NRC Publications Archive Archives des publications du CNRC

Optimization of measurement angles for soot aggregate sizing by elastic light scattering, through design-of-experiment theory

Burr, D. W.; Daun, K. J.; Thomson, K. A.; Smallwood, G. J.

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below. / Pour consulter la version de l'éditeur, utilisez le lien DOI ci-dessous.

Publisher's version / Version de l'éditeur:

<https://doi.org/10.1016/j.jqsrt.2011.12.004>

Journal of Quantitative Spectroscopy and Radiative Transfer, 113, 5, pp. 355-365, 2011-12-09

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=3cd7ec40-8cea-4623-aa13-e76148a29562>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=3cd7ec40-8cea-4623-aa13-e76148a29562>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

Elsevier Editorial System(tm) for Journal of Quantitative Spectroscopy and Radiative
Transfer
Manuscript Draft

Manuscript Number: JQSRT-D-11-00186R2

Title: Optimization of Measurement Angles for Soot Aggregate Sizing by Elastic Light Scattering,
through Design-of-Experiment Theory

Article Type: Full Length Article

Keywords: Combustion diagnostics; light scattering; particles; inverse analysis

Corresponding Author: Dr. Kyle Daun,

Corresponding Author's Institution: University of Waterloo

First Author: Kyle Daun

Order of Authors: Kyle Daun; David W Burr; Kevin A Thomson; Greg J Smallwood

Highlights

- Inferring soot aggregate sizes from the angular distribution of scattered light is ill-posed
- Design-of-experiment theory is used to find angles that minimize ill-posedness
- Optimized angles do not improve reconstruction accuracy

Optimization of Measurement Angles for Soot Aggregate Sizing by Elastic Light Scattering, through Design-of-Experiment Theory

D. W. Burr^a, K. J. Daun^{a*}, K. A. Thomson^b, and G. J. Smallwood^b

^aDepartment of Mechanical and Mechatronics Engineering, University of Waterloo, 200 University Ave. W., Waterloo ON, Canada, N2L 3G1

^bInstitute for Chemical Process and Environmental Technology, National Research Council Canada, 1200 Montreal Rd. Ottawa ON, Canada, K1A 0R6

*corresponding author
email: kjdaun@uwaterloo.ca
tel: (519) 888 4567 x37871

Abstract

In multiangle elastic light scattering (MAELS) experiments, the morphology of aerosolized particles is inferred by shining collimated radiation through the aerosol and then measuring the scattered light intensity over a set of angles. In the case of soot-laden-aerosols MAELS can, in principle, be used to recover the size distribution of soot aggregates, although this involves solving an ill-posed inverse problem. This paper presents a design-of-experiment methodology for identifying the set of angles that maximizes the information content of the angular scattering measurements, thereby minimizing the ill-posedness of the underlying inverse problem. While the optimized angles highlight the physical significance of the scattering regimes, they do not improve the accuracy of size distributions reconstructed from simulated experimental data.

Keywords: Combustion diagnostics, light scattering, particles, inverse analysis

Introduction

In most combustion processes pyrolyzed fuel molecules coalesce into nanospheres between 10 and 100 nm in diameter, called primary particles, which in turn agglomerate to form soot aggregates. Soot has long been a focus of combustion research, in large part because its formation within gas turbines and automotive engines is closely linked to the performance of these devices [1]. Accordingly, designing the next generation of clean and efficient combustion technology relies on improved soot models, which in turn are predicated on the availability of soot measurements carried out within these devices.

Soot also strongly impacts human health and the environment through mechanisms that depend strongly on aggregate size, which is often quantified by the number of primary particles per aggregate, N_p . For example, small soot particles penetrate more deeply into the lungs and can even cross the pleural membrane into the bloodstream [2]. Climatologists and atmospheric scientists have also assessed soot to be an extremely potent factor in climate change, second only to carbon dioxide [3]; soot deposited on glaciers increases their absorption of sunlight thereby hastening glacial melting [4], for example, and soot in the atmosphere promotes formation of clouds that shield the Earth from solar irradiation, which may drastically alter local climatic patterns [5]. Accordingly, instrumentation for measuring both the size and quantity of soot produced by a combustion device is crucial for assessing its impact on human health and the environment, and its compliance to emissions regulations.

While it is possible to infer the size distribution of aggregates within a soot-laden aerosol through electron microscopy of extracted soot aggregates, such as the image shown in Fig. 1, this process has a number of drawbacks. Perhaps foremost, characterizing the soot aggregate size distribution requires processing thousands of electron micrographs, an extremely time-intensive endeavor that effectively disallows analysis of transient processes. Also, inferring three-

dimensional structural information from 2-D projections of soot aggregates induces certain biases into the results [6]. Finally, obtaining the physical access needed to probe the aerosol is often difficult (for example within a combustion chamber) and the probe itself may have a perturbing effect on the physical and chemical processes occurring in the aerosol.

Optical diagnostics overcome many of the above drawbacks. One such technique is multi-angle elastic light scattering (MAELS) (e.g. [7-11]), which is shown schematically in Fig. 2. In this experiment collimated light, usually from a laser, is shone through the aerosol, and the aggregate size distribution is then inferred from the angular distribution of scattered light. The angular distribution of the scattered light intensity, $g(\theta)$, and the aggregate size distribution, $P(N_p)$, are related by a Fredholm integral equation of the first-kind [11],

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

where C is a scaling coefficient that depends on the experimental apparatus, the aggregate number density, and the optical properties of soot, $K(\theta, N_p)$ is the kernel function derived from electromagnetic theory, and $P(N_p)$ is the probability density for the number of primary particles per aggregate. Predicting the angular distribution of scattered light, $g(\theta)$, for a specified $P(N_p)$ can be done by simply carrying out the integration; this is called the *forward problem*. Solving the *inverse problem*, i.e. recovering $P(N_p)$ from $g(\theta)$, on the other hand, is mathematically ill-posed due to the smoothing action of $K(\theta, N_p)$. When carrying out the integral in Eq. (1), moderate variations in $P(N_p)$ are smoothed by $K(\theta, N_p)$ into comparatively smaller changes in $g(\theta)$; this is particularly the case for changes in $P(N_p)$ at small N_p , which are dominated by light scattered from large aggregates. As a consequence, small perturbations in $g(\theta)$, inevitable in an experimental setting, are amplified into large variations in $P(N_p)$ by the inversion process.

Until recently MAELS experiments were limited to few measurement angles, and consequently a distribution shape (often lognormal, e.g. [8-11], or self-preserving [12]) had to be assumed for the aggregate size distribution. The unknown distribution parameters are then found either through nonlinear regression, or by plotting the normalized scattering intensity as a function of the modulus of the scattering wave vector, $q(\theta) = 4\pi\sin(\theta/2)/\lambda$ where λ is the detection wavelength and identifying features of this curve that correspond to different scattering regimes. This approach is far from ideal, however, since it biases the inferred distribution towards the distribution shape assumed by the analyst, which may not necessarily be correct.

Recent innovations in experimental apparatus [10, 11, 13] have led to the possibility of obtaining a much larger set of angular measurements, which in principle can be used to relax the need to assume a distribution shape. In a previous work [11], we demonstrated that $P(N_p)$ can be estimated by solving a matrix analogue of Eq. (1), $\mathbf{Ax} = \mathbf{b}$, where $\mathbf{b}$ contains the angular scattering measurements, $\mathbf{x}$ is a discrete form of $P(N_p)$ equivalent to a normalized histogram, and $\mathbf{A}$ is derived from $K(\theta, N_p)$. The smoothing property of $K(\theta, N_p)$ makes $\mathbf{A}$ ill-conditioned, which amplifies the noise contaminating the data into a very large error component in the recovered aggregate sizes. In ref. [11] we showed that this issue can be addressed through regularization, in the form of Bayesian priors that promote smoothness, non-negativity, and distribution shapes. (Enforcing a distribution shape, as described above, is an extreme form of regularization.) While regularization stabilizes deconvolution of the MAELS data, it is undesirable since it biases the recovered solution towards the analyst's expectations.

While it has long been known that scattered light measurements made at different angles contain varying degrees of information about $P(N_p)$, a systematic and rigorous method for selecting measurement angles has yet to be proposed. Instead, most researchers either choose

these angles heuristically by trial-and-error, or more simply use uniformly-spaced angles over the angular range afforded by the apparatus. By writing the MAELS problem in matrix form, however, it is clear that the ill-posedness of the underlying experiment can be quantified by the ill-conditioning of $\mathbf{A}$, which in turn depends on the measurement angles. This observation can be used to transform the experimental design problem into a multivariate mathematical minimization/maximization problem, where the goal is to find the set of measurement angles that maximizes the information content of the data, or equivalently, to minimize the ill-posedness of the underlying problem. This paper presents a methodology for finding this set of measurement angles, and compares the performance of these measurement angles to sets that are uniformly-spaced in the θ and q domains. The optimized measurement angles indeed reduce the ill-posedness of the underlying deconvolution problem, and confirm the link between the ill-conditioning of the deconvolution problem, the information content of the light scattering measurements, and the location of the angles with respect to the soot aggregate scattering regimes. Unfortunately, the deconvolution problem is so ill-posed that these improvements are “filtered out” by the minimum regularization needed to obtain a solution under normal experimental conditions.

Derivation of the Governing Equation

The scattered light intensity incident on the detector, $g(\theta)$, is obtained by solving the radiative transfer equation (RTE) [14] for the geometry shown in Fig. 2. This analysis is simplified by a number of assumptions: emission of light by the medium is small relative to the scattered light intensity; the laser intensity, $i_{\lambda 0}$, is vertically polarized and undergoes a single scattering event between the laser and detector, which is reasonable for most laboratory-scale flames; background intensity is negligible; and finally, the laser beam is assumed to have a top-hat profile, i.e. a constant intensity across the beam width.

Combined, these assumptions reduce the RTE to

$$g(\theta) = \sigma_{s,\lambda} \frac{w}{\sin \theta} \frac{\Phi_{\lambda}(\theta)}{4\pi} \Delta\omega i_{\lambda 0} \quad (2)$$

where $\Delta\omega$ is the solid angle subtended by the laser viewed from the measurement volume defined by the intersection of the laser and detection optics, w is the width of the laser beam, $w/\sin\theta$ is the chord length of the ray traveling into the detector subtended by the laser beam, and $\sigma_{s,\lambda}$ and $\Phi_{\lambda}(\theta)$ are the bulk scattering coefficient and scattering phase function of the aerosol. The phase function and scattering coefficient are found by integrating the relevant scattering cross-sections of a soot aggregate containing N_p primary particles over all possible aggregate size classes, weighted by the probability density of the aggregate size class, $P(N_p)$.

The scattering cross-section for an aggregate containing N_p primary particles is approximated using Rayleigh-Debye-Gans Polyfractal Aggregate (RDG-PFA) theory [8], which is premised on the assumption that the primary particles are much smaller than the wavelength of laser light so that each primary particle “sees” a uniform wave field, and that the primary particles scatter independently, i.e. no intracuster multiple scattering. Through these

simplifications Eq. (2) can be rewritten in the form of Eq. (1) with a kernel that depends on N_p and θ [11]

$$K(\theta, N_p) = \frac{N_p^2 f[q(\theta), R_g(N_p)]}{\sin(\theta)} \quad (3)$$

where $R_g(N_p)$ is the radius of gyration of the aggregate, $f(\cdot)$ is the scattering form factor, and $q(\theta)$ is the modulus of the scattering wave vector. Due to the nature of diffusion-limited agglomeration the soot aggregates have a self-similar, fractal-like structure and consequently R_g is related to N_p by a power-law relationship

$$R_g(N_p) = \frac{d_p (N_p/k_f)^{1/D_f}}{2} \quad (4)$$

where d_p is the primary particle diameter, which usually obeys a comparatively narrow distribution in most soot-laden aerosols [15] and can therefore be approximated as constant, and $k_f = 2.4$ and $D_f = 1.72$ are typical values of the fractal prefactor and dimension of soot formed in laminar flames as well as overfire soot formed in turbulent flames.

The form factor has been derived in a number of different ways over the Guinier and power-law scattering regimes in the literature. The model chosen for this work is [16]

$$f[q(\theta), R_g(N_p)] = \left[1 + \frac{8(qR_g)^2}{3D_f} + (qR_g)^8 \right]^{-D_f/8} \quad (5)$$

The remaining terms related to the primary particle size parameter ($\pi d_p/\lambda$), aggregate number density, experimental apparatus, and the bulk optical properties of soot are absorbed into the coefficient C [11].

Solution by Linear Algebra

The majority of techniques described in the literature for recovering an aggregate size distribution from MAELS data rely on specifying a distribution shape for $P(N_p)$ and then performing a nonlinear regression to recover the distribution parameter (e.g.[10]), or by plotting the normalized out-scattering intensity $i_\lambda(\theta)/i_{\lambda 0}$ as a function of the modulus of the scattering wave vector, shown schematically in Fig. 3. (An unscaled out-scattering intensity is found by normalizing the signal, $g(\theta)$ by $\sin(\theta)$, to adjust for the fact that the pathlength of the detection ray transecting the laser beam changes with the detection angle, as shown in Fig. 2.) This curve reveals that light scattering occurs in three distinct regimes: the Rayleigh regime; the Guinier regime; and the power-law regime. An additional transition regime is often identified between the Guinier and power-law regimes. Most information related to the aggregate size distribution is found from the Guinier regime, in which the normalized scattering intensity obeys

$$\frac{i_{\lambda 0}}{i_\lambda(\theta)} \simeq 1 + \frac{1}{3} R_{g,meas}^2 q^2(\theta) \quad (6)$$

where $R_{g,meas}$ would be the soot aggregate radius of gyration that would best explain the MAELS data if the aerosol were assumed monodisperse. The polydispersity of N_p affects the measured scattering in the Guinier and transition regimes [8], and in particular the distribution width can be estimated from the relationship [17]

$$R_{g,meas}^2 = \left(\frac{d_p}{2} \right)^2 k_f^{-2/D_f} \frac{M_{2+2/D_f}}{M_2} \quad (7)$$

where the n th moment of $P(N_p)$ is given by

$$M_n = \int_1^\infty N_p^n P(N_p) dN_p \quad (8)$$

By plotting the normalized scattering intensity and specifying a distribution shape, Eqs. (6) and (7) can be used to recover the unknown distribution parameters.

It would be preferable, however, to recover $P(N_p)$ without imposing a distribution type, since doing so may over-regularize the solution to this inverse problem. In our previous paper [11] we showed that this can be done by first specifying a maximum aggregate size, $N_{p,max}$, beyond which $P(N_p)$ is assumed to be zero. Next the domain of N_p is discretized into n subdomains of uniform width ΔN_p , as shown in Fig. 4, over each of which $P(N_p)$ is assumed to be uniform. If the scattered light is measured over a set of m angles, the deconvolution problem reduces to solving $\mathbf{Ax} = \mathbf{b}$ where $b_i = g(\theta_i)$, $x_j = P(N_{p,j})$ and $\mathbf{A}$ is an $(m \times n)$ matrix having elements defined as

$$A_{ij} = C \int_{N_{p,j}-\Delta N_p/2}^{N_{p,j}+\Delta N_p/2} K(\theta_i, N_p^*) dN_p^* \quad (9)$$

In this paper we choose $n = m$, to simplify the analysis and avoid an over-determined system.

Unfortunately the underlying ill-posedness of the deconvolution problem causes $\mathbf{A}$ to be ill-conditioned. In the context of the discrete system, inferring $P(N_p)$ from experimental MAELS data requires solution of

$$\mathbf{Ax} = \mathbf{A}(\mathbf{x}^{exact} + \delta\mathbf{x}) = (\mathbf{b}^{exact} + \delta\mathbf{b}) = \mathbf{b} \quad (10)$$

where $\mathbf{x}^{exact}$ is $P(N_p)$ evaluated at the center of each ΔN_p domain, $\mathbf{b}^{exact} = \mathbf{Ax}^{exact}$, and $\delta\mathbf{b}$ contains measurement noise and discretization error caused by assuming $P(N_p)$ to be uniform over each subdomain of N_p when deriving $\mathbf{A}$. (Discretization error will make Eq. (10) slightly inconsistent

even in the absence of measurement noise.) The ill-conditioning of $\mathbf{A}$ amplifies $\delta\mathbf{b}$ into a very large error term, $\delta\mathbf{x}$, relative to $\|\mathbf{x}\|$.

The extent of ill-conditioning can be quantified through a singular value decomposition of $\mathbf{A}$, $\mathbf{A} = \mathbf{U}\mathbf{\Sigma}\mathbf{V}^T$, where the column vectors of $\mathbf{U}$ and $\mathbf{V}$ form an orthonormal basis for the data space and the state space (containing $\mathbf{b}$ and $\mathbf{x}$, respectively), and the diagonal matrix $\mathbf{\Sigma}$ contains the singular values, $\{w_j\}$, arranged in decreasing order. Since the inverse of an orthonormal matrix is simply its transpose, the solution to Eq. (10) can be written explicitly as

$$\mathbf{x} = \sum_{j=1}^n \frac{\mathbf{u}_j \cdot \mathbf{b}}{w_j} \mathbf{v}_j = \sum_{j=1}^n \frac{\mathbf{u}_j \cdot \mathbf{b}^{exact}}{w_j} \mathbf{v}_j + \sum_{j=1}^n \frac{\mathbf{u}_j \cdot \delta\mathbf{b}}{w_j} \mathbf{v}_j \quad (11)$$

where $\mathbf{v}_j$ and $\mathbf{u}_j$ are column vectors of $\mathbf{V}$ and $\mathbf{U}$, respectively. The rank-nullity theorem guarantees that all singular values are strictly positive as long as $m \geq n$ and the scattering measurements are independent (which is generally satisfied for distinct measurement angles) but the smoothing property of $K(\theta, N_p)$ causes some of these singular values to be very small. Physically, Eq. (11) reconstructs the low frequency components of $\mathbf{x}$ first and progressively higher-frequency components with additional terms, so the higher $\{\mathbf{u}_j \cdot \mathbf{b}\}$ terms, called Fourier coefficients, should tend to zero if $\mathbf{x}$ is smooth. In order to have a bounded solution, the discrete Picard condition [18] requires the Fourier coefficients to decay faster than the singular values with increasing j . While this is generally satisfied for the unperturbed data, i.e. $\{\mathbf{u}_j \cdot \mathbf{b}^{exact}\}$, the discrete Picard condition is not satisfied for discrete ill-conditioned problems when $\mathbf{b}$ is contaminated with noise. Instead, these small singular values produce an error term (the second term on the right-hand-side of Eq. (11)) that dominates $\mathbf{x}$. However, the fact that the discrete Picard condition is satisfied for the unperturbed condition, and error amplification occurs when

the $\{\mathbf{u}_j \delta \mathbf{b}\}$ terms overwhelm the “true” data, $\{\mathbf{u}_j \mathbf{b}^{exact}\}$, when these later terms become small implies that this information can be excluded from the reconstruction. This approach, a form of regularization, is discussed later in this paper.

The small singular values arise from the fact that the information content of $\mathbf{A}$ is barely sufficient to uniquely specify $\mathbf{x}$ from the observed angular scattering data in $\mathbf{b}$, making the solution susceptible to measurement noise. (Letting $n > m$ results in a singular matrix, in which case $\mathbf{A}$ fails to specify a unique solution.) It is therefore necessary to use regularization, which adds extra information based on the expected solution attributes including smoothness, small magnitude, and non-negativity, to reduce this ambiguity and eliminate the small singular values. The drawback of regularization, however, is that it biases the outcome towards the analyst’s expectations, with little indication in the form of an elevated residual due to the ill-conditioning of $\mathbf{A}$. Consequently, in inverse analysis it is often difficult to discern how reliant the recovered solution is on *a priori* assumptions about the presumed solution attributes.

Optimal Design of Experiments

Expressing the MAELS deconvolution problem in the form of Eq. (11) reveals that the extent of ill-conditioning is determined entirely by $\mathbf{A}$, which in turn depends on the set of measurement angles, $\boldsymbol{\theta}$, once N_p has been specified. It follows, then, that the choice of $\boldsymbol{\theta}$ may play an important role in the performance of the MAELS experiment. Despite this, however, many researchers simply use heuristically-chosen measurement angles that lie between the minimum and maximum measurement angles afforded by the experimental apparatus; an obvious candidate is uniform angular increments between $\theta_{\min}$ and $\theta_{\max}$. In his 2001 review paper, Sorensen [8] notes that $q(\theta)$ has far more physical significance compared to θ , since $1/q(\theta)$

provides a length scale for R_g . Following this argument a second possibility is to choose measurement angles with uniform increments in q , between $q(\theta_{\min})$ and $q(\theta_{\max})$.

The fact that the extent of ill-conditioning can be predicted from the singular values suggests that there may be a more rigorous way to choose measurement angles. The first step is to define an objective function, $F(\boldsymbol{\theta})$, that quantifies the extent of ill-conditioning of the $\mathbf{A}$ matrix. The most obvious approach is to maximize the condition number, $\text{Cond}(\mathbf{A}) = w_{\max}/w_{\min}$, by maximizing the smallest singular value (called *E*-optimality [19]) although this approach only considers the ratio of the largest and smallest singular values, and neglects covariance between the parameters.

A more rigorous choice is *D*-optimality [19], which accounts for parameter covariance. This technique can be derived from the χ -squared function; if the measurements in $\mathbf{b}$ are mutually-independent and each obeys an unbiased normal distribution having a standard deviation σ_i ,

$$\chi^2(\mathbf{x}) = \sum_{i=1}^n \left[\frac{\mathbf{a}_i \cdot \mathbf{x} - b_i}{\sigma_i} \right]^2 \quad (12)$$

where $\mathbf{a}_i$ is the i th row of the $\mathbf{A}$ matrix. Minimizing $\chi^2(\mathbf{x})$ is equivalent to maximizing the likelihood,

$$p(\mathbf{b}|\mathbf{x}) = \prod_{i=1}^n \frac{1}{\sqrt{2\pi}\sigma_i} \exp \left[-\frac{(b_i - \mathbf{a}_i \cdot \mathbf{x})^2}{2\sigma_i^2} \right] \quad (13)$$

while the value of $\chi^2(\mathbf{x}^*)$ indicates the degree of agreement between the data and $\mathbf{b}$ and the most probable solution, $\mathbf{x}^* = \text{argmin}[\chi^2(\mathbf{x})]$; in other words, $\chi^2(\mathbf{x}^*)$ indicates the agreement between modeled and measured data, in the context of unbiased, normally-distributed error contaminating $\mathbf{b}^{exact}$.

If the rows in $\mathbf{Ax} = \mathbf{b}$ are scaled so the data is homoscedastic, i.e. distribution widths of each b_i are equal, then Eq. (12) becomes

$$\chi^2(\mathbf{x}) = \frac{1}{\sigma^2} (\mathbf{Ax} - \mathbf{b})^T (\mathbf{Ax} - \mathbf{b}) \quad (14)$$

which is minimized by $\mathbf{x}^*$. Substituting $\mathbf{x} = \mathbf{x}^* + \delta\mathbf{x}$ into Eq. (14) and simplifying gives

$$\Delta\chi^2(\delta\mathbf{x}) = \chi^2(\mathbf{x}^* + \delta\mathbf{x}) - \chi^2(\mathbf{x}^*) = \frac{1}{\sigma^2} \delta\mathbf{x}^T \mathbf{A}^T \mathbf{A} \delta\mathbf{x} \quad (15)$$

which defines a confidence interval traced out by the vector $\delta\mathbf{x}$ with its tail on $\mathbf{x}^*$ for a given value of $\Delta\chi^2$ corresponding to a tabulated probability. For a specified value of $\Delta\chi^2$, the confidence interval is a hyperellipse in n space; the hyperellipse volume indicates to what extent the measurement noise in $\delta\mathbf{b}$ is amplified into a solution error, $\delta\mathbf{x}$, so the objective of this analysis is to design the experiment so the hyperellipse volume is minimized. Since σ^2 and $\Delta\chi^2$ are constants, the vector $\delta\mathbf{x}$ can only be made small by making $\mathbf{A}^T \mathbf{A}$ as large as possible. This can be done by minimizing the objective function

$$F(\boldsymbol{\theta}) = -\det[\mathbf{A}^T(\boldsymbol{\theta}) \mathbf{A}(\boldsymbol{\theta})] \quad (16)$$

which is equivalent to increasing the singular values of $\mathbf{A}$ through the identity

$$\det[\mathbf{A}^T \mathbf{A}] = \prod_{j=1}^n w_j^2 \quad (17)$$

Geometrically, the hyperellipse corresponding to $\Delta\chi^2 = 1$ has principal axes in the direction of the column vectors of $\mathbf{V}$, scaled by the inverse of the corresponding singular value. Hence, making the singular values as large as possible minimizes the hyperellipse volume.

An instructive 2D example is shown in Fig. 5, corresponding to the linear problem

$$\mathbf{A}\mathbf{x} = \begin{bmatrix} 4 & 2+\varepsilon \\ 2 & 1 \end{bmatrix} \begin{bmatrix} x_1^{exact} + \delta x_1 \\ x_2^{exact} + \delta x_2 \end{bmatrix} = \begin{bmatrix} 2 + \delta b_1 \\ 1 + \delta b_2 \end{bmatrix} = \mathbf{b} \quad (18)$$

where δb_1 and δb_2 are sampled from an unbiased normal distribution of width $\sigma = 0.1$, and ε is a heuristic parameter used to adjust $\mathbf{A}$; changing ε represents changes to the experimental configuration. The case of $\varepsilon = 0$ corresponds to a singular $\mathbf{A}$ matrix since the first row is twice the second row, and the $\Delta\chi^2$ contours in Fig. 5 (a) show that the information content of $\mathbf{A}$ is inadequate to specify a unique solution for $\mathbf{x}$; instead there exists a locus of solutions along the dotted red line that makes $\Delta\chi^2(\delta\mathbf{x}) = 0$. Setting $\varepsilon = 0.002$, shown in Fig. 5 (b), admits a unique solution for $\mathbf{x}$, but the singular values of $\mathbf{A}$ are very small and consequently $\delta\mathbf{x}$ is very large. Making $\varepsilon = 2$, as shown in Fig. 5 (c), increases the singular values of $\mathbf{A}$ and decreases the ellipse volume. The geometric relationship between the ellipse volume corresponding to $\Delta\chi^2 = 1$, the column vectors of $\mathbf{V}$, and the singular values, are shown in Fig. 5 (d).

Optimization of MAELS Measurement Angles

The above methodology is applied to optimize a set of 23 measurement angles for the MEALS experiment described in refs. [10] and [11]. The objective function defined in Eq. (16) is minimized starting from an initial angle set $\boldsymbol{\theta}^0$ filled with uniform angles between 10° and 160° , the minimum and maximum angles permitted by the apparatus. These values are imposed as bound constraints on the minimization procedure.

We initially attempted to minimize $F(\boldsymbol{\theta})$ using Newton's method, but this approach failed due to the multimodal objective function topography. This can be seen in Fig. 6, which is a plot of $F(\boldsymbol{\theta})$ with all angles fixed at their nominal values, $\boldsymbol{\theta}^0$, except θ_{20} , which ranges between 10° and 160° . Accordingly, we switched to simulated annealing; in contrast to gradient-based

methods, which always produce a descent direction at each iteration based on the local objective function curvature, simulated annealing periodically accepts an uphill direction with a probability that increases with a specified annealing temperature. This parameter is initially large enough to allow the algorithm to “hop” out of shallow local minima, but is progressively reduces as the solution hones in on a deep local minimum. The effectiveness of this algorithm is verified by using it to minimize $F(\boldsymbol{\theta})$ with θ_{20} as the only free variable and the rest held at $\boldsymbol{\theta}^0$. Figure 6 shows that simulated annealing appears to find the global minimum of this univariate minimization problem.

The optimal set of angles found by multivariate minimization of $F(\boldsymbol{\theta})$ are shown in Table 1, along with measurement angle sets that are uniformly spaced in the θ and q domains. The singular values of the $\mathbf{A}$ matrix formed by each of the angular sets are plotted in Fig. 7, which verifies that the optimized angles produce larger singular values compared to the other two angular sets; a comparison of the singular values further suggests that using measurement angles uniformly-spaced in the q -domain provide more information about $P(N_p)$ than the same number of measurement angles uniformly-spaced in the θ -domain. The larger singular values should correspond to a smaller hyperellipse volume and less measurement noise amplification.

It is interesting to note that the optimization procedure is independent of the soot aggregate size distribution, which are contained in $\mathbf{b}$, since the underlying ill-conditioned nature of the problem is described entirely by $\mathbf{A}$. At first this may seem counter-intuitive, since in practice experimentalists often choose measurement angles (and detection wavelengths) based on the anticipated aggregate size in the aerosol. The analyst removes this parameter, however, by specifying a maximum aggregate size, $N_{p,max}$, beyond which $P(N_p)$ is expected to be zero. In

other words, the measurement angle optimization seeks to maximize the information content of the data up to $N_{p,max}$, regardless of the true distribution shape.

Evaluation of MAELS Measurement Angles

The performance of the three measurement angle sets are compared by solving synthetic MAELS experiments on two candidate distributions shown in Fig. 8: a lognormal distribution fit to a histogram of aggregate sizes derived from electron micrographs of sampled particles [11, 15]; and a bimodal distribution composed of two superimposed lognormal distributions. The soot aggregates are assumed to have uniform primary particle diameters of $d_p = 29$ nm based on the TEM analysis of Tian et al. [15], and the fractal parameters are $k_f = 2.4$ and $D_f = 1.72$, which as noted above, are typical for many combustion systems. The laser wavelength is taken to be 527 nm.

The analysis is carried out on three different data sets for each of the distributions. First, an “exact” dataset, $\mathbf{b}^{exact}$ is derived by setting $\mathbf{b}^{exact} \equiv \mathbf{A}^{exact} \mathbf{x}^{exact}$, where the elements of $\mathbf{x}^{exact}$ is the specified probability densities, $P(N_p)$, evaluated at the center of each strip as shown in Fig. 4. We define $\mathbf{b}^{exact}$ in this way so that the resulting linear system, $\mathbf{A} \mathbf{x}^{exact} = \mathbf{b}^{exact}$, is consistent, i.e. $\|\mathbf{A} \mathbf{x}^{exact} - \mathbf{b}^{exact}\| = 0$. A second set of data, $\mathbf{b}^{meas}$, is found by substituting $P(N_p)$ into Eq. (1) and carrying out the integration at each measurement angle. The data is then contaminated with random iterates sampled from unbiased normal distributions having a 3% standard deviation. This level of measurement noise, corresponding to a signal-to-noise ratio of 30.65 dB, was found to be typical of the MAELS experiment described in [10] and [11]. The error term, $\delta \mathbf{b} = \mathbf{b}^{meas} - \mathbf{b}^{exact}$, contains both discretization error and measurement error, although the measurement error is much larger in magnitude.

We first attempt to reconstruct $P(N_p)$ for the bimodal distribution using the unperturbed data, $\mathbf{b}^{exact}$, and truncated singular value decomposition (TSVD). This approach is based on Eq. (11), except the last p terms corresponding to the p smallest singular values, are excluded (truncated) from the reconstruction, i.e.

$$\mathbf{x}^p = \sum_{j=1}^{n-p} \frac{\mathbf{u}_j \cdot \mathbf{b}}{\sigma_j} \quad (19)$$

The system produced by the optimal measurement angles requires no truncation ($p = 0$) to reconstruct $\mathbf{x}^{exact}$, but due to perturbations induced by numerical precision the systems produced using uniform increments in the θ - and q -domains require truncation of 8 ($p = 8$) and 7 ($p = 7$) terms, respectively, to reconstruct both distributions. Figure 9 illustrates the relationship between the level of regularization and the reconstruction for the measurement angles uniformly spaced in the θ -domain.

These result supports the hypothesis that the information content of the data collected from optimized measurement angles are superior to the other two measurement sets, and furthermore, measurements obtained using uniform increments in q contain more information about $P(N_p)$ compared to measurements found using uniform θ increments. Some physical insight into why the optimized angles outperform the other two sets is found by superimposing the angles over a plot of the normalized scattering intensity, $i_\lambda(\theta)/i_{\lambda,0}$ versus $q(\theta)$. Figure 10 shows that the uniformly spaced measurement angles in the θ and q domains are clustered towards larger values of q , corresponding to power-law scattering. Scattering in this regime should appear as a straight-line when plotted on a log-log graph, and can be defined from relative few measurements. Therefore, multiple measurements made in this regime provide nearly redundant information. The optimized measurement angles, in contrast, are more densely

concentrated within the Guinier and transition regimes, in which the angular variation of scattering intensity reveals more information about the shape of $P(N_p)$.

Next, we attempt to reconstruct the distributions using the artificial measurement data, $\mathbf{b}^{meas}$, which is far more challenging due to discretization and measurement noise error. In this case we use standard Tikhonov regularization, which augments $\mathbf{Ax} = \mathbf{b}$ with a second equation, $\mathbf{Ix} = 0$, thereby promoting a solution having a small Euclidean norm. (More informative priors, such as the smoothness and distribution shape priors used in [11], are also possible.) The aggregate size distribution is then recovered by solving the linear least-squares problem

$$\mathbf{x}_\lambda = \arg \min \left\| \begin{bmatrix} \mathbf{A} \\ \alpha \mathbf{I} \end{bmatrix} \mathbf{x} - \begin{bmatrix} \mathbf{b} \\ 0 \end{bmatrix} \right\|_2^2 \quad (20)$$

where the regularization parameter, α , determines the influence of the prior assumptions, in this case a small solution norm, on the recovered solution relative to the scattering data. Equation (20) can be written in a form similar to Eq. (11) [18],

$$\mathbf{x}^\alpha = \sum_{j=1}^n \phi_j(\alpha) \frac{\mathbf{u}_j \cdot \mathbf{b}}{w_j} \mathbf{v}_j = \sum_{j=1}^n \frac{w_j}{\alpha + w_j} \frac{\mathbf{u}_j \cdot \mathbf{b}}{w_j} \mathbf{v}_j \quad (21)$$

where $\phi_j(\alpha)$ are filter factors. This formulation highlights the similarity between standard Tikhonov and TSVD, since $\phi_j \approx 1$ when $w_j \gg \alpha$ and $\phi_j \approx 0$ when $w_j \ll \alpha$.

When regularizing a discrete inverse problem, a major challenge is to select a regularization parameter (p or α) that provides a good trade-off between perturbation error, the error associated with amplification of $\delta \mathbf{b}$ due to the ill-conditioning of $\mathbf{A}$, and regularization error, due to the fact that the prior information is inconsistent with the problem physics. If the exact solution is known, the regularization and perturbation errors can be determined through a perturbation analysis of $\mathbf{Ax} = \mathbf{b}$ [18]

$$\mathbf{x}^{exact} - \mathbf{x}^\alpha = \mathbf{x}^{exact} - \mathbf{A}_\alpha^\# (\mathbf{b}^{exact} + \delta \mathbf{b}) \quad (22)$$

which leads to an expression for the total error of $\mathbf{x}$

$$\delta \mathbf{x} = (\mathbf{x}^{exact} - \mathbf{A}_\alpha^\# \mathbf{b}^{exact}) - \mathbf{A}_\alpha^\# \delta \mathbf{b} \quad (23)$$

where $\mathbf{x}^{exact}$ is the true solution, $\mathbf{x}^\alpha = \mathbf{A}_\alpha^\# \mathbf{b} = \mathbf{A}_\alpha^\# (\mathbf{b}^{exact} + \delta \mathbf{b})$ is the regularized solution, and $\mathbf{A}_\alpha^\#$ is the regularized pseudoinverse of the augmented matrix for a given α , which can be found from a singular value decomposition. The perturbation error is given by $\varepsilon_{pert} = \|\mathbf{A}_\alpha^\# \delta \mathbf{b}\|$, the regularization error by $\varepsilon_{reg} = \|\mathbf{x}^{exact} - \mathbf{A}_\alpha^\# \mathbf{b}^{exact}\|$, and the total error by $\varepsilon_{tot} = \|\delta \mathbf{x}\|$. The perturbation error also includes discretization error due to the discretization scheme used in Eq. (9) to form $\mathbf{A}$.

Figure 11 shows the perturbation error, regularization error, and total error found using various levels of standard Tikhonov regularization to recover the bimodal distribution shown in Fig. 8 for a sample dataset, $\mathbf{b}^{meas}$, while Fig. 12 shows the solution recovered using the optimal level of regularization, α^* . As noted above, perturbation error decreases monotonically with increasing α , while regularization error follows the opposite trend. The total error, which is approximately the sum of these two components, has a minimum at an intermediate value of α .

We compare the performance of the three sets of measurement angles by using Tikhonov regularization to recover the solution from randomly perturbed datasets. In practice one would employ a parameter selection technique to choose the regularization parameter, for example L-curve curvature [20], the discrepancy principle [21], or generalized cross-validation [22], but in this case since $\mathbf{x}^{exact}$ is known we find α^* by minimizing the total error, which represents the best possible scenario for Tikhonov regularization. Table 3 shows the average error components for the three angular sets and both trial distributions generated from 1000 randomly-perturbed data vectors, while Fig. 13 shows a scatter plot of α^* versus $\varepsilon_{tot}(\alpha^*)$ for the bimodal distribution. In

contrast to the unperturbed case, the optimized measurement angles present no clear advantage over the other two measurement sets when the data is perturbed with discretization error and measurement noise.

Insight into why optimizing the measurement angles fails to improve reconstruction accuracy is gained by plotting $\mathbf{u}_j \cdot \mathbf{b}^{exact}$ and the expected value of $\mathbf{u}_j \delta \mathbf{b}$, $E(\mathbf{u}_j \cdot \delta \mathbf{b})$, as shown in Fig. 14 for the case of the bimodal distribution. As noted above, both TSVD and standard Tikhonov work by filtering out high frequency components of the solution, and only retaining terms that satisfy $\mathbf{u}_j \cdot \mathbf{b}^{exact} \gg \mathbf{u}_j \cdot \delta \mathbf{b}$. Inspection of Fig. 14 shows that this condition is only satisfied for the first few terms ($j < 6$), while Fig. 7 shows that improvements to the singular values realized by optimal experiment design occurred for $j > 14$. In order to reconstruct the solution generated using $\mathbf{b}^{meas}$, then, the regularization algorithm must truncate all terms corresponding to singular values that were improved by design-of-experiment; moreover, the aggregate size distributions are likely to be smooth, due to the nature of the collision and separation processes within the aerosol, so the exact solution is unlikely to have high frequency components, which is reflected by the fact that a reasonable reconstruction can be obtained through Tikhonov regularization. A third set of lines, corresponding to $\mathbf{u}_j \cdot \delta \mathbf{b}^{disc}$ where $\delta \mathbf{b}^{disc}$ is the error caused by discretizing the problem, shows that even in the absence of measurement noise, discretization error negates any advantage gained through the optimum measurement angles.

Conclusions and Future Work

Soot aggregate sizing through MAELS involves solving an ill-conditioned matrix equation. This ill-conditioning must be suppressed through regularization, but regularization itself introduces a bias into the recovered solution based on the analyst's expectation of the solution attributes. This

paper showed how design-of-experiment theory can be used to derive an optimal set of angles that maximizes the information content of the light scattering data, thereby minimizing the ill-posedness of the underlying problem. The outcome of the design-of-experiment highlights the physical connection between the conditioning of the coefficient matrix, $\mathbf{A}$, and the distribution of the measurement angles relative to the scattering regime. Unfortunately, improvements realized through this procedure are negated by the regularization needed to recover the problem when the data is contaminated with perturbation and regularization error.

In this problem, optimizing measurement angles was ineffective due to the extreme ill-posedness of this inverse problem. In this study the design-of-experiments only considered the ill-conditioning of $\mathbf{A}$ as a design metric. In practice, however, uncertainties in the optical properties of soot, the fractal model of soot aggregates, and light scattering theory (particularly the well-known limitations of RDG-PFA theory for larger soot aggregates [23]) impose severe limitations on the accuracy of the recovered results. The methodology presented in this paper can be extended to mitigate these uncertainties. If each uncertainty is assumed to obey a specified (usually normal) distribution, it can be treated as an additional source of “effective measurement noise,” which will influence the choice of measurement angles [24]. The maximum likelihood approach used here also facilitates combination of multiple measurement techniques in a mathematically-rigorous way based on associated experimental uncertainties; we will shortly be investigating using this technique to integrate MAELS and laser-induced incandescence experiments.

References

- [1] Lefebvre AH. Gas Turbine Combustion. Boca Raton: CRC; 1999.
- [2] Pope CA, Burnett RT, Thun MJ, Calle EE, Krewski D, Ito K, Thurston GD. Lung Cancer, Cardiopulmonary Mortality, and Long-Term Exposure to Fine Particulate Air Pollution. JAMA: the journal of the American Medical Association. 2002;287:1132.
- [3] Solomon S, Qin D, Manning M, Chen Z, Marquis M, Averyt K, Tignor M, Miller H. Ipcc 2007: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. New York: Cambridge University Press; 2007.
- [4] Hansen J, Nazarenko L. Soot Climate Forcing Via Snow and Ice Albedos. Proceedings of the National Academy of Sciences of the United States of America. 2004;101:423.
- [5] Wild M. Global Dimming and Brightening: A Review. Journal of Geophysical Research. 2009;114:D00D16.
- [6] Brasil A, Farias T, Carvalho M. A Recipe for Image Characterization of Fractal-Like Aggregates. Journal of Aerosol Science. 1999;30:1379.
- [7] Sorensen C. Light Scattering by Fractal Aggregates: A Review. Aerosol Science and Technology. 2001;35:648-87.
- [8] Sorensen C, Lu N, Cai J. Fractal Cluster Size Distribution Measurement Using Static Light Scattering. Journal of Colloid and Interface Science. 1995;174:456-60.
- [9] Iyer SS, Litzinger TA, Lee SY, Santoro RJ. Determination of Soot Scattering Coefficient from Extinction and Three-Angle Scattering in a Laminar Diffusion Flame. Combustion and Flame. 2007;149:206-16.

- [10] Link O, Snelling D, Thomson KA, Smallwood GJ. Development of Absolute Intensity Multi-Angle Light Scattering for the Determination of Polydisperse Soot Aggregate Properties. *Proceedings of the Combustion Institute*. 2010;33:847-54.
- [11] Burr DW, Daun KJ, Link O, Thomson KA, Smallwood GJ. Determination of the Soot Aggregate Size Distribution from Elastic Light Scattering through Bayesian Inference. *JQSRT*. 2010;112:1099-107.
- [12] Friedlander S, Wang C. The Self-Preserving Particle Size Distribution for Coagulation by Brownian Motion. *Journal of Colloid and Interface Science*. 1966;22:126-32.
- [13] Oltmann H, Reimann J, Will S. Wide-Angle Light Scattering (WALS) for Soot Aggregate Characterization. *Combustion and Flame*. 2010;157:516-22.
- [14] Howell JR, Siegel R, Menguc MP. *Thermal Radiation Heat Transfer*, 5th Ed. Boca Raton: CRC Press; 2010.
- [15] Tian K, Liu F, Thomson KA, Snelling DR, Smallwood GJ, Wang D. Distribution of the Number of Primary Particles of Soot Aggregates in a Nonpremixed Laminar Flame. *Combustion and Flame*. 2004;138:195-8.
- [16] Yang B, Koçlu UO. Soot Processes in a Strongly Radiating Turbulent Flame from Laser Scattering/Extinction Experiments. *JQSRT*. 2005;93:289-99.
- [17] Khlebtsov NG, Melnikov AG. Structure Factor and Exponent of Scattering by Polydisperse Fractal Colloidal Aggregates. *Journal of Colloid and Interface Science*. 1994;163:145-51.
- [18] Hansen PC. *Rank-Deficient and Discrete Ill-Posed Problems: Numerical Aspects of Linear Inversion*. Philadelphia: SIAM; 1998.
- [19] Steinberg DM, Hunter WG. *Experimental Design: Review and Comment*. *Technometrics*. 1984; 26:71-97.

- [20] Hansen PC, O'Leary DP. The Use of the L-Curve in the Regularization of Discrete Ill-Posed Problems. *SIAM J Sci Comput.* 1993;14:1487-503.
- [21] Morozov VA. On the Solution of Functional Equations by the Method of Regularization. *Sov. Math. Dokl.* 1966;7:414-417.
- [22] Golub GH, Heath M, Wahba G. Generalized Cross-Validation as a Method for Choosing a Good Ridge Parameter. *Technometrics.* 1979; 21:215-223.
- [23] Farias TL, Köylü, ÜÖ, Carvalho MG, Range of Validity of the Rayleigh-Debye-Gans Theory for Optics of Fractal Aggregates, *Applied Optics.* 1996; 35: 6560-6567.
- [24] Emery A, Fadale T. Design of Experiments Using Uncertainty Information. *Journal of Heat Transfer.* 1996;118:532-9.

List of Tables

Table 1: MAELS measurement angles.

Table 2: Minimum regularization required to reconstruct $\mathbf{x}$ with $\delta\mathbf{b} = 0$.

Table 3: Reconstruction error components using optimal amount of standard Tikhonov regularization.

List of Figures

Fig. 1: Transition electron micrograph of a soot aggregate.

Fig. 2: Schematic of a MAELS experiment.

Fig. 3: Plot of the normalized scattering intensity versus the modulus of the scattering wave vector.

Fig. 4: Discretization of $P(N_p)$.

Fig. 5: Example of optimal design of experiments, corresponding to Eq. (18).

Fig. 6: Plot of $f(\theta)$ allowing θ_{20} to vary while other angles are held at their nominal values.

Fig. 7: Singular values of $\mathbf{A}$ matrices generated using measurement angle sets.

Fig. 8: Aggregate size distributions used to evaluate the measurement angle sets: (a) lognormal, and (b) bimodal distributions.

Fig. 9: Reconstructions of the bimodal distribution obtained from $\mathbf{b}^{exact}$ using various levels of TSVD regularization, for uniformly-spaced measurement angles.

Fig. 10: Plot of measurement angles (as q) versus normalized scattering intensity: (a) uniform θ increments; (b) uniform q increments; and (c) optimized angles.

Fig. 11: Perturbation, regularization, and total errors found using various levels of Tikhonov regularization to recover the bimodal distribution.

Fig. 12: Solution recovered from $\mathbf{b}^{meas}$ using standard Tikhonov regularization with optimal parameter selection, for the bimodal distribution.

Fig. 13: Total errors and optimal regularization parameters for reconstructing the bimodal distribution using noised data.

Fig. 14: Fourier coefficients and singular values for the bimodal distribution.

Table 1

Uniform θ increments [degrees]	Uniform q increments [degrees]	Optimized [degrees]
10	10	10
23.0	14.5	11.8
29.6	19.3	13.7
36.1	24.2	15.3
42.6	29.1	16.7
49.1	34.1	18.1
55.7	39.2	19.5
62.2	44.4	20.8
68.7	49.6	22.2
75.2	54.9	23.7
81.7	60.4	25.4
88.3	66.1	27.1
94.8	71.9	29.1
101.3	78.0	31.4
107.8	84.3	34.0
114.3	90.9	37.1
120.9	98.0	40.9
127.4	105.7	45.8
133.9	114.0	52.2
140.4	123.5	61.3
147.0	134.6	75.6
153.5	149.4	102.0
160	160	160

Table 2

	Lognormal	Bimodal
Uniform θ	$p = 8$	$p = 8$
Uniform q	$p = 7$	$p = 7$
Optimal	$p = 0$	$p = 0$

Table 3

	Meas. angles	ϵ_{pert}	ϵ_{reg}	ϵ_{tot}
Normal distribution	uniform θ	0.0088	0.0174	0.0179
	uniform q	0.0083	0.0186	0.0194
	optimal	0.0084	0.0202	0.0208
Bimodal distribution	uniform θ	0.0032	0.0108	0.0107
	uniform q	0.0032	0.0108	0.0107
	optimal	0.0032	0.0109	0.0108

Fig 1

Fig2

Fig 3

Fig4

Fig 5

Fig 6

Fig 7

Fig 8

Fig 9

Fig 10

Fig 11

Fig 12

Fig 13

Fig 14