

The Effect of Varying Aerosol Concentrations on Visibility and Particle Size Distribution in Urban Atmosphere: Validating OPAC Results Using MERRA Satellite Aerosol Data

ABSTRACT

In this work, comparison and validation was carried out using MERRA averaged Angstrom Exponents (α) and averaged visibilities for ten urban countries against simulated OPAC urban components. The simulations of OPAC were performed at 0%, 50%, 70%, 80%, 90%, 95%, 98% and 99% relative humidities (RH) while the MERRA satellite data were extracted at an average of 78% relative humidity. results based on α values showed that OPAC was only able to simulate well the insoluble and soot aerosol particle size distributions of India's urban atmosphere such that it had approximately the same insoluble and soot aerosol particle size distributions as India. For the rest of the nine countries (USA, Brazil, Indonesia, China, Japan, Mexico, Nigeria, Pakistan and Ethiopia), OPAC was seen to either overestimate or underestimate water soluble, insoluble and soot aerosol particle size distributions. For visibility and water soluble or insoluble aerosol concentrations, OPAC showed good simulated values that were approximately the same as those found within Ethiopia's and Japan's urban atmosphere. OPAC also had visibility value and insoluble aerosol concentration that were approximately the same as those found within China's atmosphere. For all other countries (India, USA, Brazil, Indonesia, Mexico, Nigeria and Pakistan) OPAC either overestimated their visibilities and underestimated their water soluble, insoluble and soot aerosol concentrations or vice versa. With both OPAC and MERRA, the presence of fine mode aerosol particles in an urban atmosphere was established with α values > 1 . But MERRA also showed otherwise for three countries (Nigeria, Ethiopia and Pakistan) that had α values < 1 which implied the presence of coarse mode aerosol particles. The relationship between visibility and α satisfied the direct power law for OPAC, for MERRA the relationship approximately satisfied the power law for most of the countries.

Keywords: Aerosol Concentration, MERRA, OPAC, Particle Size Distribution, Water soluble aerosols, Insoluble aerosols, Soot, Visibility.

1. INTRODUCTION

The Earth's climate characteristics are changing daily and the demand for more spatially and temporally detailed weather and climate information is on the rise [1]–[3]. It is this requirement that has led to the application of climate models and simulation packages in order to simulate past and future climate systems' evolution [4]. These simulations and models are meant to add value through a more precise representation of different atmospheric features through successful simulation of climate characteristics that are physically out of reach [4]. One of such simulation packages is the Optical Properties of Aerosols and Clouds (OPAC 4.0).

The OPAC 4.0 is a software package that provides optical properties of aerosol particles in the solar and terrestrial range [5]. The properties of aerosol particles are highly variable in time and space and in most cases the actual aerosol properties are not known, this makes it difficult to model aerosols in detail. OPAC achieves this by using data sets of internally mixed aerosol components at varying concentrations and particle size distributions. The aerosol components can be externally mixed making it possible to simulate optical properties of atmospheric particulate constituents that are relevant for climate calculations. With OPAC it is possible to define aerosol mixtures for calculation of optical properties. Aerosol height distribution can also be specified during the data input stage of the simulation. For comparison and validation purposes, results from models and simulation packages can be compared to real time satellite aerosol data like those of the Moderate Resolution Imaging Spectroradiometer (MODIS) and Modern Era Retrospective Analysis for Research and Applications (MERRA) provided by the NASA EarthData network.

This work is one of two parts, here the aerosol optical properties of extinction coefficient and angstrom exponent (α) were extracted from MERRA to compare and validate results obtained with the results obtained from the analysis carried out on the effects of varying aerosol concentrations on visibility and particle size distribution in urban atmosphere using OPAC by Abdulkarim and Tijjani [6]. Hess [5] indicated that the OPAC is a software whose given aerosol components are adapted to average conditions and their size distributions might not necessarily apply to actual conditions [5]. Therefore

simulations using OPAC 4.0 must be compared and validated against real time satellite observations or other models and datasets in order to be confident in the models' predictions thus MERRA was used.

The Modern Era Retrospective Analysis for Research and Applications (MERRA) is an aerosol data assimilation from the NASA global modeling and assimilation office GMOA [7]. The MERRA assimilates long term global reanalysis of space-based observations of aerosols and represents their interactions with other physical processes in the climate system. MERRA also includes bias-corrected Aerosol Optical Depth (AOD) from Advanced Very High Resolution Radiometer (AVHRR) instruments AOD retrievals from the Multiangle Image SpectroRadiometer (MISR) over bright surfaces [8] and Aerosol Robotic Network (AERONET) direct measurements of AOD level 2 [9,7]. It was generated with version 5.2.0 of the Goddard Earth Observing System (GEOS) atmospheric model and Data Assimilation System (DAS). The MERRA also used the best features available from models and real world biased observations to produce 4D gridded outputs that combine model continuity. This work chose MERRA because one of the performance drivers of the GEOS DAS products has historically been temperature and moisture. Although this work did not include temperature components, it considered atmospheric moisture in terms of relative humidity. MERRA data were used to validate results obtained using OPAC in the work presented as first progress by Abdulkarim and Tijjani [6].

2. METHODS

This research considered ten countries with their extracted total Angstrom Exponents (α) at and total aerosol extinctions at 550nm. MERRA provides time series area averaged map for Nigeria (8W 9S 8E 9N) through 12 years (1997 to 2009), for China (104W 35S 104E 35N), India (78W 20S 78E 20N), USA (95W 37S 95E 37N) through 13 years (1997 to 2000) and Brazil (131.51W 14S 51E 14N), Ethiopia (40W 9S 40E 9N), Japan (138W 36S 138E 36N), Pakistan (180W 90S 180E 90N), Indonesia (113W 0S 113E 0N) and Mexico (102W 23S 102E 23N) through 37 years. Data for China, India, USA, Brazil, Ethiopia, Japan, Pakistan, Indonesia and Mexico were scaled down to 12 years (1997 to 2009) for uniformity of data.

These ten countries were chosen based on their Worldometer ranking as the most populous countries. This research chooses the world's most populous countries because it is expected that more populous urban regions will experience the most atmospheric pollutions. From downloaded MERRA data total

extinction for each of the ten countries considered were averaged. The averaged extinctions were calculated for each country based on the Koschmieder formula that relates extinction coefficient to visibility. Visibility was calculated using the averaged total extinction coefficients at 550nm. The angstrom exponent (α) values were also averaged for each country. A time series analysis was performed on data for each country to show that the model fits the observed data using angstrom exponent (α) level and seasonality effects. From the OPAC 4.0 analysis, visibility results for each model of the WASO (water soluble), INSO (insoluble) and SOOT component at 0- 99% relative humidity were extracted at 550nm wavelength. Extraction of Angstrom parameters at 550nm, 0- 99% relative humidity were also carried out from the OPAC simulation for all models. These extracted Angstrom parameters are averaged for better comparison and validation with the averaged Angstrom parameters extracted from MERRA for each country.

3. RESULTS AND DISCUSSIONS

This section presents results obtained from analysis carried out on MERRA Angstrom Exponent (α) and visibility data. The results were compared to the α values obtained from OPAC simulations for three components of WASO (water soluble), INSO (insoluble) and SOOT. Level and seasonality effects on MERRA Angstrom Exponent (α) are also presented. While varying OPAC aerosol concentrations for WASO, INSO and SOOT aerosol components, five models (Model 1 to Model 5) were considered for each component. For this work, one model (Model 1) is presented for each component. For the OPAC INSO component, Model 1 is selected because it presents the highest Angstrom Exponent (α) values at all the eight relative humidities considered. The Angstrom Exponent (α) also shows the same behavior across relative humidities for the rest of the models (Model 2 to Model 5). Additionally for the OPAC WASO component Model 1 is selected likewise for the SOOT component. This selection is made because all five models for both components present constant Angstrom Exponent (α) values as the aerosol concentrations increased from Model 1 to Model 5.

Table 1: Model Statistics and Exponential Smoothing Model Parameters β (Level) and δ (Season) from time series forecast for Angstrom Exponent (α) of ten most populous countries

Model Statistics			Exponential Smoothing Model Parameters		
Country	R ²	Sig.	Model	Estimate	Sig.
Brazil	0.89766245	0.00024485	β (Level)	0.59969641	0.00000000
			δ (Season)	0.00002629	0.99910854
China	0.83607637	0.09059628	β (Level)	0.30015856	0.00000013
			δ (Season)	0.00000467	0.99992872
Ethiopia	0.85092047	0.01321676	β (Level)	0.70023586	0.00000000
			δ (Season)	0.00002272	0.99943411
India	0.94534777	0.04181863	β (Level)	0.20030091	0.00002207
			δ (Season)	0.00001255	0.99967043
Indonesia	0.85363950	0.00017391	β (Level)	0.99900003	0.00000000
			δ (Season)	0.00007494	0.99999368
Japan	0.81356568	0.00004011	β (Level)	0.75999153	0.00000000
			δ (Season)	0.37766882	0.00001940
Mexico	0.89168038	0.00074289	β (Level)	0.78651862	0.00000000
			δ (Season)	0.50619887	0.00006907
Nigeria	0.85426974	0.00624526	β (Level)	0.09982499	0.01028376
			δ (Season)	0.00004547	0.99908184
Pakistan	0.90388185	0.00018597	β (Level)	0.70011726	0.00000000
			δ (Season)	0.00005313	0.99882640
USA	0.88163753	0.29104558	β (Level)	0.49989803	0.00000000
			δ (Season)	0.00006009	0.99930835

Table 1 presents the results of a time series analysis forecast for MERRA extracted angstrom exponent values for ten countries. The level of the angstrom exponent is signified by β (Level) while the seasonality effects in terms of particle size distributions is signified by δ (Season). For all countries considered, the R² values show that the model fits the observed data well. R² values of 90% for Brazil, Mexico and Pakistan shows statistical significance. This indicates that 90% of the data fit the model and 90% of the variation in the Angstrom Exponent (α) is explained by the season and time of the year. Ethiopia, Indonesia and Nigeria have R² values of 84%, 94%, 81% and 88% respectively. The R² values for Brazil, Mexico and Pakistan show the best fit over R² values for China and the USA which show no statistical significance.

Table 2: MERRA extracted average Visibility and Angstrom Exponent (α) at 550nm and 78% RH for Ten Countries

Country	Calculated, Averaged MERRA Visibility at 550nm and 78% RH	Averaged MERRA Angstrom Exponent (α) at 550nm and 78% RH
Brazil	29.56391794	1.342156919
China	14.56502993	1.008823473
India	13.50291219	1.045158614
Indonesia	21.90235478	1.215147471
Nigeria	8.645397543	0.508421448
Pakistan	13.12494174	0.673450947
Usa	26.70555392	1.198213718
Ethiopia	20.24253496	0.804081727
Japan	15.30235656	1.125407922
Mexico	26.13860893	1.338033052

3.1 β (LEVEL) EFFECTS ON ANGSTROM EXPONENT (α)

From table 1, it is observed that Brazil, China, India, Indonesia, United States, Japan and Mexico have α values greater than 1, signifying the presence of fine mode aerosol particles within the atmosphere at 78% relative humidity. From Table 2 it can be seen that there is a statistical significance in the estimates of the β (Level) values, this indicates that the level of dominance of fine mode aerosol particles within the atmosphere has not changed much over the years for those countries at that given relative humidity. Table 1 also shows Nigeria Pakistan and Ethiopia having α values of less than 1 indicating the presence of coarse mode aerosol particles within the atmosphere. From table 2, there is a statistical significance in the estimates of the β (Level) values and this indicates that the level of dominance for coarse mode aerosols within the atmosphere of the above mention three countries at 78% RH has not changed over the years.

3.2 δ (SEASON) EFFECTS ON ANGSTROM EXPONENT (α)

Considering the seasonality effects of Angstrom Exponent (α) on the particle size distribution through the span of 12 years, Table 1 shows that the seasonality effects for all the countries with the exception of Japan and Mexico show no statistical significance. This non significance can be attributed to fluctuations in α and the fluctuations are not due to actual changes in the particle size distribution but could be attributed to fluctuations found within the model. Normal seasonality forecast for α and constant particle size distributions dominated by fine mode aerosol particles are observed for the countries Brazil, USA, China, Indonesia and India. Normal seasonality forecast for α and particle size distribution and the dominance of coarse mode aerosol particles are also observed for the countries Nigeria, Ethiopia and Pakistan. Since fluctuations are not due to actual changes in the particle size distribution, the distribution is assumed to remain within previous forecast values.

Table 3: OPAC Angstrom Exponent (α) at 550nm and eight Relative Humidities for WASO, INSO and SOOT aerosol components

		OPAC Angstrom Exponent (α)							
Comp	Model	0%	50%	70%	80%	90%	95%	98%	99%
Inso									
	Model 1	1.32000	1.31500	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 2	1.30500	1.30500	1.29500	1.28000	1.23000	1.16000	1.04200	0.95150
	Model 3	1.29000	1.30000	1.29000	1.27500	1.23000	1.15900	1.04250	0.95100
	Model 4	1.28500	1.29000	1.28000	1.26500	1.22500	1.15300	1.03600	0.95050
	Model 5	1.27000	1.28500	1.27500	1.26000	1.21500	1.15200	1.03500	0.95050
Waso									
	Model 1	1.32000	1.32000	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 2	1.32000	1.32000	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 3	1.32000	1.32000	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 4	1.32000	1.32000	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 5	1.32000	1.32000	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
Soot									
	Model 1	1.32000	1.31500	1.30500	1.28500	1.23500	1.16500	1.04250	0.95650
	Model 2	1.32000	1.31500	1.30500	1.29000	1.23500	1.16500	1.04250	0.95750
	Model 3	1.32000	1.31500	1.30500	1.29000	1.23500	1.16500	1.04250	0.95750
	Model 4	1.32000	1.31500	1.30500	1.29000	1.23500	1.16500	1.04250	0.95750
	Model 5	1.32000	1.31500	1.30500	1.29000	1.23500	1.16500	1.04250	0.95750

From Table 3, the OPAC Angstrom Exponent (α) decreases as INSO aerosol concentration increases from Model 1 to Model 5 indicating an increase in particle size distribution. Angstrom Exponent (α) also shows a decrease as atmospheric relative humidity (RH) increases from 0-99% RH. This indicates that from 0-99% (RH), the aerosol particles are increasing in size and they reach their largest size at saturation point as they absorb atmospheric moisture this aerosol particle size increase is attributed to the water soluble component of the mixture. The WASO component of the OPAC aerosol mixture also shows decreasing Angstrom Exponent (α) values as relative humidity increases indicating aerosol particle growth. At each relative humidity however, Angstrom Exponent (α) is constant indicating that particle size distribution does not change for a particular relative humidity as WASO concentration is increased across the five models. The SOOT OPAC aerosol component shows similar behavior as the WASO component having constant Angstrom Exponent (α) across the five models. For all three components of INSO,

WASO and SOOT the total aerosol mixture is dominated by fine mode aerosol particles from 0-98% relative humidity, for WASO component, these particles became more coarse as RH increased.

Table 4: OPAC Visibilities at 550nm and eight Relative Humidities for WASO, INSO and SOOT aerosol components

		OPAC Visibility (Km)							
Comp	Model	0%	50%	70%	80%	90%	95%	98%	99%
Inso									
	Model 1	20.15456	12.77179	12.77179	11.07901	8.33227	5.96887	3.76879	2.77054
	Model 2	19.97957	12.70130	12.70130	11.02593	8.30221	5.95343	3.76154	2.76662
	Model 3	19.80760	12.63158	12.63158	10.97335	8.27236	5.93807	3.75793	2.76466
	Model 4	19.63855	12.56262	12.56262	10.92127	8.24273	5.92279	3.75072	2.76076
	Model 5	19.47237	12.49441	12.49441	10.86969	8.21331	5.90758	3.74354	2.75687
Waso									
	Model 1	20.07183	12.70542	12.70542	11.01972	8.28463	5.93177	3.74354	2.75299
	Model 2	19.98978	12.63974	12.63974	10.96105	8.23579	5.89512	3.71863	2.73375
	Model 3	19.91853	12.57474	12.57474	10.89997	8.18924	5.85980	3.69754	2.71667
	Model 4	19.83773	12.51039	12.51039	10.84257	8.14152	5.82403	3.67324	2.69793
	Model 5	19.75758	12.45067	12.45067	10.78577	8.09603	5.78870	3.64925	2.68129
Soot									
	Model 1	20.04098	12.72609	12.72609	11.04461	8.31279	5.95887	3.76516	2.76858
	Model 2	19.92868	12.68071	12.68071	11.01041	8.29341	5.94891	3.76154	2.76662
	Model 3	19.81763	12.63158	12.63158	10.97643	8.27411	5.93897	3.75793	2.76466
	Model 4	19.70781	12.58687	12.58687	10.94266	8.25491	5.92907	3.75432	2.76271
	Model 5	19.59920	12.54248	12.54248	10.90605	8.23579	5.91920	3.75072	2.75882

From Table 4, OPAC visibilities at 550nm were considered. It is seen that as INSO aerosol concentration was increased across models, visibility decreased. This happens across all the models at each relative humidity such that visibility is highest at the lowest INSO concentration and lowest at the highest INSO concentration. As relative humidity increased for each model visibility is seen to decrease. The same is seen to apply for both the WASO and SOOT components. Comparison between α values from Table 3 and visibility values from Table 4 showed that for the INSO component increasing aerosol concentration across models decreased visibility and increasing RH increased aerosol particle size also decreasing visibility. For WASO and SOOT components, constant aerosol concentration across models decreased visibility and increasing RH also decreased visibility. However, INSO component shows overall higher visibility values across models as compared to WASO and SOOT components.

3.3 WASO (WATER SOLUBLE) AEROSOL COMPONENT

The Angstrom Exponents (α) for OPAC WASO aerosol component at eight relative humidities (0-99%) were compared against the MERRA averaged Angstrom Exponents (α) for ten countries. The visibilities at the said eight relative humidities were also compared to the MERRA calculated visibility values for the ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India). Five models were considered for the OPAC WASO component but only the first model (Mode 1) is presented in this section for both Angstrom Exponent (α) and visibility respectively.

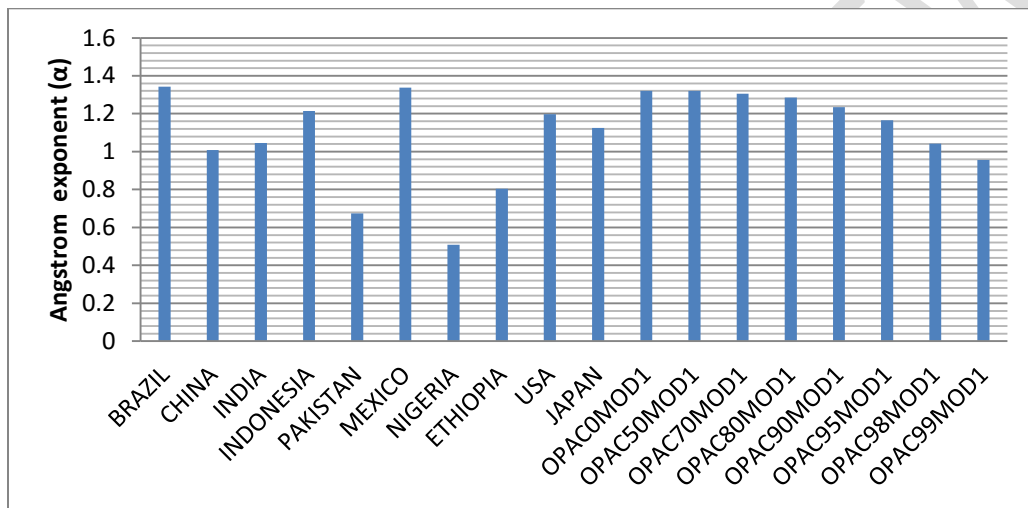


Fig 1. Histogram plot for OPAC WASO Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

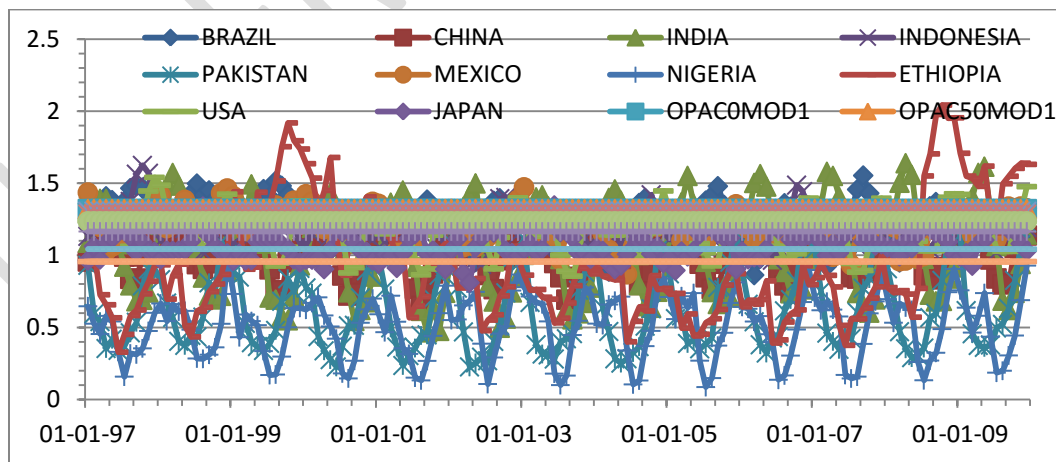


Fig 2. Angstrom Exponent (α) plot for OPAC WASO Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

Fig. 1 and 2 show an Angstrom Exponent (α) plot for ten countries and OPAC WASO aerosol component at eight relative humidities (0-99%). Three countries (Nigeria, Pakistan and Ethiopia) have Angstrom Exponent (α) values < 1 which indicates that their atmosphere is composed of coarse mode aerosol particles with the coarsest particles found within Nigeria, followed by Pakistan then Ethiopia. The α values for these countries are less than OPAC α values for WASO aerosol component.

It can also be seen that the atmospheric aerosols for china have α values >1 indicative of fine mode particles, this value is less than OPAC α values at 0-98% RH but greater than OPAC α values at 99% RH. This indicates that china has less water soluble aerosol particles within its atmosphere than those of OPAC at 0-98% RH and more water soluble aerosol particles than those of OPAC at 99% RH. The α value for India indicate the dominance of fine mode particles. The α values for India are less than the α values for OPAC at 0-95% RH but greater than the α values for OPAC at 98-99% RH. This indicates that India has atmospheric aerosol particles that are less water soluble than OPAC aerosol particles at 0-95% RH but more soluble than OPAC atmospheric aerosols at 98-99% RH. Japan is mostly dominated by fine mode atmospheric aerosol particles. Its particles are less water soluble than those of OPAC at 0-95% RH but more water soluble than those of OPAC at 98-99% RH because Japans α values are greater than OPAC α values at 98-99% RH and less than OPAC α values at 0-95% RH. Additionally, Indonesia, USA, Brazil and Mexico have their atmospheric aerosols dominated by fine mode aerosol particles. Indonesia and USA however have their atmospheric aerosol α values $<$ than OPAC α values at 0-80% RH but $>$ than OPAC α values at 90-99% RH. Thus Indonesia and USA have more water soluble particles than those of OPAC at 90-99% RH and less water soluble particles than those of OPAC at 0-80% RH. Brazil and Mexico have their atmospheric aerosol α values $>$ than OPAC α values at all relative humidities meaning they contain greater water soluble aerosols within their atmosphere.

In summary, it is seen that Nigeria Ethiopia and Pakistan have coarse mode aerosol particles and these particles are coarser than those found within the OPAC WASO component. On the other hand, the seven countries (USA, Brazil, Indonesia, India, Mexico, Japan and China) have fine mode aerosol particles. OPAC at lower relative humidities has finer aerosol particles than the above mentioned seven countries. Additionally, OPAC at higher relative humidities has aerosol particles that are not as fine as those found

within the atmospheres of the seven countries. From α values, all ten countries have been observed to have lower water soluble aerosol particle size distributions within their atmospheres when compared to OPAC at lower relative humidities and higher water soluble aerosol particle size distributions when compared to OPAC at higher relative humidities.

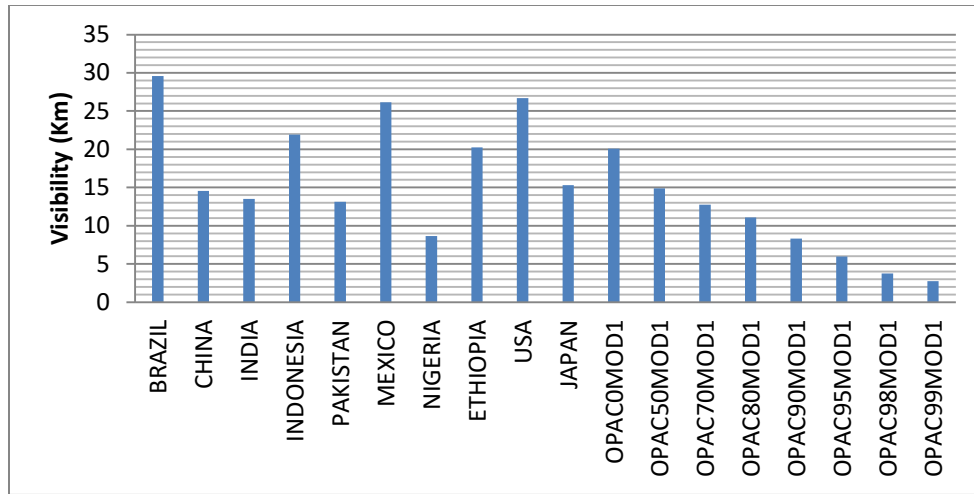


Fig 3. Histogram plot for OPAC WASO Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

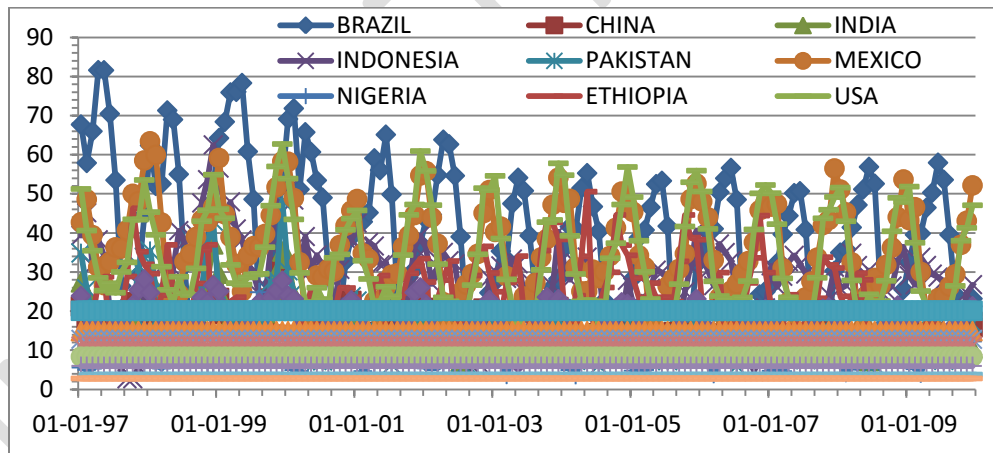


Fig 4. Visibility plot for OPAC WASO Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and Indian)

Visibility for ten countries is calculated from MERRA supplied total extinction coefficient at 550nm wavelength. These visibility plots can be seen from Fig. 3 and 4 where they are compared with OPAC WASO aerosol component visibilities at eight relative humidities (0-99%). Out of the ten countries, Nigeria

has the lowest visibility value and this visibility value is lower than OPAC WASO component visibility value at 0-80% RH but higher than OPAC WASO component visibility value at 90-99% RH. India and Pakistan show higher visibility values than OPAC visibility at 70-99% RH and lower visibility values than OPAC visibility at 0-50% RH. China atmospheric visibility is however found to be lower than OPAC WASO visibility at 0-50% RH but higher than OPAC WASO visibility at 70-99% RH. Japan shows atmospheric visibility approximately the same as that of OPAC WASO visibility at 50% RH. It also shows a visibility value lower than OPAC visibility at 0%RH and higher visibility value than OPAC visibility at 70-99% RH. The four countries Brazil, Indonesia, Mexico and USA have their visibility values higher than OPAC WASO visibility values at all the eight relative humidities. Ethiopia on the other hand has visibility values equivalent to OPAC WASO visibility value at 0% RH but greater visibility value than OPAC WASO visibility values at 50-99% RH.

In summary, at lower relative humidities, OPAC was seen to have lower water soluble aerosol concentrations and higher visibilities than the countries Nigeria, India, Pakistan and China but at higher relative humidities OPAC had higher water soluble aerosol concentrations and lower visibilities than the said countries above. As compared against the country Japan, at lower relative humidities OPAC had approximately the same water soluble concentration and approximately the same visibility values than the country Japan. At higher relative humidities however, OPAC had higher water soluble aerosol concentrations and lower visibilities than those of Japan. OPAC at all relative humidities was seen to have higher water soluble aerosol concentrations and lower visibilities than the countries Brazil, Indonesia, Mexico and the USA. Also OPAC at the lowest relative humidity of 0% had approximately the same water soluble aerosol concentration and visibility values with the country Ethiopia but showed higher aerosol concentrations and lower visibility values at relative humidity of 50-99% than Ethiopia.

3.4 INSO AEROSOL COMPONENT

This section presents plots for OPAC Angstrom Exponent (α) and MERRA averaged exponents for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India). OPAC visibility plots and MERRA calculated and averaged visibilities are also presented. From the five OPAC INSO aerosol component models for Angstrom Exponent (α) and visibility, Model 1 was considered.

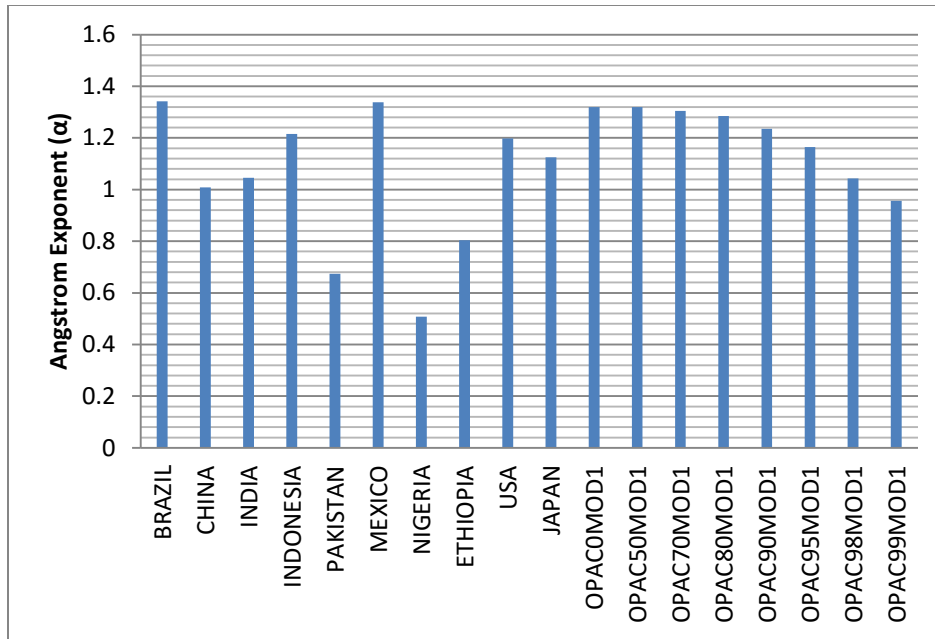


Fig 5. Histogram plot for OPAC INSO Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

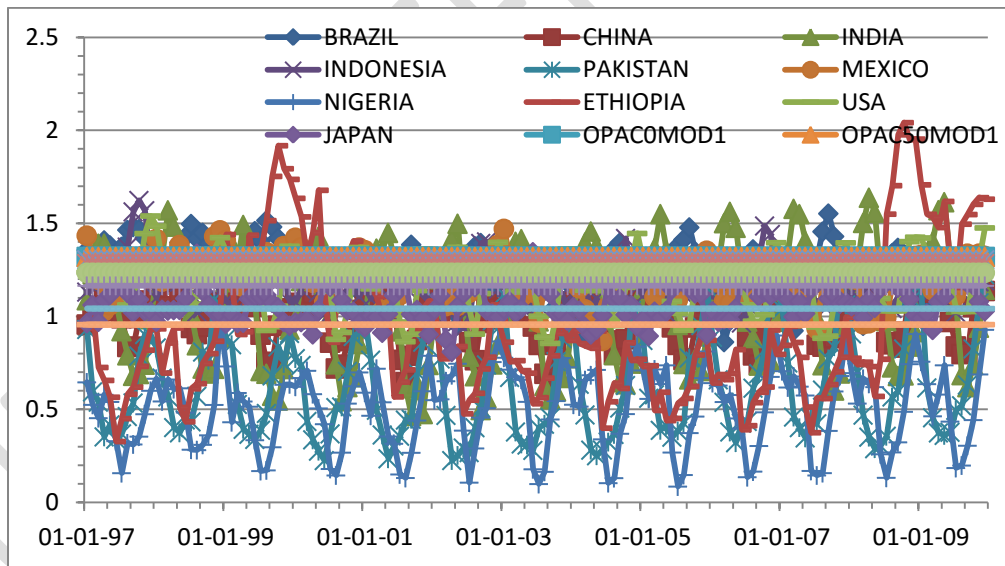


Fig 6. Angstrom Exponent (α) plot for OPAC INSO Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

The INSO OPAC aerosol component at eight relative humidities was compared to those of ten countries and these plots are presented in Fig. 5 and 6 for Angstrom Exponent (α). Three countries (Nigeria, Pakistan and Ethiopia) have Angstrom Exponent (α) values < 1 which indicates that their atmosphere is composed of coarse mode aerosol particles with the coarsest particles found within Nigeria, followed by Pakistan then Ethiopia. The α values for these countries are less than OPAC α values for INSO aerosol component. It can also be seen that the atmospheric aerosols for china have α values > 1 indicative of fine mode particles, this value is less than OPAC α values at 0-98% RH but greater than OPAC α values at 99% RH. This indicates that china has less insoluble aerosol particles within its atmosphere than those of OPAC at 0-98% RH and more insoluble aerosol particles than those of OPAC at 99% RH.

The α value for India indicate the dominance of fine mode particles. The α values for India were approximately the same with those of OPAC at 98% RH but greater than the α values for OPAC at 99% RH. Japan is mostly dominated by fine mode atmospheric aerosol particles. Its particles are less insoluble than those of OPAC at 0-95% RH but more insoluble than those of OPAC at 98-99% RH because Japans α values are greater than OPAC α values at 98-99% RH and less than OPAC α values at 0-95% RH. Additionally, Indonesia, USA, Brazil and Mexico have their atmospheric aerosols dominated by fine mode aerosol particles. Indonesia and USA have their atmospheric aerosol α values $<$ than OPAC α values at 0-90% RH but $>$ than OPAC α values at 95-99% RH. Thus Indonesia and USA have more insoluble particles than those of OPAC at 95-99% RH and less insoluble aerosol particles than those of OPAC at 0-90% RH. Brazil and Mexico have their atmospheric aerosol α values $>$ than OPAC α values at all relative humidities meaning they contain greater insoluble aerosols within their atmosphere. Angstrom Exponent (α) for the Indian atmosphere show dominance of fine mode particles and it was also observed that the α values for India were approximately the same as the α values for OPAC INSO aerosol component at 98% RH but greater than OPAC α values at 99% RH. This implies that OPAC INSO aerosol component at 98% RH had approximately the same insoluble aerosol particle size distributions as the Indian atmosphere.

In summary, OPAC INSO aerosol component at 98% RH simulates well the particle size distribution for insoluble aerosols found within the Indian urban atmosphere. None the less OPAC appears to have lower insoluble aerosol particle size distribution at 99% RH than those found within the Indian atmosphere.

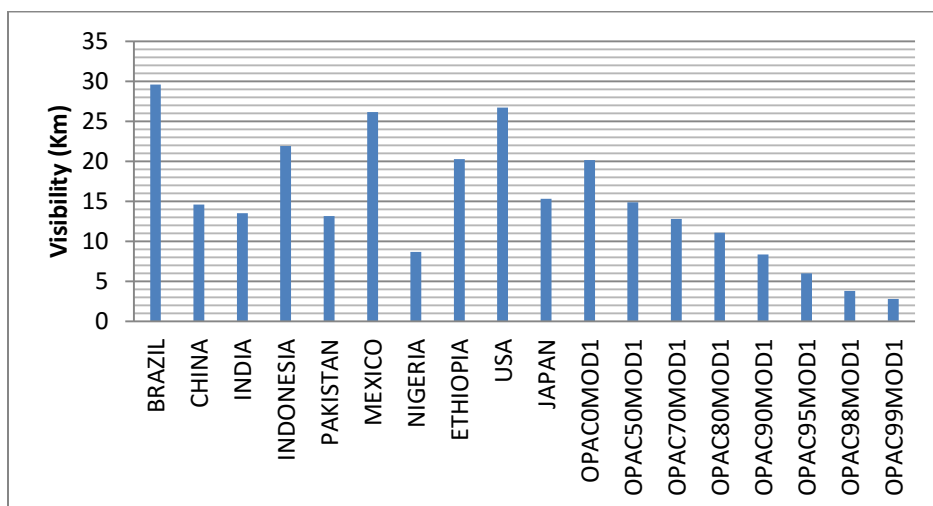


Fig 7. Histogram plot for OPAC INSO Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

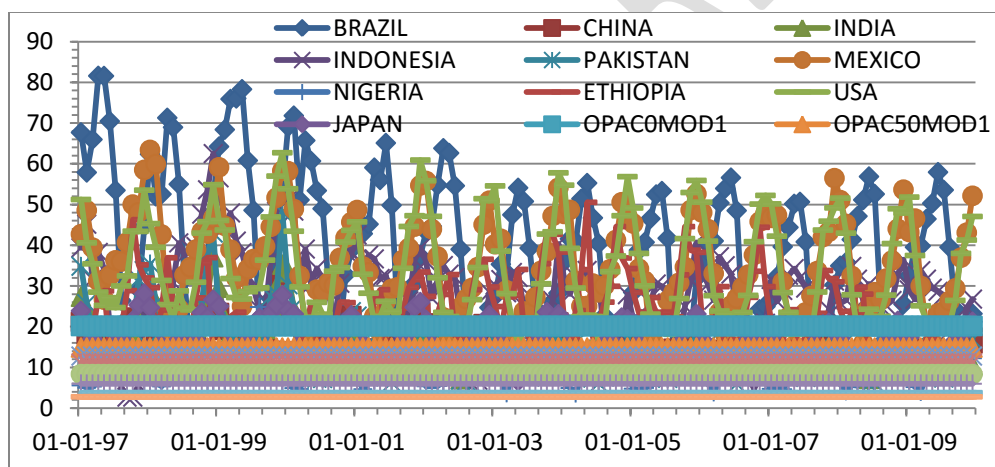


Fig 8. Visibility plot for OPAC INSO Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

Fig. 7 and 8 present visibility comparisons between OPAC INSO aerosol component visibilities at eight relative humidities and MERRA calculated and averaged visibilities. The visibility comparisons for INSO OPAC component and MERRA visibility values for nine countries are similar to visibility comparisons presented under the section for WASO aerosol component. An exception is seen for the country China. China's atmospheric visibility is observed to be approximately the same as OPAC INSO aerosol component visibility at 50% RH and greater than OPAC INSO aerosol component visibility at 70-99% RH.

In summary, Nigeria has lower atmospheric visibility due to higher insoluble aerosol concentrations than those of OPAC at low RH but higher atmospheric visibility due to lower insoluble aerosol concentrations at higher relative humidity. India and Pakistan have higher visibility due to lower insoluble aerosol concentrations at low relative humidity and lower visibility due to higher insoluble aerosol concentrations at high relative humidity. Japan and Ethiopia were seen to have approximately the same visibility and the same insoluble aerosol concentrations at low relative humidity. The two countries have high visibility and low insoluble aerosol concentrations at high relative humidity. USA, Mexico, Brazil and Indonesia however have higher visibility and lower insoluble aerosol concentrations than all OPAC visibilities at all relative humidities. OPAC simulates China's atmospheric visibility well at 50% relative humidity but overestimates the visibilities and presents lower insoluble aerosol concentrations at higher relative humidities.

3.5 SOOT AEROSOL COMPONENT

The Angstrom Exponents (α) for OPAC SOOT aerosol components at eight relative humidities (0-99%) were compared against the MERRA averaged Angstrom Exponents (α) for ten countries. Also OPAC SOOT aerosol component visibilities at the said eight relative humidities were compared to the MERRA calculated visibility values for the ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India). Five models were considered for the OPAC SOOT component but only the first model (Mode 1) is presented in this section for both Angstrom Exponent (α) and visibility respectively.

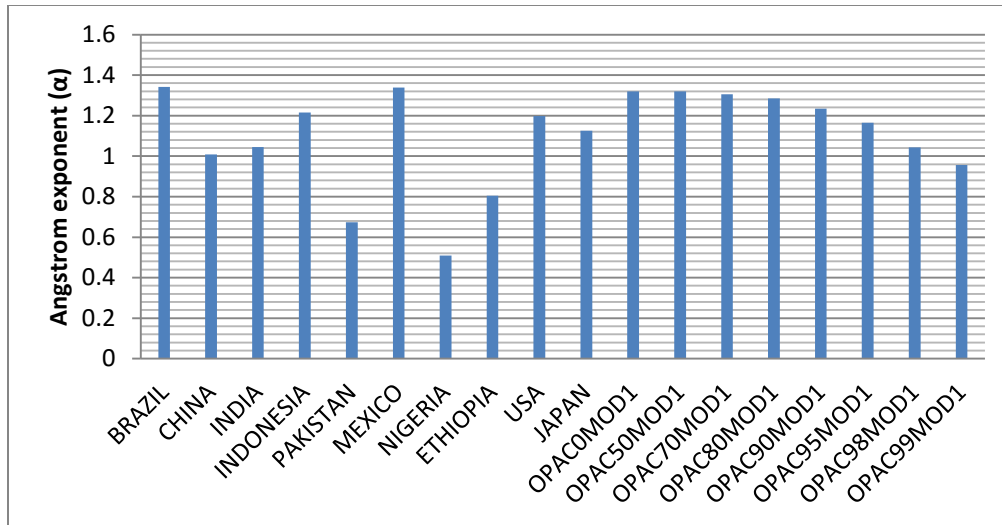


Fig 9. Histogram plot for OPAC SOOT Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

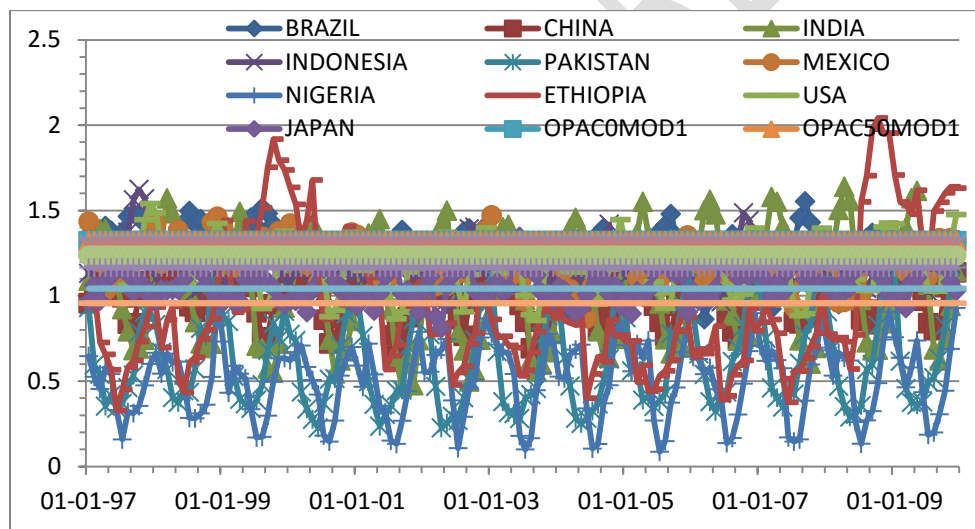


Fig 10. Angstrom Exponent (α) plot for OPAC SOOT Model 1 Angstrom Exponent (α) and MERRA averaged Angstrom Exponent (α) for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

Fig. 9 and 10 present Angstrom Exponent (α) plots for OPAC SOOT aerosol components at eight relative humidities and ten countries. The presentations for MERRA α values and OPAC SOOT component are similar to those presented for the WASO and INSO aerosol components (sections 3.3 and 3.4) for seven countries (Nigeria, Brazil, China, Japan, Ethiopia, Mexico and Pakistan). Different observations are made for three countries (India, Indonesia and USA) such that though their atmospheric aerosols are dominated

by fine mode particles, India has α values approximately the same as those of OPAC SOOT aerosol component α values at 98% RH. India's α values are also greater than OPAC SOOT α values at 99% RH. It is also observed that India's aerosol α values are less than OPAC SOOT α values at 0-95% RH. The OPAC SOOT aerosol models at 98% RH present approximately the same particle size distribution for soot aerosols found within the Indian atmosphere while it underestimates its values at 99% RH and overestimates the α values at 0-95% RH. Indonesia however has α value < OPAC SOOT α values at 0-90% RH and α values > OPAC SOOT α values at 95-99% RH. Additionally, USA also has α values < OPAC SOOT α values at 0-90% RH and α values > OPAC SOOT α values at 95-99% RH This indicates that the atmosphere of the two countries Indonesia and the USA present lower soot aerosol particle size distributions than OPAC SOOT models at 0-90% RH and higher soot aerosol particle size distributions than those of OPAC SOOT models at 95-99% RH.

In summary from α values all ten countries are seen to have lower soot aerosol particle size distributions within their atmospheres when compared to OPAC at lower relative humidities and higher soot aerosol particle size distributions when compared to OPAC at higher relative humidities.

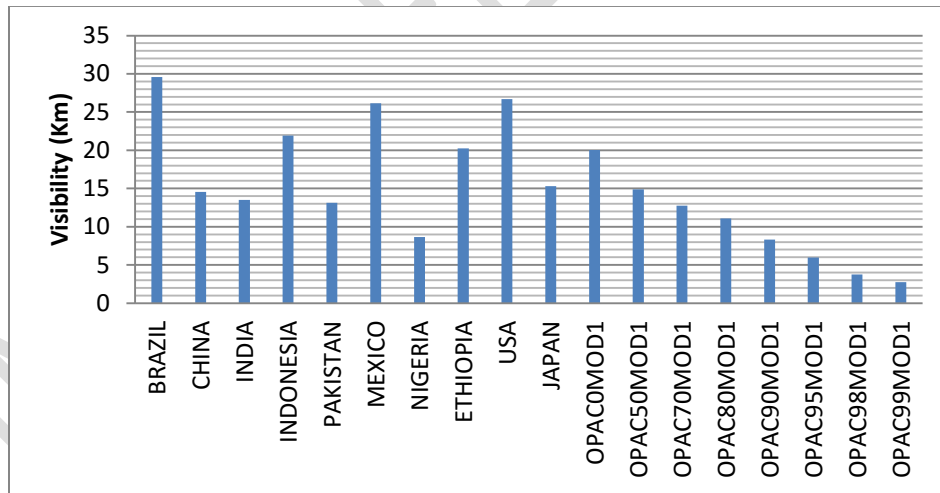


Fig 11. Histogram plot for OPAC SOOT Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

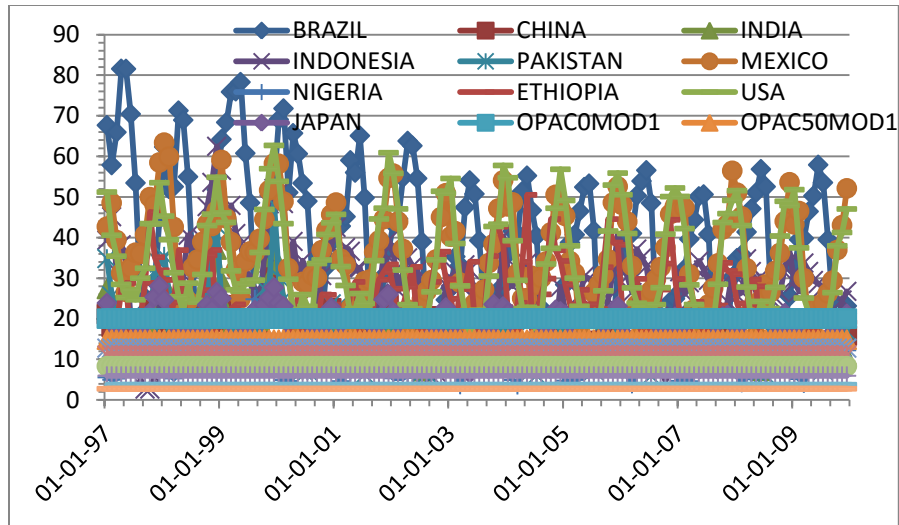


Fig 12. Visibility plot for OPAC SOOT Model 1 Visibilities and MERRA averaged visibilities for ten countries (Nigeria, Ethiopia, Brazil, USA, Pakistan, Mexico, Japan, China, Indonesia and India)

Fig. 11 and 12 shows that the visibility presentation for SOOT OPAC aerosol component and MERRA calculated visibilities are similar for those observed under WASO and INSO OPAC components. However China's atmospheric visibility is observed to be lower than OPAC SOOT component visibility at 0-50% RH and higher than OPAC SOOT aerosol component visibility at 70-99% RH.

In summary, China is said to have higher soot aerosol concentrations than OPAC SOOT component at lower relative humidities and lower soot aerosol concentrations than OPAC SOOT component at higher relative humidities.

4. SUMMARY AND CONCLUSION

This work used aerosol extinction coefficient and Angstrom Exponents (α) extracted from MERRA satellite data to validate results obtained from simulations carried out using OPAC 4.0 in the work done by Abdulkarim and Tijjani (2021). From the effect of varying aerosol concentrations and relative humidity on visibility and particle size distribution in urban atmosphere [6], five models were considered for each OPAC aerosol component of WASO (water soluble), INSO (insoluble) and SOOT. All the OPAC models showed Angstrom Exponent (α) values > 1 at 0-98% RH indicating the dominance of fine mode aerosol particles. These fine mode particles become larger as relative humidity increased from 0-98% RH. The increase in particle size was indicated by α values tending towards values < 1 . At 99% RH however, $\alpha < 1$ indicated the largest aerosol particles within the model. As OPAC concentrations of water soluble and

soot aerosol particles across the five models increased, the Angstrom Exponent (α) values remained constant which indicated constant aerosol particle size distributions. In addition, when the OPAC concentration of INSO (insoluble) aerosol particles across the five models increased the particle size distributions decreased this is indicated by a decrease in Angstrom Exponent (α) values across the five models. Additionally, when OPAC WASO, INSO and SOOT aerosol component concentrations increased across the five models, visibility decreased. Visibility also decreased as relative humidity increased due to aerosol particle growth. From the MERRA satellite data, Angstrom Exponent (α) time series analysis was carried out for each of the ten countries (Nigeria, Japan, China, Brazil, Mexico, India, Indonesia, USA, Ethiopia and Pakistan) for the span of 12 years. Model statistics showed that R^2 values fit the observed data well. β (level) effects on Angstrom Exponent (α) showed the presence of fine mode aerosol particles within the atmospheres of Brazil, China, India, Indonesia, USA, Japan and Mexico which didn't change over the years. They also showed the presence of coarse mode aerosol particles within the atmospheres of Nigeria, Pakistan and Ethiopia. δ (season) effects on Angstrom Exponent (α) showed that for the countries Japan and Mexico, fluctuations in α were not due to the actual changes in particle size distributions but due to fluctuations found within the model.

From Angstrom Exponent values, OPAC has identified the urban atmosphere to be only characterized by fine mode aerosol particles as α values were seen to be > 1 . From MERRA it has been observed that not all urban atmospheres are characterized by fine mode aerosol particles but some urban atmospheres are characterized by coarse mode aerosol particles also as Angstrom Exponent values for some countries were observed to be < 1 .

For all ten countries, OPAC is seen to overestimate the aerosol particle size distributions of water soluble aerosols at low relative humidities and underestimate the aerosol particles size distributions of water soluble aerosols at high relative humidities for the urban atmosphere. For insoluble aerosols within an urban atmosphere, OPAC was only able to simulate well the particles size distributions of insoluble aerosols for India at 98% relative humidity because at 98% relative humidity it was observed to have approximately the same insoluble aerosol particle size distributions as those found within the urban atmosphere of India. However for the rest of the nine countries (USA, Brazil, Indonesia, China, Japan,

Mexico, Nigeria, Pakistan and Ethiopia), OPAC was seen to overestimate insoluble aerosol particle size distributions at low relative humidities and underestimate insoluble aerosol particle size distributions at high relative humidities. When compared to the OPAC SOOT aerosol component, OPAC at 98% relative humidity is observed to simulate well the soot aerosol particle size distribution of the Indian urban atmosphere. For the rest of the nine countries on the other hand, OPAC is seen to have overestimated their soot aerosol particle size distributions at low relative humidities and underestimate their soot aerosol particle size distributions at high relative humidities.

In terms of the relationship between Angstrom Exponent and urban atmospheric visibility, it was observed that it satisfied the direct power law for OPAC and approximately satisfied the direct power law for most of the countries. For four countries (Nigeria, India, Pakistan and China) OPAC overestimates their visibilities at low relative humidities and underestimates their visibilities at high relative humidities. This indicates low water soluble aerosol concentrations at low relative humidities and high water soluble aerosol concentrations at high relative humidities. At 0% and 50% OPAC relative humidities respectively, OPAC is seen to simulate approximately the same visibility values and water soluble aerosol concentrations for Ethiopia (at 0% RH) and Japan (at 50% RH). The rest of the four countries (Brazil, Indonesia, Mexico and USA) however are seen to have their visibility and water soluble aerosol concentrations underestimated at all relative humidities by OPAC. There is a clear underestimation by OPAC of visibility and overestimation of insoluble aerosol concentration at low relative humidity for the Nigerian urban atmosphere. At high relative humidities OPAC also overestimates visibility and underestimates insoluble aerosol concentrations. Two countries India and Pakistan have their visibilities overestimated and insoluble aerosol concentrations underestimated at low relative humidities. OPAC is also observed to underestimate visibilities therefore overestimating aerosol concentrations at high relative humidities. OPAC simulates and shows approximately the same visibility and insoluble aerosol concentrations at low RH when compared to Ethiopia's and Japan's atmosphere and also when compared to China's atmosphere at 50% relative humidity. An overestimation of visibility and insoluble aerosol concentrations is seen at all relative humidities for the countries USA, Mexico, Brazil and Indonesia. All other comparisons for visibility and soot aerosol concentrations were found to be similar with insoluble aerosol concentrations for all the ten countries except for China. It was seen that OPAC underestimates visibility

and overestimates soot aerosol concentrations for the country china at low relative humidities while it overestimates visibility and underestimates aerosol concentrations at high relative humidities.

According to Hess (1998), OPAC is a software simulation package that has aerosol components adapted to average conditions. Its main aim was to be able to simulate optical properties like the Angstrom exponent (α) and visibility for atmospheric aerosol components and their mixtures in order to enhance climate analyses and calculations. It can be concluded that OPAC is still yet to completely capture the characteristics of atmospheric particulate constituents of the real urban atmospheres in terms of visibility, aerosol particle size distributions and atmospheric water soluble, insoluble or soot aerosol concentrations.

REFERENCES

- [1] Stanier, C. O. Khlystov, A. Y., Chan, Wanyu R., Mandiro, M.. (2004) Aerosol Science and Technology, 38(S1):215–228. *American Association for Aerosol Research*. DOI: 10.1080/02786820390229525
- [2] Srivastava, P. Dey, S. Kumar, A. Singh, S. Mishra, S. K. and Tiwari, S. (2017). Science of the Total Environment Importance of aerosol non-sphericity in estimating aerosol radiative forcing in Indo-Gangetic Basin. *Science of the Total Environment* Vol (599-600), 655-662 <https://10.1016/j.scitotenv.2017.04.239>
- [3] Cheng, Y.F., et al., (2008). Relative humidity dependence of aerosol optical properties and direct radiative forcing in the surface boundary layer at Xinken in Pearl. *Atmospheric Environment* (2008), doi:10.1016/j.atmosenv.2008.04.009
- [4] Rienecker, M., Suarez, M. J., Todling, R., Bacmeister, J., Takacs, L., Liu, H.-C., Gu, W., Sienkiewicz, M., Koster, R. D., Gelaro, R., Stajner, I., and Nielsen, J. E. (2008). The GEOS-5 Data Assimilation System-Documentation of Versions 5.0.1, 5.1.0, and 5.2.0., Technical Report Series on Global Modeling and Data Assimilation, 104606, 27, 2008.
- [5] Hess, M., Koepke, P., and Schult, I. (1998): Optical properties of aerosols and clouds: The software package OPAC, *B. Am. Meteor. Soc.*, 79, 831–844.
- [6] Abdulkarim, U. & Tijjani, B., (2021). Effect of Varying Aerosol Concentrations and Relative Humidity on Visibility and Particle Size Distribution in Urban Atmosphere. *Journal of Atmospheric Science Research* . Vol. 4 (3), 14-28. DOI: <https://doi.org/10.30564/jasr.v4i3.3430>
- [7] Rienecker, M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G.-K., Bloom, S., Chen, J., Collins, D., Conaty, A., da Silva, A., Gu, W., Joiner, J., Koster, R. D., Lucchesi, R., Molod, A., Owens, T., Pawson, S., Pegion, P., Redder, C. R., Reichle, R., Robertson, F. R., Ruddick, A. G., Sienkiewicz, M., and Woollen, J. (2011) MERRA – NASA’s Modern-Era Retrospective Analysis for Research and Applications, *J. Climate*, 24, 3624–3648, doi:10.1175/JCLI-D-11-00015.1
- [8] Kahn, R., Gaitley, B., Martonchik, J., Diner, D., Crean, K., and Holben, B.(2005) Multiangle Imaging Spectroradiometer (MISR) global aerosol optical depth validation based on 2 years of coincident Aerosol Robotic Network (AERONET) observations, *J. Geo-phys. Res.*, 110, D10S04, doi:10.1029/2004JD004706

- [9] Holben, B. N., Eck, T., Slutsker, I., Tanre, D., Buis, J. P., Set-zer, A., Vermote, E., Reagon, J. A., Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A.(1998) AERONET – A federated instrument network and data archive for aerosol characterization, Remote Sens. Environ., 66, 1–16
- [10] Textor, C., J. E., Putaud, J.-P., Schulz, M., van der Werf, G. R., and Wilson, J.(2006) Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmospheric Chemistry and Physics, 6, 4321–4344, <https://doi.org/10.5194/acp-6-4321-2006>, <https://www.atmos-chem-phys.net/6/4321/2006/>

UNDER PEER REVIEW