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Detailed study on the role of forward scattering in particle sizing based on spherical particle model

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ABSTRACT

It is detailed studied that whether the forward scattering is suitable for particle sizing by both of the theory analysis and numeric simulation based on spherical particle model. The detailed theory analysis indicates that usually the kernel matrix in the forward region is more ill-conditioned than that in other regions. The numerical simulation also indicates that under the same conditions, the particle sizing results from the forward region are generally much worse than those from other scattering regions. Therefore, we think that in the future particle sizing related studies the forward scattering should be avoided.

1. Introduction

Size-distribution (SD) is a very important parameter of particles. There are many methods to find the SD, among which the scattering method is quite attractive due to its advantages such as quickness and non-contact [1–3].

When a non-polarized parallel incident beam, with the intensity and wavelength being I_0 and λ_{inp} respectively, is incident on the undetermined spherical particles, the scattered radiation distribution can be determined by [4]

$$F_{\lambda_{inp}}(\theta) = I_0 \int_{r_{\min}}^{r_{\max}} P_{\lambda_{inp}}(r,\theta) n(r) dr \tag{1}$$

where θ is the scattering angle of the scattered radiation, r is the particle radius with minimum and maximum values of $r_{\min}$ and $r_{\max}$, respectively, $P_{\lambda_{inp}}(r,\theta)$ is the scattering phase function of the single particle calculated using the Mie scattering theory, and $n(r)$ is the undetermined SD. The most commonly used retrieval model is the independent model [3], which assumes the radius section $[r_{\min}, r_{\max}]$ is divided into M subsections. In every subsection, $n(r)$ is a constant n_i ($i = 1,2,\cdots,M$), so Eq. (1) can be rewritten as (I_0 is supposed to be 1 for simplicity)

$$\begin{pmatrix} F_{\lambda_{inp}}(\theta_1) \\ F_{\lambda_{inp}}(\theta_2) \\ \vdots \\ F_{\lambda_{inp}}(\theta_s) \end{pmatrix} = \begin{pmatrix} P_{\lambda_{inp}}(r_1,\theta_1) & P_{\lambda_{inp}}(r_2,\theta_1) & \cdots & P_{\lambda_{inp}}(r_M,\theta_1) \\ P_{\lambda_{inp}}(r_1,\theta_2) & P_{\lambda_{inp}}(r_2,\theta_2) & \cdots & P_{\lambda_{inp}}(r_M,\theta_2) \\ \vdots & \vdots & \ddots & \vdots \\ P_{\lambda_{inp}}(r_1,\theta_s) & P_{\lambda_{inp}}(r_2,\theta_s) & \cdots & P_{\lambda_{inp}}(r_M,\theta_s) \end{pmatrix} \begin{pmatrix} n_1 \\ n_2 \\ \vdots \\ n_M \end{pmatrix} \tag{2}$$

here the first matrix on the right is the so called "kernel matrix" of this equation (it is called as "P matrix" in this paper). We can measure $F_{\lambda_{inp}}(\theta)$ using a detector, which means n_i can theoretically be retrieved as long as $s \geq M$. However, the P matrix is typically ill-conditioned [4,5], which would cause the retrieval results being very unstable. That is to say, a very small disturbance in the measurement of $F_{\lambda_{inp}}(\theta)$ would cause a very large change in the retrieval of n_i.

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It is obvious that detecting the $F_{\lambda_{inp}}(\theta)$ accurately is very important. It is always regarded that the forward scattering region is an "irreplaceable" region to detect the $F_{\lambda_{inp}}(\theta)$, one reason of which is due to the opinion that "the forward scattering contains a large amount of valuable information of the particle size-distribution" [3,6–8]. Some researchers proposed that the forward region for particle sizing is usually problematic [9], but the forward scattering is still widely used in most the particle sizing related studies and particle analyzers [3,6–8]. Therefore, it is necessary to make clear such a controversial issue that whether the forward scattering is suitable for particle analysis. In this paper, we study this problem in detail through both of theory analysis and numerical simulation based on the spherical particle model.

2. Theory study

2.1. Condition number (CN) analysis

It is known that the CN is a very important parameter that reflects the condition of a matrix, and the larger the CN, the more ill-conditioned the matrix. In our study, it is found that the different angle ranges lead to very different CN values of the P matrix, that is to say, the CN is very sensitive to the angle range selection. In order to study it quantitatively, we suppose four kinds of spherical "sample particles" as follows for detailed study:

Sample particle 1: The complex index is 1.3–0.001i (negative imaginary part represents the absorption of the particles [3]), the radius range is 0.1–5 μm. And according to most references, the radius range [0.1, 5] is divided into 25 sub-sections logarithmically, that means the [lg0.1, lg5] is divided to 25 sub-sections uniformly.
Sample particle 2: The complex index is 1.3–0.001i, the radius range is 0.1–10 μm, and the radius range [0.1, 10] is divided 25 sub-sections logarithmically.
Sample particle 3: The complex index is 1.3–0.001i, the radius range is 0.1–20 μm, and the radius range [0.1, 20] is divided 25 sub-sections logarithmically.
Sample particle 4: The complex index is 1.3–0.001i, the radius range is 0.1–100 μm, and the radius range [0.1, 100] is divided 25 sub-sections logarithmically.

Because the radius range of all the sample particles is divided into 25 sub-sections ($M=25$), therefore, 25 angles are needed for independent retrieval model. Supposing that the incident wavelength is 0.4 μm, with the 25 angles being in different angle ranges, the different CN values of P matrix can be got, as shown in Table 1.

From Table 1, it can be seen that to all the sample particles, the CNs of the P matrices using the forward angles are much larger than that using the other angle ranges.

2.2. Angle optimization analysis

Table 1 also indicates that the CN is so sensitive to the selection of angles, therefore, it is expected to get a better conditioned P matrix by optimizing the angles. Here we optimize the angles by using Genetic Algorithm [10]. First, randomly select 1000 angles in the range of 1°–180° as candidates. The candidates are randomly divided into 40 sets as the initial groups of the GA, with 25 angles in every group. The other evolution conditions of the GA are floating point coding, random crossing points, random mating ratios, a 1% mutation ratio, and the remainder selection rule. The fitness function is

$$Fitness = \min\{cond(P)\} \tag{3}$$

where $cond(P)$ corresponds to the CN of the P matrix and min{} forces fitness to the minimum CN value. For the simulation in this paper, the groups evolved through 800 generations before accepting the angles as optimized. The output angles are shown in Fig. 1. Using the angles in Fig. 1, the CNs of the P matrices of sample particle 1, sample particle 2, sample particle 3 and sample particle 4 can be decreased to be 7.2×10^4, 6.4×10^5, 6.6×10^5 and 5.0×10^6 respectively, which are better than those in Table 1.

From Fig. 1 it can be seen that to all the sample particles, angles in forward region are all discarded by the GA program, indicating that the forward region can worsen the condition of the P matrix.

Table 1
The condition number of P matrix in different angle ranges.

A	S			
	Sample particle 1 C	Sample particle 2	Sample particle 3	Sample particle 4
1°–25° (step size: 1°)	1.2×10^{15}	2.3×10^{15}	7.4×10^{14}	2.9×10^{16}
26°–50° (step size: 1°)	1.2×10^{12}	2.9×10^{12}	4.5×10^{13}	6.4×10^{13}
51°–75° (step size: 1°)	2.4×10^{11}	3.1×10^{11}	1.1×10^{12}	1.8×10^{12}
76°–100° (step size: 1°)	6.7×10^{10}	3.1×10^{11}	1.5×10^{11}	2.1×10^{10}
101°–125° (step size: 1°)	1.4×10^{10}	4.4×10^{10}	9.4×10^{10}	1.3×10^{11}
126°–150° (step size: 1°)	7.4×10^{10}	8.8×10^{11}	9.6×10^{12}	4.7×10^{13}
151°–175° (step size: 1°)	8.3×10^{10}	9.4×10^{11}	8.0×10^{11}	2.4×10^{13}
1°–145° (step size: 6°)	1.5×10^{9}	1.1×10^{10}	6.1×10^{9}	1.1×10^{9}
31°–175° (step size: 6°)	6.2×10^{5}	7.0×10^{8}	6.8×10^{7}	6.8×10^{8}

S—sample particles; C—condition number; A—angle range.