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Determination of the soot aggregate size distribution from elastic light scattering through Bayesian inference

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ABSTRACT

Recovering the size distribution of aerosolized soot aggregates from multiangle elastic light scattering data requires the inversion of an integral equation, which is a mathematically ill-posed problem. This paper demonstrates how maximum a posteriori (MAP) inference can be used to stabilize the inversion by introducing prior information about the size distribution of the soot aggregates. Results show that the size distribution can be recovered using only simple smoothness and non-negativity priors if the aggregate number density is known, but otherwise it is necessary to specify additional information about the presumed distribution shape.

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1. Introduction

In most combustion processes unburned pyrolyzed fuel forms nanospheres called primary particles, which in turn agglomerate into polydisperse fractal soot aggregates. The impact of these aggregates on human health [1] and the environment [2] is a function of their transport properties and absorption and scattering cross-sections. Given the dependence of these attributes on aggregate morphology, especially the number of primary particles per aggregate, there is a need for instruments that quickly and accurately characterize the size distribution of aerosolized soot aggregates.

One way to do this is by extracting aggregates from the aerosol directly through thermophoresis, and then imaging them using transmission electron microscopy (TEM). This is a time consuming process, however, and the results may be

biased since certain aggregate sizes may be preferentially attracted to the collection slide, among other factors [3]. An alternative is to shine collimated, monochromatic light through the aerosol and infer the aggregate size distribution from the observed angular distribution of scattered light [4]. This method potentially gives greater spatial and temporal refinement than physical probing, and a properly calibrated instrument provides results much faster than can be obtained through TEM imaging.

Most often, determining aggregate sizing through multiangle elastic light scattering is carried out by first assuming that the aggregates obey a prescribed (usually lognormal) distribution type, and then finding the distribution parameters that best explain the data. This is generally done either by analyzing features of the experimental curve formed by plotting the measured angular scattering intensity versus the modulus of the scattering wave vector [4–7], or through a least-squares fitting of the distribution type to experimental data [8,9]. Unfortunately, the solutions produced by these procedures are most often ambiguous, in the sense that a wide range of parameters can accurately reconstruct the data within experimental

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accuracy. This situation arises because the soot aggregate size distribution is related to angular scattering measurements by a Fredholm integral equation of the first-kind

$$g(\theta) = C \int_1^\infty P(N_p) K(\theta, N_p) dN_p \quad (1)$$

where $g(\theta)$ is the measured scattering data, N_p is the number of primary particles per aggregate, $P(N_p)$ is the unknown size distribution of the soot aggregates, $K(\theta, N_p)$ is a kernel function derived from light scattering theory, and C is a scaling parameter. Depending on the properties of the kernel, integral equations of this form often lack a unique solution; instead, an infinite number of candidate aggregate size distributions could be substituted into the integral to recover the observed data.

Consequently, this ill-posed problem must be solved by augmenting the deficient information provided by multi-angle light scattering with assumed attributes of $P(N_p)$, called priors. Since priors are often only a “guess,” and may not describe $P(N_p)$ exactly, they must be assigned an appropriate influence on the recovered solution. For example, enforcing a lognormal distribution for $P(N_p)$ is not ideal since the true distribution may not be exactly lognormal; due to the ill-posed nature of Eq. (1), however, a lognormal distribution may indeed exist that explains $g(\theta)$, leading one to the potentially erroneous conclusion that $P(N_p)$ is lognormal.

A few studies have avoided constraining $P(N_p)$ to a presumed distribution type by transforming Eq. (1) into a matrix equation, $\mathbf{Ax}=\mathbf{g}$, where $\mathbf{x}$ is a discrete form of $P(N_p)$. The ill-posed nature of Eq. (1) makes $\mathbf{A}$ ill-conditioned, hence $\mathbf{x}$ must be solved using regularization. Twomey [10] was the first to do this by developing his own linear and then iterative regularization techniques; the latter technique was consequently improved by Markowski [11]. More recently di Satsio et al. [12] used Tikhonov regularization [13] to recover soot primary particle size distributions from small angle X-ray scattering data.

The main purpose of this paper is to elucidate the ill-posed nature of inferring soot aggregate size distributions from multiangle scattering. In an attempt to address this ill-posedness, a formalized Bayesian methodology called maximum a posteriori (MAP) inference [14] is used to recover a soot aggregate size distribution from multiangle elastic light scattering data. Unlike other methods (e.g. [4–9]) MAP inference explicitly assigns a value to the influence that each prior has on the recovered solution. We start by deriving Eq. (1), follow with a brief review of MAP inference, and then validate the inverse methodology by recovering a prescribed distribution using a synthetic data set. Finally, the method is applied to experimental data and the recovered distribution is compared to TEM data collected under identical operating conditions.

2. Multiangle light scattering from a soot-laden aerosol

Eq. (1) is derived from the radiative transfer equation for the experimental geometry shown in Fig. 1. Signal trapping between the measurement volume and the detector is assumed to be negligible relative to the intensity of scattered light, which is reasonable for optically thin

Fig. 1. Schematic of multiangle elastic light scattering experiment.

flames [8]. The background bleed-through intensity due to soot incandescence is also negligible at the detection wavelengths; accordingly, the detector signal is due only to out-scattering of laser light into the detector direction within the sample volume formed by the intersection of the laser beam and the detector viewing cone

$$g(\theta) = C_{exp} \sigma_{s,\lambda} \frac{w}{\sin(\theta)} \frac{\Phi_\lambda(\theta)}{4\pi} \Delta\omega_{laser} i_{\lambda,laser} \quad (2)$$

where $\Phi_\lambda(\theta)$ is the spectral scattering phase function, $\sigma_{s,\lambda}$ is the spectral scattering coefficient, $i_{\lambda,laser}$ is the laser intensity (assumed here to be vertically-polarized), θ is the angle between the detection and the laser propagation directions, w is the beam width, and $\Delta\omega_{laser}$ is a small but finite solid angle subtended by the laser beam. The constant C_{exp} relates the incident intensity to the detector signal, and accounts for the collection optics and photoelectric conversion efficiencies. Although Eq. (2) assumes a spatially uniform beam profile, the error caused by using a beam with a Gaussian intensity distribution is negligible.

We now relate $\Phi_\lambda(\theta)$ and $\sigma_{s,\lambda}$ to the soot particle morphology through Rayleigh–Debye–Gans Polydisperse Fractal Aggregate (RDG-PFA) theory [7,15]. For vertically polarized laser light, the phase function and scattering coefficient are given by

$$\Phi_\lambda(\theta) = 4\pi \frac{\bar{C}_{vv,\lambda}^{agg}(\theta)}{\bar{C}_{s,\lambda}^{agg}} \quad (3)$$

and

$$\sigma_{s,\lambda} = N_{agg} \bar{C}_{s,\lambda}^{agg} \quad (4)$$

where N_{agg} is the aggregate number density and $\bar{C}_{s,\lambda}^{agg}$ and $\bar{C}_{vv,\lambda}^{agg}$ are the average scattering and vertical polarization cross-sections of the soot aggregates. The product $\sigma_{s,\lambda} \cdot \Phi_\lambda(\theta)$ then becomes

$$\sigma_{s,\lambda} \Phi_\lambda(\theta) = 4\pi N_{agg} \int_1^\infty \bar{C}_{vv}^{agg}(N_p, \theta) P(N_p) dN_p \quad (5)$$

From RDG-PFA theory, the vertical polarization cross-section of a soot aggregate is defined by [7,15]

$$\bar{C}_{vv}^{agg}(N_p, \theta) = N_p^2 C_{vv}^p f[q(\theta) R_g(N_p)] \quad (6)$$

where $q(\theta) \equiv 2\eta \sin(\theta/2)$ is the modulus of the scattering wave vector, R_g is the radius of gyration, $C_{vv}^p = x_p^6 F(\mathbf{m})/\eta^2$ is the polarization cross-section of a primary particle, $\eta \equiv 2\pi/\lambda$ is the wavenumber, λ is the laser wavelength,

$x_p \equiv (\pi d_p)/\lambda$ is the size parameter, d_p is the primary particle diameter, $F(\mathbf{m}) = |(\mathbf{m}^2 - 1)/(\mathbf{m}^2 + 1)|^2$ is the complex scattering function, $\mathbf{m}$ is the complex index of refraction, and $f[\cdot]$ is the form factor.

Collecting all the constants together shows that the detector intensity is related to the size distribution by Eq. (1), where the kernel contains terms that depend on N_p and θ ,

$$K(\theta, N_p) = \frac{N_p^2 f[q(\theta) R_g(N_p)]}{\sin(\theta)} \quad (7)$$

and the remaining terms are absorbed into the coefficient,

$$C = C_{\text{exp}} w \Delta\omega_{\text{laser}} i_{\text{laser}} \frac{N_{\text{agg}} x_p^2 F(\mathbf{m})}{\eta^2} \quad (8)$$

The radius of gyration indicates the size of the soot aggregates. Most flame-synthesized soot aggregates obey the fractal relationship, $N_p = k_g (2R_g/d_p)^{D_f}$, with $k_g = 2.4$ and $D_f = 1.72$ being typical values of the fractal prefactor and dimension [5,9]. This expression is rearranged to give

$$R_g(N_p) = \frac{d_p (N_p/k_g)^{1/D_f}}{2} \quad (9)$$

The form factor, $f[q(\theta) R_g(N_p)]$, has been derived in a number of different ways over the Guinier and power-law regimes. The model for the form factor chosen for this work is [16]

$$f[q(\theta) R_g(N_p)] = \left[1 + \frac{8(qR_g)^2}{3D_f} + (qR_g)^8 \right]^{-D_f/8} \quad (10)$$

which is valid over both regimes. This was compared with the hypergeometric form factor recommended in [4] and found to be orders of magnitude faster in terms of computation time with negligible differences in value.

A comparison of Eqs. (1) and (8) shows that this is a linear inverse problem only if N_{agg} is known. While N_{agg} can be measured directly using a condensation particle counter [17], it is often instead inferred indirectly from the soot volume fraction, f_v , obtained from a line-of-sight attenuation measurement. These two parameters are related by

$$N_{\text{agg}} = \frac{f_v}{\pi d_p^2 N_p} = \frac{f_v}{\pi d_p^2 \int_1^\infty N_p P(N_p) dN_p} \quad (11)$$

The dependence of N_{agg} on $P(N_p)$ means C is also a function of N_p , hence the problem is nonlinear when C is inferred in this way.

Moreover, many of the other parameters in Eq. (8) are often difficult to quantify; C_{exp} and $\Delta\omega_{\text{laser}}$ can be determined by running the experiment without a soot source and calibrating the angular scattering signal to the expected background Rayleigh scattering caused by nitrogen molecules [9], but this is a time-consuming endeavor. A recent survey of the literature [18] also reveals that a large degree of uncertainty prevails over the complex index of refraction, $\mathbf{m}$, and consequently $F(\mathbf{m})$. For these reasons, it is often desirable to treat C as an additional unknown, which again makes the inverse problem nonlinear. We show, later in the paper, how this can be done within the MAP framework.

It should also be noted that RDG-PFA theory only approximates the exact scattering physics by: treating primary particles as Rayleigh scatterers; assuming their electromagnetic fields are spatially uniform and unaffected

by other primary particles, and; by neglecting multiple scattering events [3,19]. Farias et al. [19] compared RDG-PFA to the more accurate integral equation formulation for scattering (IEFS), and concluded that RDG-PFA computes scattering to within 10% accuracy for $|\mathbf{m} - 1| < 1$ and $x_p < 0.3$; for the present case where $x_p \approx 0.35$, uncertainty can exceed 10%. This result is supported by Liu and Snelling [20], who compared RDG-PFA with the more accurate generalized multiparticle Mie (GMM) method [21]. While more accurate IEFS [19], GMM [20], and T-matrix [22,23] simulations have been carried out for select aggregate sizes, however, generation of a sufficient number of aggregates to represent an actual polydisperse aerosol is prohibitively computationally expensive. Furthermore, generating synthetic aggregates also introduces uncertainties and can produce artifacts in the scattering simulation based on how the fractal dimension of the aggregates is maintained during growth. For these reasons RDG-PFA theory remains a standard tool for analyzing multiangle light scattering measurements, although the present method is sufficiently modular that more exact but computational tractable kernels could be used in place of RDG-PFA theory as they become available.

3. Maximum a posteriori inference

One popular method that has been employed to reconstruct the aggregate size distribution is by fitting a lognormal distribution to the elastic light scattering data. The general form of the lognormal distribution is

$$P(N_p) = \frac{1}{N_p \ln(\sigma_g) \sqrt{2\pi}} \exp \left[-\frac{(\ln(N_p) - \ln(N_{p,g}))^2}{2 \ln(\sigma_g)^2} \right], \quad (12)$$

where σ_g is the width of the distribution and $N_{p,g}$ is the geometric mean. In this procedure a set of artificial data, $\mathbf{g}(\Phi)$, is generated by substituting Eq. (12) into Eq. (1) for a given set of distribution parameters, $\Phi = \{N_{p,g}, \sigma_g\}^T$, and then numerically integrating over all possible size classes at the discrete measurement angles, $\{\theta_i\}$. An objective function is then formed from the 2-norm of the residual between the experimental and artificial data

$$f(\Phi) = \|\mathbf{g}(\Phi) - \mathbf{g}^{\text{meas}}\|_2^2 \quad (13)$$

which is minimized using nonlinear programming to yield Φ_{LS} , containing the distribution parameters that best explain the data. Unfortunately the recovered distributions may vary widely due to the inherent ill-posedness of the problem; this can be seen from the plot of $f(\Phi)$ in Fig. 2(a), which reveals a long, narrow valley of solutions, plotted in Fig. 2(b). The inset of Fig. 2(b) shows that, when substituted into Eq. (1), all these solutions reproduce the data within experimental accuracy.

An alternative method is needed to make use of light scattering data to recover the aggregate size distribution in a way that directly addresses the underlying ill-posedness of the problem. Working within the framework laid out in the previous section, Eq. (1) can be transformed into a matrix equation, $\mathbf{Ax} = \mathbf{g}$, by setting an upper limit for the aggregate size, $N_{p,\text{max}}$, and then discretizing N_p into n uniform strips over each of which $P(N_p)$ is assumed to be

uniform, as shown in Fig. 3. Each element of $\mathbf{A}$ is found by numerically integrating over a strip of width ΔN_p

$$A_{ij} = C \int_{N_{p,j}-\Delta N_p/2}^{N_{p,j}+\Delta N_p/2} K(\theta, N_p^*) dN_p^* \quad (14)$$

with $g_i = g(\theta_i)$ and $x_j = P(N_{p,j})$.

Two distinct attributes of this problem complicate its solution: first, the underlying ill-posedness of Eq. (1) causes the $\mathbf{A}$ matrix to be ill-conditioned, such that a small amount of noise in the data can cause large variations in the solution; second, because the number of unknowns, n , usually exceeds the number of measurement angles, m , the $\mathbf{A}$ matrix has a non-trivial nullspace (or “nullity”), which implies that there exists a set of nonzero vectors $\{\mathbf{x}_n\}$ such that $\mathbf{A}\mathbf{x}_n = 0$, giving $\mathbf{A}(\mathbf{x}_{LS} + \mathbf{x}_n) = \mathbf{g}$, where $\mathbf{x}_{LS}$ is the smallest solution that minimizes $\|\mathbf{A}\mathbf{x} - \mathbf{g}\|_2^2$. These characteristics of $\mathbf{A}$ are evident from its singular value decomposition (SVD)

$$\mathbf{A} = \mathbf{U}\mathbf{\Sigma}\mathbf{V}^T = \mathbf{U} \begin{bmatrix} \sigma_1 & & & & \\ & \sigma_2 & & & \\ & & \ddots & & \\ & & & \sigma_m & \\ & & & & 0 \\ & & & & & \ddots \\ & & & & & & 0 \end{bmatrix} \mathbf{V}^T \quad (15)$$

Fig. 2. (a) Contour plot of least-squares objective function and (b) lognormal aggregate size distributions that belong to the locus of solutions. The inset shows reconstructed elastic light scattering data.

Fig. 3. Deconvolution problem is converted into a linear problem by discretizing $P(N_p)$.

where $\mathbf{\Sigma}$ is a diagonal matrix containing the singular values. This decomposition is a powerful tool for diagnosing ill-conditioned linear systems, since the decomposed matrix can be rearranged to give

$$\mathbf{x} = \sum_{j=1}^n \frac{\mathbf{u}_j^T \mathbf{g}}{\sigma_j} \mathbf{v}_j \quad (16)$$

where $\mathbf{u}$ and $\mathbf{v}$ are column vectors of the orthonormal matrices $\mathbf{U}$ and $\mathbf{V}$. The SVD of an $\mathbf{A}$ matrix generated from Eq. (14) for 50 bins and 23 measurement angles reveals that it is rank-deficient, since it only has 23 nonzero singular values that decay continuously over several orders of magnitude, characteristic of a matrix equation generated from a Fredholm integral equation of the first-kind as shown in Fig. 4. In an experimental setting, the data is contaminated with noise, $\mathbf{g} = \mathbf{g}^{exact} + \delta\mathbf{g}$, and

$$\mathbf{x} = \mathbf{x}^{exact} + \delta\mathbf{x} = \sum_{j=1}^n \frac{\mathbf{u}_j^T \mathbf{g}^{exact}}{\sigma_j} \mathbf{v}_j + \sum_{j=1}^n \frac{\mathbf{u}_j^T \delta\mathbf{g}}{\sigma_j} \mathbf{v}_j \quad (17)$$

hence the smallest singular values cause the last perturbation term to dominate the solution. The remaining 27 null singular values (not shown) indicate the nontrivial nullspace of $\mathbf{A}$, and must be excluded from the summation to avoid division by zero.

These factors combined prevent this problem from being solved using conventional linear algebra tools.

Fig. 4. Singular values of $\mathbf{A}$ reveal that it is ill-conditioned and rank-deficient.

Instead, the information provided by $\mathbf{Ax}=\mathbf{g}$ must be augmented with prior assumptions about $P(N_p)$. Maximum a posteriori (MAP) inference provides a formalized and generic way to do this. The basis of this method is Bayes' theorem

$$P(\mathbf{x}|\mathbf{g}) = \frac{P(\mathbf{g}|\mathbf{x})P_{\text{model}}(\mathbf{x})}{P(\mathbf{g})} \quad (18)$$

where $P(\mathbf{x}|\mathbf{g})$ is the probability of $\mathbf{x}$ being correct given an observed data set, $\mathbf{g}$, $P(\mathbf{g}|\mathbf{x})$ is the probability of the data in $\mathbf{g}$ actually occurring for a given $\mathbf{x}$ (also called the likelihood function), $P(\mathbf{g})$ is the marginal probability of the data, and $P_{\text{model}}(\mathbf{x})$ is the probability that the solution is correct based on assumed priors. Combining the likelihood function with prior probabilities results in *posterior probability*, $P(\mathbf{x}|\mathbf{g})$; the goal, then, is to find $\mathbf{x}^*$ that maximizes the posterior probability, usually through nonlinear programming. (As $P(\mathbf{g})$ only scales Eq. (18) it can be neglected.)

Relying solely on multiangle elastic light scattering data without imposing any additional assumptions about the nature of $P(N_p)$ is equivalent to setting $P_{\text{model}}(\mathbf{x})=1$ and maximizing the likelihood function. For a linear model, which assumes $\mathbf{g}=\mathbf{Ax}$, and positing that the data in $\mathbf{g}$ is contaminated with Gaussian-distributed error having the same variance σ^2 for each angle (which can be achieved by rescaling)

$$P(\mathbf{g}|\mathbf{x}) \propto \exp \left[-\frac{1}{2\sigma^2} \|\mathbf{Ax}-\mathbf{g}\|_2^2 \right] \quad (19)$$

Thus $P(\mathbf{g}|\mathbf{x})$ is maximized by minimizing $\|\mathbf{Ax}-\mathbf{g}\|_2^2$, but again, because $\mathbf{A}$ is ill-conditioned there exists a large set of solution distributions that almost minimize $\|\mathbf{Ax}-\mathbf{g}\|_2^2$.

The solution of $\mathbf{x}^*$ must therefore be stabilized by adding informed priors to Eq. (18); since $P(\mathbf{x}|\mathbf{g})$ is the union of the likelihood function and $P_{\text{model}}(\mathbf{x})$, the priors are simply multiplied on to Eq. (18) as they are introduced into the problem. The Gibbs prior is appropriate for problems in which $\mathbf{x}$ should obey a smooth distribution

$$P_{\text{Gibbs}}(\mathbf{x}) \propto \exp \left[-\beta \|\mathbf{Lx}\|_2^2 \right] \quad (20)$$

where $\mathbf{L}$ is a smoothing matrix. One common choice for $\mathbf{L}$ is a discrete approximation of the derivative operator [13], maximizing Eq. (20) when all the elements of $\mathbf{x}$ are identical. The parameter β scales the importance of this prior relative to the likelihood function and other priors.

The elements of the distribution $\mathbf{x}$ should also be strictly non-negative, which is enforced using

$$P_{\text{nonneg}}(\mathbf{x}) = \prod_{j=1}^n H(x_j) \quad (21)$$

where the Heaviside step function, $H(x_j)$, is zero if $x_j < 0$ and is otherwise taken to be unity.

One final source of information is that the aggregation dynamics in the soot aerosol generates a self-preserving distribution, which usually resembles a lognormal distribution [4,24]. Instead of forcing the distribution to be lognormal, as is done in [4–9], we define a prior that promotes a lognormal distribution [25]

$$P_{\text{dist}}(\mathbf{x}) \propto \exp \left[-\alpha \|\mathbf{x}-\mathbf{x}_{\text{dist}}(\Phi^*)\|_2^2 \right] \quad (22)$$

where Φ^* are the parameters of the lognormal distribution that best matches the current estimate of $\mathbf{x}$, found by minimizing the Kolmogorov-Smirnov goodness-of-fit statistic [26]. The discrete lognormal distribution, $\mathbf{x}_{\text{dist}}(\Phi^*)$ is generated by evaluating the best-fit distribution at the center of each strip shown in Fig. 3. Eq. (22) quantifies how closely $\mathbf{x}$ resembles a lognormal distribution, while α determines the emphasis this prior has relative to the likelihood function and the other priors.

Combining these priors with the likelihood function, and defining $\lambda^2 \equiv 2\sigma^2\beta$ and $\gamma^2 \equiv 2\sigma^2\alpha$, gives

$$P(\mathbf{g}|\mathbf{x}) \propto \exp \left[-\|\mathbf{Ax}-\mathbf{g}\|_2^2 - \lambda^2 \|\mathbf{Lx}\|_2^2 - \gamma^2 \|\mathbf{x}-\mathbf{x}_{\text{dist}}(\Phi^*)\|_2^2 \right] \prod_{j=1}^n H(x_j) \quad (23)$$

Due to the monotonicity of the logarithm function, maximizing Eq. (23) is equivalent to solving the constrained nonlinear least-squares minimization problem

$$\mathbf{x}^* = \arg \min_{\mathbf{x}} \left\| \begin{bmatrix} \mathbf{A} \\ \lambda \mathbf{L} \\ \gamma \mathbf{I} \end{bmatrix} \mathbf{x} - \begin{bmatrix} \mathbf{g} \\ 0 \\ \gamma \mathbf{x}_{\text{dist}}(\Phi^*) \end{bmatrix} \right\|_2^2 \quad \text{s.t. } \mathbf{x} \geq 0 \quad (24)$$

where $\mathbf{I}$ is the identity matrix. Eq. (24) is actually a generalization of other methods used to solve this problem: augmenting the likelihood function with Eq. (20) alone is Tikhonov regularization [13]; augmenting the likelihood function with Eq. (22) and solving $\lim_{\gamma \rightarrow \infty} \mathbf{x}^*$, with an initial guess that is a lognormal distribution, is equivalent to least-squares fitting a presumed distribution to experimental scattering data in $\mathbf{g}$.

The types of priors and their influence relative to the likelihood function needed to successfully recover a robust estimate of $P(N_p)$ depends on the quality and quantity of experimental data. In general, these priors should be used only as necessary to stabilize the inverse problem, since they inherently introduce a bias into the solution based on analyst's expectation of the solution characteristics, e.g. that $P(N_p)$ should be lognormal. To that end, a good strategy

is to attempt a solution using the least-presumptuous priors first (non-negativity and smoothness) and turn to an assumed distribution type as a last resort. In the following sections we consider two scenarios: first, one in which C (and hence N_{agg}) is known; and second, one where C is unknown and is instead inferred along with $\mathbf{x}$.

4. Deconvolution of artificial data

First consider the case where C is characterized by some independent means (most likely involving a condensate particle counter, as noted in Section 2, and calibration to match the measured signal to the absolute scattering intensity), in which case Eq. (1) is a linear integral equation of the first-kind. To demonstrate this situation we attempt to recover a prescribed aggregate size distribution, $P(N_p)$ representative of the soot aggregate size distribution we would expect to see in the flame [8] used in the experimental portion of this study, to be discussed later. This distribution was based on TEM images of soot sampled from an ethylene laminar diffusion flame at the location and under the operating conditions described in [8]. A histogram approximation of $P(N_p)$ was generated by analyzing 3300 TEM images of soot aggregates [27] using image processing software and the projected area method [3]. The bins were aggregated to form approximate points on the cumulative distribution function (CDF), to which a lognormal CDF was fitted by least-squares minimization. The resulting lognormal distribution has parameters $\Phi_{TEM} = [N_{p,g,TEM}, \sigma_{g,TEM}]^T = [23.14, 3.35]^T$.

Artificial angular scattering data was generated by substituting the fitted distribution into Eq. (1) and numerically integrating at uniformly spaced measurement angles between 10° and 160° . This data was then contaminated with artificial noise sampled from a normal distribution having a 3% standard deviation relative to the magnitude of each data point, typical of measurement noise observed in the experiment. The $\mathbf{A}$ matrix was formed by taking $N_{p,max}=500$ and splitting the domain into 55 uniformly-spaced bins. The rows of $\mathbf{Ax}=\mathbf{g}$ were then rescaled so that the variances, σ^2 , of the data were uniform.

MAP inference was then implemented with the Gibbs and nonnegativity priors, but without promoting a distribution type through Eq. (22), which is equivalent to first-order Tikhonov regularization with a nonnegativity constraint. While λ can be chosen heuristically, we instead used the L-Curve Criterion method [28], which works by finding the value of λ^* corresponding to the point of maximum curvature on a parametric plot of $\log_{10}\|\mathbf{Lx}_\lambda\|_2^2$ versus $\log_{10}\|\mathbf{Ax}_\lambda - \mathbf{g}\|_2^2$, where $\mathbf{x}_\lambda$ is the constrained Tikhonov solution obtained using a given λ . This value of λ^* was then substituted into Eq. (24), with $\gamma=0$, to find $\mathbf{x}^*$.

Fig. 5 shows that the recovered distribution closely matches the assumed distribution at large N_p , but departs the assumed distribution at smaller N_p . The kernel function is strongly weighted by large aggregate sizes due to the quadratic dependence on N_p in the numerator, implying that less information is conveyed by smaller particles, a well-known limitation of multiangle elastic light scattering experiments [4]. Nevertheless, this result shows that a good estimate $P(N_p)$ may be obtained without assuming a distribution type, as long as C is known.

5. Deconvolution of experimental data

The inverse methodology was next applied to experimental data collected at the same location and under the same experimental conditions described in [8], using the optical apparatus shown in Fig. 6. The laser source was a diode-pumped, Q-switched Nd:YLF laser with a wavelength of 527 nm and average power of 400 mW at 5 kHz. The laser beam was expanded to a 3 mm diameter and passed through a waveplate that controlled the laser energy. The beam then traveled through a polarizer and a second waveplate to control the final polarization on the laser side of the setup, which was vertical. A final lens focused the laser beam to a diameter between 150 and 200 μm . Lastly, before reaching the flame, the beam passed through a shielding apparatus to prevent any errant light scattered by the optical components from reaching the soot source.

Fig. 5. (a) A comparison of specified lognormal size distribution to the distribution recovered using constrained Tikhonov regularization and (b) the L-curve used to choose λ^* .

Fig. 6. Experimental apparatus.

The detection optics was mounted on a rotation stage and aligned to view the center of rotation. The laser beam was steered, via two mirrors, to also intersect the center of rotation. The flame was positioned so that the feature of interest was contained within a sample volume formed by the intersection of the laser beam and the viewing field of the optics. The detector angle was controlled by an aperture of 17 mm diameter mounted 14 cm from the center of rotation, which in turn was followed by a collection lens that collimated the light. This was followed by a polarizer to ensure that the final polarization was vertical before traveling through a focusing lens of the same focal length as the collection lens, focusing the light down to pass through a 50 μm slit aperture. The light then passed through a bandpass filter for 527 nm before reaching the photomultiplier. Scattering data was collected at angles chosen to be linearly spaced in the q -domain ($q=2\eta \sin(\theta/2)$), taking values in $q=1$ intervals from $q=3$ to $q=23$. For completeness, $q=2.08$ ($\theta=10^\circ$) and $q=23.48$ ($\theta=160^\circ$) were also included to make full use of the view angles available, for a total of 23 angular measurements.

As noted above, specifying C in this experiment is often difficult due to the illusive nature of N_{agg} and the large degree of uncertainty associated with the remaining parameters in Eq. (8). Accordingly, in this treatment we consider it an additional unknown to be inferred through the Bayesian procedure.

Following [9], a size distribution was first recovered assuming a lognormal $P(N_p)$, by minimizing Eq. (13); as noted in Section 3, this is equivalent to minimizing Eq. (24) with $\lambda=0$ and $\gamma \rightarrow \infty$. The lognormal distribution found by this procedure is defined by $\Phi_{LS}=[N_{p,g,LS}, \sigma_{g,LS}]^T=[19.4, 1.94]^T$; Fig. 7 shows that, due to the underlying ill-posedness of Eq. (1), this minimum is surrounded by a long,

Fig. 7. Contour plot of the least-squares objective function constructed using experimental data.

shallow valley containing solutions that explain the observed data almost as well as this solution, including the lognormal distribution inferred from the TEM analysis. Also note that Fig. 7 only represents the set of plausible lognormal distributions that could explain the observed data; other distribution shapes are also possible.

Eq. (24) was minimized for $\mathbf{x}$ and C , with $\gamma=0$. Unfortunately, Fig. 8(a) shows that this function lacks a distinct local minimum for C , regardless of λ . In other words, the smoothness and non-negativity priors are insufficient to obtain a robust estimate for $\mathbf{x}^*$ and C^* ; instead, an infinite set of solutions for $\mathbf{x}$ and C exist that explain the observed data subject to these priors, and it is therefore necessary to impose more prior information through Eq. (22). Accordingly, the process was repeated with $\lambda=0.01$ (chosen heuristically) and various values of γ . Fig. 8(b) shows that

robust estimate for C^* emerges for $\gamma > 1$. The corresponding distribution for $\mathbf{x}^*$ is shown in Fig. 9(a), along with the best-fit least-squares distributions to the angular scattering data and the TEM histogram. Both show that the recovered solution is close to the distribution obtained by minimizing Eq. (13), which one would expect given that both rely on the optical scattering data and a unique MAP solution is found only by promoting a lognormal distribution.

The parameters corresponding to the best-fit lognormal to the MAP-inferred solution, i.e. the final Φ^* from the prior in Eq. (22), are also plotted in Figs. 7 and 9. (These coordinates are labeled Φ_{MAP}). Although Fig. 8 shows that robust estimates for C^* and $\mathbf{x}^*$ can be found only by promoting a distribution type, unlike other implementations in which $\mathbf{x}^*$ is forced to be lognormal, MAP inference allows $\mathbf{x}^*$ to deviate from the presumed distribution if information provided by the angular light scattering data and other priors contradicts a lognormal distribution. The agreement between the MAP-derived solution, $\mathbf{x}^*$, and the closest lognormal solution specified by Φ_{MAP} shows that $\mathbf{x}^*$ is

essentially lognormal, however, meaning the prior in Eq. (22) is satisfied.

Fig. 9(b) also shows that the best lognormal fits to the TEM data (Φ_{TEM}), the least-squares lognormal fit to the angular scattering data (Φ_{LS}), and the MAP-derived solution, $\mathbf{x}^*$, produce angular scattering data indistinguishable from the original experimental data when substituted back into Eq. (1); this is also reflected in Fig. 7 by the flat topography in the vicinity of Φ_{TEM} , Φ_{LS} , and the closest lognormal to $\mathbf{x}^*$, Φ_{MAP} . This result underscores the fact that angular scattering data by itself is insufficient to uniquely specify $P(N_p)$, and it is crucial to introduce additional information to the problem through priors.

6. Conclusions

Inferring an aggregate size distribution from multiangle elastic light scattering data involves solving an ill-posed integral equation. In this paper, the integral equation is transformed into an ill-conditioned matrix equation and

Fig. 8. Bayesian objective function $f(\mathbf{x}(C))$ constructed using only smoothness and non-negative priors (a) lacks a distinct minimum, while adding a prior that promotes a lognormal distribution (b) resolves this ambiguity.

Fig. 9. (a) Distributions obtained using Bayesian inference, $\mathbf{x}^*$, Eq. (24); the closest lognormal distribution, Φ_{MAP} ; least-squares minimization assuming a lognormal distribution, Φ_{LS} , Eq. (13), and the TEM histogram; and (b) artificial angular scattering data found by substituting these distributions into Eq. (1) and integrating. Discrete points show experimental data.

solved using MAP inference, which stabilizes the inversion by adding prior information about the solution characteristics. While it is possible to recover $P(N_p)$ using only smoothness and nonnegativity priors if the coefficient C is known, an additional prior that promotes a specified distribution type is required if this parameter is unknown. Accordingly, MAP inference is used to recover a size distribution by promoting a lognormal distribution. The recovered distribution is similar to a distribution obtained by least-squares fitting to angular scattering data, and similar to one obtained from a TEM-derived histogram of aggregate sizes; substituting these distributions into the governing equation produces artificial angular light scattering data nearly indistinguishable from the experimental data.

This similarity highlights the importance of using additional information to mitigate the inherent ambiguity of this inverse problem. In the future more formal ways of combining information from multiple sources, such as TEM data and aerosol dynamics theory, into a MAP inference formalism for recovering soot aggregate size distributions will be derived, which may relax the requirement of a prescribed distribution type. Since these information sources contain their own biases, however, it will be crucial to ensure this is reflected in their influence on the posterior probability estimate.

An additional advantage of transforming the integral equation into an ill-conditioned matrix problem is that it allows direct quantification of the ill-posedness of the inverse problem through the singular values of the matrix, or an equivalent decomposition. In the near future we hope to use this attribute in conjunction with design-of-experiment techniques to develop a tool that finds the set of experimental parameters, such as laser wavelength and detector angles, which maximizes the information content of the light scattering data and minimizes the ill-posedness of the inverse problem.

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