



# EVALUATION OF THE EFFECTS OF AEROSOL CONCENTRATIONS AND RELATIVE HUMIDITY ON POLARIZABILITY AND PHASE FUNCTION OF URBAN AEROSOLS VALIDATED WITH MODIS SATELLITE DATA

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## Article History

Received: 07.03.2026

Accepted: 15.04.2026

Published: 06.05.2026

**Abstract:** This work numerically evaluates urban aerosol optical properties using OPAC 4.0 across 0.4–0.8  $\mu\text{m}$  wavelengths, focusing on insoluble (INSO), soot (SOOT), and water-soluble (WASO) components at 0% and 50% relative humidity (RH). Fifteen aerosol models were developed with five concentration gradients per component. The Ångström exponent ( $\alpha_a$ ) systematically decreases with increasing INSO (1.376-1.273) and increases with SOOT (1.376-1.386) and WASO (1.376-1.433). A critical non-linear hygroscopic response was identified:  $\alpha$  increases from 0% to 50% RH at low WASO concentrations but decreases at high concentrations due to hygroscopic saturation and coarse-mode transition. Phase function forward scattering enhances with RH, most pronounced for WASO (1.626-1.785). MODIS validation across ten countries shows OPAC-derived  $\alpha_a$  (1.27-1.43) matching anthropogenically influenced regions (Germany  $\alpha_a=1.27$ , Ethiopia  $\alpha_a=1.45$ ), while extreme values (South Africa  $\alpha_a=2.39$ ; Egypt  $\alpha_a=0.12$ ) represent aerosol types beyond OPAC's urban scope.

**Keywords:** Aerosol optical properties; Ångström exponent; Relative humidity; OPAC; MODIS.

## Cite this article:

Aliyu, A., Tijjani, B. I., and Gana, U. M., (2026). EVALUATION OF THE EFFECTS OF AEROSOL CONCENTRATIONS AND RELATIVE HUMIDITY ON POLARIZABILITY AND PHASE FUNCTION OF URBAN AEROSOLS VALIDATED WITH MODIS SATELLITE DATA. *ISAR Journal of Science and Technology*, 4(5), 15-25.

## 1. Introduction

Atmospheric aerosols constitute a major uncertainty in climate assessments, directly influencing Earth's radiative budget through scattering and absorption (Stocker *et al.*, 2013). Urban aerosols exhibit particular complexity due to heterogeneous sources: fossil fuel combustion, industrial activities, biomass burning, and resuspended mineral dust. Each component manifests distinct optical characteristics-INSO (mineral dust) shows weak spectral dependence, SOOT demonstrates strong absorption, and WASO components (sulfates, nitrates, organics) undergo pronounced hygroscopic growth (Seinfeld & Pandis, 2016).

The interaction between aerosols and water vapor critically alters optical properties as hygroscopic particles absorb water with increasing RH, shifting phase functions toward stronger forward scattering (Svenningsson *et al.*, 2006). However, this response varies non-linearly with concentration and chemical species.

The OPAC (Optical Properties of Aerosols and Clouds) software package (Hess *et al.*, 1998; version 4.0, Koepke *et al.*, 2015) enables computation of spectral optical properties for user-defined aerosol mixtures. Despite its widespread application, three research gaps persist: (1) component-specific  $\alpha$  response to RH across concentration gradients remains unquantified; (2) validation against satellite-derived  $\alpha_a$  across diverse geographical regions is limited; and (3) OPAC's validity boundaries for urban mixtures require rigorous establishment.

This study addresses these gaps by: (1) numerically evaluating concentration and RH effects on polarizability and phase function using OPAC 4.0; (2) deriving component-specific  $\alpha_a$  values and identifying non-linear hygroscopic responses; and (3) validating simulations against MODIS satellite retrievals across ten countries spanning dust-dominated, biomass-burning, and anthropogenically-influenced aerosol regimes.



## 2. Theoretical Background

### 2.1 Aerosol Size Distribution

Atmospheric aerosols follow a multimodal lognormal distribution (Whitby and McMurry, 1997):

The number size distribution is:

$$\frac{dN(r)}{d\log(r)} = \sum \left\{ \left[ \frac{N_i}{(\sqrt{2\pi})\log\sigma_i} \right] \exp \left[ -\frac{[\log(r)-\log(r_{mi})]^2}{2\log\sigma_i^2} \right] \right\} \quad (1)$$

where  $r$  = particle radius,  $r_{mi}$  = mode radius,  $\sigma_i$  = geometric standard deviation,  $N_i$  = total number concentration of mode  $i$ . For  $i=1, 2, 3 \rightarrow$  mono-, bi-, or trimodal distribution.

### 2.2 Polarizability and Phase Function

Polarizability  $\alpha$  relates induced dipole moment to external electric field:  $\vec{P} = \alpha \vec{E}$  (Bohren & Huffman, 1983). For mixtures,

$$\alpha_{\text{eff}} = \sum_{i=1}^n V_i \alpha_i \quad (2)$$

where  $V_i$  = volume mixing ratio,  $\alpha_i$  = polarizability of component  $i$ . Larger molecules/particles are generally more polarizable

The phase function  $F_{11}(\theta)$  describes angular scattering distribution normalized as

$$\frac{1}{2} \int_0^\pi \sin(\theta) F_{11}(\theta) d\theta = 1 \quad (3)$$

The ratio  $-F_{12}/F_{11}$  gives the degree of linear polarization for unpolarized incident light (Mishchenko *et al.*, 2002).

### 2.3 Hygroscopic Growth

Hygroscopic growth is the ratio of the particle diameter at any relative humidity (RH) to the particle diameter at RH = 0% (Tijjani, Shaaibu and Aliyu, 2014).

Hygroscopic growth factor at relative humidity (RH) is defined as (Tang, 1996):

$$g(\text{RH}) = \frac{r(\text{RH})}{r(\text{RH}_0)} \quad (4)$$

For a mixture, the effective growth factor is:

$$g_{\text{eff}}(\text{RH}) = \left( \sum_k x_k g_k^3(\text{RH}) \right)^{1/3} \quad (5)$$

where  $x_k$  = volume mixing ratio of component  $k$ ,  $g_k$  = its growth factor (Quinn *et al.*, 2005).

### 2.4 Ångström Power Law

The spectral dependence is characterized by:  $\tau(\lambda) = \beta \lambda^{-\alpha_a}$  (6)

linearized as

$$\ln \tau(\lambda) = \ln(\beta) - \alpha_a \ln(\lambda), \quad (7)$$

where  $\alpha_a$  (Ångström exponent) indicates particle size (larger  $\alpha_a$  = fine-mode dominance) and  $\beta$  (turbidity coefficient) represents aerosol loading (Moorthy *et al.*, 2001).

## 3. Methodology

### 3.1 OPAC 4.0 Numerical Simulations

OPAC 4.0 computed optical properties across the visible spectrum (0.4–0.8  $\mu\text{m}$ ) for fifteen aerosol models (Tables 1–3). Five concentration gradients per component (INSO, SOOT, WASO) were simulated at 0% RH (dry) and 50% RH (moderately humid). Mie theory was applied for spherical particles (WASO, SOOT) and T-matrix method for spheroids (INSO).

**Table 1:** INSO concentration gradient models (WASO=28,000  $\text{cm}^{-3}$ , SOOT=130,000  $\text{cm}^{-3}$  fixed)

| Component                 | Model 1 | Model 2 | Model 3 | Model 4 | Model 5 |
|---------------------------|---------|---------|---------|---------|---------|
| INSO ( $\text{cm}^{-3}$ ) | 1.50    | 1.95    | 2.40    | 2.85    | 3.30    |

**Table 2:** SOOT concentration gradient models (INSO=1.50  $\text{cm}^{-3}$ , WASO=28,000  $\text{cm}^{-3}$  fixed)

| Component                 | Model 1 | Model 2 | Model 3 | Model 4 | Model 5 |
|---------------------------|---------|---------|---------|---------|---------|
| SOOT ( $\text{cm}^{-3}$ ) | 130,000 | 169,000 | 208,000 | 247,000 | 286,000 |

**Table 3:** WASO concentration gradient models (INSO=1.50  $\text{cm}^{-3}$ , SOOT=130,000  $\text{cm}^{-3}$  fixed)

| Component                 | Model 1 | Model 2 | Model 3 | Model 4 | Model 5 |
|---------------------------|---------|---------|---------|---------|---------|
| WASO ( $\text{cm}^{-3}$ ) | 28,000  | 36,400  | 44,800  | 53,200  | 61,600  |

### 3.2 MODIS Satellite Data Validation

MODIS Collection 6.1 Level-2 aerosol products (MOD04/MYD04, 1997–2010) were obtained for ten countries.

Monthly mean AOD values at 0.41, 0.49, 0.55, and 0.67  $\mu\text{m}$  were extracted within defined bounding boxes. Ångström exponent and turbidity coefficient were derived via linear regression using SPSS 22.0.

## 4. Results and Discussion

### 4.1 INSO Component Results

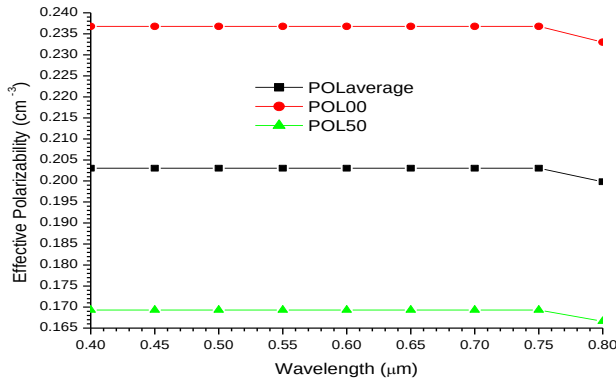


Figure 1: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for INSO Model 1

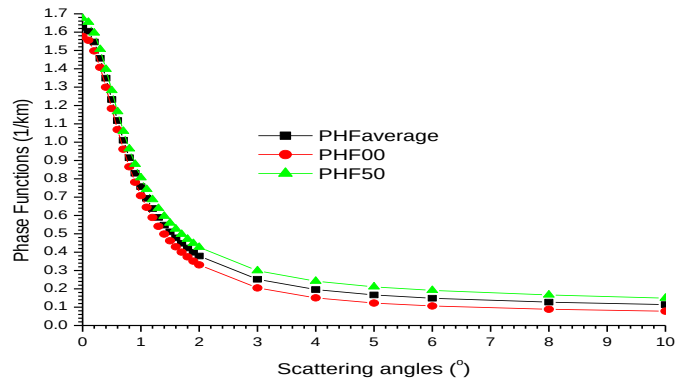


Figure 2: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, INSO Model 1 at phase angle 0° to 10°

- **Figure 1 (INSO Model 1, average of 0% and 50% RH):** Polarizability decreases monotonically from 0.203 at 0.4  $\mu\text{m}$  to 0.200 at 0.8  $\mu\text{m}$ . The 0% RH curve shows slightly lower values than the 50% RH curve, indicating hygroscopic enhancement.
- **Figure 2 (INSO Model 1, average of 0% and 50% RH):** Forward peak = 1.626 at 0°, decreasing sharply to 0.004 at 90°.

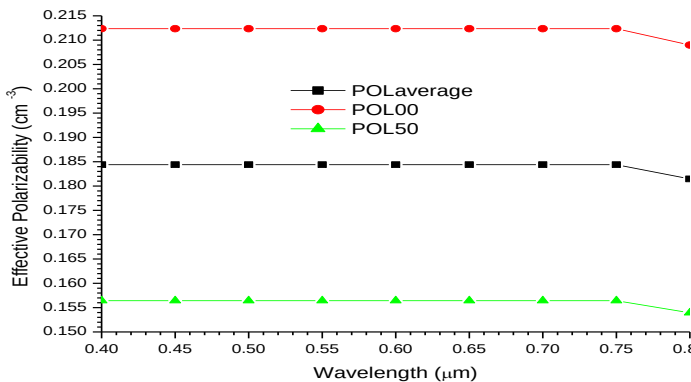


Figure 3: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for INSO Model 2

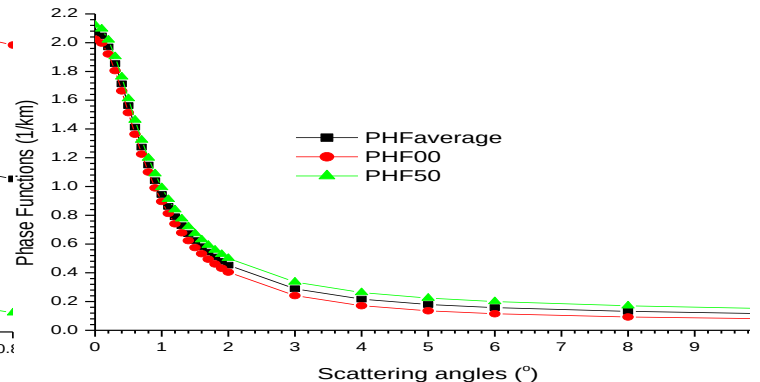


Figure 4: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, INSO Model 2 at phase angle 0° to 10°

- **Figure 3 (INSO Model 2, average of 0% and 50% RH):** Polarizability decreases from 0.184 at 0.4  $\mu\text{m}$  to 0.181 at 0.8  $\mu\text{m}$ , representing a reduction of approximately 9.4% from Model 1.
- **Figure 4 (INSO Model 2, average of 0% and 50% RH):** Forward peak = 2.073, showing enhanced forward scattering.

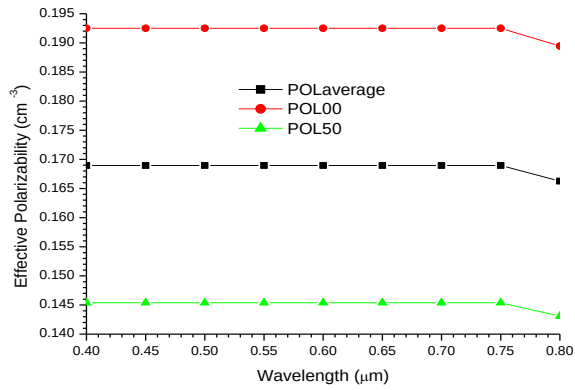


Figure 5: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for INSO Model 3

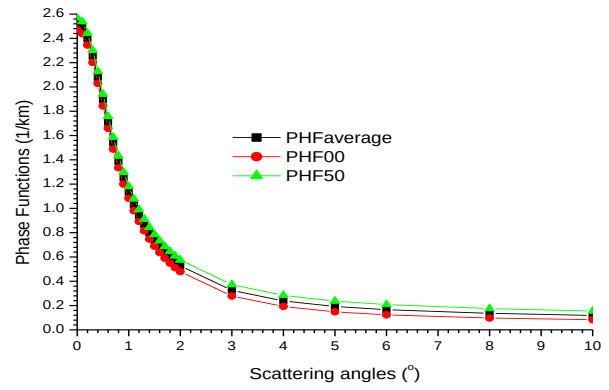


Figure 6: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, INSO Model 3 at phase angle 0° to 10°

- **Figure 5 (INSO Model 3, average of 0% and 50% RH):** Polarizability decreases from 0.169 at 0.4 μm to 0.166 at 0.8 μm.
- **Figure 6 (INSO Model 3, average of 0% and 50% RH):** Forward peak = 2.521, with oscillations appearing beyond 120°.

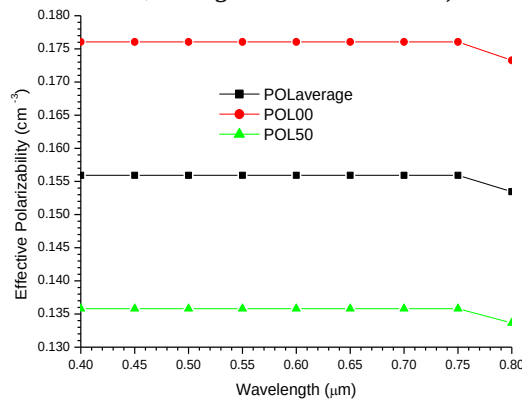


Figure 7: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for INSO Model 4

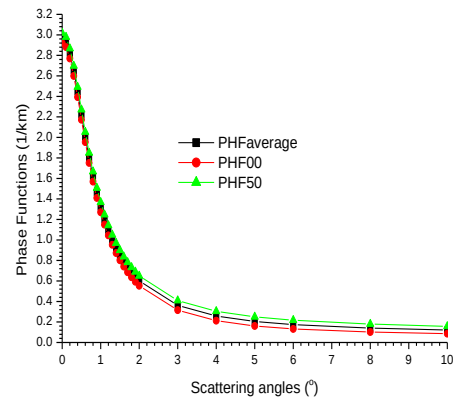


Figure 8: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, INSO Model 4 at phase angle 0° to 10°

- **Figure 7 (INSO Model 4, average of 0% and 50% RH):** Polarizability decreases from 0.156 at 0.4 μm to 0.153 at 0.8 μm.
- **Figure 8 (INSO Model 4, average of 0% and 50% RH):** Forward peak = 2.969, with pronounced glory-like feature near 180°.

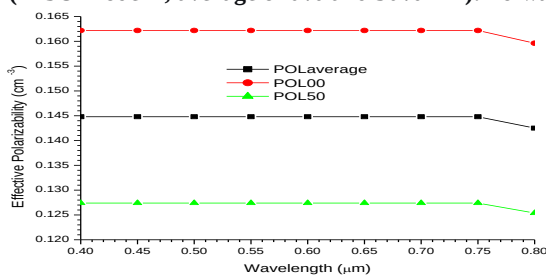


Figure 9: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for INSO Model 5

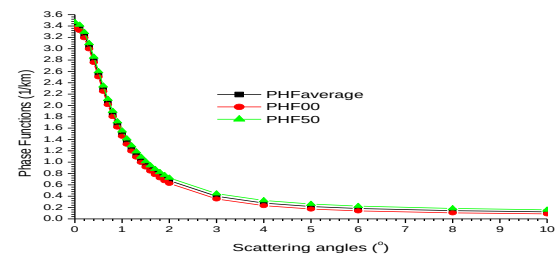


Figure 10: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, INSO Model 5 at phase angle 0° to 10°

- **Figure 9 (INSO Model 5, average of 0% and 50% RH):** Polarizability decreases from 0.145 at 0.4 μm to 0.143 at 0.8 μm. The 50% RH curve consistently lies above the 0% RH curve across all wavelengths.
- **Figure 10 (INSO Model 5, average of 0% and 50% RH):** Forward peak = 3.416, showing dense Mie oscillations.

**4.1.1 Polarizability:** Effective polarizability decreases systematically with increasing INSO concentration, from 0.203 at 0.4  $\mu\text{m}$  (Model 1) to 0.145 at 0.4  $\mu\text{m}$  (Model 5) at average RH (0%+50%). The 50% RH curve consistently lies above the 0% RH curve across all wavelengths, indicating hygroscopic enhancement of coexisting WASO components.

**4.1.2 Phase Function:** Forward scattering peak at  $0^\circ$  increases from 1.626 (Model 1) to 3.416 (Model 5) at average RH. Side scattering at  $90^\circ$  remains low (0.0038–0.0042), while backscattering increases progressively (0.0038→0.0042). Distinct Mie oscillations appear in Models 3–5 at  $120^\circ$ – $180^\circ$ .

#### 4.1.3 INSO Regression

**Table 4: Ångström Regression Parameters for INSO Aerosol Models (Average of 0% and 50% RH)**

| Model          | Avg $R^2$ | Avg Sig ( $R^2$ ) | Avg Alpha | Avg Alpha Sig | Avg Beta (Turbidity) | Avg Beta Sig |
|----------------|-----------|-------------------|-----------|---------------|----------------------|--------------|
| <b>Model 1</b> | 0.9987    | 2.99E-11          | 1.3758    | 2.99E-11      | 0.0993               | 2.00E-14     |
| <b>Model 2</b> | 0.9989    | 1.57E-11          | 1.3481    | 1.57E-11      | 0.1027               | 1.28E-14     |
| <b>Model 3</b> | 0.9991    | 9.81E-12          | 1.3219    | 9.81E-12      | 0.1061               | 7.71E-15     |
| <b>Model 4</b> | 0.9992    | 6.11E-12          | 1.2972    | 6.11E-12      | 0.1095               | 4.94E-15     |
| <b>Model 5</b> | 0.9993    | 3.54E-12          | 1.2726    | 3.54E-12      | 0.1130               | 2.85E-15     |

**Ångström Exponent:**  $\alpha$  decreases from 1.376 (Model 1) to 1.273 (Model 5) at average RH, confirming that increasing insoluble dust concentration shifts the aerosol population toward coarser mode.  $\beta$  increases from 0.099 to 0.113.

#### 4.2 SOOT Component Results

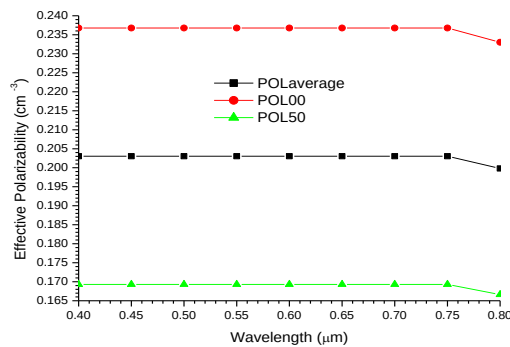


Figure 11: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for SOOT Model 1

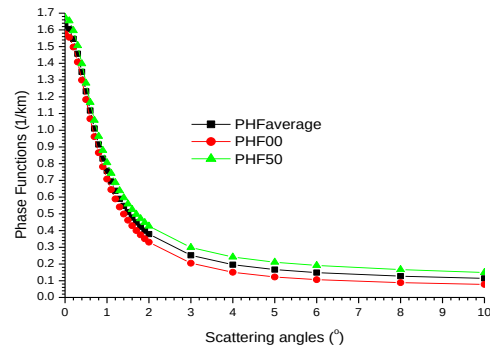


Figure 12: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, SOOT Model 1 at phase angle  $0^\circ$  to  $10^\circ$

- **Figure 11 (SOOT Model 1, average of 0% and 50% RH):** Polarizability decreases from 0.203 at 0.4  $\mu\text{m}$  to 0.200 at 0.8  $\mu\text{m}$ .
- **Figure 12 (SOOT Model 1, average of 0% and 50% RH):** Forward peak = 1.626, nearly isotropic at larger angles.

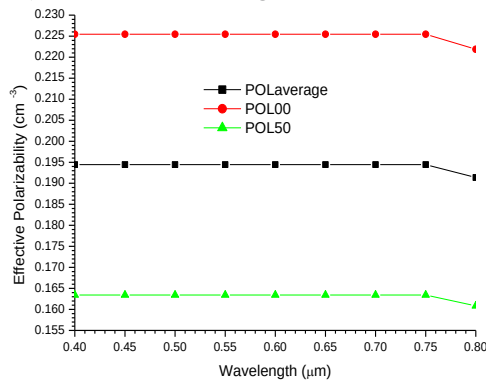


Figure 13: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for SOOT Model 2

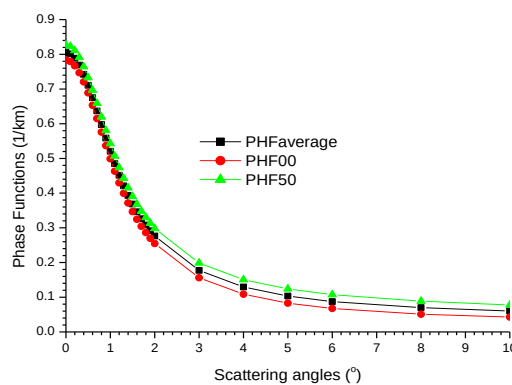


Figure 14: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, SOOT Model 2 at phase angle  $0^\circ$  to  $10^\circ$

- **Figure 13 (SOOT Model 2, average of 0% and 50% RH):** Polarizability decreases from 0.194 at 0.4  $\mu\text{m}$  to 0.191 at 0.8  $\mu\text{m}$ .
- **Figure 14 (SOOT Model 2, average of 0% and 50% RH):** Forward peak = 0.805

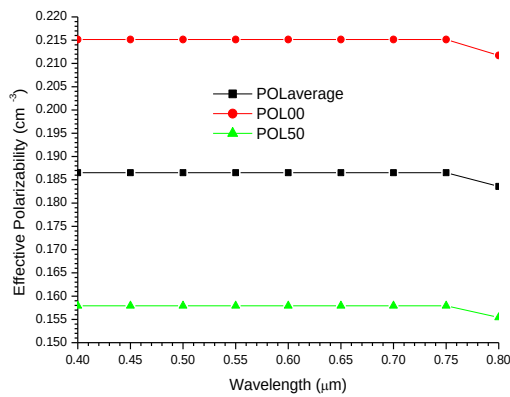


Figure 15: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for SOOT Model 3

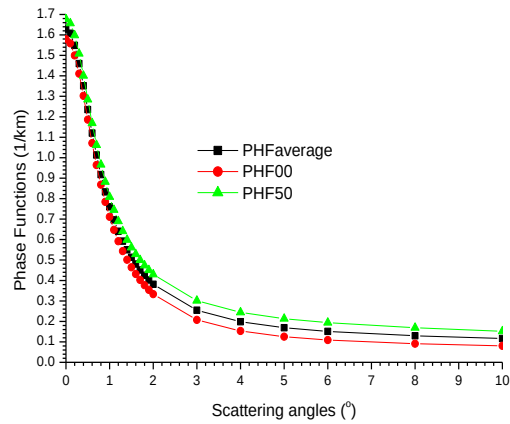


Figure 16: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, SOOT Model 3 at phase angle 0° to 10°

- **Figure 15 (SOOT Model 3, average of 0% and 50% RH):** Polarizability decreases from 0.187 at 0.4  $\mu\text{m}$  to 0.184 at 0.8  $\mu\text{m}$ .
- **Figure 16 (SOOT Model 3, average of 0% and 50% RH):** Forward peak = 1.628, with featureless angular distribution.

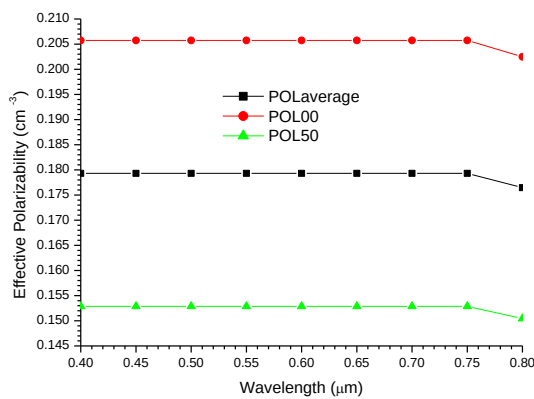


Figure 17: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for SOOT Model 4

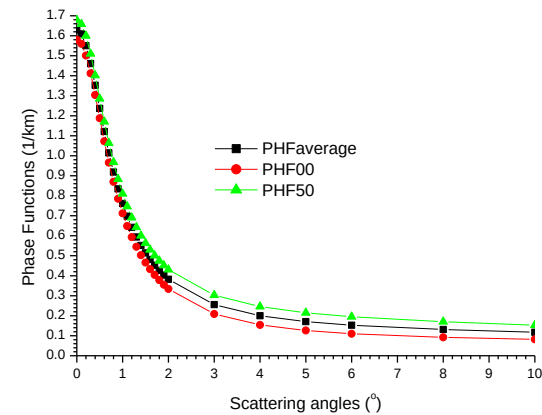


Figure 18: A graph of Phase functions against scattering angles at average 0% RH and 50% RH, SOOT Model 4 at phase angle 0° to 10°

- **Figure 17 (SOOT Model 4, average of 0% and 50% RH):** Polarizability decreases from 0.179 at 0.4  $\mu\text{m}$  to 0.176 at 0.8  $\mu\text{m}$ .
- **Figure 18 (SOOT Model 4, average of 0% and 50% RH):** Forward peak = 1.630.

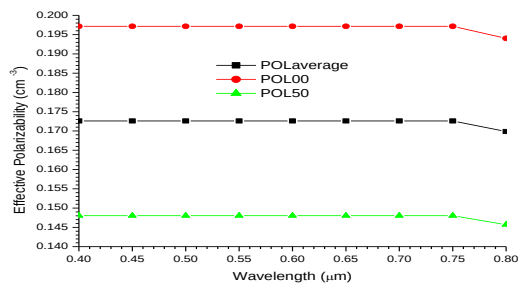


Figure 19: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for SOOT Model 5

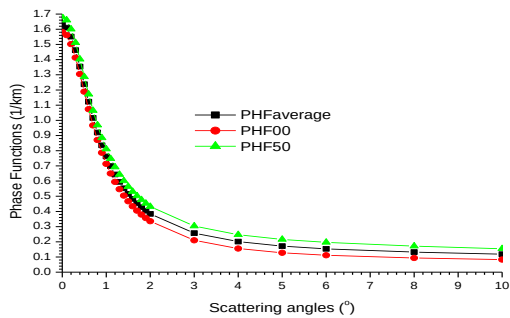


Figure 20: A graph of Phase functions against scattering angles at average 0% RH and 50% RH SOOT Model 5 at phase angle 0° to 10°

- **Figure 19 (SOOT Model 5, average of 0% and 50% RH):** Polarizability decreases from 0.173 at 0.4  $\mu\text{m}$  to 0.170 at 0.8  $\mu\text{m}$ . The 50% RH curve shows marginally higher values due to hygroscopic growth of the coexisting WASO component.
- **Figure 20 (SOOT Model 5, average of 0% and 50% RH):** Forward peak = 1.631, confirming minimal concentration dependence.

**4.2.1 Polarizability:** SOOT models show lower polarizability than WASO due to strong absorption. Values decrease from 0.203 (Model 1) to 0.173 (Model 5) at 0.4  $\mu\text{m}$ . Minimal RH response reflects weak soot hygroscopicity.

**4.2.2 Phase Function:** Suppressed forward scattering (1.626–1.631) compared to INSO and WASO, with remarkably smooth angular distributions lacking oscillations—characteristic of strongly absorbing particles.

#### 4.2.3 4.2.3 SOOT Regression

**Table 5: Ångström Regression Parameters for SOOT Aerosol Models (Average of 0% and 50% RH)**

| Model          | Avg R <sup>2</sup> | Avg Sig (R <sup>2</sup> ) | Avg Alpha | Avg Alpha Sig | Avg Beta (Turbidity) | Avg Beta Sig |
|----------------|--------------------|---------------------------|-----------|---------------|----------------------|--------------|
| <b>Model 1</b> | 0.9987             | 2.99E-11                  | 1.3758    | 2.99E-11      | 0.0993               | 2.00E-14     |
| <b>Model 2</b> | 0.9989             | 1.51E-11                  | 1.3788    | 1.51E-11      | 0.1086               | 1.64E-14     |
| <b>Model 3</b> | 0.9990             | 9.98E-12                  | 1.3815    | 9.98E-12      | 0.1179               | 1.42E-14     |
| <b>Model 4</b> | 0.9992             | 6.33E-12                  | 1.3834    | 6.33E-12      | 0.1272               | 1.15E-14     |
| <b>Model 5</b> | 0.9992             | 4.68E-12                  | 1.3855    | 4.68E-12      | 0.1365               | 1.09E-14     |

**Ångström Exponent:**  $\alpha_a$  increases modestly from 1.376 (Model 1) to 1.386 (Model 5). Minimal  $\alpha$  increase from 0% to 50% RH (1.373→1.379 for Model 1; 1.384→1.387 for Model 5) confirms weak hygroscopicity of soot particles.

#### 4.3 WASO Component Results (Critical Findings)

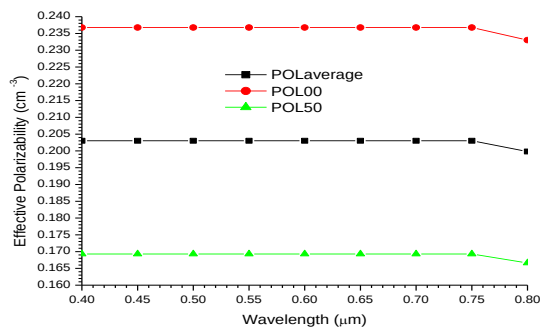


Figure 21: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for WASO Model 1

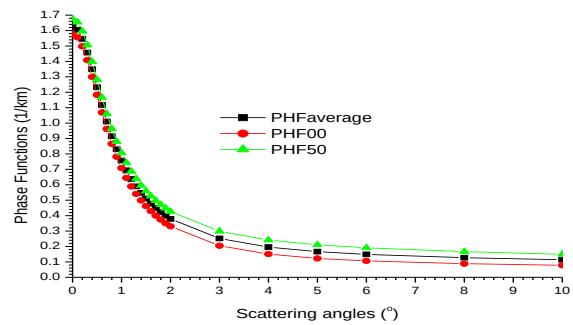


Figure 22: A graph of Phase functions against scattering angles at average 0% RH and 50% RH WASO Model 1 at phase angle 0° to 10°

- **Figure 21 (WASO Model 1, average of 0% and 50% RH):** Polarizability decreases from 0.203 at 0.4  $\mu\text{m}$  to 0.200 at 0.8  $\mu\text{m}$ .
- **Figure 22 (WASO Model 1, average of 0% and 50% RH):** Forward peak = 1.626, smooth angular distribution.

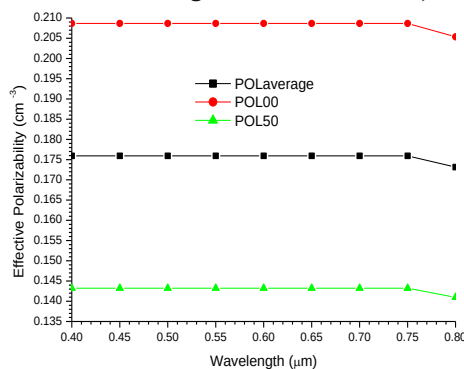


Figure 23: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for WASO Model 2

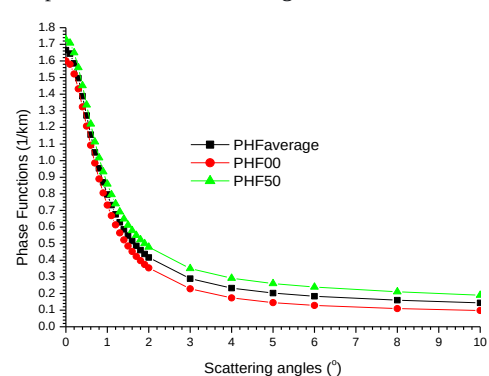


Figure 24: A graph of Phase functions against scattering angles at average 0% RH and 50% RH WASO Model 2 at phase angle 0° to 10°

- **Figure 23 (WASO Model 2, average of 0% and 50% RH):** Polarizability decreases from 0.176 at 0.4  $\mu\text{m}$  to 0.173 at 0.8  $\mu\text{m}$ .
- **Figure 24 (WASO Model 2, average of 0% and 50% RH):** Forward peak = 1.665, developing forward lobe.

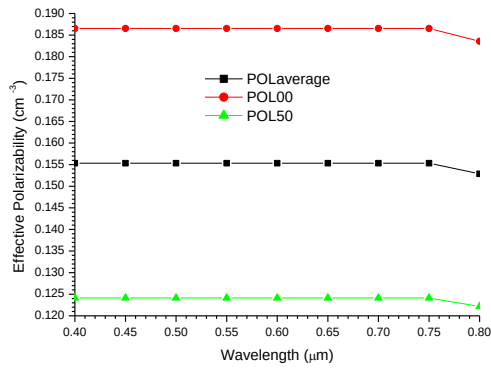


Figure 25: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for WASO Model 3

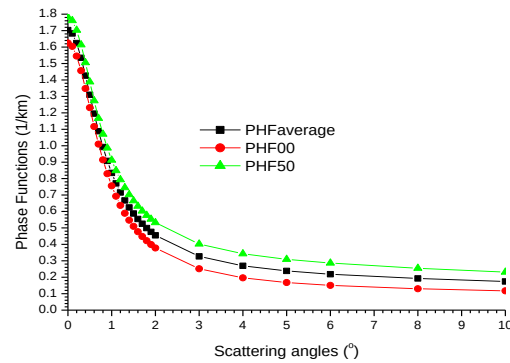


Figure 26: A graph of Phase functions against scattering angles at average 0% RH and 50% RH WASO Model 3 at phase angle 0° to 10°

- **Figure 25 (WASO Model 3, average of 0% and 50% RH):** Polarizability decreases from 0.155 at 0.4  $\mu\text{m}$  to 0.153 at 0.8  $\mu\text{m}$ .
- **Figure 26 (WASO Model 3, average of 0% and 50% RH):** Forward peak = 1.703, weak oscillations appearing.

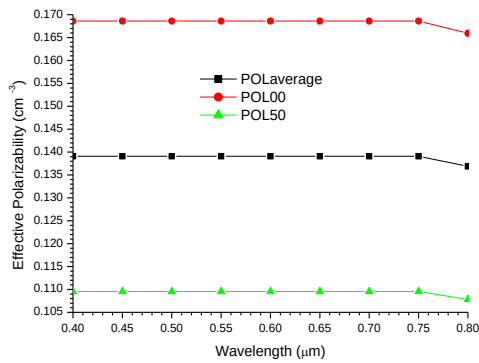


Figure 27: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for WASO Model 4

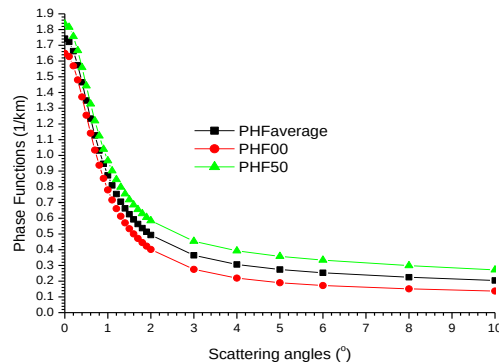


Figure 28: A graph of Phase functions against scattering angles at average 0% RH and 50% RH WASO Model 4 at phase angle 0° to 10°

- **Figure 27 (WASO Model 4, average of 0% and 50% RH):** Polarizability decreases from 0.139 at 0.4  $\mu\text{m}$  to 0.137 at 0.8  $\mu\text{m}$ .
- **Figure 28 (WASO Model 4, average of 0% and 50% RH):** Forward peak = 1.742, clear oscillations in backward hemisphere.

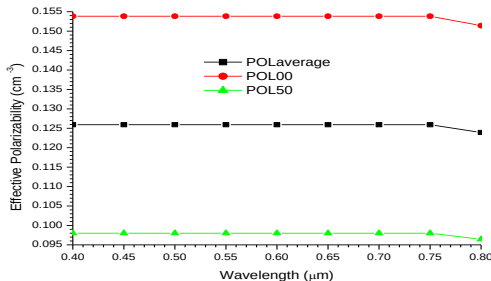


Figure 29: A graph of Effective Polarizability against Wavelength at average 0 and 50 RH for WASO Model 5

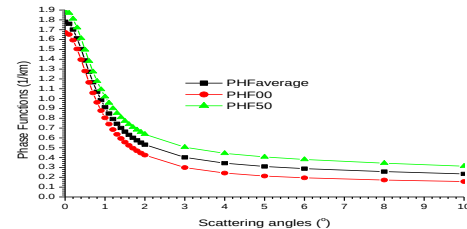


Figure 30: A graph of Phase functions against scattering angles at average 0% RH and 50% RH WASO Model 5 at phase angle 0° to 10°

- **Figure 29 (WASO Model 5, average of 0% and 50% RH):** Polarizability decreases from 0.126 at 0.4  $\mu\text{m}$  to 0.124 at 0.8  $\mu\text{m}$ . The polarizability spectra for WASO show the most pronounced RH dependence, with the 50% RH curve lying significantly above the 0% RH curve, reflecting the strong hygroscopic growth of water-soluble particles.

- **Figure 30 (WASO Model 5, average of 0% and 50% RH):** Forward peak = 1.781, dense Mie oscillations across all angles.

**4.3.1 Polarizability:** WASO models demonstrate the most pronounced RH dependence. At 0.4  $\mu\text{m}$ , polarizability decreases from 0.203 (Model 1) to 0.126 (Model 5). The 50% RH curve lies significantly above the 0% RH curve, reflecting strong hygroscopic growth.

**4.3.2 Phase Function:** Forward scattering enhancement with RH is most pronounced for WASO. Model 5 forward peak increases from 1.781 (average) to 1.785 (50% RH). Side scattering at 90° increases systematically (0.0038→0.0073), with distinct backward-hemisphere oscillations from Model 3 onward.

#### 4.3.3 WASO Regression

**Table 6: Ångström Regression Parameters for WASO Aerosol Models (Average of 0% and 50% RH)**

| Model          | Avg R <sup>2</sup> | Avg Sig (R <sup>2</sup> ) | Avg Alpha | Avg Alpha Sig | Avg Beta (Turbidity) | Avg Beta Sig |
|----------------|--------------------|---------------------------|-----------|---------------|----------------------|--------------|
| <b>Model 1</b> | 0.9987             | 2.99E-11                  | 1.3758    | 2.99E-11      | 0.0993               | 2.00E-14     |
| <b>Model 2</b> | 0.9984             | 5.71E-11                  | 1.3961    | 5.71E-11      | 0.1164               | 8.72E-14     |
| <b>Model 3</b> | 0.9981             | 1.00E-10                  | 1.4115    | 1.00E-10      | 0.1335               | 2.66E-13     |
| <b>Model 4</b> | 0.9978             | 1.45E-10                  | 1.4232    | 1.45E-10      | 0.1507               | 6.40E-13     |
| <b>Model 5</b> | 0.9976             | 1.99E-10                  | 1.4325    | 1.99E-10      | 0.1678               | 1.41E-12     |

A critical finding emerges from comparing WASO Regression. The Ångström exponent ( $\alpha_a$ ) exhibits distinct behavior across WASO concentration gradients:

#### 4.3.4 Ångström Exponent – Non-linear Hygroscopic Response:

**Table 7: Summary of Ångström Exponent ( $\alpha_a$ ) Behavior Across WASO Concentration Gradients Illustrating Non-Monotonic RH Response**

| Model | Concentration | $\alpha_a$ (0% RH) | $\alpha_a$ (50% RH) | $\Delta\alpha_a$ | Behavior      |
|-------|---------------|--------------------|---------------------|------------------|---------------|
| 1     | Baseline      | 1.3729             | 1.3788              | 0.0059           | Increase      |
| 2     | 30%           | 1.3960             | 1.3962              | 0.0001           | Near-constant |
| 3     | 60%           | 1.4141             | 1.4090              | -0.0051          | Decrease      |
| 4     | 90%           | 1.4280             | 1.4183              | -0.0097          | Decrease      |
| 5     | 120%          | 1.4393             | 1.4257              | -0.0136          | Decrease      |

**Explanation:** At low WASO concentrations (Models 1–2), hygroscopic growth increases particle size within the accumulation mode (0.1–1.0  $\mu\text{m}$ ), enhancing spectral dependence and increasing  $\alpha_a$ . At high concentrations (Models 3–5), deliquesced particles grow beyond the accumulation mode into the coarse mode (>1.0  $\mu\text{m}$ ), where scattering becomes wavelength-independent, causing  $\alpha_a$  to decrease—a phenomenon termed "hygroscopic saturation" (Day and Malm, 2001).

#### 4.4 MODIS Validation Results

**Table 8: MODIS-derived Ångström exponent and turbidity coefficients by country**

| Country      | $\alpha_a$ | $\beta$ | R <sup>2</sup> | Dominant Aerosol Type    |
|--------------|------------|---------|----------------|--------------------------|
| South Africa | 2.390      | 0.003   | 0.655          | Biomass burning          |
| USA          | 1.842      | 0.122   | 0.920          | Urban-industrial/biomass |
| Ethiopia     | 1.446      | 0.039   | 0.666          | Mixed biomass/dust       |
| Germany      | 1.271      | 0.030   | 0.997          | Urban-industrial         |
| Serbia       | 1.003      | 0.078   | 0.999          | Mixed urban/dust         |
| India        | 0.847      | 0.061   | 0.810          | Anthropogenic/dust       |
| France       | 0.761      | 0.031   | 0.997          | Marine/urban             |
| Mexico       | 0.627      | 0.015   | 0.467          | Mixed urban/biomass/dust |
| Nigeria      | 0.566      | 0.054   | 0.888          | Dust/biomass             |
| Egypt        | 0.120      | 0.099   | 0.574          | Saharan dust             |

**Comparison with OPAC:** Germany ( $\alpha_a=1.27$ ) matches WASO Models 3–4 ( $\alpha_a=1.41\text{--}1.42$ ) and Ethiopia ( $\alpha_a=1.45$ ) matches WASO Models 2–3, confirming OPAC's validity for anthropogenically-influenced regions. Extreme values (South Africa  $\alpha_a=2.39$ , Egypt  $\alpha_a=0.12$ ) represent aerosol types beyond OPAC's urban-focused component mixtures.

#### 4.5 Synthesis

**Table 9: Four-Parameter Interpretation for Urban Aerosols under Varying Composition and Relative Humidity Conditions**

| Scenario           | Polarizability | Phase Function      | $\alpha_a$ | $\beta$      | Interpretation         |
|--------------------|----------------|---------------------|------------|--------------|------------------------|
| High WASO, low RH  | High           | Strong forward      | 1.44       | High         | Fine-mode dominated    |
| High WASO, high RH | Very high      | Very strong forward | 1.43       | Very high    | Hygroscopic saturation |
| High SOOT          | Low            | Suppressed          | 1.39       | Low-moderate | Absorbing aerosol      |
| High INSO          | Moderate       | Moderate forward    | 1.27       | Moderate     | Dust influence         |

**Key pattern:** Higher polarizability and stronger forward phase function  $\rightarrow$  higher  $\beta$ ;  $\alpha_a$  peaks at medium WASO/low RH, then declines with high RH or dust/soot influence.

#### 4.6 OPAC vs. MODIS Comparison

**Table 10: Validation of OPAC 4.0 Simulations Against MODIS Satellite Data by Country**

| Country      | MODIS Average $\alpha_a$ | OPAC Equivalent                          | Dominant Aerosol Type    | Match Quality           |
|--------------|--------------------------|------------------------------------------|--------------------------|-------------------------|
| Germany      | 1.271                    | WASO Model 3-4 (1.412-1.423)             | Urban-industrial         | Excellent               |
| France       | 0.761                    | INSO Model 5 (1.273)                     | Marine/urban             | Poor (marine influence) |
| India        | 0.847                    | INSO Model 5 (1.273)                     | Mixed anthropogenic/dust | Moderate                |
| Egypt        | 0.12                     | No match (dust-dominated)                | Saharan dust             | N/A                     |
| South Africa | 2.39                     | No match (extreme biomass)               | Biomass burning          | N/A                     |
| USA          | 1.842                    | Between SOOT Model 5 (1.386) and biomass | Mixed urban/biomass      | Moderate                |
| Ethiopia     | 1.446                    | WASO Model 2-3 (1.396-1.412)             | Mixed biomass/dust       | Good                    |
| Serbia       | 1.003                    | Between INSO Model 5 (1.273) and lower   | Mixed urban/dust         | Moderate                |
| Mexico       | 0.627                    | Below INSO Model 5                       | Mixed urban/biomass/dust | Poor                    |
| Nigeria      | 0.566                    | Below INSO Model 5                       | Dust/biomass             | Poor                    |

Germany shows excellent agreement with OPAC WASO Models 3–4 ( $\alpha_a = 1.41\text{--}1.43$ ), confirming the urban-industrial aerosol signature dominated by water-soluble components from fossil fuel combustion (Putaud *et al.*, 2010). Ethiopia ( $\alpha_a = 1.45$ ) matches WASO Models 2–3. South Africa exhibits the highest  $\alpha_a$  (2.39), characteristic of intense biomass burning aerosols from savanna fires (Eck *et al.*, 2003). Egypt exhibits the lowest  $\alpha_a$  (0.12), characteristic of Saharan dust aerosols with large effective radius (Papayannis *et al.*, 2008).

## 5. Conclusions

- Component-specific  $\alpha_a$  behavior:**  $\alpha_a$  systematically decreases with INSO (1.376 $\rightarrow$ 1.273), increases with SOOT (1.376 $\rightarrow$ 1.386), and increases strongly with WASO (1.376 $\rightarrow$ 1.433).
- Non-linear hygroscopic response:**  $\alpha$  increases from 0% to 50% RH at low WASO concentrations but decreases at high concentrations due to hygroscopic saturation and coarse-mode transition—a critical finding for remote sensing applications.
- Phase function enhancement:** Forward scattering at  $0^\circ$  increases with RH across all models, most pronounced for WASO (1.626 $\rightarrow$ 1.785 at Model 5).
- MODIS validation:** OPAC-derived  $\alpha_a$  (1.27–1.43) matches anthropogenically-influenced regions (Germany, Ethiopia), confirming OPAC 4.0 validity. Extreme  $\alpha_a$  values (South Africa 2.39, Egypt 0.12) require additional aerosol components (biomass burning, pure dust) beyond OPAC's urban scope.

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