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USING THE DDA (DISCRETE DIPOLE APPROXIMATION) METHOD IN DETERMINING THE EXTINCTION CROSS SECTION OF BLACK CARBON

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Abstract

BC (Black Carbon), which can be found in the atmosphere, is characterized by a large value of the imaginary part of the complex refractive index and, therefore, might have an impact on the global warming effect. To study the interaction of BC with light often computer simulations are used. One of the methods, which are capable of performing light scattering simulations by any shape, is DDA (Discrete Dipole Approximation). In this work its accuracy was estimated in respect to BC structures using the latest stable version of the ADDA (vr. 1.2) algorithm. As the reference algorithm the GMM (Generalized Multiparticle Mie-Solution) code was used. The study shows that the number of volume elements (dipoles) is the main parameter that defines the quality of results. However, they can be improved by a proper polarizability expression. The most accurate, and least time consuming, simulations were observed for IGT_SO. When an aggregate consists of particles composed of ca. 750 volume elements (dipoles), the averaged relative extinction error should not exceed ca. 4.5%.

Keywords: black carbon, discrete dipole approximation, light scattering, fractal-like aggregates.

1. Introduction

BC (Black Carbon) is a product of an incomplete combustion of carbon-based fuels. Due to its absorption properties it is assumed to have an impact on the global warming effect [1, 2]. The interaction of BC aggregates with light is a matter of concern for many climate scientists. One of ways of studying this phenomena is to perform light scattering simulations. However, many common algorithms are limited to aggregates composed of spherical, non-overlapping particles only. This is a significant drawback because, in reality, BC aggregates shortly after emission interact with both organic and inorganic matter and form much more complex structures [3]. One of the algorithms capable of performing light scattering simulations by any shape is DDA (Discrete Dipole Approximation) [4, 5]. It was introduced by Purcell and Pennypacker and was based on the interaction between volume elements (dipoles) of a scatterer [6]. One of the most important limitations of this method is the fact that structures must be decomposed into a significant number of volume elements (dipoles) and, therefore, simulations might be very time consuming. Furthermore, their accuracy decreases when strongly absorbing materials are used [7]. The main goal of this work was to approximate the DDA accuracy for BC aggregates, which are the core element of more complex, atmospheric structures.

The research was divided into two parts. First, the optimal parameters for DDA simulations were estimated. Next, the accuracy of the DDA method for BC aggregates was investigated. As the reference algorithm the GMM (Generalized Multiparticle Mie-Solution) code by Xu was used [8]. When a single particle is considered it simplifies to the solution by Mie which is assumed to be exact. However, for more complex structures results might be slightly erroneous.

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For this reason, extinction values for BC aggregates used in the second part of this work were compared to those calculated with the T-Matrix algorithm by Mackowski [9]. The maximum relative error was not larger than 0.06% in any case. This proved that GMM is reliable and, therefore, can be used as the reference code for every shape used in this work.

2. Results and discussion

In the first part of the study the optimal DDA settings for simulating the light scattering by BC particles were determined. Their diameter is dependent on the generation conditions and usually varies from dp=10 nm to dp=50 nm [10–12]. As the reference particle a single sphere, characterized by the diameter of dp=30 nm, was generated. Then, it was decomposed into different meshes of volume elements (dipoles) required for DDA simulations. They are presented in Fig. 1 and described in Tab. 1 in more detail. To keep a constant distance between volume elements (dipoles) in a mesh, the volume correction procedure was not used. There are no universal rules on how to predict the required mesh size. However, a few guidelines exist. One of them states that DDA should not be used for materials characterized by a large value of the imaginary part of the complex refractive index m, i.e. [7]:

$$|m - 1| < 2. \tag{1}$$

This condition is true for every refractive index used in this work. However, recent studies prove that even materials with very high m can be investigated [13, 14]. In this case the number of volume elements (dipoles) should be significantly increased to avoid inaccuracies and simulation errors [13, 15]. The next DDA rule states that the mesh should be able to precisely describe the shape of the structure [7]. It is recommended to place at least 10 volume elements (dipoles) along its smallest dimension. One of the meshes used in this work did not meet this requirement, i.e. M0 (see Fig. 1). It was generated for a theoretical study only and should not be used in extensive simulations. The third DDA rule defines the minimum distance between volume elements (dipoles) [7]:

$$d < \frac{\lambda}{10|m|}, \tag{2}$$

where λ is the incident wavelength. However, for absorbing materials the mesh should be much more compact [7]. More information about the DDA accuracy can be found elsewhere, e.g. in the work by Penttila et al. [16].

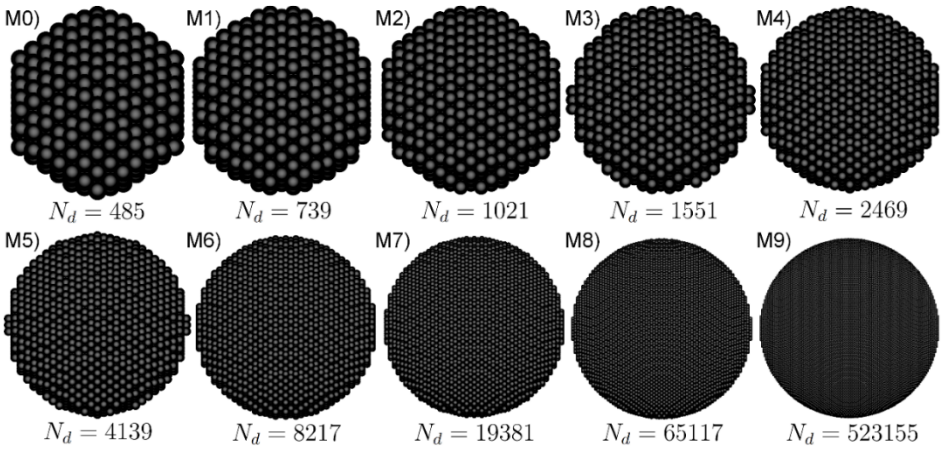

Fig. 1. Different DDA meshes composed of Nd volume elements (dipoles). The smallest one, namely M0, was used for a theoretical study only and is not recommended for extensive DDA simulations. All shapes were symmetrical and the volume correction procedure was not used, i.e. the distance between volume elements (dipoles) was always constant.

Table 1. The description of the DDA meshes used in the accuracy study (Fig. 1). d is the distance between volume elements (dipoles) and Nds defines their number along the smallest dimension.

Mesh	M0	M1	M2	M3	M4
N_d	485	739	1021	1551	2469
d [nm]	3.0	2.7	2.4	2.1	1.8
N_{ds}	9	11	13	15	17
Mesh	M5	M6	M7	M8	M9
N_d	4139	8217	19381	65117	523155
d [nm]	1.5	1.2	0.9	0.6	0.3
N_{ds}	19	25	33	49	99

There are many algorithms which allow to perform DDA simulations. Very popular ones are open source projects: DDScat and ADDA. Both of them are widely recognized by the light scattering community and, in many cases, can be used interchangeably. In this work mostly ADDA was used but a short comparison with DDScat was also performed. Simulations were performed on a middle-class PC with a 16 GB RAM and a 64bit i7-3930K processor. Only one thread was used and the GPU was inactive. As the representative parameter for determining the accuracy the extinction cross section C_{ext} was used.

A variety of different refractive indices of BC have been published [10]. In this work the one by Chang and Charalampopoulos was used [17]. It is valid for particles generated in a propane-oxygen flame with the fuel equivalence ratio ϕ=1.8. In preliminary studies HAB (Height Above Burner) was assumed to be 10 mm. The chosen refractive index is in agreement with the criterion proposed by Bond et al. [10]. In the study the incident wavelength varied from λ=300nm to λ=900nm with the step $\Delta\lambda$=10 nm. Neither configurational nor orientational averaging was performed [18].

The first step of the comparison procedure was associated with the changeable parameters implemented in the latest stable version of ADDA (vr 1.2). Nine different configuration sets, which are presented in Tab. 2, were generated. They consisted of three elements: the polarizability expression, the formulation for the interaction term and the procedure for calculating scattering quantities. The most important parameter was the polarizability expression, namely LDR (Lattice Dispersion Relation), CLDR (Corrected Lattice Dispersion Relation), CM (Clausius-Mossotti elation), RRC (Radiative Reaction Correction), DGF (Digitized Green's Function), LAK (formulation by Lakhtakia) and IGT_SO (approximate Integration of Green's Tensor over the dipole).

Table 2. The description of nine sets used for determining the most accurate polarizability expression used in DDA simulations.

Set	Polarizability	Interaction term	Scattering quantities
1	LDR	POI	DR
2	CLDR	POI	DR
3	CM	POI	DR
4	RRC	POI	DR
5	DGF	POI	DR
6	LAK	POI	DR
7	IGT_SO	IGT_SO	IGT_SO
8	FCD	FCD	DR
9	SO	SO	SO

Set 8 defined the FCD (Filtered Coupled Dipoles) routine which is recommended for materials with large values of the imaginary part of the refractive index [13, 15]. Set 9 defined the SO (Second Order) procedure, which is still under development and, therefore, no detailed description is available. POI (Interaction of Point Dipoles) is the default formulation for the interaction term and DR (Draine's Formulation) is the default procedure for calculating scattering quantities. Other ADDA parameters were set to their default values. The aim of this work was not to provide a detailed overview of the DDA method and, therefore, more information about the polarizability formulations and other DDA parameters can be found elsewhere [7, 13, 19].

The results are presented in Tab. 3. The amount of RAM used was defined by the largest mesh, i.e. M9. The simulation time was measured for the same shape for the incident wavelength of λ=600 nm. It is located exactly in the middle of the investigated electromagnetic spectrum and, therefore, it was used as the reference value. The averaged relative extinction error was defined by the following equation:

$$\delta C_{ext} = \frac{\sum_{i=0}^{60} \delta C(300+(i\cdot 10))_{ext}}{61}, \tag{3a}$$

$$\delta C(\lambda)_{ext} = \left| \frac{C(\lambda)_{ext,ADDA} - C(\lambda)_{ext,XU}}{C(\lambda)_{ext,XU}} \cdot 100\% \right|. \tag{3b}$$

Table 3. The results of the accuracy study for different polarizability expressions. The amount of RAM used was defined by the largest mesh, i.e. M9. The simulation time was measured for the same shape for the reference wavelength (λ=600 nm).

Set (see Tab. 2)	1	2	3	4	5	6	7	8	9
RAM [MB]	489.1	489.1	489.1	489.1	489.1	489.1	489.2	489.1	1034.9
Time [s]	31	31	30	31	31	31	29	31	36
Mesh	Averaged relative error [%]								
M0	4.196	4.196	4.233	4.233	4.188	4.188	4.446	4.113	4.461
M1	5.630	5.630	5.596	5.597	5.636	5.636	5.493	6.120	5.479
M2	2.706	2.706	2.681	2.681	2.712	2.712	2.556	3.146	2.546
M3	4.691	4.691	4.671	4.671	4.695	4.695	4.494	4.992	4.486
M4	4.107	4.107	4.093	4.093	4.110	4.110	3.996	4.509	3.990
M5	0.967	0.967	0.961	0.962	0.969	0.969	0.832	1.007	0.829
M6	2.006	2.006	2.000	2.000	2.007	2.007	1.908	2.137	1.905
M7	1.166	1.166	1.163	1.163	1.167	1.167	1.089	1.197	1.087
M8	0.355	0.355	0.354	0.354	0.355	0.355	0.304	0.348	0.304
M9	0.347	0.347	0.347	0.346	0.347	0.347	0.319	0.348	0.319

The most accurate procedure was SO. However, due to the amount of RAM used, a long simulation time and the fact that it is still under development it was not used in further study. The least accurate procedure was FCD what was rather surprising. Nevertheless, the results prove that the main parameter which affects the accuracy is the number of volume elements (dipoles) and not the polarizability expression. The averaged relative error does not decrease gradually with the number of volume elements (dipoles), which might be an important information when the assumed DDA accuracy is based on their number only. This phenomenon might be associated with mesh generating procedures. Although those used in this study were not erroneous, different distances between volume elements (dipoles) can cause significant changes in the particle shape and affect simulations. The worst case was observed for M1 and a very good agreement between the accuracy and the simulation time was achieved for M5 (4.2

MB of RAM used, the simulation time was ca. 257 ms). Nevertheless, ca. 4000 volume elements (dipoles) for a single sphere when an aggregate is composed of a few hundred particles might be too much for a standard PC. The lowest recommended number of volume elements (dipoles), i.e. ca. 700, should be associated with the averaged extinction error of ca. 6%. The only mesh that underestimated the scattering cross section was M0. The amount of RAM used was 489.1 MB for the largest mesh for almost all sets. It was slightly increased for IGT_SO and reached its maximum when the SO expression was used. The least time consuming procedure was IGT_SO. However, when the simulation speed is considered, the difference between sets is almost negligible. The extinction diagrams are presented in Fig. 2 and in Fig 3 is the error chart. In the further study the CM expression was used. Mostly because it is the most basic one, it does not require external table files (like IGT_SO and SO), provides a good accuracy and a reasonable simulation time. Additional tests proved that the alternative procedure for calculating scattering quantities, namely FIN (Finite Dipole Correction), does not improve the accuracy. Furthermore, changing the stopping criterion from EPS=5 to EPS=10 doubles the simulation time but has no impact on the results.

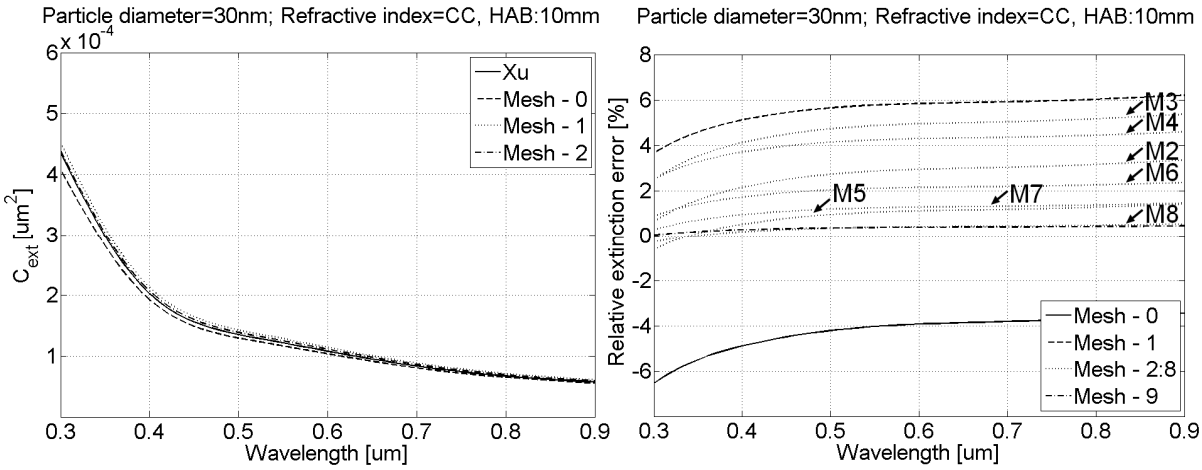

Fig. 2. Left) The extinction diagrams for the CM polarizability expression and different meshes of volume elements (dipoles). Meshes M2-M9 are positioned between the exact value (Xu) and M1. Right). The relative error for different meshes of volume elements (dipoles).

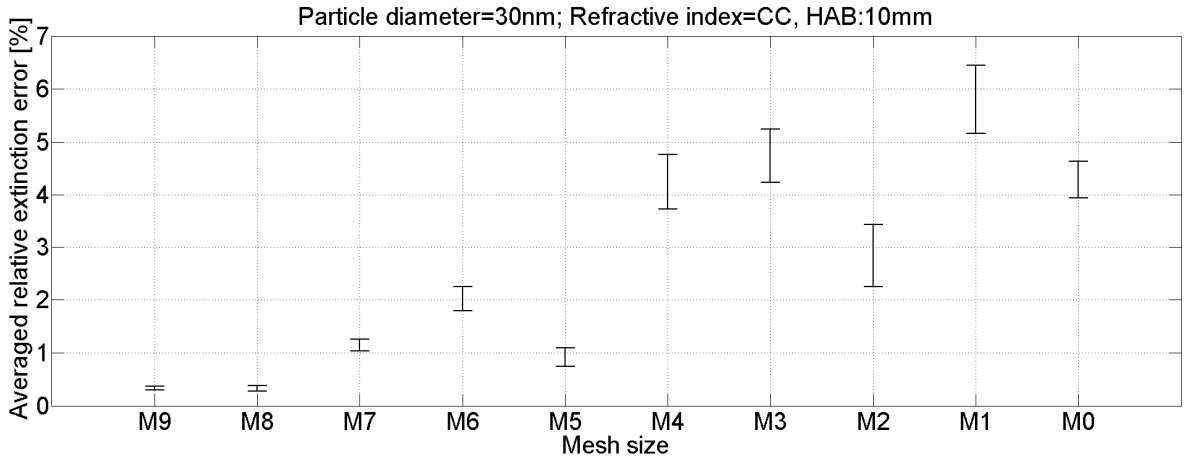

Fig. 3. The minimum and the maximum values of the averaged extinction error for all polarizability expressions used in this study (see Tab. 2).

The polarizability expression is not the only changeable parameter that can be used to increase the quality of DDA simulations. In the next step different iterative solvers, namely: QMR2, CSYM, CGNR, BICG and BICGSTAB, were used. The default procedure was QMR and its performance is presented in Tab 3. In this study, the polarizability expression was CM and the remaining ADDA parameters were set to their default values. The results are presented in Tab 4. This time only the amount of RAM used and the simulation time are included in the table because there was almost no difference in the results (the relative extinction error differed by no more than 0.001%). Therefore, when a single BC particle is considered, all expressions can be considered as equal.

Table 4. The results of the accuracy study for different iterative solvers. The amount of RAM used was defined by the largest mesh, i.e. M9. The simulation time was measured for the same shape for the reference wavelength (λ=600 nm).

Iterative solver	QMR2	CSYM	CGNR	BICG	BCGS2	BICGSTAB
RAM [MB]	465.2	465.2	417.3	417.3	513.1	489.1
Time [s]	32	64	66	31	34	33

Next, the extinction error for different refractive indices was calculated (Fig. 4). Three of them were adapted from the paper by Chang and Charalampopoulos [17] and each one represented a different HAB (Height Above Burner) value. The forth one was taken from the OPAC database [20]. In the simulations the CM polarizability expression was used, other parameters were set to their default values.

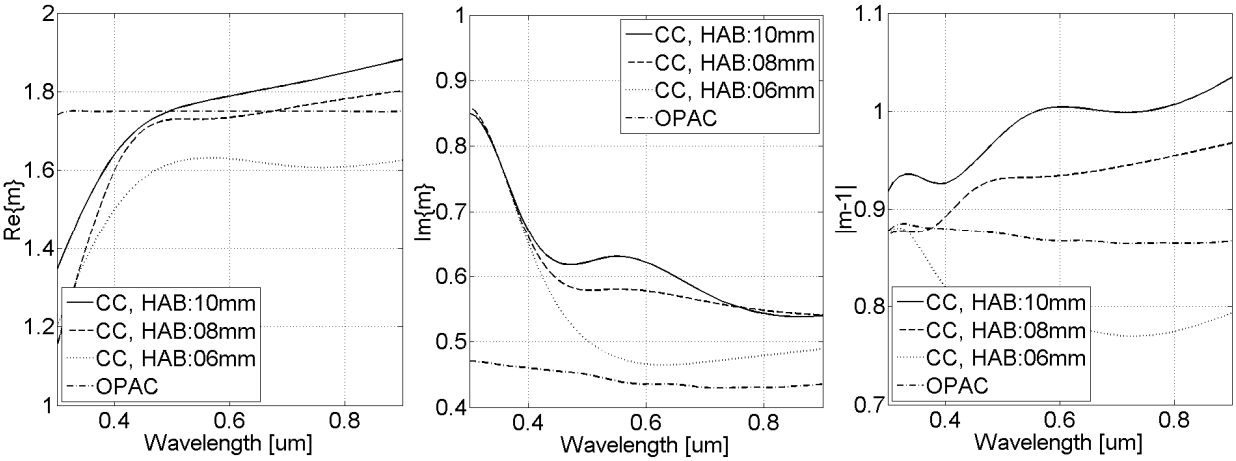

Fig. 4. Refractive indices used in this study. CC defines the work by Chang and Charalampopoulos [17, 20]. OPAC is the acronym for the Optical Properties of Aerosols and Clouds database [20].

Table 5. The results of the accuracy study for different refractive indices.

Refractive index	CC HAB=10 mm	CC HAB=8 mm	CC HAB=6 mm	OPAC
Mesh	Averaged relative error [%]			
M0	4.2329	4.6419	5.4085	4.5309
M1	5.5964	5.2255	4.5512	5.3926
M2	2.6809	2.3274	1.6054	2.4531
M3	4.6712	4.2539	3.4920	4.4381
M4	4.0928	3.7882	3.2339	3.9243

M5	0.9613	0.8264	0.3234	0.7412
M6	1.9997	1.7866	1.3977	1.8791
M7	1.1662	1.0082	0.6949	1.0673
M8	0.3550	0.3033	0.1073	0.2607
M9	0.3466	0.2980	0.1868	0.3124

One of the reasons for choosing the CC (HAB=10 mm) refractive index as the reference one was the fact that it was characterized by the largest absolute value for visible wavelengths. The DDA accuracy is associated with the expression |m-1| and, therefore, when a different refractive index is used, the quality of the results should be improved. The aim of this work was to compare the extinction error for the worst case.

In spite of the fact that in this study d_p=30 nm represents the average size, particles characterized by different dimensions are common. For the next part of this study particles with following diameters: d_p=10 nm, d_p=30 nm, d_p=50 nm, d_p=70 nm, d_p=90 nm and d_p=110 nm were generated. This set covered a wide range of BC particle shapes that can be found in the atmosphere. DDA simulations were performed using the CM polarizability expression and other parameters were set to their default values. The size of volume elements (dipoles) was constant but the distance between them differed. The results are presented in Tab. 6. They show that the deviation in simulation times is very small. The DDA quality increases along with the particle size. However, this tendency does not apply to M0. The results show that it is not necessary to increase the number of volume elements (dipoles) along with the particle size. Furthermore, results for larger particles can be even more accurate.

Table 6. The results of the accuracy study for different particle diameters. The simulation time was measured for the largest mesh, i.e. M9, for the reference wavelength (λ=600 nm).

Particle diameter [nm]	10	30	50	70	90	110
Time [s]	32	30	31	31	32	30
Mesh	Averaged relative error [%]					
M0	4.025	4.233	4.641	5.175	5.724	6.199
M1	5.615	5.596	5.535	5.404	5.194	4.931
M2	2.750	2.681	2.529	2.303	2.031	1.712
M3	4.704	4.671	4.582	4.419	4.187	3.913
M4	4.103	4.093	4.053	3.963	3.816	3.629
M5	1.000	0.961	0.887	0.790	0.683	0.579
M6	2.018	2.000	1.953	1.870	1.757	1.627
M7	1.182	1.166	1.116	1.044	0.953	0.847
M8	0.366	0.355	0.330	0.298	0.262	0.225
M9	0.353	0.347	0.331	0.308	0.280	0.246

Finally, to prove that the above presented results are genuine, a comparison with another DDA algorithm, namely DDScat, was performed. The polarizability expression for DDScat were D1 (Draine and Goodman), D2 (Draine and Gutkowicz-Krusin) and FCD (Filtered Couple Dipole). The iterative solver was PBCGS2 (BiConjugate Gradient with Stabilization). The only non-default ADDA parameter was the polarizability expression, i.e. CM. The light scattering results are presented in Tab. 7. They show that in spite of the fact that both algorithms behave similarly, some differences can be observed. During the ADDA simulations the distance between volume elements (dipoles) was constant, what affected the particle volume. On the contrary, during DDScat simulations the particle volume was exact, what had an impact on the distance between volume elements (dipoles). What is important, the accuracy does not

necessarily increases with the number of volume elements (dipoles). In some cases, it might even lead to worse results.

The accuracy for BC particles should not be treated as exact, but rather rough data. To achieve more reliable results the orientational averaging procedure should be implemented. Furthermore, different decomposition algorithms must be tested. When a mesh is sparse, e.g. M0, even a small displacement can alter the particle shape, surface and volume. The aim of this part was to estimate the most accurate simulation parameters. In the following section, because of the fact that aggregates are composed of a significant number of primary particles and each one is characterized by a slightly different mesh of volume elements (dipoles), the extinction error can be assumed to be much more accurate.

Table 7. The comparison of two different algorithms based on the DDA method: DDScat (vr. 7.3) and ADDA (vr. 1.2).

Mesh	M0	M1	M2	M3	M4	M5	M6	M7	M8	M9
	Averaged relative error [%]									
ADDA	4.233	5.596	2.681	4.671	4.093	0.961	2.000	1.166	0.355	0.347
D1	3.579	2.605	2.876	3.007	2.176	2.161	1.553	1.228	0.854	0.435
D2	3.579	2.605	2.876	3.007	2.175	2.161	1.554	1.228	0.854	0.435
FCD	3.587	3.016	3.266	3.264	2.542	2.183	1.672	1.251	0.843	0.434

The second part of the study was devoted to BC fractal-like aggregates. Its aim was to check whether the approximated accuracy is valid for large structures. For this reason four different aggregates were generated with the algorithm by Filippov [20, 21]. They were composed of N_p=25, N_p=50, N_p=75 and N_p=100 particles, respectively, and are presented in Fig. 5. The fractal dimension, which is the main parameter defining the shape of the structure, was D_f=2.2. This value was based on the work by Adachi et al. [2], who used the ET (Electron Tomography) procedure to retrieve morphological parameters of real fractal-like structures [23]. Naturally, many different methods for analysing such parameters exist, e.g. TEM (Transmission Electron Microscopy), but they were not considered in this work. More information about them can be found elsewhere [24-26]. Furthermore, light based techniques can be used to study different structures, like erythrocytes or fibres [27-32]. In this study primary particles, characterized by the diameter of d_p=30 nm, were not overlapping and there was no neck between them [33, 34].

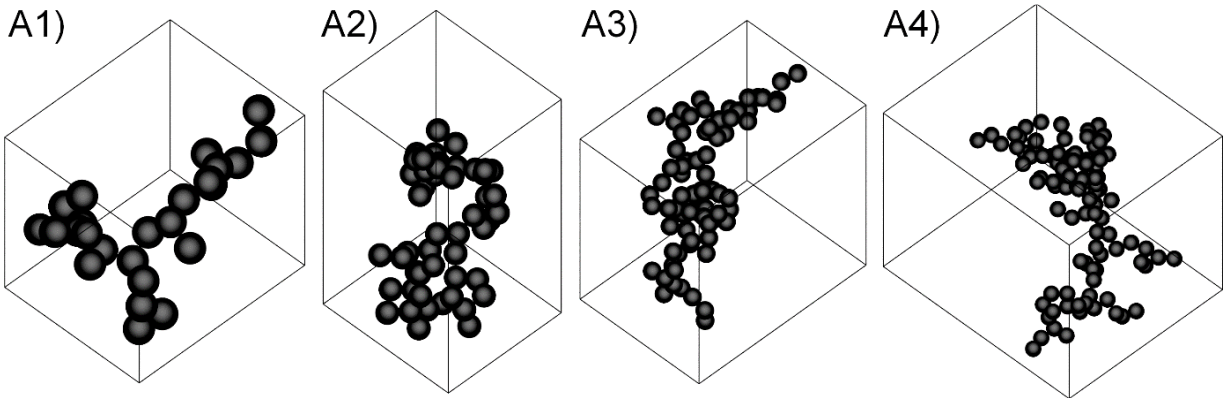

Fig. 5. Fractal-like aggregates composed of Np=25, Np=50, Np=75 and Np=100, respectively, primary particles with the diameter dp=30nm. The fractal dimension is Df=2.2.

For DDA simulations M2, M3 and M5 meshes were used. Larger grids were not applicable for a standard PC due to a significant number of volume elements (dipoles). Moreover, BC fractions of real atmospheric soot aggregates is estimated as ca. 7% [3]. When other components are included, i.e. organic/inorganic matter, the size and, therefore, the number of volume elements (dipoles), must be significantly increased. The scattering results were not averaged and were compared with the algorithm by Xu. During the DDA simulations the CM equation was used and remaining ADDA parameters were set to their default values. The incident wavelength varied from λ=300 nm to λ=900 nm with the step $\Delta\lambda$=10 nm. The results are less accurate than those for a single particle (Tab. 8). Nevertheless, this proves that the simulation parameters approximated for a single sphere might not always be suitable for large aggregates. Furthermore, the extinction error decreases monotonically along with the number of volume elements (dipoles). Next, the simulations were repeated using the most and the less accurate polarizability expression determined in the previous study, i.e. IGT_SO and FCD, respectively. The results are presented in Tab. 9 and in Tab. 10 Once again, the IGT_SO polarizability expression turned out to be the most accurate and least time consuming.

Table 8. The results of the accuracy study for the CM polarizability expression. The amount of RAM used was associated with the largest mesh. The simulation time was measured for the same shape for the reference wavelength (λ=600 nm). pA and pB define the polarization state.

Aggregate	25 pA	25 pB	50 pA	50 pB	75 pA	75 pB	100 pA	100 pB
Time [s]	60		147		356		812	
RAM [MB]	671		1465		3128		6348	
Dipoles	104705		209398		314176		418812	
Mesh	Averaged relative error [%]							
M1	3.404	3.646	3.421	3.065	3.459	3.243	3.293	3.471
M3	2.713	2.841	3.158	3.027	2.781	2.745	2.782	2.821
M5	2.398	2.303	2.318	2.184	2.174	2.214	2.250	2.204

Table 9. The results of the accuracy study for the IGT_SO polarizability expression. The amount of RAM used was associated with the largest mesh. The simulation time was measured for the same shape for the reference wavelength (λ=600 nm). pA and pB define the polarization state.

Aggregate	25 pA	25 pB	50 pA	50 pB	75 pA	75 pB	100 pA	100 pB
Time [s]	52		127		351		736	
RAM [MB]	672		1466		3129		6348	
Dipoles	104705		209398		314176		418812	
Mesh	Averaged relative error [%]							
M1	3.180	3.381	3.153	2.836	3.216	2.978	3.039	3.226
M3	2.548	2.655	2.952	2.846	2.597	2.555	2.589	2.635
M5	2.274	2.182	2.178	2.059	2.049	2.086	2.120	2.075

Table 10. The results of the accuracy study for the FCD polarizability expression. The amount of RAM used was associated with the largest mesh. The simulation time was measured for the same shape for the reference wavelength (λ=600 nm). pA and pB define the polarization state.

Aggregate	25 pA	25 pB	50 pA	50 pB	75 pA	75 pB	100 pA	100 pB
Time [s]	55		133		342		744	
RAM [MB]	671		1465		3129		6348	
Dipoles	104705		209398		314176		418812	
Mesh	Averaged relative error [%]							
M1	4.154	4.500	2.476	3.830	4.215	4.015	4.049	4.249
M3	3.061	3.316	3.464	3.337	3.088	3.071	3.108	3.148
M5	2.585	2.571	2.476	2.356	2.337	2.390	2.432	2.385

3. Conclusions

The aim of this work was to approximate the extinction error of DDA simulations when BC aggregates are considered. This knowledge might lead to more accurate results and improve the modelling process which can help to understand many physical phenomena [35]. The study revealed that the number of volume elements (dipoles) is the main parameter that defines DDA accuracy (Tab. 3, Tab. 8, Tab. 9 and Tab. 10). Other elements, like the polarizability expression, are not as influential, but might improve the results and reduce the simulation time (Tab. 3, Tab. 8, Tab. 9 and Tab. 10). The IGT_SO routine slightly increases the RAM used but reduces the simulation time and is the most accurate polarizability expression (Tab. 3, Tab. 9). The least reliable routine turned out to be FCD (Tab. 3, Tab. 10). These findings apply for a single BC particle as well as large fractal aggregates. The DDA accuracy should not be based on results for a single sphere only. In some cases, the differences in accuracy between a single sphere and a large aggregate might not be negligible (Tab. 3, Tab. 9). Furthermore, when small scatterers are considered, an increase in the number of volume elements (dipoles) does not necessarily increases the quality (Tab. 3). Such procedure might affect the particle shape and, therefore, lead to more erroneous results. The study shows that there is no need to increase the number of volume elements (dipoles) when large spheres are used (Tab. 6). The distance between volume elements (dipoles) can be related to the size of the structure. The accuracy decreases with increasing the absolute value of the complex refractive index (Tab. 5), what was expected. Different DDA algorithms can be used interchangeably, although the accuracy might slightly differ (in this work DDScat and ADDA were compared, Tab. 7). Finally, when an aggregate consists of particles composed of ca. 750 volume elements (dipoles), the averaged relative extinction error should not exceed the value of ca. 4.5% for any polarizability expression for both polarization states. For ca. 1550 volume elements (dipoles) it decreases to 3.4% and for 4150 to 2.6%. These values does not apply for simulations of a single particle (Tab. 8, Tab. 9 and Tab. 10). Note, that the results might be valid for BC structures only.

References

[1] Bond, T.C. (2001). Spectral dependence of visible light absorption by carbonaceous particles emitted from coal combustion. *Geophys. Res. Lett.*, 28(21), 4075–4078.

[2] Adachi, K., Buseck, P.R. (2008). Internally mixed soot, sulfates, and organic matter in aerosol particles from Mexico City. *Atmos. Chem. Phys.*, 8(21), 6469–6481.

[3] Adachi, K., Chung, S.H., Buseck P.R. (2010). Shapes of soot aerosol particles and implications for their effects on climate. *J. Geophys. Res.*, 115(15), D15206.

[4] Yurkin, M.A., Hoekstra, A.G. (2007). The discrete dipole approximation: An overview and recent developments. *J. Quant. Spectrosc. Radiat. Transf.*, 106(1), 558–589.

[5] Draine, B.T., Flatau, P.J. (1994). Discrete-dipole approximation for scattering calculations. *J. Opt. Soc. Am. A*, 11(4), 1491–1499.

[6] Purcell, E.M., Pennypacker, C.R. (1973). Scattering and absorption of light by nonspherical dielectric grains. *Astrophys. J.*, 186, 705–714.

[7] Yurkin, M.A., Hoekstra, A.G. (2011). The discrete-dipole-approximation code ADDA: Capabilities and known limitations. *J. Quant. Spectrosc. Radiat. Transf.*, 112(13), 2234–2247.

[8] Xu, Y.-L, Gustafson, B.A.S. (2001). A generalized multiparticle mie-solution: further experimental verification. *J. Quant. Spectrosc. Radiat. Transf.*, 15(4), 395–419.

[9] Mackowski, D.W., Mishchenko, M.I. (2011). A multiple sphere T-matrix FORTRAN code for use on parallel computer clusters. *J. Quant. Spectrosc. Radiat. Transf.*, 112(13), 2182–2192.

[10] Bond, T.C., Bergstrom, R.W. (2006). Light Absorption by Carbonaceous Particles: An Investigative Review. *Aerosol. Sci. Tech.*, 40(1), 27–67.

[11] Andreae, M.O., Galencser, A. (2006). Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols. *Atmos. Chem. Phys.*, 6(10), 3131–3148.

[12] Wentzel, M., Gorzawski, H., Naumann, K.H., Saathoff, H., Weinbruch, S. (2003). Transmission electron microscopical and aerosol dynamical characterization of soot aerosols. *J. Aerosol. Sci.*, 34(10), 1347–1370.

[13] Chaumet, P.C., Sentenac, A., Rahmani, A. (2004). Coupled dipole method for scatterers with large permittivity. *Phys. Rev. E Stat. Nonlin. Soft. Matter. Phys.*, 70(32), 036606-1-036606-6.

[14] Yurkin, M.A, Min, M., Hoekstra, A.G. (2010). Application of the discrete dipole approximation to very large refractive indices: Filtered coupled dipoles revived. *Phys. Rev. E Stat. Nonlin. Soft. Matter. Phys.*, 82(3), 036703.

[15] Yurkin, M.A., De Kanter, D., Hoekstra, A.G. (2010). Accuracy of the discrete dipole approximation for simulation of optical properties of gold nanoparticles. *J. Nanophotonics*, 4(1), 041585.

[16] Penttila, A., Zubko, E., Lumme, K., Muinonen, K., Yurkin, M.A., Draine, B., Rahola, J., Hoekstra, G., Shkuratov, Y. (2007). Comparison between discrete dipole implementations and exact techniques, *J. Quant. Spectrosc. Radiat. Transf.*, 106(1), 417–436.

[17] Charalampopoulos, T.T., Chang H. (1990). Determination of the wavelength dependence of refractive indices of flame soot. *Proc. Math. Phys. Sci.*, 430, 577–591.

[18] Riefler, N., Di Stasio, S., Wriedt, T. (2004). Structural analysis of clusters using configurational and orientational averaging in light scattering analysis. *J. Quant. Spectrosc. Radiat. Transf.*, 89(1), 323–342.

[19] Piller, N.B., Martin, O.J.F. (1998). Increasing the Performance of the Coupled-Dipole Approximation: A Spectral Approach. *IEEE Trans. Antennas Propag.,* 46(8), 1126–1137.

[20] Hess, M., Koepke, P., Schult, I. (1998). Optical properties of aerosols and clouds: The software package OPAC. *B. Am. Meteorol. Soc.*, 79(5), 831–844.

[21] Filippov, A.V., Zurita M., Rosner D.E. (2000). Fractal-like aggregates: Relation between morphology and physical properties. *J. Colloid Interface Sci.*, 229(1), 261–273.

[22] Skorupski, K., Mroczka, J., Wriedt, T., Riefler, N. (2014). A fast and accurate implementation of tunable algorithms used for generation of fractal-like aggregate models. *Physica A*, 404, 106–117.

[23] Van Poppel, L.H., Friedrich, H., Spinsby, J., Chung S.H., Seinfeld J.H., Buseck P.R. (2005). Electron tomography of nanoparticle clusters: Implications for atmospheric lifetimes and radiative forcing of soot. *Geophys. Res. Lett.*, 32(24), 1–4.

[24] Mroczka, J., Wozniak, M., Onofri, F.R.A. (2012). Algorithms and methods for analysis of the optical structure factor of fractal aggregates. *Metrol. Meas. Syst.*, 19(3), 459–470.

[25] Wozniak, M., Onofri, F.R.A., Barbosa, S., Yon, J., Mroczka, J. (2012). Comparison of methods to derive morphological parameters of multi-fractal samples of particle aggregates from TEM images. *J. Aerosol Sci.*, 47, 12–26.

[26] Mroczka, J., Szczuczynski, D. (2013). Improved technique of retrieving particle size distribution from angular scattering measurements. *J. Quant. Spectrosc. Radiat. Transf.*, 129, 48–59.

[27] Wriedt, T., Hellmers, J., Eremina, E., Schuh, R. (2006). Light scattering by single erythrocyte: Comparison of different methods. *J. Quant. Spectrosc. Radiat. Transf.*, 100(1), 444–456.

[28] Nilsson, A.M.K., Alsholm P., Karlsson A., Andersson-Engels S. (1998). T-matrix computations of light scattering by red blood cells. *Appl. Optic*s, 37(13), 2735–2748.

[29] Mroczka, J., Wysoczanski, D. (2000). Plane-wave and Gaussian-beam scattering on an infinite cylinder. *Opt. Eng.*, 39(3), 763–770.

[30] Girasole, T., Gouesbet, G., Grehan, G., Le Toulouzan, J.N., Mroczka, J., Ren, K.F., Wysoczanski, D. (1997). Cylindrical fibre orientation analysis by light scattering. Part 2: Experimental aspects. *Part. Part. Syst. Char*, 14(5), 211–218.

[31] Girasole, T., Bultynck, H., Gouesbet, G., Grehan, G., Le Meur, F., Le Toulouzan, J.N., Mroczka, J., Wysoczanski, D. (1997). Cylindrical fibre orientation analysis by light scattering. Part 1: Numerical aspects. *Part. Part. Syst. Char.*, 14(4), 163–174.

[32] Girasole, T., Le Toulouzan, J.N., Mroczka, J., Wysoczanski D. (1997). Fiber orientation and concentration analysis by light scattering: Experimental setup and diagnosis. *Rev. Sci. Instrum.*, 68(7), 2805–2811.

[33] Skorupski, K., Mroczka, J. (2014). Effect of the necking phenomenon on the optical properties of soot particles. *J. Quant. Spectrosc. Radiat. Transf.*, 141, 40–48.

[34] Skorupski, K., Mroczka, J., Riefler, N., Oltmann, H., Will, S., Wriedt, T. Impact of morphological parameters onto simulated light scattering patterns (2013). *J. Quant. Spectrosc. Radiat. Transf.*, 119, 53–66.

[35] Mroczka, J. (2013). The cognitive process in metrology. *Measurement*, 46(8), 2896–2907.