**Appendix B: A Pseudocode of the Spectral APCSA Method**

The following pseudocode is the main part of the spectral APCSA method. It must be completed with a subroutine to evaluate the first values of the parameters being optimized and inserted within one of two external cycles, to obtain a set of solutions from which the final average values and standard deviations are calculated. In the step 1 of the pseudocode, we indicate in parenthesis the values we used for those parameters fixed at the beginning of the optimization.

**Pseudocode Start** [36-39]:

1. Several parameters need to be fixed at the beginning of the optimization process: mmax is the maximum number of allowed temperature cycles (mmax=5000), jmax is the maximum number of random variations of the parameters being optimized (jmax=1500000), at each temperature cycle, d is the numerical tolerance for the evaluation of weighted partition functions (initially d=0.04), h is a numerical tolerance used to test the solidification criterion (h=5.0x10-6), ND is a number larger than unity used to discretize the space of search (ND=1.15), po is a number somewhat less than unity used to evaluate the initial temperature (po=0.9), Np is the number of parameters per wavelength being optimized (Np=2), N is the number of spectral points (N=301), L is an integer trial number used to generate MP=LNp random variations involved in the evaluations of the frequencies of change, at the beginning of the optimization and after finishing each temperature cycle (L=5). The average number of moves accepted through the Metropolis algorithm is sa. A minimum value must be initially assigned (sa=10). The test of the partition function convergence (line 18) is carried out when sm is larger than sa. In that case, the sa is updated taking the average over the consecutive temperature cycles carried out. Q defines the sensitivity threshold of the steps (Q=0.005). When D(k,i)/P(k,i)<Q the corresponding step involved in the moves is not changed.
2. An initial estimation of the parameters to optimize must be carried out. In the application reported here, Lorenz-Mie theory was used to obtain a first evaluation of msca and mabs for the visible wavelengths considered. The matrix of unknown parameters is initially set: Po(1,i)=msca(li) and Po(2,i)=mabs(li) with i=1,2,…,N. Then, the boundaries of the search domain are defined by means of the matrices Pl(k,i) and Pu(k,i), with k=1,2,…,Np. The boundaries can be treated as fixed or mobile through the whole optimization process.
3. The domain is discretized through the matrix D(k,i)=[Pu(k,i)-Pl(k,i)]/ND with k=1,2, …,Np and i=1,2,…,N.
4. Generate Mp arbitrary initial configurations within the search domain previously defined.
5. Determine an initial average value of the M-merit-function for these Mp evaluations. This value is referred as <Eo> or initial energy-value. Determine the initial energy dispersion of the Mp evaluations (so) around the mean value. The initial temperature of the system is evaluated: To=-so/ln(po) (see Equation (6) in [36]).
6. Calculate an initial matrix of parameters Po(k,i) for k=1,2,…,Np and i=1,2,…,N, and its corresponding energy Eo.
7. Calculate the initial values of the frequencies of change matrix, F(k,i) for k=1,2,…,Np and i=1,2,…,N. By varying each optimization parameter, with the other kept fixed, the average energy value is obtained with the corresponding deviation. The maximum deviation value must be obtained, to evaluate the matrix of frequency of change from the ratio between the dispersion for the kth parameter and the maximum deviation (see Equation (5) in [37]). Normalize to unity the frequencies of change. The normalization is carried out in such a way that, for each parameter to be optimized, the sum of the N frequencies of change is equal to unity.
8. Initialize m=1, j=1, Ej-1=Eo, Emin(m)=1010
9. Do while mmmax 
10. Initialize n=0 (at the preceding temperature)
11. Do k=1,Np 

Do i=1,N 

Generate a random number w[0,1]

If F(k,i)>w generate a random variation (a move) : P(k,i)=Po(k,i)+rD(k,i) where r

is a random number between -1 and 1

End do 

End do 

1. Evaluate the energy of the new configuration Ej and the change of energy DE=Ej-Ej-1
2. If DE<0 then 

the new state (merit function value) is accepted

pj=1 (the acceptance probability is set to unity)

pn=0

Go to 17

Else

1. Apply Metropolis algorithm to determine whether to accept new state
2. Generate pj=exp(-DE/Tm-1) and a random number s[0,1]
3. If pj<s then (the move is rejected) 

P(k,i)=Po(k,i) for k=1,2,…,Np and i=1,2,…,N

Go to line 11

1. Else (the move is accepted)

Po(k,i)=P(k,i) for k=1,2,…,Np and i=1,2,…,N

Ej-1=Ej

nn+1

pn=1

if Ej<Emin(m) then Emin(m)=Ej

Gn=pj and Cn=Ej (vectors G and C are being updated)

If n>3 then 

e1=En-2

e2=En-1

e3=En

End if 

sm=0

Do l=1,n 

sm=sm+pl

End do 

Evaluate the partition function Dn=(1/n)  [(<Em-1>-E*l*)/Tm-1]

If sm<sa Go to 11

sa=0

Do l=1,m 

sa=sa+sm

End do 

sa=integer[sa/m]

1. If n>2, b=|1-Dn/Dn-1|>d and j<jmax then 

jj+1

Go to line 11 (the inner constant temperature cycle continues)

Else

Go to line 19 (out of the temperature cycle)

End if 

1. Statistical analysis from the n-components of the vectors G and C:

Evaluate their average values (<G> and <Em>) and corresponding deviations

(sG and sC)

1. Evaluate DM and compare with DEm,m-1=<Em>-<Em-1>
2. If DEm,m-1>DM and D(k,i)/P(k,i)>Q for k=1,2,…,Np and i=1,2,…,N then 

D(k,i)D(k,i)/ND

Else

d10d

End if 

1. Update the temperature: Tm=-sC/lnpm with pm=poexp(-m2/2sG2)
2. If m>3 then 

If e1<h, e2<h, and e3<h then

Save the matrix elements P(k,i) for k=1,2,…,Np and i=1,2,…,N

Stop

Else

mm+1

Update and normalize to unity the frequency of change matrix F(k,i)

Go to line 10

End if

End if 

End if 

End if 

End do 

**Pseudocode End**

The evaluation of b from the partition functions Dn and Dn-1 (line 18) requires special care. When the temperature decreases and for positive arguments of the exponential function involved in the evaluation of Dn, a numerical overflow can happen. To avoid this, we have written the evaluation of b in the following way [39]:

 . (B1)

The use of an adjustable average value of the number of moves accepted through the Metropolis algorithm (sa), from an initial trial value, allows to prevent the occurrence by chance of movements with low energy dispersion, at the initial states of optimization. When this fact occurs, the temperature corresponding to the first cycle (m=1) is quite low which delays the entire optimization process.