

# FLEXAOD: A CHEMISTRY-TRANSPORT MODEL POST-PROCESSING TOOL FOR A FLEXIBLE CALCULATION OF AEROSOL OPTICAL PROPERTIES

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## ABSTRACT

The theoretical basis of the FlexAOD package under development are reported here. In its current version, FlexAOD is a software that may calculate aerosol optical properties offline from the output of a chemistry-transport model with bulk mass approach. It is applied here to the GEOS-Chem global model and the libRadtran radiative transfer model. Future developments include different mixing rules options and AERONET-like column output.

## 1. INTRODUCTION

Aerosol in the atmosphere originates from primary emissions of natural (soil dust, sea salt, volcanoes, wildfires, plant debris, ...) and anthropogenic (fossil fuel combustion, agricultural practices, ...) sources and from secondary formation via condensation of semivolatile gaseous species [1]. It results a complex and continuously evolving mixture of aerosol compositions and shapes, which affect the size distribution, hygroscopicity and complex refractive index.

Remote sensing of atmospheric aerosol properties uses retrieval algorithms that often relies on one or more "aerosol models" [2]. These models define the optical properties of a "typical" or "average" aerosol layer present at the scene of observation, assuming a certain range of chemical compositions and size distributions. Aerosol optical depths and other physical and optical properties are then retrieved minimizing the difference of observed radiances with the available spectrum of simulated radiances, calculated by means of Radiative Transfer Models (RTM). Moreover, these aerosol models may also be preliminarily used in the calculations of atmospheric correction, when Earth' surface reflectance is the target of the retrieval process.

The calculation of aerosol optical properties in Chemistry-Transport Models (CTM) is generally required for photolysis rates computation and to compare/integrate results with optical observations, such as the Aerosol Optical Depth (AOD) or the extinction profile. In General Circulation Models (GCM) and Numerical Weather Prediction (NWP) models optical properties of aerosol are also needed to simulate the propagation of radiation, e.g. for the radiation budget in the governing equations and for climate forcing assessment (direct and indirect radiative effects).

there is currently a lack of interface between the aerosol remote sensing and the modelling community, mostly

because of the peculiar difficulties in dealing with reciprocal algorithms. Here we describe a new tool aimed at filling this gap. FlexAOD offers an easy and customizable way to (1) calculate aerosol optical properties, such as aerosol optical depth, extinction profile, LIDAR backscattering, single scattering albedo, etc., from CTM model output, and (2) calculate input for radiative transfer models (e.g. asymmetry factor, phase function, etc.) for specific locations and times. The package is applied here to the GEOS-Chem [3] global model output and it is used in comparison with multi-spectral aerosol measurements from satellites and AERONET network for several test cases.

## 2. SOFTWARE DESCRIPTION

In its current version and default setup, FlexAOD replicates offline the aerosol optics calculations performed online within the GEOS-Chem global CTM. The original methodology for aerosol mass simulation and related conversion to optical depth [4] is derived from the GOCART model [5]. GEOS-Chem simulates aerosol with a bulk mass approach, i.e. it does not simulate explicitly the size distribution, but only the composition of aerosol in terms of total mass. Exceptions are sea salts, for which two size bins are simulated (accumulation and coarse mode, SSA and SSC), and dust, simulated in 4 size bins (DST1-4). Other simulated species are inorganic secondary ions (sulphate, nitrate, ammonium, SULF), organic (primary and secondary, OC) and black carbon (BC). For SSA, SSC, SULF, OC and BC "optical species" log-normal size distributions of number concentration per size interval  $n(r)$  are assigned [6]:

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{r \ln \sigma_g} \exp\left[-\frac{(\ln r - \ln r_0)^2}{2 \ln^2 \sigma_g}\right] \quad (1)$$

where  $r$  is the aerosol radius,  $N_0$  is the total number concentration calculated by the model, while  $r_0$  and  $\sigma_g$  are the modal radius and the geometric standard deviation, respectively, taken from the OPAC database [7] with some modifications [6].

For dust, gamma distributions are assumed [4, 6]:

$$n(r) = N_0 r^{(1-3b)/b} \exp\left(-\frac{r}{ab}\right), \quad b \in (0, 0.5) \quad (2)$$

Moreover, as a legacy of the interface to the RTM used

in GEOS-Chem for photolysis rates calculations, the finer dust size bin is split into 4 bins, resulting in a total of 7 simulated dust “species” for optical calculations. The mass of the finer bin is evenly distributed among the 4 subbins. In Fig. 1 we show the assumed dry size distributions (RH = 0%). Dust bins are taken over the same gamma distribution, but they only contribute to the optical calculations in limited size ranges as shown in the figure. OCPI and BCPI refer to the hydrophilic fraction of OC and BC. The hydrophobic fraction, also simulated into the model, is assumed on the same log-normal distributions.

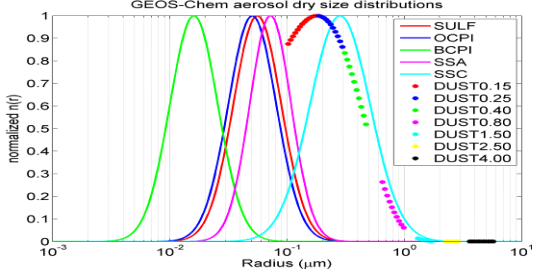


Figure 1. Aerosol dry size distributions assumed in the GEOS-Chem model (and default FlexAOD setup) for optical calculations.

Optical calculations are done under the assumption of spherical particles, thus the Mie theory applies. Mie calculations are carried out using NASA’s *spher* code [8]. For a given size-distribution the aerosol optical depth  $\tau$  for each model layer is approximated as [5, 9]:

$$\tau = \sum_{i=1}^{N_{ext}} \frac{3}{4} \frac{\overline{Q}_{e,dry,i} f_{RH,i}}{r_{e,dry,i} \rho_i} M_i = \sum_{i=1}^{N_{ext}} \beta_i M_i \quad (3)$$

where  $N_{ext}$  is the number of externally mixed aerosol components,  $\overline{Q}_{e,dry,i}$  is the Mie extinction efficiency averaged over the dry size-distribution,  $\rho_i$  is the species density,  $M_i$  is the column mass concentration in the layer ( $\text{g}/\text{m}^2$ ),  $r_{e,dry,i}$  is the dry effective radius, that for log-normal distributions is [9]:

$$r_e = r_0 \exp\left(\frac{5}{2} \ln^2 \sigma_g\right) \quad (4)$$

The term  $f_{RH}$  in Eq. 3 is a scaling factor used to account for hygroscopic growth [4-6], calculated as follows for all species but dust, which is assumed to be non-hygroscopic ( $f_{RH} = 1$ ). Parameters of Eq. 3 are pre-tabulated at five relative humidity bins (0, 50, 70, 80, and 90%) and then interpolated to ambient RH for each scene. The  $f_{RH}$  scaling factor is calculated as:

$$f_{RH,i} = \frac{\overline{Q}_{e,wet,i} \left( \frac{r_{e,wet,i}}{r_{e,dry,i}} \right)^2}{\overline{Q}_{e,dry,i}} \quad (5)$$

where  $\overline{Q}_{e,wet,i}$  and  $r_{e,wet,i}$  are the extinction efficiency and effective radius, respectively, interpolated at ambient RH from the look-up table.

The terms multiplying the mass concentrations on the r.h.s. of Eq. 3 are often grouped in one variable  $\beta_i$  ( $\text{m}^2/\text{g}$ ), called mass extinction efficiency. The RH dependence of  $\beta_i$  for GEOS-Chem/FlexAOD species is shown in [6].

Aerosol characterization for radiative transfer calculations generally also requires the knowledge of other variables in addition to extinction, specifically [10]:

- the single scattering albedo:

$$\omega_{0,i} = \sigma_{sca,i} / \sigma_{ext,i} \quad (6)$$

which is the ratio of scattering to total (scattering plus absorption) extinction;

- the Legendre expansion coefficients of the phase function [8]:

$$P_i(\Theta) = \sum_{s=1}^{s_{max}} \alpha_{s,i} P_s(\cos \Theta), \quad \alpha_{0,i} \equiv 1 \quad (7)$$

where  $\Theta$  is the scattering angle,  $P(\Theta)$  is the phase function,  $P_s$  are generalized spherical functions,  $\alpha_s$  are the expansion coefficients, and  $s_{max}$  is the order of truncation of the expansion:

- and the asymmetry parameter:

$$g_i = \frac{1}{2} \int_0^\pi \cos \Theta P_i(\Theta) \sin \Theta d\Theta \quad (8)$$

The above parameters are not available from the standard GEOS-Chem output, but may be calculated offline with FlexAOD and saved in output in a format suitable for input to libRadtran RTM [10].

For comparison with measurements, FlexAOD also adds the following diagnostic variables:

- the aerosol number concentration  $N_i$  ( $\#/\text{cm}^3$ ):

$$N_i = \frac{\chi_i M_{w,i} n_{air}}{N_{avo} \rho_i V_i} \quad (9)$$

where  $\chi_i$  is the mixing ratio of species  $i$ ,  $M_{w,i}$  its molar weight,  $n_{air}$  the air number

concentration,  $N_{avo}$  Avogadro's number, and  $\bar{V}_i$  is the average particle volume;

- the LIDAR ratio [11]:

$$S = \frac{\sigma_{ext}(km^{-1})}{\sigma_{back}(km^{-1}sr^{-1})} = \frac{4\pi}{P(180^\circ)} \quad (10)$$

which express the ratio of total to backscatter extinction.

In the standard online calculations, the 9 optical species are assumed to be externally mixed, thus  $N_{ext} = 9$  in Eq. 3, i.e. the total aerosol optical depth of a layer is given by the sum of the optical depths calculated for individual species. The same applies to the total extinction coefficient and the total number concentration. The average values of other properties must be weighted by the extinction or the scattering coefficients, as follows [12]:

- $$\bar{\omega}_0 = \frac{\sum_i \sigma_{ext,i} \bar{\omega}_{0,i}}{\sum_i \sigma_{ext,i}} \quad (11)$$

- $$\bar{\alpha}_s = \frac{\sum_i \sigma_{sca,i} \alpha_{s,i}}{\sum_i \sigma_{sca,i}} \quad (12)$$

- $$\bar{g} = \frac{\sum_i \sigma_{sca,i} g_i}{\sum_i \sigma_{sca,i}} \quad (13)$$

### 1.1. Internal mixing

Although for optical properties purposes aerosol are often assumed in external mixtures into models, in the atmosphere they are usually coated in various types of internal mixtures. External mixing assumption is computationally convenient, because one can rely on tabulated size distributions and refractive indices for each species. Accounting for the internal mixing nature of aerosol is computationally much more expensive, because the refractive index of the mixture changes at every location and time-step. Indeed, in first approximation, the refractive index of an internal mixture of aerosol may be calculated as the volume-weighted average of components refractive indices as [12]:

$$m_j = \frac{\sum_i v_i m_i}{\sum_i v_i} \quad (14)$$

where  $m_i$  is the refractive index of the  $i$ -th species and  $v_i$  its total volume concentration:

$$v_i = \int \frac{4}{3} \pi r^3 n(r) dr \quad (15)$$

The refractive index of the internal mixture thus changes because it depends on the concentration of the components, which is not the case for external mixtures. The mixing rule given in Eq. 14 is only one the possible, but the best mixing rule depends on the geometric arrangement of the components [9]. When insoluble particles (inclusion) are suspended into a solution (e.g. black carbon into aqueous sulphate particles), the Maxwell-Garnett effective dielectric constant approximation should be used:

$$\varepsilon_{MG} = \varepsilon_2 \left[ \frac{\varepsilon_1 + 2\varepsilon_2 + 2f_1(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1 + 2\varepsilon_2 - f_1(\varepsilon_1 - \varepsilon_2)} \right] \quad (16)$$

where  $\varepsilon = \sqrt{m}$  is the dielectric constant, subscript 1 and 2 refer to inclusion and solution, respectively, and  $f_1$  is the volume fraction of the inclusion.

When the insoluble particles are interdispersed by dry components, then the Bruggeman approximation apply, and the effective dielectric constant is the solution of:

$$f_1 \left( \frac{\varepsilon_1 - \varepsilon_B}{\varepsilon_1 + 2\varepsilon_B} \right) + f_2 \left( \frac{\varepsilon_2 - \varepsilon_B}{\varepsilon_2 + 2\varepsilon_B} \right) = 0 \quad (17)$$

The choice of the mixing rule is especially important for the determination of the imaginary part of the refractive index, related to the absorption properties of the medium: differences as high as 15-20% may arise for different mixing rules, that will directly reflect into the subsequent optical properties calculations.

The internal mixing case is being implemented into FlexAOD and it is not available in current version of the code.

### 3. SAMPLE APPLICATION

In Fig. 2 we show the comparison of the annual average AOD as observed by MISR onboard Terra at three wavelengths (443, 555, 670 nm) and calculated from GEOS-Chem aerosol fields. AOD is calculated offline using FlexAOD from the GEOS-Chem simulation. A spectral model to observation comparison is often avoided, just because one need to repeat a time-consuming full CTM simulation. FlexAOD overcomes this difficulty and allow to look at model bias at different wavelengths: for example, one can see that the model bias over Africa is positive at shorter wavelengths and negative at longer wavelengths. Thus one may attribute the bias not only to the calculation of bulk mass, but also to the uncertainty introduced by assumptions made for optical calculations.

In Fig. 3 we show the same comparison over equatorial Africa. The model AOD slope with increasing wavelength is steeper than observations and this might be simply due to a wrong assumption in the size distribution (in particular too fine in this case), even if the mass was simulated correctly.

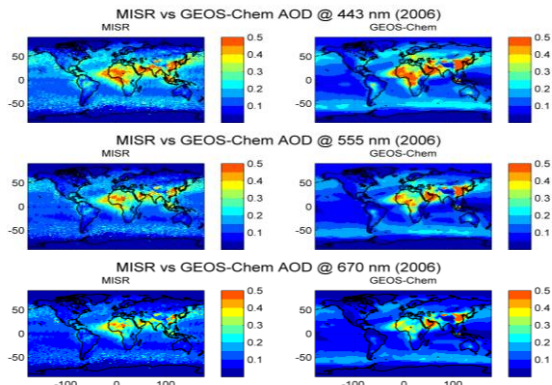


Figure 2. Spectral comparison of annual average AOD observed by MISR and simulated with GEOS-Chem. Model AOD are calculated offline using FlexAOD.

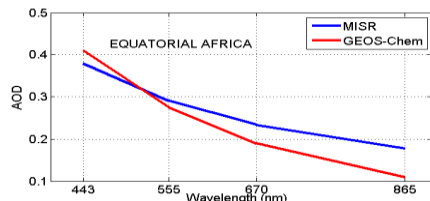


Figure 3. MISR vs GEOS-Chem spectral AOD over equatorial Africa.

#### 4. ONGOING DEVELOPMENTS

The following FlexAOD code developments are undergoing:

- Internal mixing options (see sec. 1.1)
- AERONET like output: the world-wide sunphotometers network provide inversion products of column-averaged aerosol properties [13]. A column-average model output is being implemented for direct comparison with those data.
- Idealized profiles, suitable for more conceptual sensitivity tests on optical calculations.
- More flexible input/output, in terms of customizable aerosol mixtures and kind of output variables.
- Interface with general CTM, i.e. extend the applicability also to sectional and modal aerosol models.

#### 5. ACKNOWLEDGEMENTS

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