# Adjoint Data Assimilation for Aerosol Dynamic Equations

Adrian Sandu*, Dacian Dăescu† and Gregory R. Carmichael‡

*Department of Computer Science, Michigan Technological University, Houghton, MI 49931.
†Applied Mathematical and Computational Sciences and The Center for Global and Regional Environmental Research, The University of Iowa, Iowa City, IA 52246.
‡Department of Chemical and Biochemical Engineering and The Center for Global and Regional Environmental Research, The University of Iowa, Iowa City, IA 52246.

**Abstract**

This paper presents an application of adjoint modeling to retrieve the initial distribution of the aerosol population from measurements at later times. A general framework is given for the discretization of particle dynamics equation by projection methods. The adjoint of the discrete model is constructed. Adjoint modeling succesfully retrieves the initial distribution, even if the measurements are restricted to only specific size bins.

## 1 Introduction

As our understanding expands, new processes are incorporated into air quality computer models. One example is the particulate matter (aerosol) processes, the importance of which is now widely recognized. Aerosols are now a priority focus area in environmental science due to the leading role they play as a cause of adverse human health, and their ability to scatter and absorb incoming solar radiation and thus modify warming due to greenhouse gases and reduce visibility. To accurately study the effects of aerosols it is necessary to resolve aerosol number and mass distributions as a function of chemical composition and size.

## 2 The continuous particle dynamics equation

In this paper the continuous particle size distributions are considered functions of particle volume ($v$) and time ($t$). For simplicity we consider single component particles, but the techniques can be generalized to multiple components.

The size distribution function (number density) of a family of particles will be denoted by $n(v,t)$; the number of particles per unit volume of air with the volume between $v$ and $v+dv$ is $n(v,t)dv$.

Similar formulations can be given in terms of volume, surface, or mass densities [15]. However, recovering mass from a volume formulation is difficult in practice, as the densities are only approximatively known and are a function of composition and size.

The aerosol population undergoes a series of physical and chemical transformations. *Growth* processes include condensation, evaporation, deposition and sublimation (of gases to/from the particle surface). The growth of each component's volume takes place at a rate that depends on the particle's dimension and composition, $dv/dt = I_v(v,t)$. *Coagulation* forms new particles of volume $v+w$ from the collision of two smaller particles of volumes $v$ and $w$; the collision rate $\beta_{v,w}n(v)n(w)$ is proportional to the number of available small particles. *Nucleation* of gases creates small particles. *Emissions* increase the number of particles of a specific composition and size, while *deposition* processes remove particles from the atmosphere. In addition, the constituents interact chemically inside each particle, changing the chemical composition (but not the number) of particles.

Under the above physical transformations the number density changes according to [4]

$$\begin{array}{rcl} \partial n(v,t)/\partial t & = & -\partial \left[ I_v(v)\, n(v,t) \right] / \partial v \\ & & +\frac{1}{2} \int_0^v \beta_{v-w,w}\, n(v-w,t) n(w,t) dw - n(v,t) \int_0^\infty \beta_{v,w}\, n(w,t) dw \\ & & +S(v,t) \ , \\ n(v,0) = n_0(v) \ , & & n(0,t) = 0 \ . \end{array} \tag{1}$$

The different terms in equation (1) describe, in order, the modification in the number of particles due to growth, creation of particles of volume $v$ by coagulation, loss of volume $v$ particles due to coagulation, increase in particle number due to nucleation, emissions and depositions (sources and sinks). Each of the terms will be explained in detail below. The equation is subject to a specified initial condition $n_0$, and the boundary condition of no zero volume particles.

For simplicity, we treat here only the case of single-component particles.

# 3 Previous work

Three major approaches are used to represent the size distribution of aerosols: continuous, discrete and parametrized. In this paper we focus on continuous models (i.e. continuous size distributions and the general dynamic equations in continuous form).

For computational purposes one needs to use finite-dimensional approximations of the continuous size distributions. In the *sectional approach* the size domain $v \in [0, \infty]$ is divided into size bins $v \in [V_i^{\text{low}}, V_i^{\text{high}})$. In each size bin $i$ there are $n_i$ particles per unit volume, all of them having the same mean volume $V_i$. variations of this approach include the *full-moving* structure, the *quasi-stationary approach*, as well as the *moving-center* structure [9].

The integro-differential coagulation equation is difficult to solve accurately, due to the quadratic terms under the integral. The algorithms proposed in the literature for the coagulation equation include semi-implicit solutions, finite element method, collocation, J-space transformations, analytical solutions [9, Section 16], [12] etc.

The growth equation in number densities has the form of an advection equation, with the "flow speed" equal to the time derivative of the volume [16, Section 12]. A nice survey of several popular numerical methods for the growth equations is given in Zhang et. al. [20]. Different solution of the growth equations were proposed in [2, 8, 10, 11].

Many models include different processes successively, using a time splitting scheme. This enables the use of numerical methods tuned to each subprocess but introduces hard-to-quantify splitting errors. Simultaneous solutions of all dynamic processes are given in [4, 5, 6, 7, 13, 15, 19].

# 4 Numerical solution of the dynamics equation

The approach taken here consists of a discretization in size using a projection method followed by a discretization in time. We will solve this equation by a semi-discretization in size, followed by a time integration of the resulting system of ordinary differential equations. The semi-discretization in size is done by projecting the solution on a finite-dimensional subspace $\text{span}\{\phi_1(v), \cdots, \phi_s(v)\}$; this generalizes the sectional approach. The dynamic equation is imposed to hold exactly in a certain subspace (projection) or at a certain set of nodes (collocation).

## 4.1 Discretization of the particle size distribution

The continuous number distribution is given a finite-dimensional approximation. Let $\{\phi_i\}_{1 \le i \le s}$ be a set of continuous basis functions; then

$$n(v,t) = \sum_{i=1}^{s} n_i(t)\, \phi_i(v) \ , \quad \phi_i(x) = \text{basis function} \ . \tag{2}$$

The set of time-dependent expansion coefficients

$$n(t) = [\, n_1(t), \cdots, n_s(t) \,]^T \ , \tag{3}$$

will be determined from the dynamics equation.

In logarithmic scaled coordinates the basis function arguments are changed accordingly, $\phi_i(\log[v/V_0])$.

The representation (2) places the problem in the general framework of *projection methods* [1]. For example $\phi_i$ can be piecewise polynomials or can be orthogonal polynomials. The result is a continuous distribution $n(v,t)$. Higher order approximations can be obtained by increasing the order of the basis functions without changing the number of bins $s$.

We note in passing that the full-stationary sectional approach can be formally cast into the form (2) by using Dirac* basis functions $\phi_i(v) = \delta(v - V_i)$. For this reason we extend the sectional interpretation and call span$\{\phi_i\}$ the *size bin* $i$, and refer to $n_i$ as the number of particles in *bin* $i$.

## 4.2 Coagulation

The theoretical coagulation equation for single-component particles is [9, Section 16]

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2}\int_0^v \beta_{v-w,w}\, n(v-w,t) n(w,t) dw - n(v,t)\int_0^\infty \beta_{v,w}\, n(w,t) dw \ . \tag{4}$$

To obtain a discrete form of the coagulation equation one inserts (2) into (4):

$$\begin{aligned} \sum_{i=1}^s n_i'(t)\phi_i(v) &= \frac{1}{2}\sum_{k=1}^s\sum_{m=1}^s n_k(t) n_m(t) \int_0^v \beta_{v-w,w}\phi_k(v-w)\phi_m(w)\, dw \\ &\quad - \sum_{k=1}^s\sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \beta_{v,w}\phi_k(v)\phi_m(w)\, dw \ . \end{aligned}$$

The resulting equation is multiplied by the test function $\xi_j(v)$ $(j = 1 \cdots s)$ and integrated from $v = 0$ to $\infty$ to obtain a system of $s$ ordinary differential equations

$$\begin{aligned} \sum_{i=1}^s n_i'(t)\int_0^\infty \phi_i(v)\xi_j(v) dv &= \frac{1}{2}\sum_{k=1}^s\sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \left(\int_0^v \beta_{v-w,w}\phi_k(v-w)\phi_m(w) dw\right) \xi_j(v) dv \\ &\quad - \sum_{k=1}^s\sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \left(\int_0^\infty \beta_{v,w}\phi_m(w) dw\right) \phi_k(v)\xi_j(v) dv \ . \end{aligned} \tag{5}$$

We build the following matrices of integral coefficients

$$\begin{aligned} A &= \left[\int_0^\infty \phi_j(v)\xi_i(v) dv\right]_{1\le i,j\le s} \\ C^j &= \left[(1/2)\int_0^\infty \left(\int_0^v \beta_{v-w,w}\phi_k(v-w)\phi_m(w) dw\right) \xi_j(v) dv \right. \\ &\quad \left. - \int_0^\infty \left(\int_0^\infty \beta_{v,w}\phi_m(w) dw\right) \phi_k(v)\xi_j(v) dv\right]_{1\le k,m\le s} \ , \qquad 1 \le j \le s \ . \end{aligned} \tag{6}$$

If $n(t)$ is the vector of number concentrations (3), the equation (5) becomes

$$A\, n'(t) = \begin{bmatrix} n^T(t)\, C^1\, n(t) \\ \vdots \\ n^T(t)\, C^s\, n(t) \end{bmatrix} \ . \tag{7}$$

To simplify notation we introduce the matrix $D$ which is a linear function of $n$,

$$D(n) = \begin{bmatrix} n^T\, C^1 \\ \vdots \\ n^T\, C^s \end{bmatrix} \ . \tag{8}$$

The semi-discrete coagulation equation reads

$$A\, n'(t) = D(\, n(t)\,)\, n(t) \ . \tag{9}$$

*Recall that $\delta(x) = 0$ for $x \neq 0$, $\delta(0) = \infty$, and $\int_{V_i-\epsilon}^{V_i+\epsilon} f(x)\delta(x - V_i)\, dx = f(V_i)$.

**The Galerkin approach.** With a *Galerkin* type scheme [1], $\{\phi_i(v)\}$, $\{\xi_i(v)\}$ are continuous basis functions. For pure Galerkin the test and basis functions coincide ($\xi_i = \phi_i$, $\forall i$), while for Petrov-Galerkin they are different. The equation (7) is relatively expensive to solve, since one has to evaluate a large number of double integrals.

**The collocation approach.** We can solve (5) with a *collocation* method. In this approach $\{\phi_i(v)\}$ are continuous basis functions, but the test functions are deltas, $\{\xi_i(v) = \delta(v - V_i^c)\}$, with $V_i^c$ the collocation points. The resulting equation is also of form (7), but the integral coefficients to be computed simplify to

$$\begin{aligned} A &= [\phi_j(V_i^c)]_{1\le i,j\le s} \\ C^j &= \left[(1/2)\int_0^v \beta_{V_j^c - w,w}\phi_k(V_j^c - w)\phi_m(w)dw - \phi_k(V_j^c)\int_0^\infty \beta_{V_j^c,w}\phi_m(w)dw\right]_{1\le k,m\le s} \ , \ 1 \le j \le s. \end{aligned} \tag{10}$$

This approach is promising since all coefficients involve only simple integrals.

## 4.3 Growth

Growth processes include condensation, evaporation, deposition and sublimation (of gases to/from the particle surface). Consider again the case of single component particles. The growth equation in number densities

$$\frac{\partial n(v,t)}{\partial t} = -\frac{\partial}{\partial v}\left[I(v)\, n(v,t)\right] \ , \quad I(v) = \frac{dv(t)}{dt} \ , \quad n(0,t) = 0 \ , \quad n(v,0) = n^0(v) \ , \tag{11}$$

has the form of an advection equation, with the "flow speed" provided by the time derivative of the volume. This equation is to be solved subject to an initial distribution $n_0(v)$ and the boundary condition of no zero-sized particles [16, Section 12],

A similar derivation process (as presented for coagulation) leads to the semi-discrete formulation of the growth equations

$$A\, n'(t) = G\, n(t) \ , \tag{12}$$

where

$$A = \left[\int_0^\infty \phi_j(v)\xi_i(v)dv\right]_{1\le i,j\le s} \ , \qquad G = \left[\int_0^\infty I(v)\phi_j(v)\xi_i'(v)dv\right]_{1\le i,j\le s} \ . \tag{13}$$

For $G$ we have used one integration by parts and homogeneous boundary conditions at $v = 0$ and $v = \infty$. For the Galerkin approach use $\xi_i = \phi_i$. For the collocation approach one obtains

$$A = [\phi_j(V_i^c)]_{1\le i,j\le s} \ , \qquad G = \left[-\left[I\phi_j\right]'(V_i^c)\right]_{1\le i,j\le s} \ . \tag{14}$$

## 4.4 Sources and sinks

Sources and sinks (i.e. emissions, nucleation and deposition processes) have a simple mathematical formulation,

$$\frac{\partial n(v,t)}{\partial t} = S(v,t) \ . \tag{15}$$

The simplicity comes from the fact that $S$ terms are not coupled across different volumes. A finite-dimensional approximation of $S$ can be given, $S(v,t) = \sum_{i=1}^s S_i(t)\phi_i(v)$. Discrete evolution equations can be written for each bin, $n_i'(t) = S_i(t)$, $1 \le i \le s$; in vector notation

$$n'(t) = S(t) \ . \tag{16}$$

## 4.5 Simultaneous discretization of the dynamic equations

Of particular interest is the coupled solution of coagulation, growth, nucleation, emissions and deposition. The coupled approach will, for example, better capture the competition between nucleation of new particles and condensation on existing particles for gas-to-particle conversion [20].

For single component particles combining (7), (12) and (16) gives the semi-discrete aerosol dynamics equation

$$A\,n'(t) = \underbrace{G\,n(t)}_{\text{growth}} + \underbrace{D(n(t))\,n(t)}_{\text{coagulation}} + \underbrace{A\,S(t)}_{\text{nucl.+em.+dep.}} \quad . \tag{17}$$

This is a system of $s$ coupled ordinary differential equations. The discrete initial conditions

$$n(0) = n_0 \tag{18}$$

are derived by projecting the continuous initial distribution $n_0(v)$ onto the finite-dimensional solution space,

$$n_0(v) = \sum_{i=1}^{s} n_i^0\,\phi_i(v) \;, \qquad n_0 = \left[\,n_1^0, \cdots, n_s^0\,\right]^T \;.$$

In the present calculations we use linear (hat) functions and the Galerkin formulation.

### 4.6 Time integration

The system (17)-(18) can be solved by any appropriate time-stepping method. The system has a particular form: the growth term is linear, while the coagulation term is bilinear. For this reason we employ a linearized backward Euler time discretization:

$$\left(A - h\,G - h\,D\left(n(t_k)\right)\right)\, n(t_{k+1}) = A\,n(t_k) \;, \tag{19}$$

where $h = t_{k+1} - t_k$ is the discretization time step.

## 5 4D variational data assimilation

We consider the 4D variational data assimilation problem associated with the dynamical model (17) in the time interval $[0, T]$ and the initial state $n_0 = n(0)$ (18) as the set of control parameters. The discrepancy between the solution of (17)-(18) and observations $n^o(t)$ over the interval $[0, T]$ is measured by the cost function

$$J = \frac{1}{2}\int_0^T (Hn - n^o)^T R^{-1}(Hn - n^o)dt \tag{20}$$

where we assume that mapping from the state space into the observations space is done by the state independent linear operator $H$. In the stochastic interpretation, $R$ is the covariance matrix of the errors in measurements and model representativeness. In the deterministic approach $R$ is diagonal positive definite and is used to provide appropriate weights for the curve-fitting process. Information on the initial state resulting from a previous analysis may be considered by including a "background" term

$$J_b = \frac{1}{2}(n_0 - n^b)^T B^{-1}(n_0 - n^b) \tag{21}$$

into the functional $J$, where $B$ is the covariance matrix of the errors in the background estimate. For simplicity we neglect here the background term, and formulate the variational data assimilation problem as the problem of minimizing the cost functional (20). We express the solution of (17)-(18) as a function of the initial state, $n(t) = n(n_0, t)$, such that the data assimilation problem is

$$\min_{n_0} J(n_0) \tag{22}$$

The first order necessary conditions require that at a minimum point the gradient of $J$ must be zero: $\nabla J(n_0) = 0$. The adjoint model associated with (17) is

$$A^T \frac{d\lambda}{dt} = -\left[\frac{\partial}{\partial n}\left(Gn + D(n)n\right)\right]^T \lambda + H^T R^{-1}(Hn - n^o) \tag{23}$$

If we consider

$$\lambda(T) = 0 \tag{24}$$

we obtain the gradient of $J$ as

$$\nabla J(n_0) = -A^T \lambda(0) \tag{25}$$

The gradient of the cost function is computed by solving the adjoint (backward) problem (23)-(24) to obtain $\lambda(0)$, then using (25). For practical applications a discrete form of (20) must be considered. If we assume a sequence of observations taken at discrete moments in time $t_1, t_2, \ldots t_m$, we define the cost function as

$$J = \frac{1}{2} \sum_{l=1}^{m} (H_l n_l - n_l^o)^T R_l^{-1} (H_l n_l - n_l^o) \tag{26}$$

which gradient $\nabla J$ is

$$\nabla_{n_0} J(n_0) = \sum_{l=1}^{m} \left( \frac{\partial n_l}{\partial n_0} \right)^T H_l^T R_l^{-1} (H_l n_l - n_l^o) \tag{27}$$

## 5.1 Implementation of the adjoint model

By recursively applying the chain rule in (27), the gradient may be computed by initializing $\nabla J = 0$, then using the backward loop:

*for l=m, 1, -1 do*

$$\nabla J = \left( \frac{\partial \mathbf{n}_l}{\partial \mathbf{n}_{l-1}} \right)^T [\mathbf{H}_l^T \mathbf{R}_l^{-1} (\mathbf{H}_l \mathbf{n}_l - \mathbf{n}_l^o) + \nabla J] \tag{28}$$

Since the observations are usually sparse in time, a sequence of intermediate steps must be taken to obtain $n_l$ from $n_{l-1}$. For consistency between the computed cost function and its gradient, we implement the adjoint model corresponding to the numerical scheme (19). Taking the derivative with respect to $n_k$ in (19) we obtain

$$-h \frac{\partial}{\partial n_k} [D(n_k) \underline{n}_{k+1}] + [A - hG - HD(n_k)] \frac{\partial n_{k+1}}{\partial n_k} = A \tag{29}$$

where the notation $\underline{n}_{k+1}$ is used to indicate that this term must be treated as a constant during the derivation. After taking the transpose in (29) and arranging the terms, it follows

$$\left( \frac{\partial n_{k+1}}{\partial n_k} \right)^T [A - hG - HD(n_k)]^T = \left[ A + h \frac{\partial}{\partial n_k} [D(n_k) \underline{n}_{k+1}] \right]^T \tag{30}$$

Consider now an arbitrary seed vector $u$. From (30) it results

$$\left( \frac{\partial n_{k+1}}{\partial n_k} \right)^T u = \left[ A + h \frac{\partial}{\partial n_k} [D(n_k) \underline{n}_{k+1}] \right]^T \left( [A - hG - hD(n_k)]^T \right)^{-1} u \tag{31}$$

To implement the backward loop (28) we compute the expression (31) in two steps. First, we solve for $x$

$$[A - hG - hD(n_k)]^T x = u \tag{32}$$

then we evaluate

$$\left( \frac{\partial n_{k+1}}{\partial n_k} \right)^T u = \left[ A + h \frac{\partial}{\partial n_k} [D(n_k) \underline{n}_{k+1}] \right]^T x \tag{33}$$

using (8). From (8) we observe that D depends linearly on $n_k$. The right hand side term in (33) depends then on $n_{k+1}$, but it is independent of $n_k$. Formulas (28), (32), (33) complete the implementation of the adjoint model.

# 6 Numerical experiments

**Test problem** For the numerical experiments we consider the test problem from [5], which admits an analytical solution.

Let $N_0$ be the total initial number of particles and $V_0$ the mean initial volume. The initial number distribution is exponential, the coagulation rate is constant, and the growth rate is linear with the volume:

$$n_0(v) = (N_0/V_0)\, e^{-v/V_0} \ , \quad \beta(v,w) = \beta_0 \ , \quad I(v) = \sigma\, v \ .$$

This test problem admits an analytical solution, which is given in [5].

We solve the dynamics equation in the volume interval $[V_{\min} = 0, V_{\max} = 10]$, for $N_0 = 1$, $V_0 = 1$, $\beta_0 = 1$, and for $\sigma = 0.1$.

The discretization (19) is based on the Galerkin formulation (6) and (13). We take $s = 20$ equidistant points $[V_1 = V_{\min}, \cdots, V_s = V_{\max}]$ and consider the space of piecewise-linear interpolants at these points (i.e. use piecewise-linear hat basis functions $\phi_i$).

**Data assimilation setting** The data assimilation is performed in the twin experiments framework using model generated data such that at the minimum point the value of the cost function must be zero. The functional to minimize is (26), and the matrix $R_l$ is the identity matrix. The observational operator H is a diagonal matrix with an entry one on the diagonal position $k$ if there is an observation of the component $k$ of $n$, and zero otherwise. The adjoint model is implemented according to (28), (32), (33) and for the minimization process we used the limited memory BFGS method. An initial guess state is obtained by introducing random errors selected from a normal distribution into the reference initial state. In the experiments we performed a fixed assimilation window with the length six hours is considered.

**Experiments and results** In the first experiment "observations" are provided for the particle size distribution in all bins, at an interval of one hour. The correctness of the adjoint model was checked against a first order finite differences method which was implemented using a perturbation $\epsilon = 10^{-6}$ for each component of the initial state. In Figure 1 we show the absolute and relative differences between the computed gradients at the initial guess point and it can be seen that the values of the computed gradients agree in the range $O(\epsilon)$. The advantage of the adjoint method is that it provides an exact value of the gradient and offers a considerable saving in CPU time. The adjoint model requires significant more storage resources to be allocated, since the forward trajectory must be stored at each time step. We obtained a ratio between the CPU time of the pure adjoint integration to compute the gradient and the CPU time to evaluate the cost function (model integration) $cpu(\nabla J)/cpu(J) \approx 1.9$ which shows the efficiency of the adjoint implementation. The evolution of the cost function and the gradient norm during the minimization process are shown in Figure 2, and the results at the end of the assimilation process are displayed in Figure 3 for the particle size distribution in several bins. The reference initial particle size distribution is recovered using the assimilation procedure.

The second experiment we perform is more challenging. We investigate the ability of the data assimilation to recover the initial particle size distribution when observations in only few bins are provided. We select observations only in the bins 1-10, taken at a time interval of one hour. In Figure 4 we display the results in for several unobserved bins after the assimilation takes place. No attempt is done here to provide an analysis as which observations should be used or may provide more information to the data assimilation. We just note the ability of the data assimilation to retrieve the evolution of the particle size distribution of unobserved bins using information from the observed bins. A full adjoint sensitivity analysis and experiments on comprehensive models should be performed before any conclusions can be drawn.

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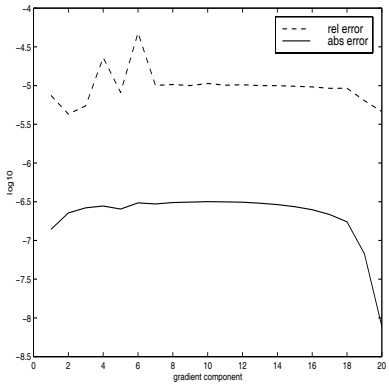


Figure 1: Absolute and relative differences between the computed gradients at the initial guess point agree within $O(\epsilon)$.

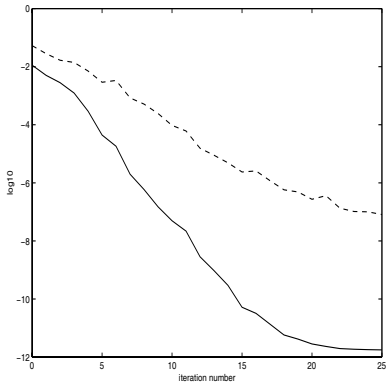


Figure 2: Evolution of the cost function and the gradient norm during the minimization process.

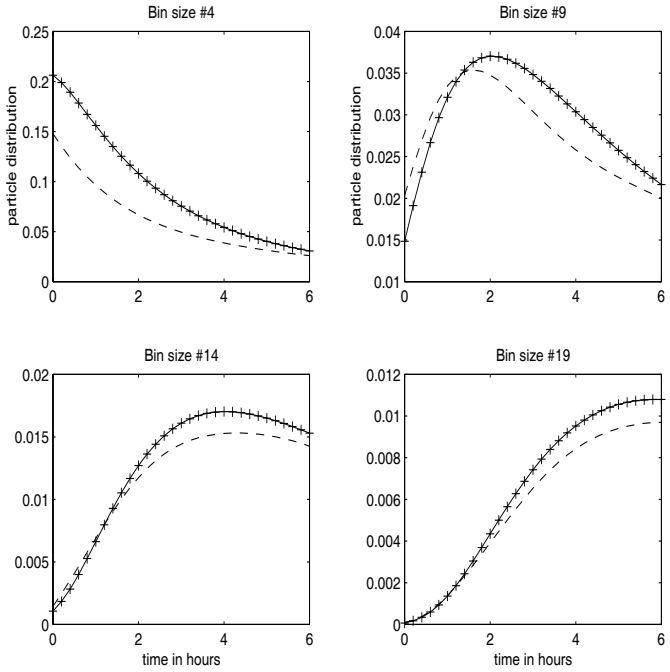


Figure 3: Results at the end of the assimilation process for several bins.

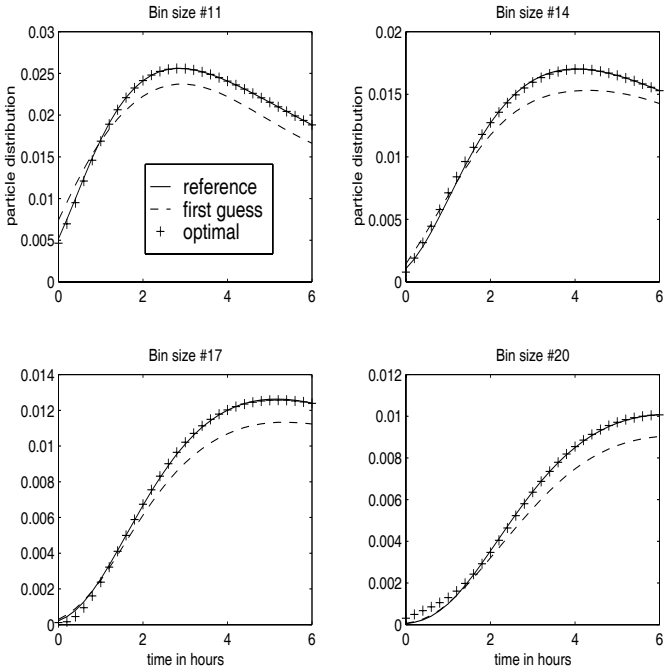


Figure 4: Results for several unobserved bins. Data assimilation is able to retrieve the evolution of the particle size distribution of unobserved bins using information from the observed bins.

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