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Comparing the quality of solution of inverse problem in nephelometric and turbidimetric measurements

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The paper presents results of the simulation research aiming at comparison of the quality of reconstruction of particle size distribution of dispersed phase in particulate systems by solving the inverse problem for nephelometric measurement data and for turbidimetric measurement data corrupted to a varying extent by random errors. In the case of both measurement techniques mathematical models based on Mie light scattering theory were applied. The results obtained demonstrated that the reconstruction on the basis of turbidimetric measurements is characterized by generally bigger accuracy compared to the reconstruction on the basis of nephelometric measurements. The advanatage of the reconstruction based on turbidimetric measurement data over the reconstruction based on nephelometric measurement data increases significantly in the case of measurement data of both kinds affected by random noise.

Keywords: inverse problem, light scattering, Mie theory, nephelometry, turbidimetry.

1. Introduction

Nephelometry and turbidimetry are measurement techniques enabling indirect determination of the size distribution function $f(a)$ of particles of the dispersed phase in a particulate system, where a denotes a volume radius of a particle defined as the radius of the sphere of the volume equal to the volume of a particle.

In nephelometry the dependence of the volume scattering function in a particulate system on scattering angle – $\beta(\theta)$ is directly measured. This quantity describes directional distribution of the scattered light intensity in the system examined.

In turbidimetry the dependence of the total extinction coefficient in a particulate system on wavelength – $c(\lambda)$ is directly measured. This coefficient characterizes the relative drop of radiance of parallel beam of monochromatic light shielded from scattered radiation incoming from another directions after traveling a differential distance in a particulate system as a result of absorption and scattering.

Calculation of the function $f(a)$ on the basis of measured function $\beta(\theta)$ or $c(\lambda)$ is an example of the inverse problem in indirect measurements. The problem is based on mathematical models of measurements, which are relations between directly measured dependences and the calculated function $f(a)$. A mathematical model of nephelometric measurements relates the function $\beta(\theta)$ to the function $f(a)$ whereas a mathematical model of turbidimetric measurements relates the function $c(\lambda)$ to the function $f(a)$.

Making proper simplifying assumptions enables formulation of mathematical models of the two types of measurement having the general form of the Fredholm integral equation of the first kind. It permits solving the problem under consideration by means of a series of effective techniques elaborated for the extensive class of inverse problems expressed using this type of equation. The model of nephelometric measurements and the model of turbidimetric measurements expressed this way differ only in the form of kernel function. Distinct properties of the function determine dissimilar fidelity of reconstruction of the function $f(a)$ based on results of nephelometric and turbidimetric measurements. The aim of simulation research discussed in the present work was to determine and compare the quality of reconstruction in both cases for various levels of measurement errors.

2. Mathematical models of measurements

Application of mathematical models providing a rigorous description of the light scattering phenomenon in particulate systems to solving the inverse problem considered in the paper is neither feasible nor expedient because of extreme complexity of these models [1, pp. 9–11]. For this purpose it is necessary to employ less complicated approximate models obtained by making numerous simplifying assumptions.

In the present work, the following simplifying assumptions were made:

– the dispersion medium of a particulate system is homogenous and isotropic,

– the dispersed phase of a particulate system is composed of homogenous and isotropic particles of the spherical shape,

– incident light is a monochromatic, unpolarized, plane wave,

– elastic, single and incohenent scattering occurs in a particulate system.

Thanks to simplifications introduced the general form of mathematical models of both kinds of measurements can be expressed by means of the following Fredholm integral equation of the first kind:

$$g(y) = \int_0^\infty K(y, a) f(a) \mathrm{d}a \tag{1}$$

where: $g(y)$ – the dependence directly measured in a given measurement technique, $K(y, a)$ – kernel function.

Mathematical model of nephelometric measurements and mathematical model of turbidimetric measurements differ only in physical interpretation of function $g(y)$ and the form of function $K(y, a)$.

The form of the function $K(y, a)$ depends on applied model of light scattering on a single particle of dispersed phase. In the present paper, Mie theory of light scattering on spherical particles was used as this model in the case of both measurement techniques considered.

2.1. Mathematical model of nephelometric measurements

In mathematical model of nephelometric measurements, function $\beta(\theta)$ acts as function $g(y)$ in Eq. (1), *i.e.*, the following identities are fulfilled:

$$y \equiv \theta, \quad g(y) \equiv \beta(\theta) \tag{2}$$

The kernel function of the integral Eq. (1) for the model discussed is given by the expression:

$$K(y, a) \equiv K(\theta, a) = N_v \frac{1}{k^2} S_{11}(a, \theta) \tag{3}$$

where: N_v – the number of particles of the dispersed phase per unit volume of the particulate system, $k = 2\pi/\lambda$ – wave number, whereas λ – the wavelength of the scattered light, $S_{11}(a, \theta)$ – an element of the Mueller matrix. In the case of dispersed phase particles of complex refractive index $N_1 = n_1 + ik_1$ and dispersion medium of complex refractive index $N_2 = n_2 + ik_2$, the element $S_{11}(a, \theta)$ of the Mueller matrix is defined by the following equations [1 (pp. 65–66), 2]:

$$S_{11}(a, \theta, m) = \frac{1}{2}\left[\left|S_1(x, \theta, m)\right|^2 + \left|S_2(x, \theta, m)\right|^2\right] \tag{4}$$

where:

$$\begin{aligned} S_1(x, \theta, m) &= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n) \\ S_2(x, \theta, m) &= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n + b_n \pi_n) \end{aligned} \tag{5}$$

In the above-mentioned equations m denotes the relative refractive index of the particle in relation to the medium and is defined by the formula [1 (p. 32), 2]:

$$m = \frac{N_1}{N_2} \tag{6}$$

whereas x denotes the particle size parameter (Mie parameter) defined by the formula [1 (p. 100), 2]:

$$x = \frac{2\pi a N_2}{\lambda} \tag{7}$$

Equations (5) are expansions of the functions $S_1(x, \theta, m)$ and $S_2(x, \theta, m)$ into infinite series of special functions π_n and τ_n dependent solely on scattering angle θ. Special functions π_n and τ_n are defined by the following formulas [1 (p. 94), 2]:

$$\pi_n(\cos\theta) = \frac{P_n^1(\cos\theta)}{\sin\theta}$$
$$\tau_n(\cos\theta) = \frac{\mathrm{d}P_n^1(\cos\theta)}{\mathrm{d}\theta} \tag{8}$$

where $P_n^1(\cos\theta)$ – associated Legendre function of the first kind with degree n and order one. The coefficients in series expansions (5) are quantities a_n and b_n called scattering coefficients. They depend solely on parameters x and m and are defined by equations [1 (p. 101), 2]:

$$a_n = \frac{m\,\psi_n(mx)\,\psi_n'(x) - \psi_n(x)\,\psi_n'(mx)}{m\,\psi_n(mx)\,\xi_n'(x) - \xi_n(x)\,\psi_n'(mx)}$$
$$b_n = \frac{\psi_n(mx)\,\psi_n'(x) - m\psi_n(x)\,\psi_n'(mx)}{\psi_n(mx)\,\xi_n'(x) - m\,\xi_n(x)\,\psi_n'(mx)} \tag{9}$$

in which $\psi_n(\rho)$ and $\xi_n(\rho)$ denotes Riccati–Bessel functions.

Values of the function $S_{11}(a, \theta)$ and intermediate quantities were computed using Bohren and Huffman numerical procedures [1 (pp. 126–129, 477–482)].

2.2. Mathematical model of turbidimetric measurements

In mathematical model of turbidimetric measurements function $c(\lambda)$ acts as function $g(y)$ in Eq. (1), *i.e.*, the following identities are fulfilled:

$$y \equiv \lambda, \qquad g(y) \equiv c(\lambda) \tag{10}$$

The kernel function of the integral Eq. (1) for the model discussed is given by the expression:

$$K(y, a) \equiv K(\lambda, a) = N_v C_{\mathrm{ext}}(a, \lambda) \tag{11}$$

where: $C_{\mathrm{ext}}(a, \lambda)$ – extinction cross-section of a single particle of dispersed phase. The quantity $C_{\mathrm{ext}}(a, \lambda)$ is defined by the formula [1 (p. 103), 2]:

$$C_{\mathrm{ext}} = \frac{2\pi}{k^2}\,\mathrm{Re}\left\{\sum_{n=1}^{\infty}(2n+1)(a_n + b_n)\right\} \tag{12}$$

Coefficients a_n and b_n occurring in expression (12) are the scattering coefficients given by Eqs. (9).

Values of the coefficients a_n and b_n as well as necessary intermediate quantities were computed using Bohren and Huffman algorithm [1 (pp. 126–129, 477–482)].

3. Inverse problem

Determination of the function $f(a)$ on the basis of relation $g(y)$ obtained as a result of direct measurements using a mathematical model of measurements is an example of the inverse problem in indirect measurements.

It can be proved that the problem considered is ill-posed both in the case of nephelometric and turbidimetric measurements. This manifests itself in instability of its solution meaning that the solution is characterized by tremendous uncertainty at even very small uncertainty of measurement data [3, 4]. In other words, a very wide range of functions $f(a)$ exist, which satisfy with given large accuracy the criterion of agreement with measurement data $g(y)$. This indicates that this criterion is insufficient for unique determination of the solution of the inverse problem. Hence, for this purpose, it is necessary to impose on the unknown function $f(a)$ an additional condition dictated by the *a priori* knowledge of the form of the solution [3]. In this connection solving the problem requires application of special mathematical techniques called inverse algorithms, which introduce various forms of *a priori* information.

Numerical inverse methods were solely used in research discussed in the present paper. These techniques seek a solution of the discretized form of the Fredholm integral equation of the first kind, which is obtained by approximating integration in the infinite interval $(0, \infty)$ by numerical integration using rectangles quadrature method in the finite interval $(a_{\min}, a_{\max})$ [2, 3, 5]:

$$\mathbf{g} = \mathbf{K}\mathbf{f} \tag{13}$$

where:

$$\mathbf{g} = \begin{bmatrix} g(y_1) & g(y_2) & \dots & g(y_m) \end{bmatrix}^T$$

$$\mathbf{f} = \begin{bmatrix} f(a_1) & f(a_2) & \dots & f(a_n) \end{bmatrix}^T$$

$$(\mathbf{K})_{ij} = K(a_j, y_i)\Delta a \tag{14}$$

$$a_i = \left(i - \frac{1}{2}\right)\Delta a$$

$$\Delta a = \frac{a_{\max} - a_{\min}}{n}$$

Ill-posedness of the inverse problem manifests itself in this case as ill-conditioning of the **K** matrix [3].

4. Inverse techniques applied

The following linear inverse techniques were employed for solving the inverse problem formulated in a discrete form by Eq. (13):

a) Twomey–Phillips method with minimization of five various measures of the lack of smoothness of the solution sought:

– the square of the Euclidean norm of the vector **f**,

– the sum of squares of the first differences of the vector **f**,

– the sum of squares of the second differences of the vector **f**,

– the sum of squares of the third differences of the vector **f**,

– the square of the Euclidean norm of the difference between the solution vector **f** and the *a priori* assumed trial solution **p**;

b) Two kinds of filtered singular value decomposition (SVD) method:

– truncated SVD method,

– Tikhonov regularization method.

Both groups of inverse algorithms were presented precisely in papers [3, 6].

5. Simulation research

The purpose of simulation research performed was to compare the quality of reconstruction of particle size distribution of dispersed phase in a particulate system realized by solving the inverse problem on the basis of outcomes of nephelometric measurements and turbidimetric measurements corrupted by various amounts of random errors.

The research proceeded in two stages. In the first phase, measurement data were generated by simulation of nephelometric and turbidimetric measurements carried out for the following parameters of a dispersed system:

a) the number of dispersed phase particles per unit volume of a dispersed system $N_v = 10^{15}$ m^{-3},

b) refractive index of dispersed phase particles $N_1 = 1.55 + 0.00i$,

c) refractive index of dispersion medium $N_2 = 1.00 + 0.00i$,

d) the sequence of radii of particles described by the quantities: $a_{\min} = 0.1$ μm, $a_{\max} = 1.0$ μm, $n = 200$,

e) test size distribution of dispersed phase particles given by the formula:

$$f_{\text{test}}(a) = 0.33 f_{\text{norm}}(a, 0.45\mu\text{m}, 0.15\mu\text{m}) + 0.17 f_{\text{norm}}(a, 0.6\mu\text{m}, 0.1\mu\text{m}) + 0.5 f_{\text{lognorm}}(a, 0.1, 0.5) \quad (15)$$

where $f_{\mathrm{norm}}(a, \mu_{\mathrm{norm}}, \sigma_{\mathrm{norm}})$ – the probability density function of the normal distribution of the variable a with the expected value μ_{norm} and the standard deviation σ_{norm}, $f_{\mathrm{lognorm}}(a, \mu_{\mathrm{lognorm}}, \sigma_{\mathrm{lognorm}})$ – the probability density function of the lognormal distribution of the variable a with the parameters μ_{lognorm} and $\sigma_{\mathrm{lognorm}}$.

The simulation of nephelometric measurements was realized on the basis of mathematical model discussed in section 2.1 for the following conditions:

a) the sequence of scattering angles described by the quantities: $m = 181$, $\theta_i = (i - 1)\cdot 1°$ for $i = 1, ..., m$,

b) wavelength $\lambda = 0.6328$ μm.

The simulation of turbidimetric measurements was realized on the basis of mathematical model discussed in section 2.2 for the sequence of $m = 161$ wavelengths: $\lambda_i = 0.25\mu\mathrm{m} + (i - 1)\cdot 0.003125\mu\mathrm{m}$ for $i = 1, ..., m$. The sequence comprised the range of wavelengths from 0.25 μm to 0.75 μm.

Vectors of the measurement data – $\boldsymbol{\beta}$ and $\mathbf{c}$ – obtained as a result of the simulation of nephelometric and turbidimetric measurements, respectively, and denoted further generally as vector $\mathbf{g}$ were subsequently corrupted by the artificially generated additive stationary and uncorrelated Gaussian noise represented by the vector $\boldsymbol{\varepsilon}$, the elements of which are uncorrelated random variables of the normal distribution with zero expected value and equal standard deviation $\sigma_\varepsilon = 1\%\cdot\max(\mathbf{g})$.

The simulations performed yielded the following data:

a) vector $\mathbf{f}_{\mathrm{test}}$ being the discrete representation of the function $f_{\mathrm{test}}(a)$,

b) two matrices $\mathbf{K}$ used in Eq. (13) for computing the vector $\mathbf{g}$ on the basis of the vector $\mathbf{f}_{\mathrm{test}}$,

c) matrix $\mathbf{K}_{\mathrm{neph}}$ corresponding with nephelometric measurements and computed using formulas (2)–(9) and (14),

d) matrix $\mathbf{K}_{\mathrm{tur}}$ corresponding with turbidimetric measurements and computed using formulas (10)–(12) and (14),

e) four vectors of simulated measurement data $\mathbf{g}$ corresponding with size distribution $f_{\mathrm{test}}(a)$:

– vector of results of nephelometric measurements not corrupted by noise $\boldsymbol{\beta}_{0\%}$,
– vector of results of nephelometric measurements corrupted by noise $\boldsymbol{\beta}_{1\%}$,
– vector of results of turbidimetric measurements not corrupted by noise $\mathbf{c}_{0\%}$,
– vector of results of turbidimetric measurements corrupted by noise $\mathbf{c}_{1\%}$.

The second phase of research comprised solving the inverse problem – determination of the vector $\mathbf{f}_{\mathrm{recons}}$, which is the discrete representation of the size distribution $f_{\mathrm{recons}}(a)$ to be reconstructed, on the basis of each of the four vectors of simulated measurement data $\mathbf{g}$ obtained at the first stage of research. In the inversion of each vector $\mathbf{g}$ the same mathematical model of measurements was applied which had been used to generate this vector within the framework of simulation. Thus, to solve the inverse problem for $\boldsymbol{\beta}_{0\%}$ and $\boldsymbol{\beta}_{1\%}$ vectors the previously used $\mathbf{K}_{\mathrm{neph}}$ matrix was applied, whereas for $\mathbf{c}_{0\%}$ and $\mathbf{c}_{1\%}$ vectors the $\mathbf{K}_{\mathrm{tur}}$ matrix was applied. Thanks to that, the obtained vector $\mathbf{f}_{\text{recons}}$ represented values of the function $f_{\text{recons}}(a)$ at exactly the same points, at which the vector $\mathbf{f}_{\text{test}}$ represented the values of the function $f_{\text{test}}(a)$.

For each of the four vectors **g** the inverse problem was solved by means of inverse techniques discussed above: Twomey–Phillips method with minimization of various measures of the lack of smoothness of solution and two schemes of filtered SVD – truncated SVD and Tikhonov regularization. Optimal values of parameters of inverse algorithms were selected individually for each case so as to achieve the best fidelity of reconstruction of the size distribution of dispersed phase particles.

The following quantity was assumed as a measure of accuracy of reconstruction of the size distribution:

$$\Delta = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(f_{\text{recons},\,i} - f_{\text{test},\,i}\right)^2} \tag{16}$$

6. Results of the simulation research

The results of the simulation research are presented in the Table.

From the data collected in the Table it follows that size distributions $f_{\text{recons}}(a)$ reconstructed on the basis of outcomes of simulated turbidimetric measurements are characterized by generally less deviation Δ from the real-test function $f_{\text{test}}(a)$ compared to distributions reconstructed on the basis of outcomes of simulated nephelometric

T a b l e. Comparison of the values of parameter Δ in μm^{-1} characterizing the accuracy of reconstruction of the particle size distribution of the dispersed phase of the dispersed system performed by solving the inverse problem for the results of simulated nephelometric and turbidimetric measurements affected by various relative errors using the inverse procedures discussed.

	Type of measurement, relative error[a]			
Inverse procedure	Nephelometric, $p = 0\%$	Nephelometric, $p = 1\%$	Turbidimetric, $p = 0\%$	Turbidimetric, $p = 1\%$
Twomey–Phillips, norm.	6.89×10^{-2}	3.47×10^{-1}	5.82×10^{-2}	2.00×10^{-1}
Twomey–Phillips, 1st order	7.47×10^{-2}	1.82×10^{-1}	9.69×10^{-2}	9.26×10^{-2}
Twomey–Phillips, 2nd order	1.03×10^{-2}	1.33×10^{-1}	2.84×10^{-3}	2.22×10^{-2}
Twomey–Phillips, 3rd order	4.30×10^{-4}	2.09×10^{-1}	4.07×10^{-4}	3.95×10^{-2}
Twomey–Phillips, aprior.[b]	6.58×10^{-2}	3.36×10^{-1}	3.95×10^{-2}	1.58×10^{-1}
Truncated SVD	4.66×10^{-2}	3.39×10^{-1}	2.50×10^{-2}	1.87×10^{-1}
SVD – Tikhonov regularization	4.65×10^{-2}	3.47×10^{-1}	2.50×10^{-2}	1.99×10^{-1}
Average	4.48×10^{-2}	2.71×10^{-1}	3.54×10^{-2}	1.28×10^{-1}

[a] The relative error p of the measurement data **g** is defined as: $p = [\sigma_\varepsilon/\max(\mathbf{g})]\cdot100\%$.
[b] The *a priori* assumed size distribution function was the constant function $f(a) = 1\ \mu m^{-1}$ represented by the n-element vector $\mathbf{p} = [1 \quad 1 \quad \ldots \quad 1]^T\ \mu m^{-1}$.

measurements. This generalization is confirmed by Δ values averaged over particular inverse techniques applied. In the case of reconstruction of $f(a)$ function on the basis of measurement data not corrupted by noise the mean error of reconstruction using the results of simulated turbidimetric measurements is over 1.26 times less than the mean error of reconstruction using results of simulated nephelometric measurements. For measurement data corrupted by noise the mean error of reconstruction on the basis of outcomes of turbidimetric measurements is over 2.1 times less than the one for reconstruction on the basis of outcomes of nephelometric measurements.

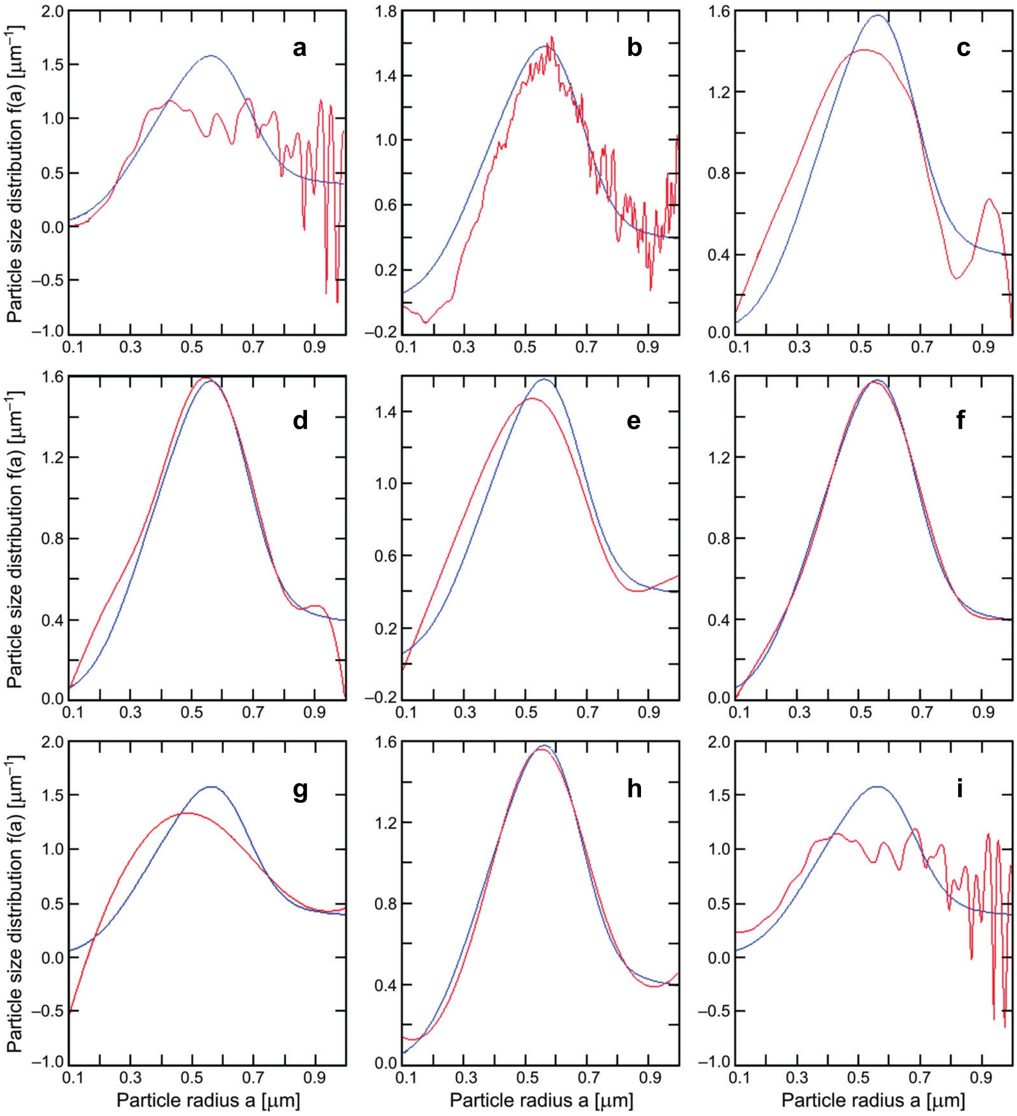

Figure. To be continued on next page.

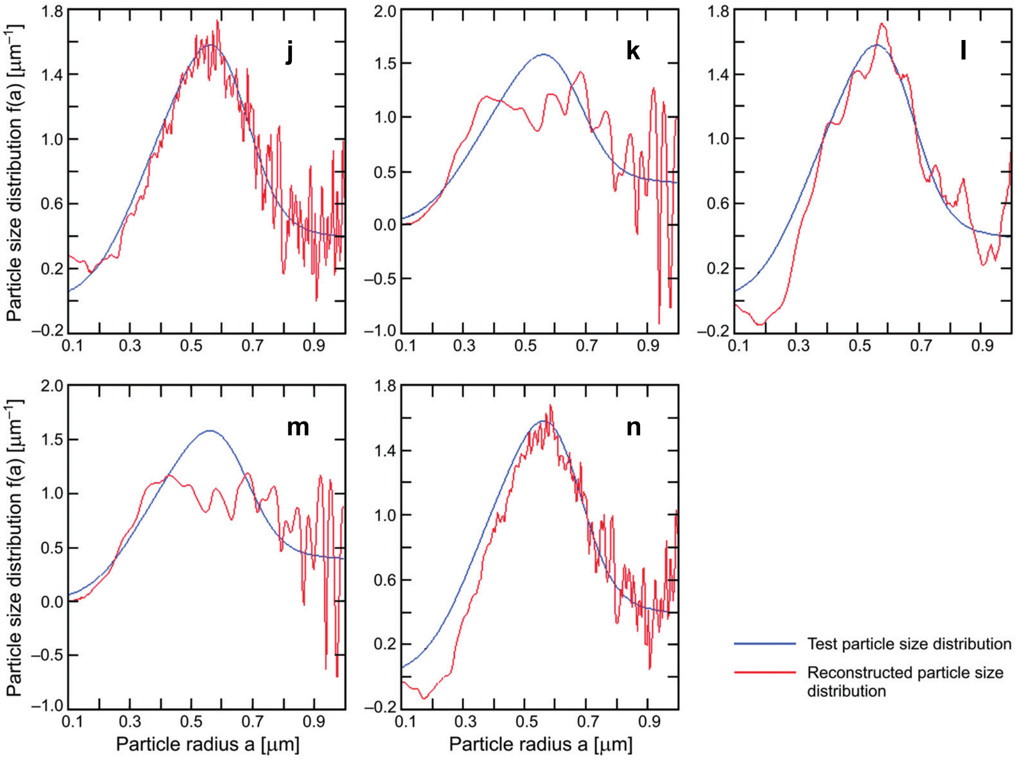

Fig. Particle size distributions of the dispersed phase of the dispersed system reconstructed by solving the inverse problem for the results of simulated nephelometric and turbidimetric measurements for $p = 1\%$ with the use of the inverse procedures discussed: **a** – nephelometric measurements, Twomey–Phillips norm., **b** – turbidimetric measurements, Twomey–Phillips norm., **c** – nephelometric measurements, Twomey–Phillips 1st order, **d** – turbidimetric measurements, Twomey–Phillips 1st order, **e** – nephelometric measurements, Twomey–Phillips 2nd order, **f** – turbidimetric measurements, Twomey–Phillips 2nd order, **g** – nephelometric measurements, Twomey–Phillips 3rd order, **h** – turbidimetric measurements, Twomey–Phillips 3rd order, **i** – nephelometric measurements, Twomey–Phillips aprior. for *a priori* size distribution function $f(a) = 1\ \mu\text{m}^{-1}$ represented by the n-element vector $\mathbf{p} = [1\ \ 1\ \ \ldots\ \ 1]^T\ \mu\text{m}^{-1}$, **j** – turbidimetric measurements, Twomey–Phillips aprior. for *a priori* size distribution function $f(a) = 1\ \mu\text{m}^{-1}$ represented by the n-element vector $\mathbf{p} = [1\ \ 1\ \ \ldots\ \ 1]^T\ \mu\text{m}^{-1}$, **k** – nephelometric measurements, truncated SVD, **l** – turbidimetric measurements, truncated SVD, **m** – nephelometric measurements, SVD Tikhonov regularization, **n** – turbidimetric measurements, SVD Tikhonov regularization.

The Figure proves that size distributions $f_{\text{recons}}(a)$ reconstructed on the basis of outcomes of simulated turbidimetric measurements render the shape of real-test size distribution $f_{\text{test}}(a)$ more accurately than size distributions $f_{\text{recons}}(a)$ reconstructed on the basis of outcomes of simulated nephelometric measurements.

Moreover, it follows both from the Table and from the Figure that the Twomey–Phillips method with minimization of the sum of squares of the first, second and third order differences of the vector **f** performs best with reconstruction of the particle size distribution for both nephelometric and turbidimetric measurements. In the case of turbidimetric measurements corrupted with noise size distributions $f_{\text{recons}}(a)$ reconstructed with use of Twomey–Phillips method with minimization of the sum of squares of the second and third order differences of the vector **f** nearly perfectly agree with real-test size distribution $f_{\text{test}}(a)$.

7. Conclusions

The paper presents the results of the simulation research aiming at comparing the quality of reconstruction of the size distribution of dispersed phase particles in particulate systems realized by solving the inverse problem on the basis of outcomes of nephelometric and turbidimetric measurements corrupted by various amounts of random errors. For both measurement techniques mathematical models based on Mie scattering theory were applied. The results of research proved that reconstruction of particle size distribution on the basis of outcomes of turbidimetric measurements is characterized by generally less error and less susceptibility to harmful effects of ill-posedness compared to reconstruction on the basis of outcomes of nephelometric measurements. The advantage of reconstruction based on results of turbidimetric measurements over reconstruction based on results of nephelometric measurements is more apparent in the case of application of measurement data corrupted by random errors.

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