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On photon correlation measurements of colloidal size distributions using Bayesian strategies

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Abstract

In this paper the evaluation of particle size distribution using photon correlation spectroscopy according to the method of regularization of first kind integral equation including Laplace transform by means of Bayesian strategy is presented. We shall convert the Laplace transform to first kind integral equation of convolution type, which is an ill-posed problem. Then we use the Bayesian regularization method to solve it. This type of problem plays an important role in the field of photon correlation spectroscopy, fluorescent decay, sedimentation equilibrium, system theory and in other areas of physics and applied mathematics. The method is applied to test problems taken from the literature and it gives a good approximation to the true solution. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

In photon correlation spectroscopy, the polydispersity effects of macromolecules in solution or colloidal suspensions have been studied extensively. There have been many approaches to analyze the autocorrelation function of quasielastically scattered light. In most experiments in the field of photon correlation, the output of the experiment is the Laplace transform of an unknown distribution function $\phi(t)$. The Laplace transform is converted to an integral equation of the first kind of convolution type and we study the regularization of integral equation by means of Bayesian technique which is similar to Phillips–Tikhonov regularization for ill-posed problems.

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Ill-posed inverse problems have become a recurrent theme in modern sciences, for example, crystallography [11], geophysics [1], medical electrocardiograms [9], meteorology [20], radio astronomy [12], reservoir engineering [13] and tomography [24]. Corresponding to this broad spectrum of fields of applications, there is a wide literature on different kinds of inversion algorithms for evaluating the inverse problems.

The basic principle common to all such methods is as follows: seek a solution that is consistent both with observed data and prior notions about the physical behavior of the phenomenon under study. Different authors have employed different methods such as the method of regularization [22], maximum entropy [12,16], quasi-reversibility [14] and cross-validation [25,15].

The problem of the recovery of a real function $\phi(t)$, $t \geq 0$, given its Laplace transform

$$\int_0^{\infty} e^{-pt} \phi(t) dt = g(p) \quad (1.1)$$

for real values of p , is an ill-posed problem and, therefore, affected by numerical instability. Regularization methods have been discussed by Varah [23], Essa and Delves [8], Wahba [25], Eggermont [7], Thompson [21], Ang [2], Rudolf [18], Beretro [3] and Brianzi [4].

2. Fredholm equation of convolution type

We shall convert the Laplace transform into the first kind integral equation of convolution type with the following substitution in Eq. (1.1):

$$p = a^x \quad \text{and} \quad t = a^{-y} \quad \text{where } a > 1. \quad (2.1)$$

Then

$$g(a^x) = \int_{-\infty}^{\infty} \log a e^{-a^{x-y}} \phi(a^{-y}) a^{-y} dy. \quad (2.2)$$

Multiplying both sides of (2.2) by a^x , we obtain the convolution equation

$$\int_{-\infty}^{\infty} K(x-y)F(y) dy = G(x), \quad -\infty \leq x \leq \infty, \quad (2.3)$$

where

$$\begin{aligned} G(x) &= a^x g(a^x) = pg(p), \\ K(x) &= \log a a^x e^{-a^x} = \log a p e^{-p}, \\ F(y) &= \phi(a^{-y}) = \phi(t). \end{aligned} \quad (2.4)$$

Eq. (2.3) occurs widely in applied sciences. K and G are known kernel and data functions, respectively, and F is to be determined. We shall assume that G, K and F lie in suitable function spaces, such as $L_2(\mathbb{R})$ so that their Fourier transforms (FTs) exist.

Note: $\hat{}$ denotes FTs and $\check{}$ denotes inverse FTs.

3. Description of the proposed Bayesian method

We assume that the support of each function F, G and K is essentially finite and contained within the interval $[0, T]$, where the period $T = N/h$, N is the number of grid points and h is the spacing.

Let T_N denote the space of trigonometric polynomials of degree at most N and period T . Let G and K be given at N equally spaced points $x_n = nh$, $n = 0, 1, 2, \dots, N - 1$ with spacing $h = T/N$. Then G and K are interpolated by G_N and $K_N \in T_N$ where

$$G_N(x) = \frac{1}{N} \sum_{q=0}^{N-1} \hat{G}_{N,q} \exp(i\omega_q x), \tag{3.1}$$

$$\hat{G}_{N,q} = \sum_{n=0}^{N-1} \exp(-i\omega_q x_n) G_N(x_n), \tag{3.2}$$

$$G(x_n) = G_n = G_N(x_n) \tag{3.3}$$

and

$$\omega_q = \frac{2\pi q}{T}.$$

Similar expressions as (3.1) and (3.2) can be obtained for K_N . In our procedure we have used cardinal B -splines and worked in Fourier space to simplify the computation.

Let F be approximated by

$$F_M(x) = \sum_{j=0}^{M-1} \alpha_j B_j(h; x), \tag{3.4}$$

where $B_j(h; x)$ are periodic cubic cardinal B -splines with period $T = Mh$ and knot spacing h . M is the number of B -splines. The vector $\alpha = (\alpha_0, \alpha_1, \dots, \alpha_{M-1})^T$ is to be determined. Following Schoenberg [19], we have

$$B_j(h, x) = Q\left(\frac{x}{h} - j - 2\right), \tag{3.5}$$

where

$$Q(x) = \frac{1}{6} \sum_{k=0}^4 (-1)^k \binom{4}{k} (x - k)_+^3 \tag{3.6}$$

since $B_0(h, x)$ is periodic on $(0, T)$. It has the Fourier series

$$B_0(h, x) = \frac{1}{T} \sum_{q=-\infty}^{\infty} \hat{B}_{0q} \exp(i\omega_q x) \tag{3.7}$$

and

$$\begin{aligned}\hat{B}_{0q} &= \int_0^T B_0(h, x) \exp(-i\omega_q x) dx \\ &= h \left[\frac{\sin(h\tilde{\omega}_q/2)}{(h\tilde{\omega}_q/2)} \right]^4 \\ \hat{\omega}_q &= \begin{cases} \omega_q, & 0 \leq q < N/2, \\ \omega_{N-q}, & 1/2N \leq q \leq N-1. \end{cases}\end{aligned}\quad (3.8)$$

Furthermore, since $B_j(h, x)$ is simply a translation of $B_0(h, x)$ by an amount jh , we have

$$\hat{B}_{jq} = \hat{B}_{0q} \exp(-i\omega_q jh), \quad q = 0, \pm 1, \pm 2, \dots, \quad j = 0, 1, 2, \dots, M-1. \quad (3.9)$$

The spline in Eq. (3.4) has the Fourier series

$$F_M(x) = \frac{1}{T} \sum_{q=-\infty}^{\infty} \hat{F}_{M,q} \exp(i\omega_q x) \quad (3.10)$$

with Fourier coefficients

$$\hat{F}_{M,q} = \sum_{j=0}^{M-1} \alpha_j \hat{B}_{jq}. \quad (3.11)$$

Consider the smoothing functional

$$C(F_M; \lambda) = \left\| \frac{1}{\sigma} [K_N(x) * F_M(x)] \right\|_2^2 + \lambda \|F_M''(x)\|_2^2, \quad (3.12)$$

where $\|\cdot\|$ denotes the inner product norm on $L_2(0, T)$ and λ is the regularization parameter to be evaluated. Since $K_N * F_M \in T_N$ for any square integrable periodic function F_M of period T , Plancherel's theorem gives

$$\left\| \frac{1}{\sigma} (K_N * F_M - G_N) \right\|_2^2 = \frac{T}{N^2 \sigma^2} \sum_{q=0}^{N-1} |\hat{K}_{N,q} \hat{F}_{M,q} - \hat{G}_{N,q}|^2 \quad (3.13)$$

and

$$\|F_M''\|^2 = \frac{1}{T} \sum_{q=-\infty}^{\infty} \tilde{\omega}_q^4 |\hat{F}_{M,q}|^2, \quad (3.14)$$

where

$$|\hat{F}_{M,q}|^2 \sim \tilde{\omega}_q^{(-8)} \quad \text{as } |q| \rightarrow \infty.$$

The infinite series clearly converges.

Now to express functional (3.12) in a matrix form, we define the matrices

$$\hat{P}(N \times N): T\hat{P}_{qr} = \frac{\sqrt{T}}{N\sigma} \delta_{qr} \quad q, r = 0, 1, 2, \dots, N-1,$$

$$\hat{K}(N \times N): \hat{K}_{qr} = \hat{K}_{N,q} \delta_{qr} \quad q, r = 0, 1, 2, \dots, N-1,$$

$\hat{B}(M \times N)$: \hat{B}_{jq} as in Eq. (3.9), $j = 0, 1, \dots, M - 1$,

$W^{(1)}(N \times M)$: $W^{(1)} = \hat{K}(\hat{B})^T$,

$W^{(2)}(N \times M)$: $W_{js}^{(2)} = \frac{1}{T} \tilde{\omega}_s^2 B_{js}$. (3.15)

We can write (3.12) as

$$C(F_M; \lambda) = C(\underline{\alpha}; \lambda) = \|\hat{P}(W^{(1)}\underline{\alpha} - \hat{G})\|_2^2 + \lambda \|W^{(2)}\underline{\alpha}\|_2^2, \tag{3.16}$$

where $\|\cdot\|_2^2$ denotes the vector 2-norm in \mathbb{C}^N and $\hat{G} = (G_{N,0}, \hat{G}_{N,1}, \dots, \hat{G}_{N,n-1})^T$ or

$$\begin{aligned} \underline{U} &= W^{(1)H}(\hat{P})^2 \hat{G}, \\ W &= W^{(1)H}(\hat{P})^2 W^{(1)}, \end{aligned} \tag{3.17}$$

and

$$V = W^{(2)H} W^{(2)}$$

$C(\underline{\alpha}; \lambda)$ has a unique minimum at

$$\underline{\alpha} = (W + \lambda V)^{-1} \underline{U}. \tag{3.18}$$

4. Special properties of W and V

It is easy to show that the r sth element of W is

$$\begin{aligned} W_{rs} &= \frac{T}{N^2 \sigma^2} \sum_{q=0}^{N-1} |\hat{K}_{N,q} \hat{B}_{0q}|^2 \exp(i\omega_q(r-s)h), \quad r, s = 0, 1, 2, \dots, M - 1 \\ &= \sum_{q=0}^{N-1} a_q \exp\left(\frac{2\pi}{M}(r-s)iq\right) \end{aligned} \tag{4.1}$$

where

$$a_q = \frac{T}{N^2 \sigma^2} |\hat{K}_{N,q} \hat{B}_{0q}|^2. \tag{4.2}$$

It follows that W is a circulant matrix. Since $W_{jk} = W_{rs}$, if $j - k = (r - s) \pmod{M}$, and W is also a hermitian matrix.

Similarly V is a circulant hermitian matrix, with

$$V_{rs} = \sum_{q=0}^{N-1} b_q \exp\left(\frac{2\pi}{M}iq(r-s)\right), \tag{4.3}$$

where

$$b_q = \frac{1}{T} |\tilde{\omega}_q^2 \hat{B}_{0q}|^2. \tag{4.4}$$

It is well known that the modal matrix ψ of any $M \times M$ circulant matrix has elements

$$\psi_{rs} = \frac{1}{\sqrt{M}} \exp\left(\frac{2\pi i}{M} rs\right) \quad (4.5)$$

under this normalization ψ is unitary,

$$\psi \psi^H = \psi^H \psi = I. \quad (4.6)$$

Thus if W and V have real eigenvalues μ_s and v_s , respectively, and $s = 0, 1, 2, \dots, M-1$, we may write

$$\begin{aligned} W &= \psi D_W \psi^H, \\ V &= \psi D_V \psi^H, \end{aligned} \quad (4.7)$$

where $D_W = \text{diag}(\mu_s)$, $D_V = \text{diag}(v_s)$.

We then have $(W + \lambda V)^{-1} = \psi \Lambda \psi^H$ where

$$\Lambda = \text{diag}\left(\frac{1}{\mu_s + \lambda v_s}\right). \quad (4.8)$$

We now show that the eigenvalues μ_s and v_s are simply related to the coefficients a_q and b_q defined in Eqs. (4.2) and (4.4). Consider the eigenvalue equation

$$\sum_{n=0}^{N-1} W_{mn} \psi_{ns} = \mu_s \psi_{ms}. \quad (4.9)$$

Using (4.1) the LHS is

$$\begin{aligned} &\sum_{n=0}^{M-1} \sum_{q=0}^{N-1} \exp\left[\frac{2\pi i}{M} q(m-n)\right] \psi_{ns} \\ &= \frac{1}{\sqrt{M}} \sum_n \sum_q a_q \exp\left[\frac{2\pi i}{M}(q(m-n)) + ns\right] \\ &= \frac{1}{\sqrt{M}} \sum_q \left\{ a_q \left(\exp\left(\frac{2\pi i}{M} mq\right) \sum_n \exp\left(\frac{2\pi i}{M}(s-q)n\right) \right) \right\}, \end{aligned}$$

since

$$\sum_{n=0}^{M-1} \exp\left[\frac{2\pi i}{M} jn\right] = \begin{cases} M, & j \equiv 0 \pmod{M} \\ 0, & \text{otherwise.} \end{cases}$$

The LHS of (4.9) is

$$M \sum_{q=0}^{N-1} a_q \psi_{mq} = \left(M \sum_{q=0}^{N-1} a_q \right) \psi_{ms},$$

where $q = s \pmod{M}$. Hence

$$\mu_s = M \sum_{q=0}^{N-1} a_q, \quad q \equiv s \pmod{M},$$

$$v_s = M \sum_{q=0}^{N-1} b_q, \quad q \equiv s \pmod{M}.$$

5. Calculation of λ and α

The r th element of vector U is

$$U_r = \sum_{q=0}^{N-1} c_q \exp \left[\frac{2\pi i}{M} qr \right], \quad r = 0, 1, 2, \dots, M - 1, \tag{5.1}$$

where

$$c_q = \frac{T}{N^2 \alpha^2} \tilde{K}_{N,q} \hat{G}_{N,q} \hat{B}_{0q}. \tag{5.2}$$

We assume that α^2 is known a priori and may be estimated by

$$\sigma^2 = \frac{1}{N(N - 2\ell)} \sum_{q=\ell}^{N-(\ell+1)} |\hat{G}_{N,q}|^2, \quad \ell \simeq \frac{N}{4}. \tag{5.3}$$

It is clear that premultiplication of a \mathbb{C}^M vector by ψ^H is equivalent to an M -dimensional DFT. We may thus write $\hat{\underline{\alpha}} = \psi^H \underline{\alpha}$ and $\hat{U} = \psi^H U$. From Eqs. (3.18) and (4.8), therefore, we have

$$\hat{\underline{\alpha}} = \hat{\underline{U}}. \tag{5.4}$$

Hence,

$$\hat{\alpha}_s = \frac{\sqrt{M} \hat{U}_s}{M(\mu_s + \lambda v_s)}, \tag{5.5}$$

where

$$\hat{U}_s = \sqrt{M} \sum_{q=0}^{N-1} c_q, \quad q = s \pmod{M}. \tag{5.6}$$

The regularization parameter λ in (5.5) is to be determined. In order to evaluate the optimal value of λ , consider the a priori c.d.f.

$$P(\underline{G}|\lambda) = \int_{\mathbb{R}_N} P(\underline{G}|\underline{\alpha}) P_\lambda(\underline{\alpha}) d\underline{\alpha}. \tag{5.7}$$

Bayes' theorem then gives a posteriori c.d.f.

$$P(\lambda|\underline{G}) = \text{Const. } P(\underline{G}|\lambda) P(\lambda) \tag{5.8}$$

in terms of an unknown a priori p.d.f. $P(\lambda)$ for λ . It can be shown that

$$P(\underline{G}|\lambda) = \left[\det \left(\frac{1}{2\pi} (\hat{P})^2 \right) \det [\lambda W (W + \lambda V)^{-1}]^{1/2} \exp \left[-\frac{1}{2} C(\underline{\hat{x}}, \lambda) \right] \right]. \tag{5.9}$$

Substituting this in Eq. (5.8), we find that a condition for a stationary point of $P(\lambda|\underline{G})$ is

$$\frac{d}{d\lambda} [\log P(\lambda)] + \text{Trace}[W(W + \lambda V)^{-1}] - \lambda \underline{\hat{x}}^H V \underline{\alpha} = 0. \tag{5.10}$$

An optimal value of λ maximizes $P(\lambda|\underline{G})$. Now if the unknown distribution $P(\lambda)$ is sufficiently “narrow”, then the effect of the first term in (5.10) is neglected and we determine λ by solving

$$\text{Trace}[W(W + \lambda V)^{-1}] - \lambda \underline{\hat{x}}^H V \underline{\alpha} = 0 \tag{5.11}$$

which reduces to [21],

$$\sum_{s=0}^{N-1} \frac{\mu_s}{\mu_s + \lambda v_s} - \lambda \sum_{s=0}^{N-1} \frac{|\hat{U}_s|^2 v_s}{(\mu_s + \lambda v_s)^2} = 0. \tag{5.12}$$

We obtain λ , the regularization parameter from (5.12). Knowing λ , $\underline{\alpha}$ may then be calculated from the inverse DFT of Eq. (5.5) as

$$\underline{\alpha} = \psi \underline{\hat{x}}.$$

6. Calculation of solution vector \underline{F}

We take $M = N/2$, the number of cardinal cubic B -splines is equal to half the number of grid points. Then

$$\hat{U}_s = \sqrt{M}(c_s + c_{M+s}),$$

$$\mu_s = M(a_s + a_{M+s}),$$

$$v_s = M(b_s + b_{M+s}), \quad 0 \leq s \leq M - 1, \quad s \equiv q \pmod{M}$$

and

$$\alpha_{-1} = \alpha_{M-1}, \quad \alpha_0 = \alpha_M \quad \text{and} \quad \alpha_1 = \alpha_{M+1},$$

$$F_M(2j) = \sum_{j=0}^{M-1} (\alpha_{j-1} + 4\alpha_j + \alpha_{j+1})/6, \tag{6.1}$$

$$F_M(2j + 1) = \sum_{j=0}^{M-1} (\alpha_{j-1} + 23\alpha_j + 23\alpha_{j+1} + \alpha_{j+2})/48.$$

7. The choice of $M = N/2$ is optimal

Natterer [17] has shown that if we discretize an integral equation of the first kind in a certain way using a very specific mesh together with projection onto a suitable space of piecewise polynomials,

then the same order of accuracy in the numerical solution may be obtained as that given by Tikhonov regularization with an optimal choice of regularization parameter. Natterer’s method has become known as “regularization by coarse discretization”. What is done in practice is to mix coarser discretization with Tikhonov regularization. A certain amount of regularization is achieved from the choice of mesh and the rest is obtained via filtering.

Since we are dealing with basis expansions, we must reduce the dimension of the spaces to coarsen the discretization. Consider $M < N$, $\lambda = 0$. Let $A(M)$ be the $N \times N$ matrix satisfying

$$\mathbf{G}_{N,0} = A(M)\mathbf{G}_N, \tag{7.1}$$

where

$$G_{N,0}(x) = \int_0^1 K_N(x - y)F_{M,0}(y) dy. \tag{7.2}$$

Taking the discrete Fourier transform (DFT) of (7.1), we have

$$\hat{\mathbf{G}}_{N,0} = \hat{A}(M)\hat{\mathbf{G}}_N,$$

where $\hat{A}(M)$ is a diagonal $N \times N$ matrix with M unit entries and zeros elsewhere.

Following Wahba [25] and Mair [15], we may minimize the predictive mean square signal error with respect to M . This means we can minimize

$$V(M) = \frac{(1/N)\mathbf{G}_N^H(I - A(M))\mathbf{G}_N}{[(1/N)\text{Trace}(I - A(M))]^2}, \tag{7.3}$$

i.e.,

$$V(M) = \frac{(1/N) \sum_{q=M}^{N-1} |\hat{G}_q|^2}{(1 - M/N)^2}. \tag{7.4}$$

Since we are dealing with FFTs, it is natural to consider $M = \frac{1}{2}N, \frac{1}{4}N, \frac{1}{8}N$, etc., where N is a power of 2. In particular, we have

$$V(\frac{1}{2}N) = \frac{4}{N} \sum_{q=\frac{1}{2}N}^{N-1} |\hat{G}_{N,q}|^2, \tag{7.5}$$

$$V(\frac{1}{4}N) = \frac{16}{9N} \sum_{q=\frac{1}{4}N}^{N-1} |\hat{G}_{N,q}|^2$$

when the decay of $|\hat{G}_{N,q}|^2$ with increasing q is sufficiently large; in particular,

$$\frac{2}{N} \sum_{q=\frac{1}{2}N}^{N-1} |\hat{G}_{N,q}|^2 < \frac{16}{9N} \sum_{q=\frac{1}{4}N}^{N-1} |\hat{G}_{N,q}|^2.$$

From (7.5) we have

$$V(\frac{1}{2}N) < V(\frac{1}{4}N) < \dots .$$

Table 1

Problem	a	T	h	σ	λ	$\ F - F_\lambda\ _\infty$	Figures
1	10.0	12.50	0.1952	0.0085	$0.3499 \cdot 10^{-12}$	0.003	1
2	5.0	11.50	0.1828	0.0074	$0.362 \cdot 10^{-9}$	0.004	2
3	10.0	12.20	0.1906	0.0027	$0.11 \cdot 10^{-5}$	0.07	3

If the successive ratios between the means

$$\frac{2}{N} \sum_{\frac{1}{2}N}^{N-1}, \quad \frac{4}{N} \sum_{\frac{1}{4}N}^{N-1}, \quad \frac{8}{N} \sum_{\frac{1}{8}N}^{N-1}, \dots$$

are each sufficiently small, we have

$$V(\frac{1}{2}N) < V(2^{-r}N). \quad (7.6)$$

It requires only a modest rate of decay for (7.6) to be satisfied. Therefore, the choice $M = \frac{1}{2}N$ is optimal out of the set $M = 2^{-r}N$.

8. Numerical result

In this section we tabulate the results of the above method applied to the test problems taken from the literature. All data functions have the property $g(p) = O(p^{-1})$ and no noise is added apart from the machine rounding error; only optimal results have been quoted in the table and demonstrated in the diagrams. In each of the test problems $N = 64$, the sample points to calculate the Fourier coefficients.

Problem 1 This problem has been taken from Cristina [6].

$$g(p) = \frac{1}{(p + 1.5)^2},$$

$$\phi(t) = te^{-1.5t}.$$

The optimal results are shown in Table 1 and Fig. 1.

Problem 2 This problem has been taken from Gabuti [10].

$$g(p) = \frac{\beta}{(p + \alpha)^2 + \beta^2},$$

$$\phi(t) = e^{-\alpha t} \sin \beta t,$$

where $\alpha = 5.0$ and $\beta = 2.2$.

The optimal results are shown in Table 1 and Fig. 2.

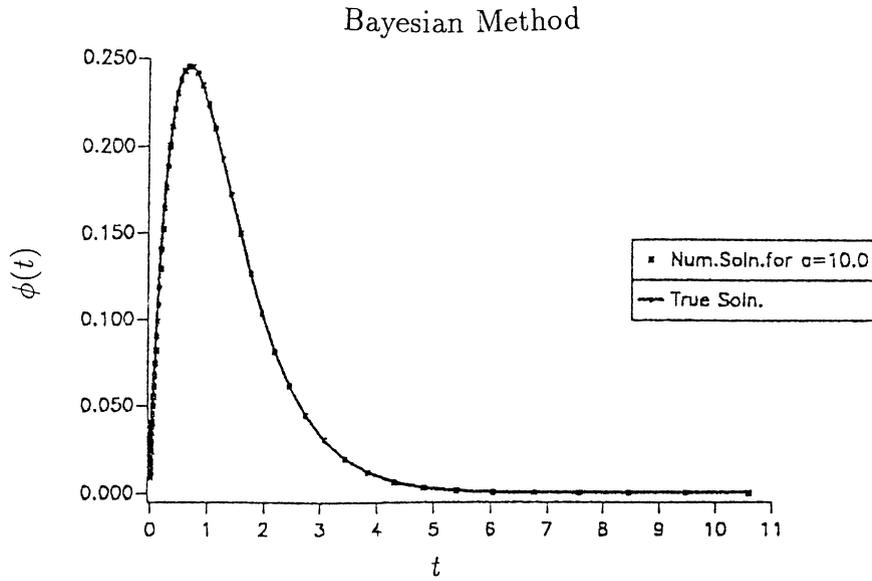


Fig. 1.

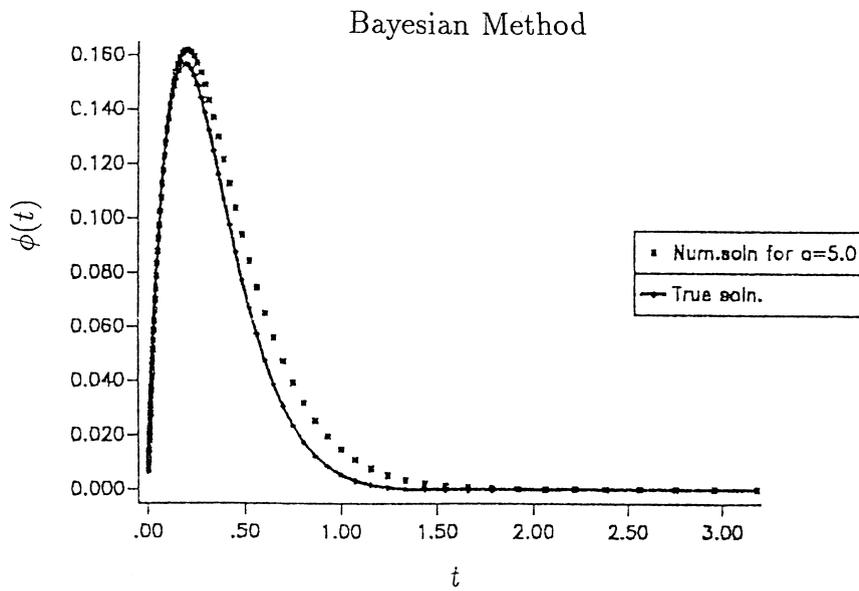


Fig. 2.

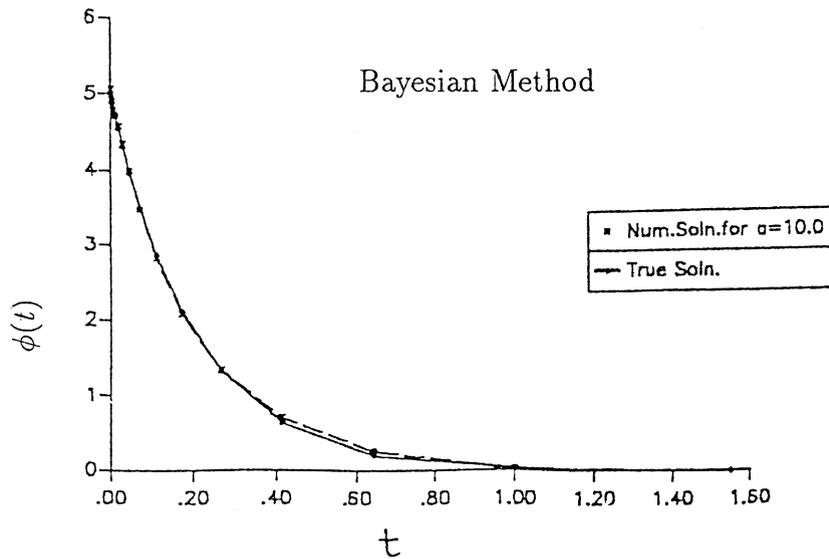


Fig. 3.

Problem 3. This problem has been taken from Chauveau [5].

$$g(p) = \frac{\lambda}{\lambda + p},$$

$$\phi(t) = \lambda e^{-\lambda t} \quad \text{for } \lambda = 5.0.$$

The optimal results are shown in Table 1 and Fig. 3.

In our numerical calculations we need to choose the two numbers x_{\min} and x_{\max} as the smallest and largest solutions of the nonlinear equation $|G(x)| < \varepsilon$ where $\varepsilon = 10^{-4}$. We may then pose deconvolution (2.3) on the interval $[0, T]$, where $T = x_{\max} - x_{\min}$. Since the size of the essential support of $G(x)$ depends upon ‘ a ’, we have for a fixed number N of equidistant data points $\{x_n\}$, $h = T/N$ and $a > 1$. We found the minimum value of λ from (5.12) and compared the L_∞ error norm of the resulting solution with the values of the true solutions.

9. Concluding remarks

Our method worked very well over all the three test problems and results obtained are shown in Figs. 1–3 and Table 1.

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