The microphysical modelling of aerosols in the Oxford-RAL Aerosol and Cloud Retrieval

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Abstract

The Oxford-RAL Aerosol and Cloud retrieval algorithm (ORAC) uses the optimal estimation of Rodgers [2000] to obtain aerosol optical depth, and aerosol effective radius from satellite measurements in the visible and near-IR by the Advanced Along Track Scanning Radiometer (AATSR).

Due to insufficient information content, all other properties of the observed aerosol distribution must be assumed. This includes assumptions about the size distribution, composition, shape, and ambient atmospheric conditions.

Work to improve the aerosol microphysical modelling of mineral dust and carbonaceous aerosol by altering the shape and composition of the particle distributions in order to better represent real atmospheric particles is presented.

The ORAC Aerosol Forward Model

In order to retrieve information from AATSR measurements, we must model what we expect the satellite to see for a certain atmospheric state. The ORAC aerosol forward model calculates top of atmosphere (TOA) relectance using DISORT [Stames et al., 1988] to obtain atmospheric transmission. The bottom two layers of this modelled atmosphere include an aerosol component whose light scattering properties are pre-computed. These properties have traditionally been based on the Optical Properties of Aerosols and Clouds database (OPAC) which assumes combinations of various aerosol modes. These individual modes are lognormal size distributions of homogeneous, spherical scatterers with uniform refractive index across the range of mode particle sizes. An example size distribution for the maritime aerosol class using properties from the OPAC database is shown in Fig. 1. Various aerosol effective radii can be generated by altering the mixing between different sized aerosol modes.

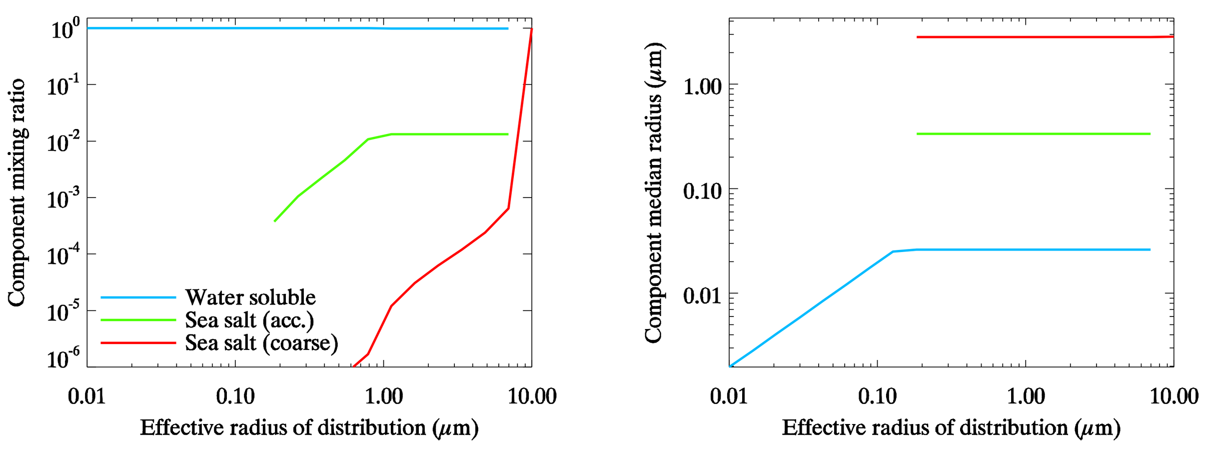

Figure 1: An example aerosol class, the maritime class. It is built up from three lognormal modes representing nucleation, accumulation, and coarse mode aerosol. In order to get a size distribution with larger or smaller effective radius, the relative mixing between components is altered.

In reality, not all aerosols are spherical and homogeneous. This work has added more exotic particles aiming to better represent those aerosols that are found in the atmosphere.

Mineral Dust Aerosol

Mineral dust aerosol is one of the most common naturally occurring aerosol types. It is brought into the atmosphere by saltation, a wind driven process where already-detached, larger particles collide with obstructions on the surface bed, projecting smaller particles into the air.

In the current ORAC model, dust is represented by four aerosol modes based on the OPAC database. From smallest to largest these are a background "water soluble" mode representing background secondary sulphate; mineral dust nucleation mode; mineral dust accumulation mode; and mineral dust coarse mode.

Size distribution investigation

The mineral dust size distribution was compared to aircraft measurements from the SAMUM campaign to investigate the appropriate of the current OPAC assumptions used. The aircraft measurements of size resolved aerosol concentration were fitted using a combination of four-mode lognormal modes, varying mixing ratios and mode properties [Weinzierl et al., 2009]. Fig. 2 shows the comparison between ORAC and the measurements, which were obtained over north-west Africa. Both the mode

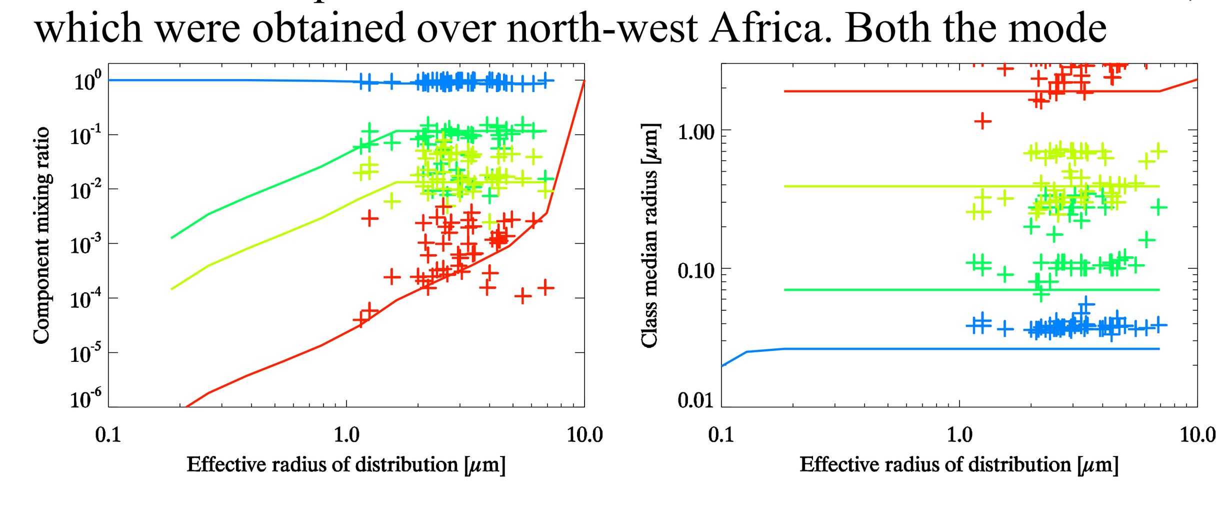

Figure 2: Comparing the ORAC representation of mineral dust aerosol to measurements of aerosol size distribution from the SAMUM campaign.

radii, and the relative mixing ratios of the four aerosol modes agree well, suggesting that current size distribution assumptions are sensible. The smallest mode in the ORAC model is possibly too small. This is the water soluble mode, with an atmospheric relative humidity of RH = 50 %. Increasing the RH would bring this mode closer to the SAMUM measurements.

Mineral dust refractive index

Dust refractive index (RI) is determined by the composition of the mineral constituents. These can vary largely with geographical region of emission, altering the light scattering properties of the dust from that area. One must then ask: does the regional variation in mineral dust refractive index lead to large variation in the aerosol light scattering properties? To answer this question, a calculation of mineral dust RI variability was undertaken, using a map of worldwide mineralogy and calculating likely emissions rates from ECMWF wind fields. Fig. 3 shows spread of imaginary RI at 550 nm. Imaginary part of RI was mainly determined by the hematite content.

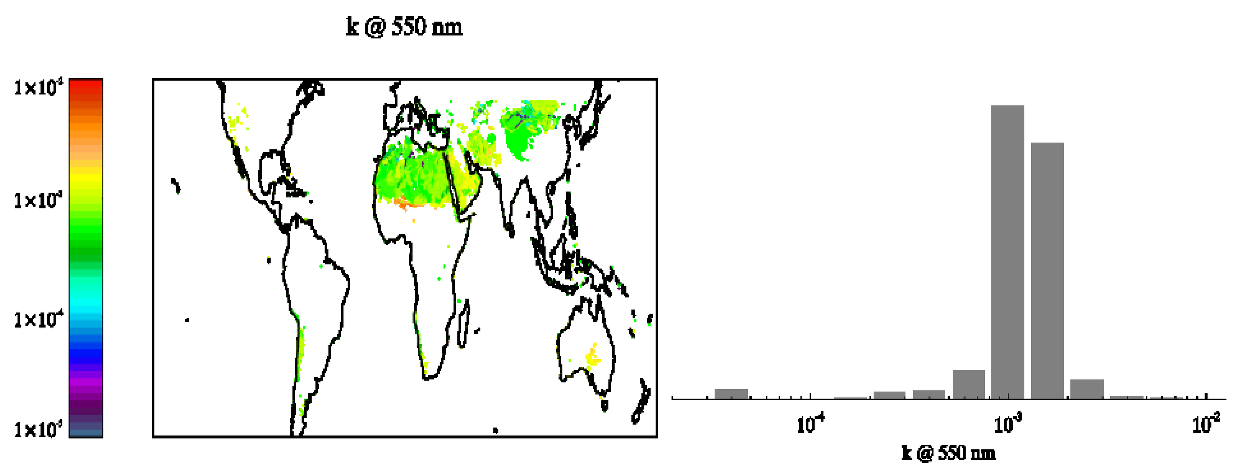

Figure 3: Estimate of worldwide mineral dust refractive index. The imaginary part of RI at 550 nm is shown. Spread of values are

Spread in refractive index was not found to dramatically alter light scattering properties when a weighted mean taking into account relative emission rates was calculated.

Non-sphericity

Electron microscopy images show mineral dust to be elongated, angular particles which are not well represented using a spherical assumption. Light scattering properties of mineral dust have been shown to be more adequately fitted by a distributions of spheroids [Dubovik et al., 2006]. This lead to dramatic changes in the phase functions of the scattering distributions. Fig. 4 shows the difference between ORAC mineral dust aerosols with and without non-spherical mineral dust. Differences are most pronounced for larger particles.

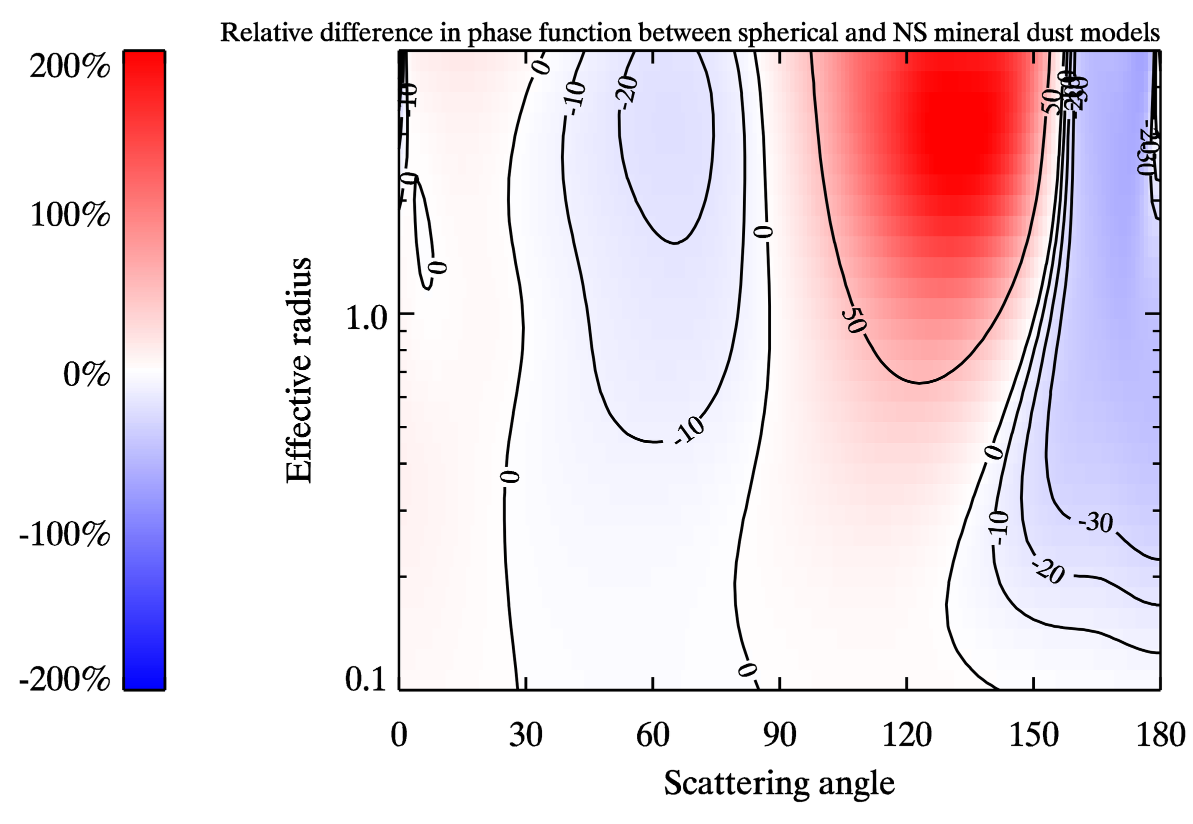

Figure 4: Phase function differences between non-spherical and spherical mineral dust modes in the ORAC mineral dust class. The ORAC mineral dust class also includes a water soluble mode, which was not altered.

A new sand model

Finally, based on observations by Kaaden et al. [2009], the fine mode mineral dust particles were made slightly hydrophilic, and modelled as coated spheres with the coating varying with relative humidity. Improvements to the retrieval are shown in Fig. 5. Residuals give the amount of radiance remaining after the retrieval has found its best solution. Thus we would like residuals to be centred about zero, suggesting that errors are random and not systematic.

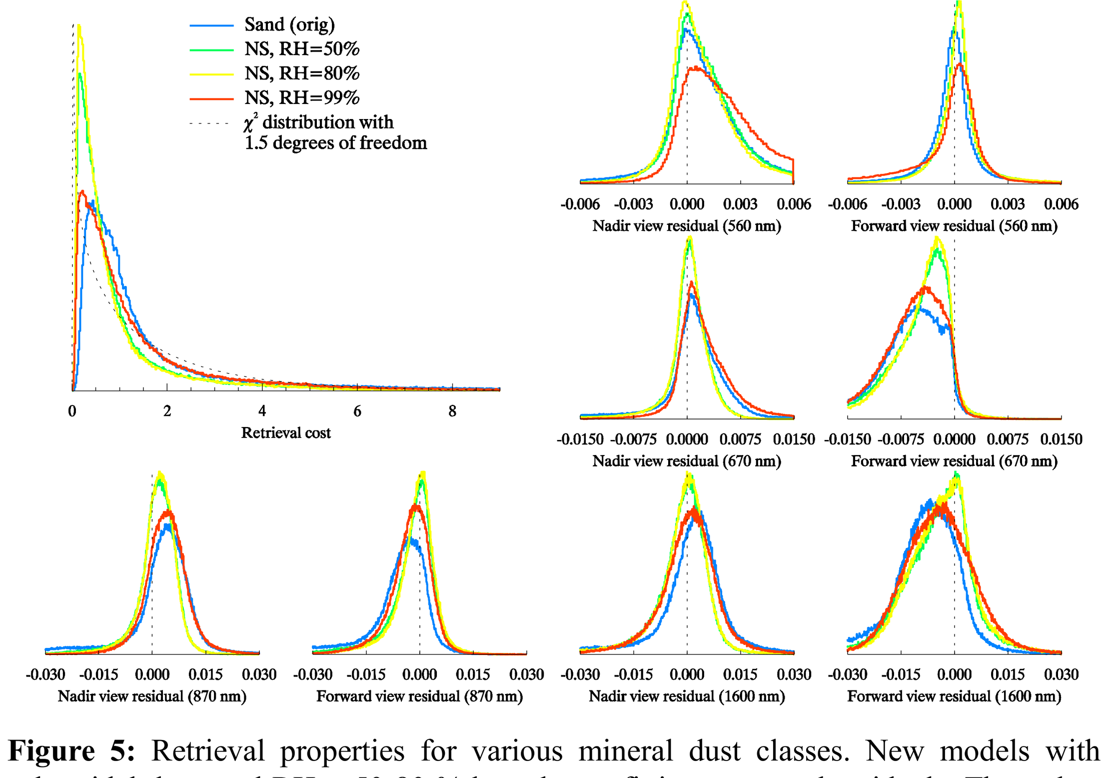

Figure 5: Retrieval properties for various mineral dust classes. New models with spheroidal dust, and RH = 50-80 % have better fitting costs and residuals. These data are from over the Sahara during a dust event in March 2006.

Carbonaceous Aerosol

Burning fuels leads to the production of black carbon (BC) aerosols. Shortly after emission, these particles are often observed to have fractal morphology [Sorensen, 2001]. Particles are made up of small "spherules" with radius of ~20 nm which agglomerate forming larger structures. The current ORAC soot model describes the particles instead as being individual spheres, each with the radius of a spherule. Due to the small sizes of the aerosol, a large number of them are required to accurately represent the mass of soot in the atmosphere. This can lead to large errors in the light scattering properties of urban aerosols.

Fractal modelling of fresh black carbon

Soot in the ORAC urban class was remodelled as a distribution of fractal particles. The lognormal distribution of individual particle radii was replaced with a lognormal distribution of equivalent-volume radii (EVR). The correct EVR is obtained by building a fractal molecule with a specific number of spherules.

Light scattering calculations for appropriate fractals are shown in Fig. 6, along with the light scattering properties for the EVR sphere. Fractals with 10 and 200 spherules are shown. Scattering calculations must be averaged over several fractal shapes with the same properties, as each shape will be unique.

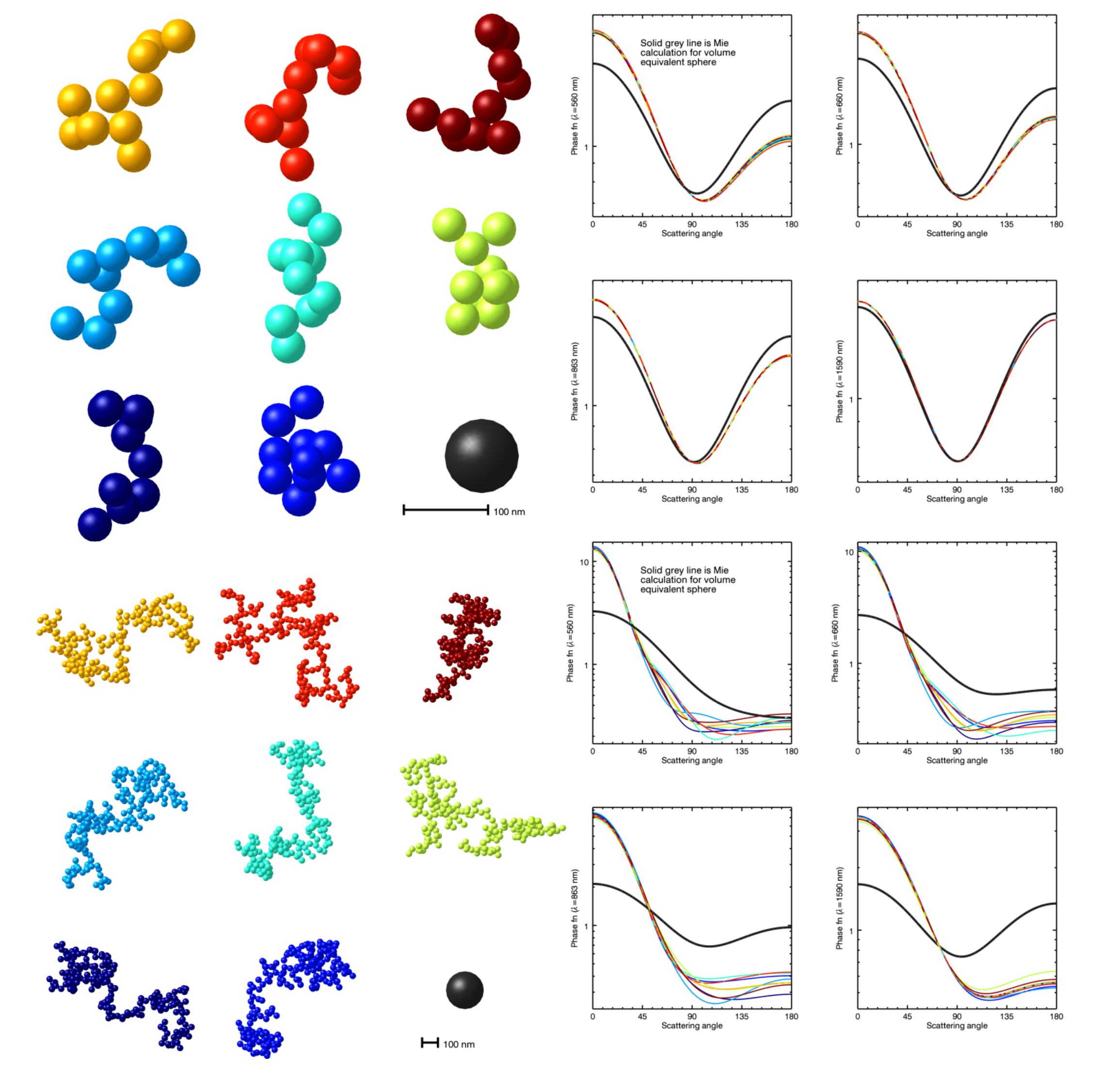

Figure 6: Example of fractal soot particles and their light scattering properties. Colours on the light scattering plots correspond to the properties from the same coloured particles on the left. The bottom right sphere is the EVR sphere. For these calcualtions, the fractal properties are: spherule radius=20 nm, D_f=1.19, k_f=1.82, RI=1.85+0.71i.

Improvements to the retrieval

The new ORAC urban aerosol class, with larger, fractal soot particles was is compared to the previous model which has smaller, spherical soot particles. Residuals are much improved, as shown in Fig. 7. Also plotted is the retrieved continental aerosol for the same scenes (which are only over urban areas). The continental aerosol class is used for background aerosol fields over land, and differs from the urban class only in the absence of a soot aerosol mode. These have very similar residuals, suggesting that both fit the measured data well. However using the urban class, retrieved optical depths are smaller.

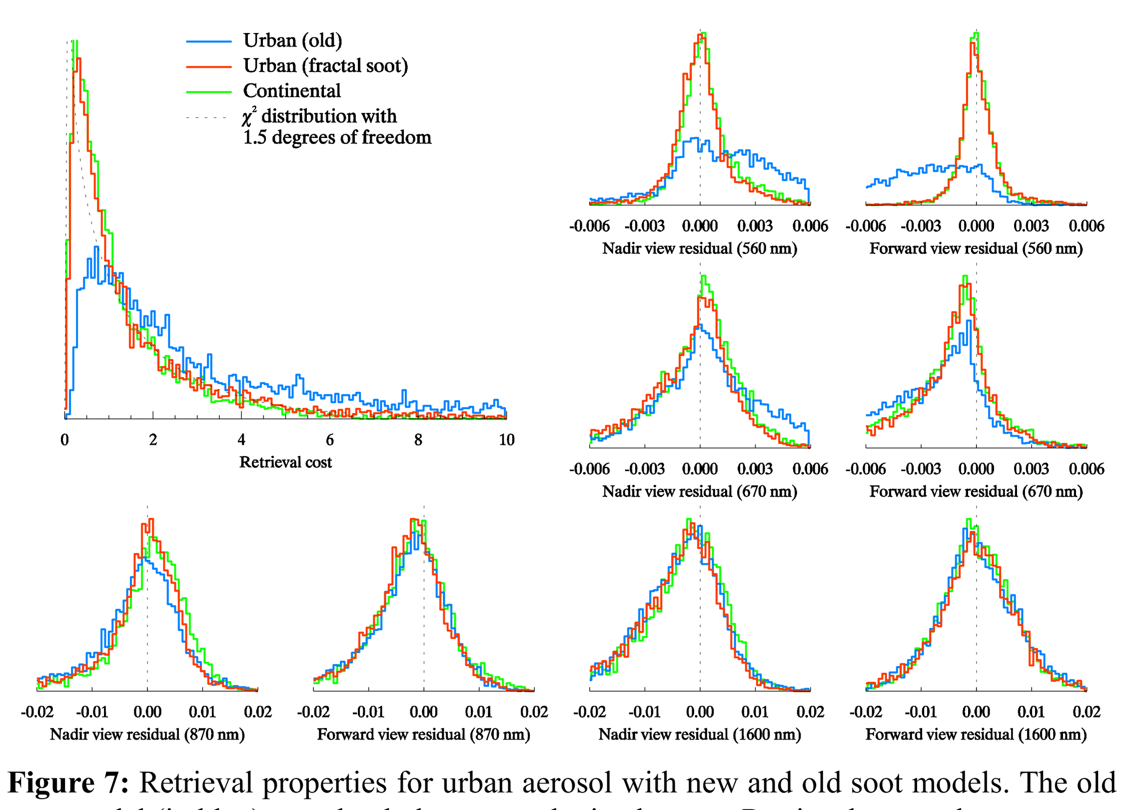

Figure 7: Retrieval properties for urban aerosol with new and old soot models. The old soot model (in blue) can clearly be seen to be inadequate. Retrievals are only over areas of urban extent. Also shown is the continental aerosol class, which represents background continental aerosol.

References

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