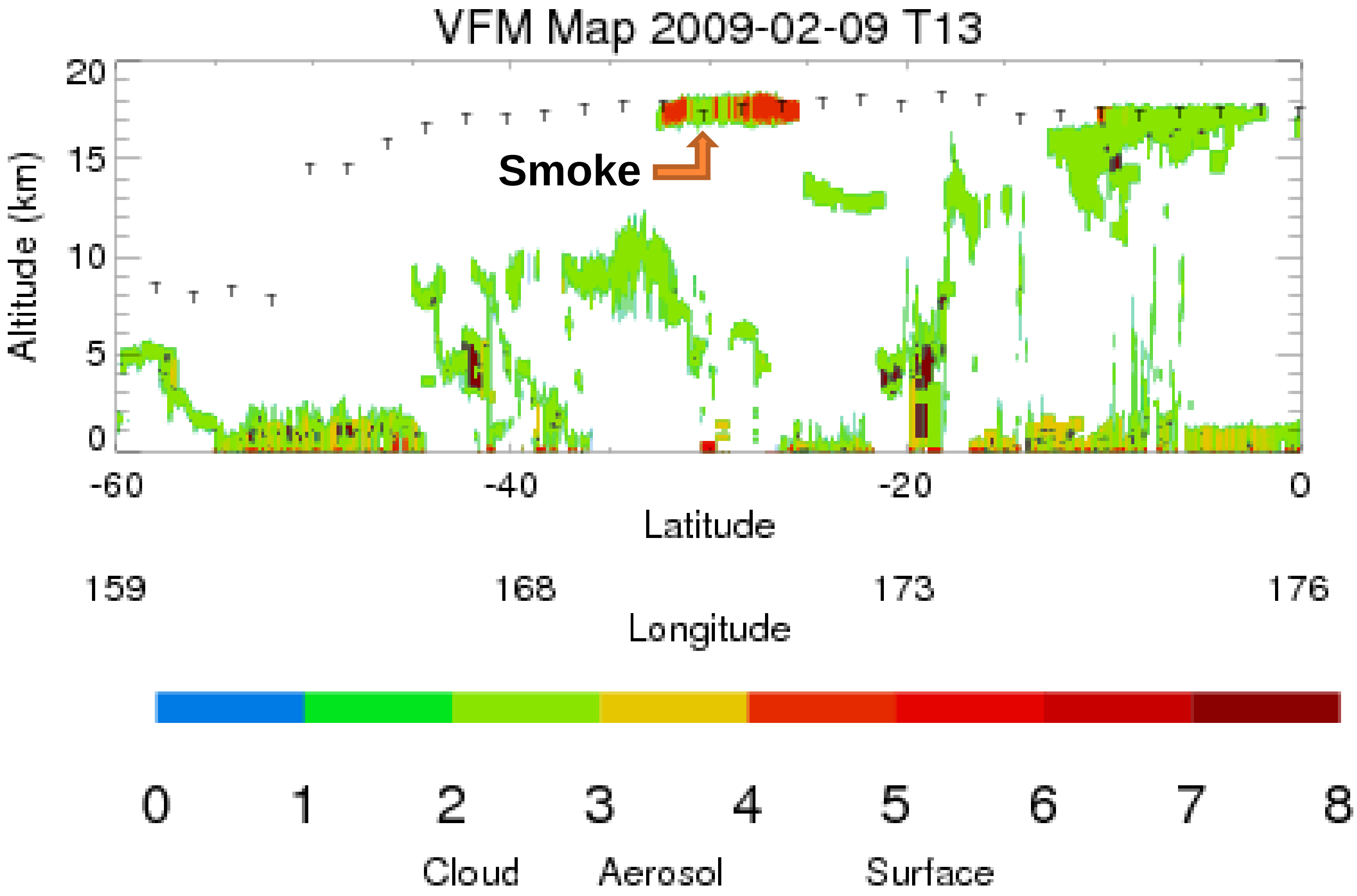
February 8 CALIPSO lidar Image



Smoke is between Australia and New Zealand at Z < 5 km

Fires started on Saturday, February 7

February 9 CALIPSO lidar image



Smoke top is at 18.5 km

T = thermal tropopause (from NCEP GFS analysis)

Absorptive Aerosol

Type	Emissions	Size
Desert dust	1500 Tg / yr	coarse
62% North Africa, 15% Asia, 11% Arabia		
Black carbon (soot)	10	fine
Biomass burning	90	fine
Primary organic aerosol	50	coarse
biological debris		
Secondary organic aerosol	70	fine
VOC = volatile organic carbon 60 Tg/yr natural VOC, 10 Tg/yr anthroprogenic VOC		

Seinfeld and Pandis, Atmospheric Chemistry and Physics, 1998

Desert Dust

Sokolik, I, and O. Toon (1999) Incorporation of mineralogical composition Into models of the radiative properties of mineral aerosol from UV to IR Wavelengths, JGR, v 104, p9423-9444.

Desert dust composition varies from place to place !

Table 2. Mineralogical Composition of Bulk Dust Samples Collected at Various Locations

	Sal Island	Barbados	Miami	Southern New Mexico	Sudan Sample 2/KH/83	Nigeria Sample 4	Nigeria Sample 9
Illite	53.8	64.3	62.9	19.0	3.8	1.17	5.6
Kaolinite	6.6	8.3	6.3	20.0	N/R	12.0	22.6
Montmorillonite	N/R	N/R	N/R	35.0	N/R	N/R	N/R
Quartz	19.6	13.8	14.2	6.0	56.3	83.3	67.4
Chlorite	4.3	4.1	4.2	N/R	N/R	N/R	N/R
Gypsum	N/R	N/R	N/R	N/R	5.0	N/R	N/R
Calcite	8.2	3.9	6.9	16.0	7.3	N/R	N/R
Hematite	N/R	N/R	N/R	N/R	6.23	N/R	N/R
Others	7.5	5.4	5.5	4.0	21.4	3.5	4.4

N/R, data were not reported. Values are in units of % by weight.

HITRAN currently has Eric Shettle's 1979 compliation of quartz, hematite, and sand indices

Absorption and the Imaginary Index

Complex Index of Refraction, $n(\lambda)$

$$n(\lambda) = n_{real} - i\, n_{imaginary}$$

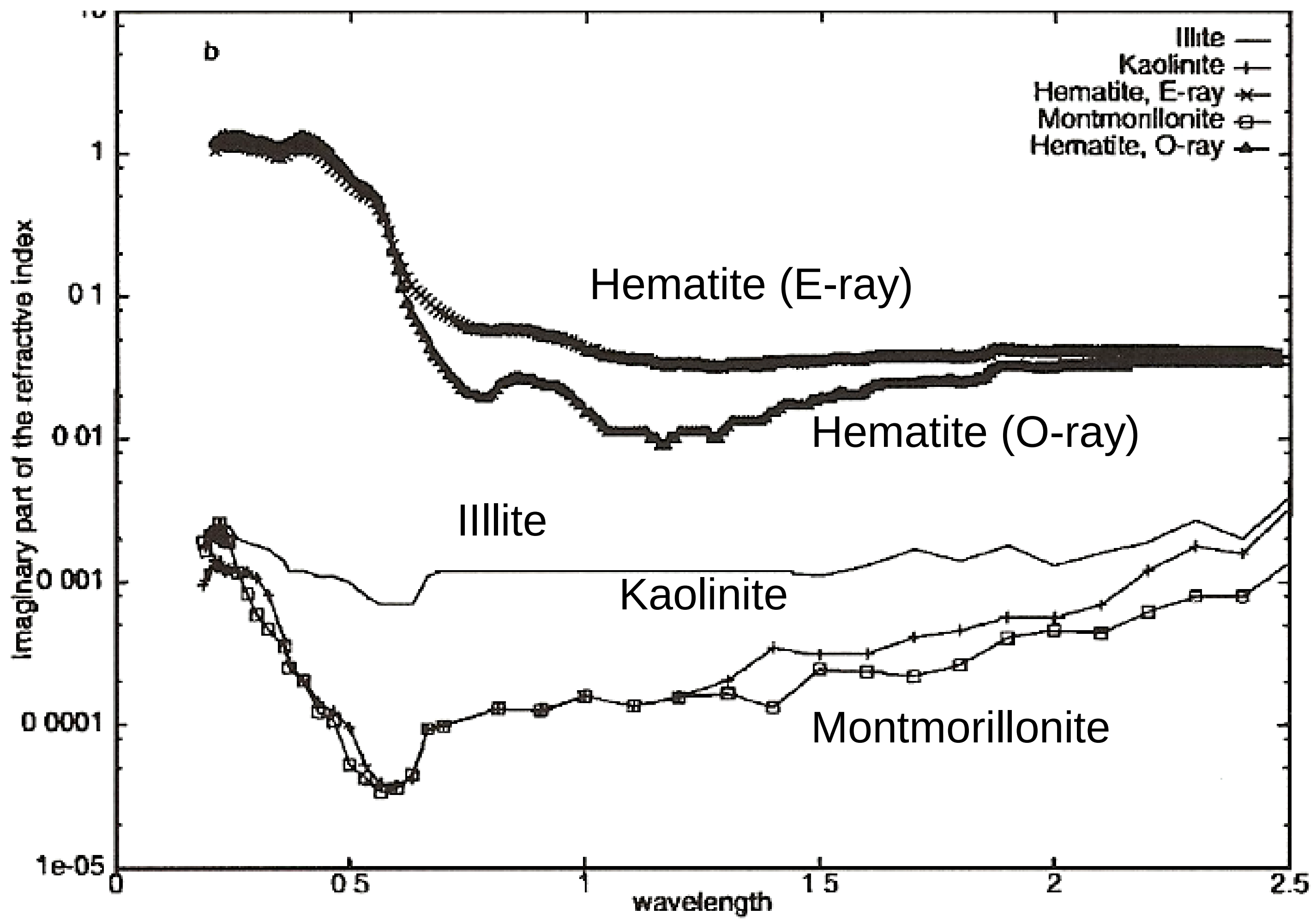
For a plane wave traveling in the z direction

$$E = \exp[-\omega\, n_{imaginary}\, z/c]\exp[i\omega(t - n_{real}\, z/c)]$$

E, ampltitude of electric field
λ, wavelength of light
c, speed of light
t, time
ω, circular frequency of the wave

Ultraviolet and visible indices

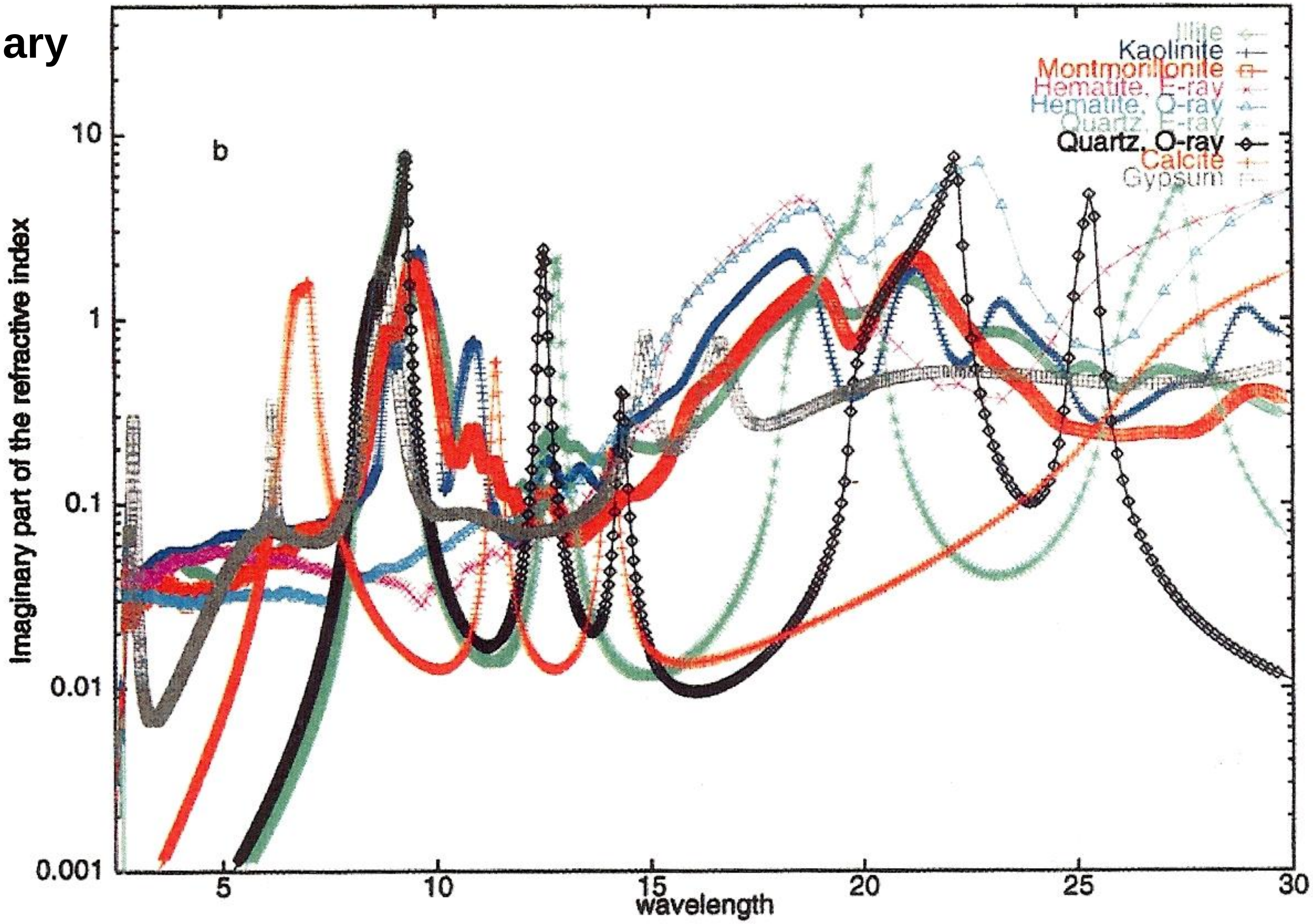
Imaginary index



Sokolik and Toon, 1999

Infrared indices

Imaginary index



Sokilik and Toon, 1999

Black Carbon (BC)

Ramanathan, V. and G. Carmichael (2008) Global and regional climate changes due to black carbon, Nature Geoscience, vol 1, 221-227.

Black Carbon (soot) ~8 Tg / yr

20% biofuels
40% fossil fuels
40% open biomass burning

Internally mixed with sulphates, nitrates, organics, dust, sea salt

An absorptive aerosol optical depth of 0.02 can enhance solar heating of the lower atmosphere by up to 50%

Global black carbon solar heating ~2.6 W / m^2

Absorption Amplification

Jacobson (2001), Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, Nature, v 409, 695-697.

Within 5 days 60% per mass of BC obtained a non-BC coating

State	Direct Radiative Forcing
Externally mixed	0.31 W / m^2
Coated Core	0.55
Well internally mixed	0.62

Evolution of Aerosol

Freshly emitted soot

Elemental carbon, organic carbon
with trace ionic and metallic species

Hydrophobic

Particles change due to coagulation with soluble aerosols

Condensation of secondary organic and inorganic aerosols

Hydrophobic content decreases with distance from source

Some models use half-life of 24 hours for this transformation

Imaginary Index (K) of Carbonaceous Materials

Twitty, J. T. and J.A. Weinman (1971), J. Appl. Meteor., Vol 10, 725-731

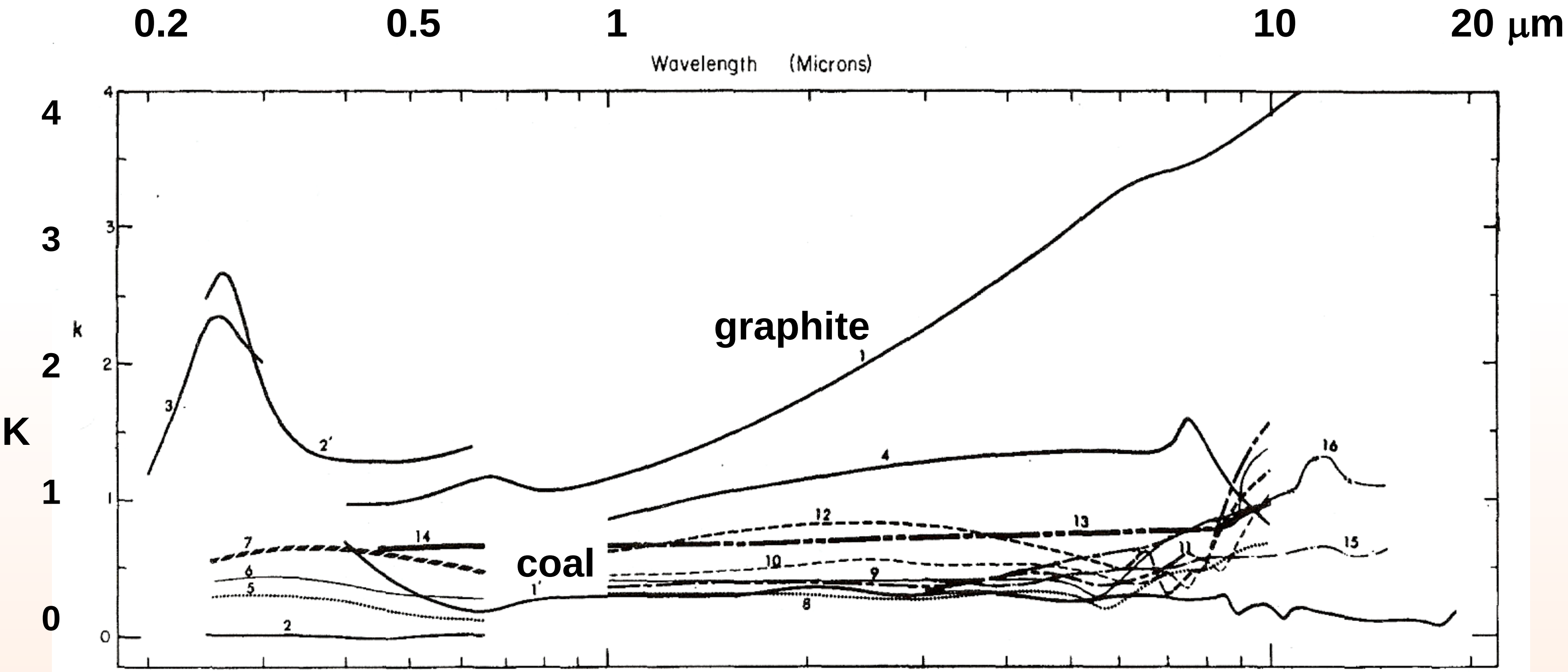
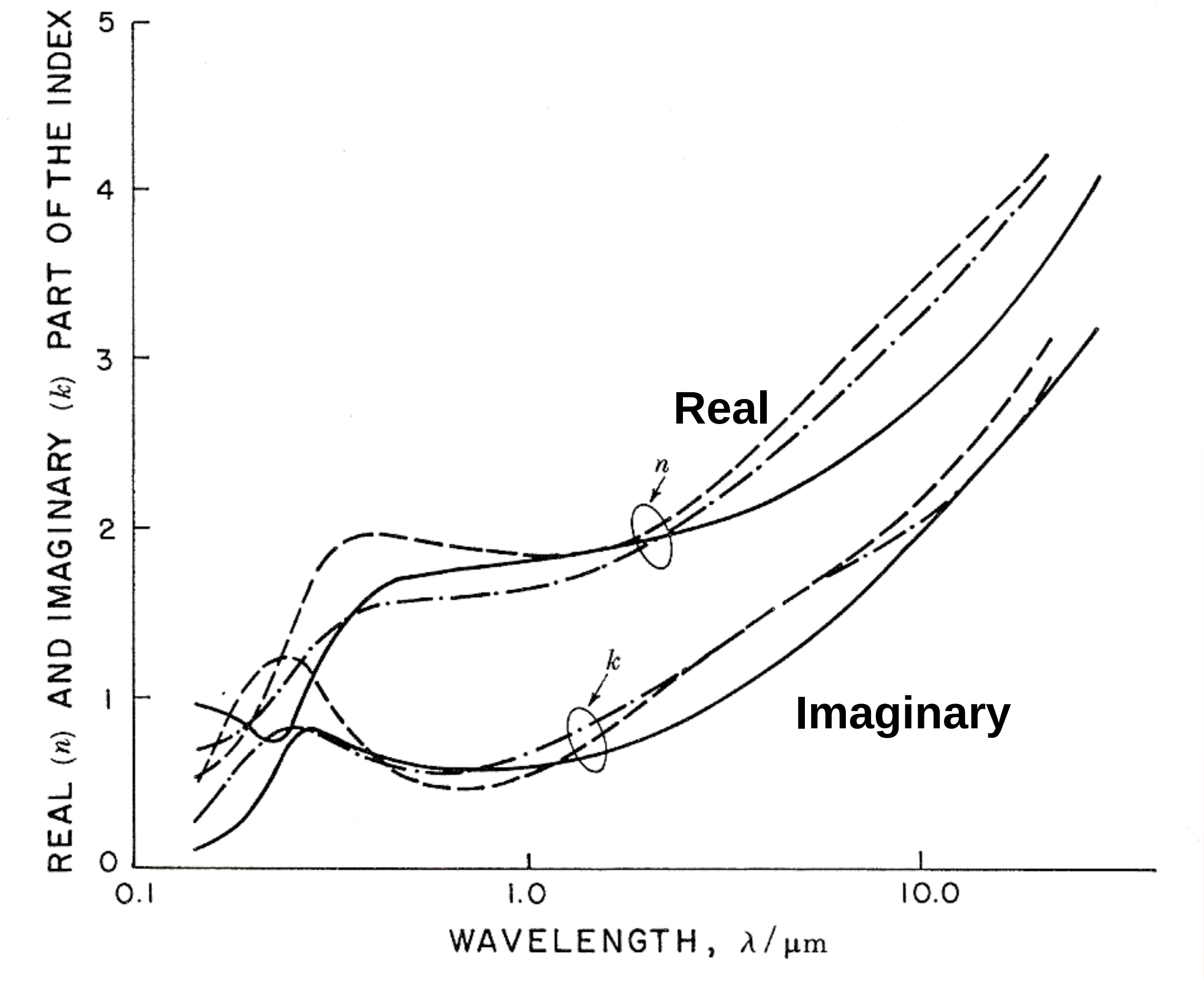


FIG. 1. Complex index of refraction of carbonaceous materials as a function of wavelength. Curves 1–4 are graphites; 5–9, coals; 10 and 11, coal burning soots; 12, oil furnace soot; 13, natural gas soot; 14, carbon black; 15, chimney soot; 16, activated charcoal. References: 1 and 1′, Lenham and Terherne (1966); 2 and 2′, Greenaway *et al.* (1969); 3, Carter *et al.* (1965); 4 and 8–13, Foster and Howarth (1968); 5–7, McCartney *et al.* (1965); 14, Senftleben and Benedict (1917); 15 and 16, Volz (1970, private communication).

HITRAN has Eric Shettle's 1979 composite curve (e.g. K=0.5 at 1 micron)

Indices of Flame Soot



Solid line: Chang and Charalampopouls, Proc R Soc Lond, v 430, p 577, 1990

2 5 10 μm

Real

Imaginary

Real

Imaginary

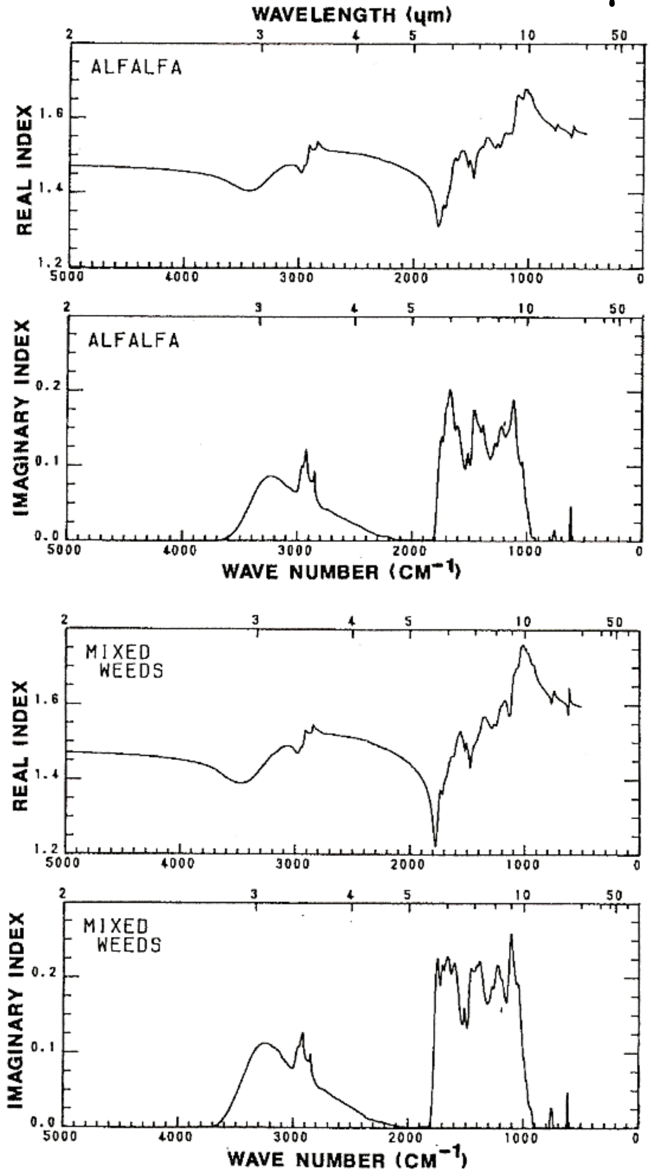


FIGURE 6. Real and imaginary refractive indexes for the alfalfa and weed-mix samples.

1.6

1.2

0.3

0.1

1.6

1.2

0.3

0.1

Sutherland and Khana, 1991
2 – 25 μm

Little variation in the vegetative indices in the IR

Sutherland indices are included in HITRAN

Optical Properties from Aircraft Measurements

Southern African Regional Science Initiative (SAFARI 2000)

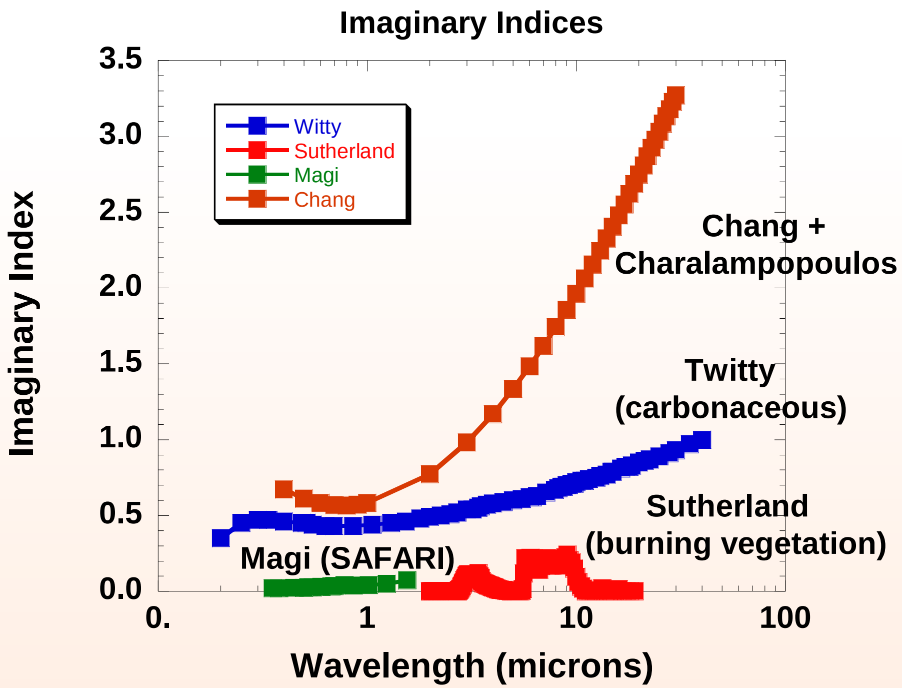
August and September aircraft flights during the dry season

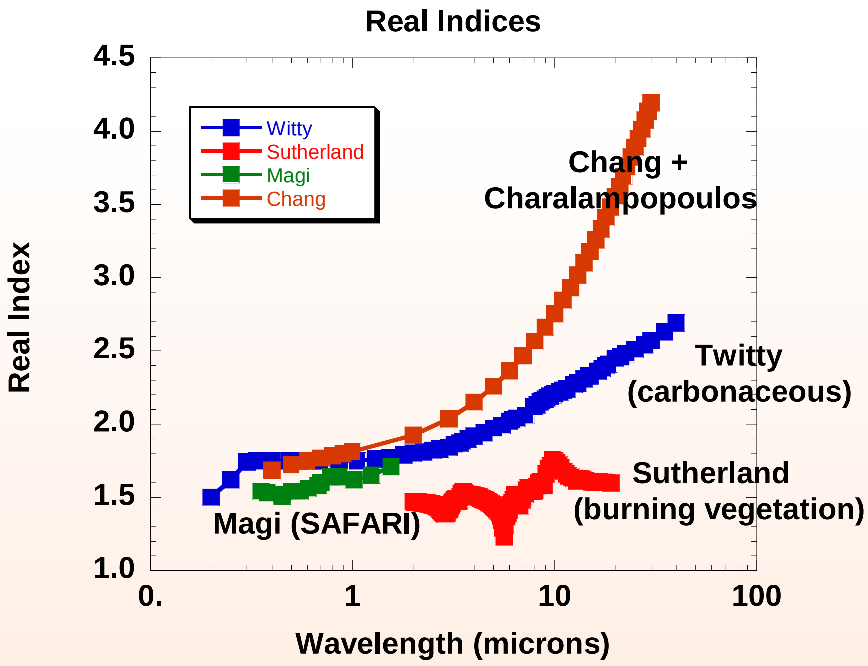
Wavelength dependent measurements
- **Extinction, single scattering albedo, backscatter ratios**
- **15 sunphotometer wavelengths from 354 to 1557 nm**
- **In-situ nephlometer measurements at 450, 500, 700 nm.**

Derive Real and Imaginary indices that best match observations

Magi, B. I., Q. Fu, and J. Redemann (2007) A methodology to retrieve self-consistent aerosol optical properties using common aircraft measurements, JGR, Vol 112, 2007.

Considerable range of index values !





Organic Acids related to Tropospheric Aerosols

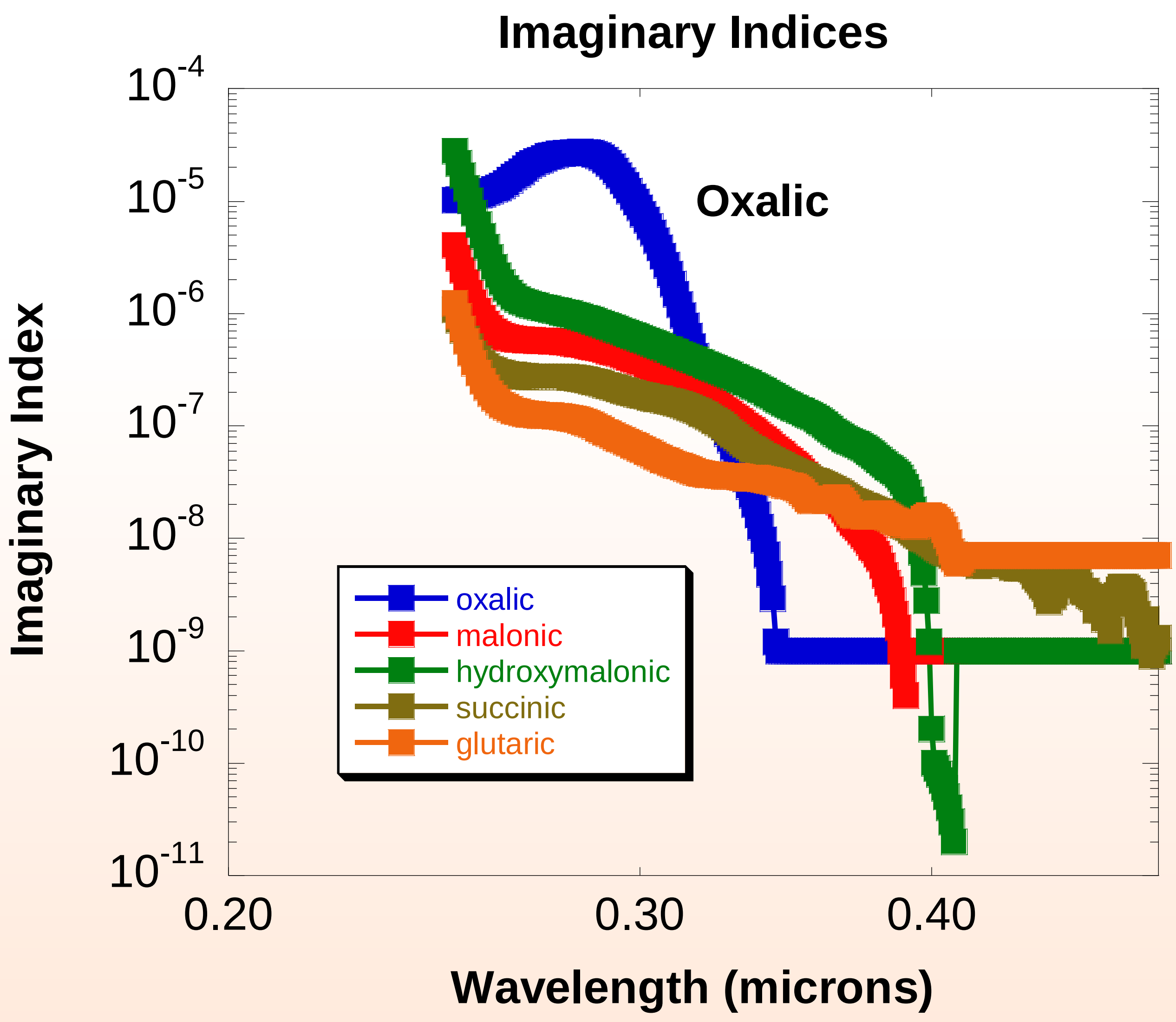
C. E. Lund Myhre and C. J. Nielsen, Optical properties in the UV and visible spectral region of organic acids relevant to tropospheric aerosols (2004) Atmos. Chem. Phys, vol 4, 1759-1769.

Wavelength range: 275 – 1100 nm, Real indices

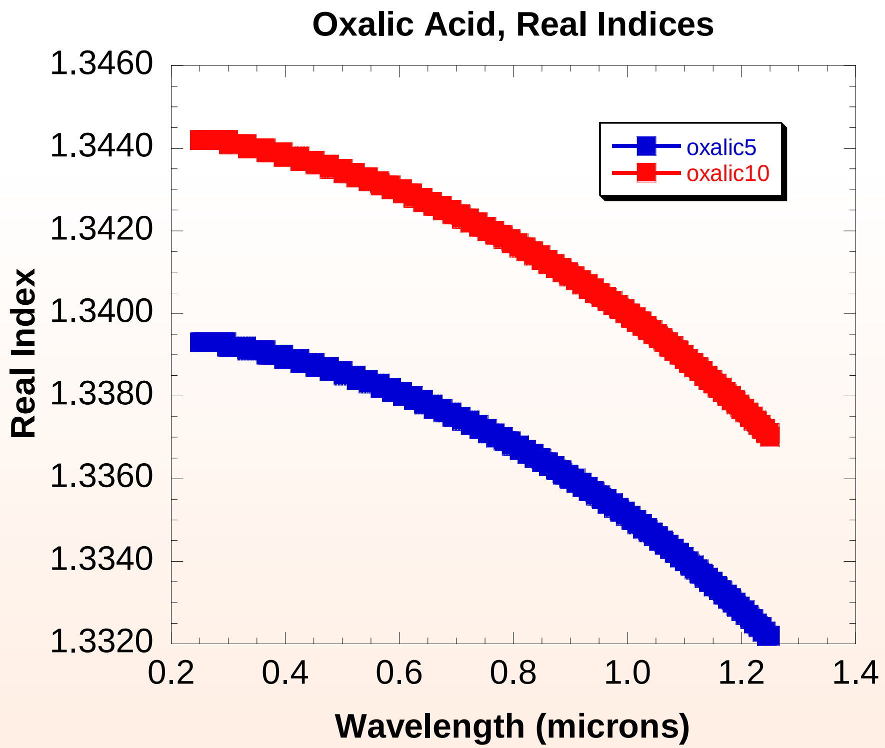
WSOC - Water Soluble Organic Carbon

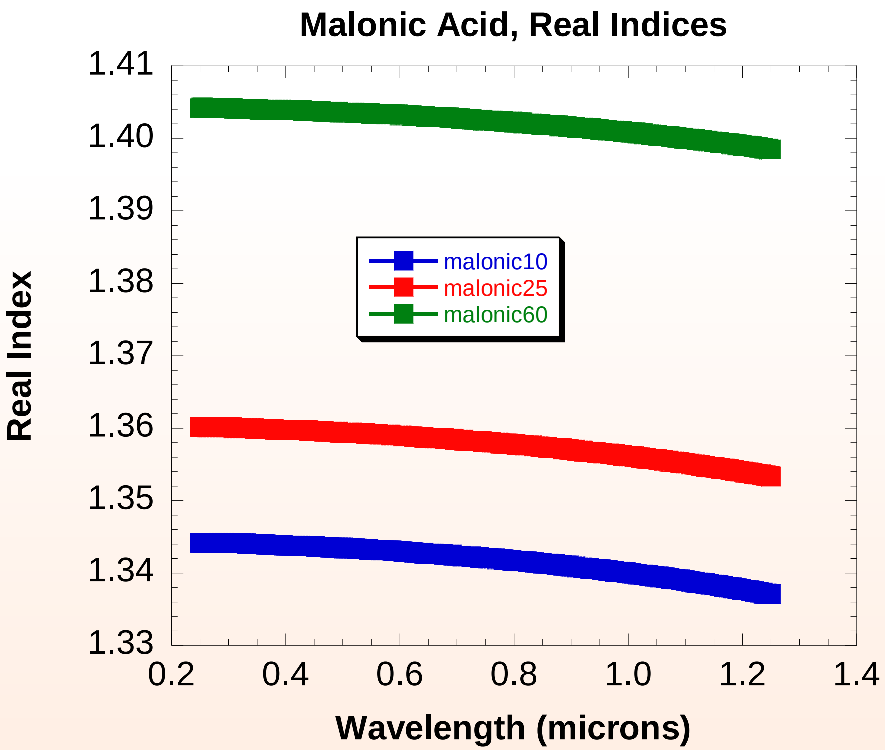
Organic Acid	Formula	Comment	
Oxalic	$C_2H_2O_4$	C2	Most abundant organic acid
Malonic	$C_3H_4O_4$	C3	High concentrations in beetroot
Tartronic	$C_3H_4O_5$	C3	Reacts with air to form mesoxalic acid
Succinic	$C_4H_6O_4$	C4	Sugar fermentation byproduct
Glutaric	$C_5H_9NO_4$	C5	Its salt is the food enhancer MSG
From oxidation of terpenes:			
Pyruvic	$C_3H_4O_3$	C3	Supplies energy to living cells
Benzoic	$C_7H_6O_2$	C7	Used as a food preservative
Pinonic	$C_{10}H_{16}O_3$	C10	Product of alpha-Pinene ($C_{10}H_{16}$)
Phthalic	$C_8H_6O_4$	C8	Abundant in the Arctic and Antarctic

Organic acids have scattering effect



Real Indices





5,10
Wt% curves

10, 25, 60
Wt% curves

Calculation of Optical Properties

Mie calculation:

Bohren and Huffman, Absorption and Scattering of Light by Small Particles, Wiley, 1983

"BHCOAT" Fortran program for coated sphere

$$\beta\ (\mathrm{km}^{-1}) = \int Q(r, \lambda, n(r))\ \pi r^2\ dN(r)/dr\ dr$$

r, particle radius

Extinction efficiency Q from Mie theory

Particle size distribution, dN(r)/dr (number $cm^{-3}\ \mu m^{-1}$)

Core: e.g. Chang flame soot indices

Coating: e.g. Lund Myhre and Nielsen oxalic indices

Conclusions and Recommendations

Current absorptive indices on HITRAN do not encompass the range of optical properties of particles that are present in the atmosphere

Add to HITRAN:

Sokolik and Toon mineralogical indices

Chang and Charalampopouls flame soot indices

Magi indices (from field data)

Lund Myhre and Nielsen organic acid indices

This talk is a plea for additional laboratory and field measurements !

Thank You

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To advance understanding of weather, climate, atmospheric composition and processes;
To provide facility support to the wider community; and,
To apply the results to benefit society.

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